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**Extensive comprehension of aluminum particles
combustion by advanced optical diagnostics
and numerical simulations**

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Compréhension approfondie de la combustion de particules d'aluminium par diagnostics optiques avancés et simulation numérique

Dans le cadre du développement de nouveaux moteurs à propulsion solide, des particules d'aluminium sont intégrées au propergol afin d'agir comme additif énergétique. Ce procédé améliore significativement les performances globales du moteur, mais il engendre également un écoulement diphasique à l'intérieur de la chambre de combustion. Cet écoulement peut être source de pertes de rendement et d'instabilités, parfois à l'origine de phénomènes vibratoires destructifs. Pour limiter ces inconvénients, il est essentiel de mieux comprendre les mécanismes mis en jeu lors de la combustion de l'aluminium, ainsi que les caractéristiques des produits de combustion. Dans cette optique, l'étude de la combustion d'une particule unique d'aluminium a été entreprise à l'aide d'un lévitateur électrodynamique. Ce dispositif permet d'isoler, de stabiliser et d'allumer une seule particule dans un environnement parfaitement contrôlé. Afin de caractériser le processus, plusieurs diagnostics optiques ont été développés. Une méthode d'extinction de la lumière, basée sur un rétro-éclairage par LED blanche, offre une mesure multispectrale de l'extinction optique du nuage d'alumine formé autour de la particule. En parallèle, la mise en place d'une méthode numérique associée à une calibration précise de la caméra permet de distinguer les émissions gazeuses de celles issues des phases condensées. Le couplage de ces deux approches rend possible la mesure de grandeurs locales au voisinage immédiat de la particule en combustion, telles que la température, la concentration en espèces condensées et la taille des particules d'alumine produites. Cette méthodologie fournit ainsi des informations inédites et cruciales sur le déroulement de la combustion d'aluminium. L'étude a été conduite dans des environnements variés afin d'identifier les facteurs influençant les mécanismes de combustion. Des diluants tels que l'azote, l'hélium ou l'argon ont été utilisés, avec des proportions d'oxygène ajustées, des pressions croissantes, et du dioxyde de carbone comme oxydant. Ces variations expérimentales permettent de mettre en évidence des paramètres clés qui pilotent la combustion et conditionnent les caractéristiques des produits formés. Les données obtenues offrent des cibles précieuses pour l'amélioration des modèles numériques. Le modèle développé par ArianeGroup a ainsi été confronté aux résultats expérimentaux. Des écarts notables ont été relevés, principalement attribués à une représentation trop simpliste du phénomène de condensation des produits de combustion. Pour remédier à cette limite, un modèle plus élaboré a été implémenté, reposant sur la théorie de la nucléation. Dans ce nouveau cadre, la condensation n'est plus décrite par une simple loi d'Arrhenius, mais par une approche combinant un taux de nucléation et un taux d'accroissement, calculés à partir des lois de la condensation homogène. Si des divergences subsistent entre expériences et simulations, les résultats obtenus confirment l'importance d'une modélisation fine des processus de formation de l'alumine. Ce travail souligne ainsi à la fois la complexité intrinsèque de la combustion de l'aluminium et l'apport essentiel d'un couplage étroit entre expériences de pointe et modélisations avancées pour progresser vers une compréhension et une maîtrise complètes de ce phénomène.

Mots clés : Combustion, Diagnostics optiques avancés, Particules d'aluminium

Extensive comprehension of aluminum particles combustion by advanced optical diagnostics and numerical simulations

In the development of new solid propulsion motors, aluminum particles are incorporated into the propellant to act as an energetic additive. This process significantly enhances the overall performance of the motor, but it also generates a two-phase flow inside the combustion chamber. Such a flow can lead to performance losses and instabilities, sometimes giving rise to destructive vibratory phenomena. To mitigate these drawbacks, it is essential to gain a better understanding of the mechanisms involved in aluminum combustion, as well as the characteristics of the resulting combustion products. With this objective, the combustion of a single aluminum particle was investigated using an electrodynamic levitator. This device makes it possible to isolate, stabilize, and ignite an individual particle in a perfectly controlled environment. To characterize the process, several optical diagnostics were developed. A light extinction method, based on white LED backlighting, provides a multispectral measurement of the optical extinction of the alumina cloud formed around the burning particle. In parallel, the implementation of a numerical method combined with precise camera calibration makes it possible to distinguish gaseous emissions from emissions arising from condensed phases. The coupling of these two approaches enables the measurement of local quantities in the immediate vicinity of the burning particle, such as temperature, condensed species concentration, and the size of the alumina particles produced. This methodology therefore provides novel and crucial insights into the combustion process of aluminum. The study was conducted under various environments to identify the factors influencing combustion mechanisms. Diluents such as nitrogen, helium, or argon were used, with adjusted oxygen proportions, increasing pressures, and, in some cases, carbon dioxide as an oxidizer. These experimental variations highlight key parameters that drive combustion and govern the characteristics of the products formed. The data obtained provide valuable targets for improving numerical models. The model developed at ArianeGroup was therefore compared with the experimental results. Significant discrepancies were observed, mainly attributed to an overly simplistic representation of the condensation phenomenon of combustion products. To address this limitation, a more advanced model based on nucleation theory was implemented. In this new framework, condensation is no longer described by a simple Arrhenius law, but by an approach combining a nucleation rate and a growth rate, both calculated from homogeneous condensation theory. Although discrepancies between experiments and simulations remain, the results confirm the importance of accurately modeling the processes involved in alumina formation. This work thus emphasizes both the intrinsic complexity of aluminum combustion and the essential contribution of a close coupling between advanced experiments and modeling in order to progress toward a complete understanding and control of this phenomenon.

Keywords : Combustion, Advanced optical diagnostics, Aluminum particles



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Contents

<i>List of Figures</i>	vii
<i>List of Tables</i>	xvii
1 Introduction & Overview	1
2 Applications of aluminum combustion	3
2.1 Aluminum in space propulsion	4
2.1.1 Introduction to rocket motors	4
2.1.2 Solid-propellant rocket motors (SRM)	6
2.1.3 The impact of aluminum	8
2.2 Aluminum as a clean energy carrier	12
2.3 Summary	14
3 Single aluminum particle combustion	17
3.1 Ignition phase	17
3.2 Combustion phase	20
3.2.1 Phenomenology - A simplified approach	20
3.2.2 Phenomenology - A more detailed approach	23
3.2.3 Temperatures	29
3.2.4 Temporal aspects	32
3.3 Extinction phase	35
3.4 Influence of the ambiance	37
3.4.1 Pressure	37
3.4.2 Diluent	40
3.4.3 Oxidizer	42
3.4.4 Electric field	43
3.5 Conclusion	46
4 Experimental Setup	49
4.1 Single particle experiments in the literature	49
4.2 General overview	50
4.3 The electrodynamic levitator	51
4.4 Optical diagnostics	54
4.4.1 Photomultipliers	54
4.4.2 Camera	55
4.4.3 Backlight	56

4.5	Ignition device	58
4.6	Summary	60
5	Development and performance assessment of an original characterization method	61
5.1	Optical diagnostics & Al combustion - state of the art	61
5.1.1	Alumina characterization	61
5.1.2	Temperature measurements	62
5.2	Hypotheses	66
5.3	Multispectral Light Extinction Method (M-LEM)	67
5.3.1	LEM: general principle	67
5.3.2	Optical depth measurement	69
5.3.3	Abel transform: Onion-peeling	73
5.3.4	Extinction cross section	75
5.3.5	Multispectral LEM	80
5.3.6	Summary	82
5.4	Modulation Absorption Emission (MAE)	83
5.4.1	Local black body emissions	83
5.4.2	Temperature evaluation	86
5.4.3	Summary	92
5.5	Limitations	93
6	Experimental Results	95
6.1	Experimental cases	95
6.2	Profiles - Atmospheric air	96
6.2.1	Alumina volume fraction	96
6.2.2	Alumina size	99
6.2.3	Temperature	100
6.2.4	Gaseous emissions	101
6.2.5	Cloud structure	103
6.3	Key combustion parameters (KCPs)	105
6.4	Influence of the medium on the KCP	106
6.4.1	Temperature	107
6.4.2	Alumina concentration	116
6.4.3	Average alumina particle size	120
6.4.4	Gaseous emissions	124
6.4.5	Bremsstrahlung emissions	125
6.5	Summary	128
7	Simulations	131
7.1	State of art	131
7.2	Description of the numerical model	135
7.2.1	System	135
7.2.2	Physical model	135
7.2.3	Alumina formation	136

7.3	Parametric study	136
7.3.1	Alumina size and condensation reversibility	136
7.3.2	Diffusivity of the oxide smoke	138
7.3.3	Pre-exponential factor of the condensation reaction	139
7.3.4	Conclusion	140
7.4	Implementation of the CNT	141
7.4.1	Nucleation rate	142
7.4.2	Nucleation rate & growth rates	142
7.5	Conclusions and perspectives	146
8	Conclusions & Perspectives	149
A	Electric field studies	163
A.1	Derivation of the electric field	163
A.2	Influence of the electric field on the combustion process	165
A.2.1	Induced magnetic field	165
A.2.2	Influence of $\vec{E}$ on the observed combustion	169
B	Spectral response graphs	171
B.1	Dichroic filters	171
B.2	Bandpass filters	173
B.3	Photomultipliers	173
B.4	Solis LED	174
C	Camera calibration	175
C.1	Spectral overlap calibration	175
C.2	Blackbody calibration	177
D	Experimental profiles	183
D.1	Temperature profiles	183
D.2	Alumina volume fraction profiles	186
D.3	Alumina particle size	189
D.4	Gaseous emissions profiles - R	192
D.5	Gaseous emissions profiles - G	195
D.6	Gaseous emissions profiles - B	198

List of Figures

1.1	Ariane 6 inaugural launch	1
1.2	Historical increase of the cataloged spacecrafts in Earth's orbit in February 2023, adapted from [1].	2
2.1	Energy density and specific energy for various gaseous fuels ($\bigcirc$), liquid fuels (∇) and solid fuels ($\square$), from [3].	3
2.2	Schematic of a propulsive system, from [5].	4
2.3	Schematic of a LRE (left) and a SRM (right), from [6]	7
2.4	Schematic of the combustion process of an aluminized HTPB/AP solid fuel from [8].	7
2.5	Image of a burning aluminum agglomerate, detached from the propellant surface, from [9].	8
2.6	Gas phase velocity profiles at the nozzle exit for pure gas flow and two-phase flow with various alumina particle sizes, from [12].	9
2.7	Schematized representations of thermo-acoustic instabilities, from [21].	11
2.8	Simulations of pressure oscillations in monophasic and two-phase flow, from [22].	12
2.9	Annual CO_2 emissions from fossil fuels and industry, from [23].	13
2.10	Aluminum-enabled carbon-free energy cycle.	14
3.1	Schematized ignition process of an aluminum particle at high and low heating rates. Pressure forces are represented by red arrows.	18
3.2	Ignition temperature as a function of aluminum particle size, from [31].	19
3.3	Combustion of an aluminum particle in a kinetically-limited regime. Blue circles: oxygen, blue arrows: oxygen flux, red arrows: heat release.	21
3.4	Combustion of an aluminum particle in a diffusion-limited regime, under the assumption of infinitely fast kinetics. The red circle segments delimit the reaction zone. Blue circles: oxygen, blue arrows: oxygen flux, gray arrows: aluminum Stefan flow, gray circles: aluminum vapor, red arrows: heat release.	22
3.5	Structure of a burning aluminum droplet, according to Ref. [48].	24
3.6	Structure of alumina aggregates collected from solid propellant combustion, from [55].	26
3.7	Driving mechanisms of alumina droplet transport.	27
3.8	Importance of heterogeneous surface reactions, as a function of pressure and droplet size, from [49].	28

3.9	Commonly accepted growth mechanism of the oxide cap during aluminum droplet combustion.	29
3.10	Aluminum droplet surface temperature as a function of aluminum vapor pressure on the surface, using Eq. 3.9.	30
3.11	Temperature profile around a burning droplet of radius $r_0 = 110\mu m$ in a O_2/Ar (21/79) % vol. mixture, from Ref [64].	31
3.12	Temperature profile simulated by Beckstead to match Bucher's data, from Ref. [47].	32
3.13	Combustion time for aluminum particles burning in air as a function of initial particle size. Comparisons with correlations from the literature are added, from Ref. [70].	33
3.14	Aluminum droplet burning rate constants in air as a function of the initial droplet diameter, from Ref. [8].	34
3.15	Qualitative size evolution of the aluminum droplet with its associated oxide cap during combustion.	36
3.16	Occurrence of a jetting/spinning phenomenon, from [75].	36
3.17	Sequence of aluminum droplet combustion in H_2O/N_2 (50/50) % vol. mixture, illustrating the coverage of the droplet by a solid phase, from [61].	37
3.18	Theoretical droplet temperature (black) and adiabatic flame temperature (red) as a function of pressure in air.	38
3.19	Aluminum droplet burning rate constants as a function of air pressure, from [77].	38
3.20	Aluminum droplet combustion time as a function of air pressure, from [78].	39
3.21	Aluminum droplet combustion sequences with increasing air pressure, from [78].	40
3.22	Aluminum droplet burning rate constants as a function of diluent nature and concentration, from [75].	42
3.23	Aluminum droplet combustion images in pure water vapor, extracted from [81] (left) and [80] (right).	43
3.24	Characteristic charge loss times as a function of temperature for different material work functions, from [82]	44
3.25	Schematics of the influence of an electric field on the charged species produced in the reaction zone of a flame.	45
4.1	Photography of the experimental setup (Top) and simplified drawing of the experimental setup (Bottom). Red lines illustrate the laser path. Blue arrows represent LED emissions. Orange arrows represent combustion emissions.	52
4.2	Schematized representation of the electrodynamic levitator.	53
4.3	Time variation of the electric potential induced by the circular electrode within the levitator. The location of the particle is represented by the grey circle. The preferred movement of the particle due to the potential is represented with red arrows.	54
4.4	Schematics of the photomultipliers (PM) system. Information of locally available wavelengths is displayed in red.	55
4.5	Example of signals from photomultipliers during aluminum combustion. Images of the particle at various times are also added.	56

4.6	Schematics of the camera-teleobjective system.	57
4.7	Spectral response of RGB matrices of the high-speed color PHANTOM T-2410 camera. The relative spectral response of the triple band filter (TBF) has been added (black).	57
4.8	Schematics of the LED system.	58
4.9	Schematics of the ignition system. The red dashed line is the laser path.	59
5.1	SEM image of a quenched particle (left) and Abel recovered CCPs relative concentration profiles (right) in the case of atmospheric air, from Ref.[52].	62
5.2	Spectral radiance of a black body, as a function of wavelength at 3000 K (red), 4000 K (green), and 5000 K (blue).	63
5.3	Representation of a beam of initial intensity I_0 passing through a medium of length l discretized in N slabs, composed of s attenuating species. The beam's final intensity is I	68
5.4	Representation of a beam of initial intensity I_0 passing through the oxide smoke of a burning Al droplet, along a path of length l , with final intensity $I + I_c$ due to combustion emissions.	70
5.5	Specific images and the conditions in which they are acquired. From top to bottom: Dark noise image; Combustion image; LED image; Backlit combustion image.	70
5.6	Sequence of an aluminum particle burning in atmospheric air. The resolution is $1.22 \mu m/px$	72
5.7	Optical depth profile surrounding an Al particle burning in atmospheric air, computed on the R,G, and B matrices of the camera.	73
5.8	Schematics of the discretized oxide smoke (left). Top view of the plane P_i from the left figure (right).	73
5.9	Image of a burning Al droplet and the line selected to apply the onion-peeling procedure.	75
5.10	Local optical depth profile surrounding an Al particle burning in atmospheric air, at the three wavelengths of interest, obtained from the deconvolution of the integrated optical depth.	76
5.11	Absorbed and scattered light due to incident light impacting a particle.	76
5.12	Extinction efficiency of an alumina particle as a function of its size, evaluated with the classical Mie theory, and with the Rayleigh approximation, for two different wavelengths (left). Relative error between extinction efficiencies computed with Mie and Rayleigh methods for two different wavelengths (right).	80
5.13	Volume fraction computed under the hypothesis of a medium of unit length, constant composition and optical depth of 0.1, evaluated with the classical Mie theory, and with the Rayleigh approximation, for two different wavelengths (left). Relative error between volume fractions computed with Mie and Rayleigh methods for two different wavelengths (right).	80
5.14	Alumina particle radius profile surrounding the burning Al droplet.	81
5.15	Alumina volume fraction profile computed under the assumption of constant alumina particle size $r_p = 75nm$ (left). Alumina volume fraction profile computed with the optimized alumina particle size (right).	82

5.16	Summary of the M-LEM.	83
5.17	Schematics of combustion emission and self absorption.	84
5.18	Normalized profiles of local black body spectral radiance multiplied by the optical coefficient for the three wavelengths of interest.	86
5.19	Profiles of the ratios of local black body emission (radiance multiplied by optical coefficient).	87
5.20	Temperature profiles from the corrected local emission ratios $\left(\frac{\alpha_R}{\beta_G}, \frac{\alpha_R}{\gamma_B}, \frac{\beta_G}{\gamma_B}\right)$	90
5.21	Normalized measured local emission profiles compared to normalized reconstructed local emission profiles from Eq. (5.66).	91
5.22	Gaseous emission profiles normalized by the respective maximum local emissions, evaluated by Eq. (5.70).	91
5.23	Summary of the MAE method.	92
6.1	Images of Al combustion in pressurized air. Camera settings are kept identical.	97
6.2	Images of Al combustion in O_2/Ar mixtures, at various O_2 concentrations and pressures. Camera settings are kept identical.	97
6.3	Images of Al combustion in an O_2/He (21/79) % vol. mixture, at various pressures. Camera settings are kept identical.	98
6.4	Image of Al combustion in a CO_2/N_2 (60/40) % vol. mixture, at 2 bar.	98
6.5	Alumina volume fraction profile and the associated 95 % confidence interval obtained in atmospheric air.	99
6.6	Alumina particle size profile and the associated 95 % confidence interval obtained in atmospheric air.	99
6.7	Temperature profile and the associated 95 % confidence interval obtained in atmospheric air.	100
6.8	Gaseous emission profiles measured at λ_R, λ_G and λ_B and the associated 95 % confidence intervals obtained in atmospheric air.	102
6.9	Gaseous emission spectrum compared to a Bremsstrahlung spectrum computed at 1.81 eV.	103
6.10	Identified cloud structure. All values are normalized with respect to their own maximum. Gaseous emissions are represented at λ_B	104
6.11	KCPs related to temperature.	105
6.12	KCPs related to alumina concentration.	106
6.13	KCPs related to gaseous emissions.	107
6.14	Maximum temperature measured for all experimental cases. Error bars represent the 95 % confidence interval. A negligible horizontal offset has been added to pressure values to facilitate the reading of the graph. The compositions are in % (vol.). Literature data has been added for comparison with similar colors to experimental data.	108
6.15	Theoretical adiabatic flame temperature (dashed lines) and maximum experimentally measured temperature (points and solid lines).	109
6.16	Theoretical conductive heat losses divided by the heat capacity of the mixture as a function of the difference between the adiabatic flame temperature and the measured flame temperature.	111

6.17	Aluminum droplet surface temperature measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).	112
6.18	Gaseous aluminum partial pressure as a function of pressure. The average value of all mole fractions is also displayed (black dashed line).	112
6.19	Radial position of the experimental maximum temperature. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).	113
6.20	SOR computed according to the classical law for hydrocarbon droplets from [116], as a function of the measured SOR.	115
6.21	SOR computed according to the law developed by Liu et al. [115] accounting for CCPs, as a function of the measures SOR.	115
6.22	Alumina peak volume fraction measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).	116
6.23	Determined correlation (dashed lines) compared to experimental results (points and solid lines).	118
6.24	Location of the alumina peak volume fraction measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).	119
6.25	Correlation between Stand-Off Ratio (SOR) and location of alumina peak volume fraction r_{fv}	119
6.26	Average alumina particle size measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).	120
6.27	Final alumina particle size computed using the CNT.	122
6.28	Computed alumina particle radii vs. measured alumina particle radii.	123
6.29	Optimized growth time vs. temperature for each experimental case.	124
6.30	Location of the maximum AIO emissions (in a unit of droplet radius) measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).	124
6.31	Experimental SOR as a function of the experimental r_{AIO}	125
6.32	r_{AIO} computed using the correlation from Eq. 7.1 vs the experimental r_{AIO}	126
6.33	Location of the maximum Bremsstrahlung emissions (in a unit of droplet radius) measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).	126
6.34	Experimental SOR as a function of the location of the maximum Bremsstrahlung emissions.	127
6.35	Electronic temperature measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The average electronic temperature is also displayed (black dotted line). The compositions are in % (vol.).	127

7.1	Temperature profiles from experiments (CO_2/N_2 (60/40) (vol.) 2 bar) and simulations from King (CO_2/N_2 (50/50) (vol.) 1 bar)	132
7.2	Normalized blue gaseous emission profile from experiments (CO_2/N_2 (60/40) (vol.) 2 bar) and simulated normalized AlO concentration profile from King (CO_2/N_2 (50/50) (vol.) 1 bar).	132
7.3	Temperature profiles from experiments and simulations from Muller and Beckstead in an O_2/Ar (21/79) (vol.) mixture at 1 bar.	134
7.4	Alumina mass fraction profiles from experiments and simulations from Muller and Beckstead in an O_2/Ar (21/79) (vol.) mixture at 1 bar.	134
7.5	Normalized blue gaseous emission profile from experiments and simulated normalized AlO concentration profile from Muller and Beckstead in an O_2/Ar (21/79) (vol.) mixture at 1 bar.	135
7.6	Experimental and simulated temperature profiles for atmospheric air.	137
7.7	Experimental and simulated alumina volume fraction profiles for atmospheric air.	138
7.8	Experimental and simulated temperature profiles for atmospheric air.	139
7.9	Experimental and simulated alumina volume fraction profiles for atmospheric air.	139
7.10	Experimental and simulated temperature profiles for atmospheric air.	140
7.11	Experimental and simulated alumina volume fraction profiles for atmospheric air.	141
7.12	Temperature profiles obtained by simulations using an Arrhenius-based condensation law, a nucleation rate-based condensation law, and from experiments.	143
7.13	Alumina volume fraction profiles obtained by simulations using an Arrhenius-based condensation law, a nucleation rate-based condensation law, and from experiments.	143
7.14	Temperature profiles obtained using a growth + nucleation based condensation rate in atmospheric air, for various nucleus sizes, and compared to experimental data.	144
7.15	Alumina volume fraction profiles obtained using a growth + nucleation based condensation rate, for various nucleus sizes, and compared to experimental data.	145
7.16	Close up version of Fig. 7.15.	145
A.1	Combustion images with the electric field (top) and without the electric field (bottom).	169
B.1	Spectral transmission and reflectance efficiency of the DMLP650R dichroic filter	171
B.2	Spectral transmission and reflectance efficiency of the DMLP735R dichroic filter	172
B.3	Spectral transmission and reflectance efficiency of the DMLP805R dichroic filter	172

B.4	Spectral transmission efficiency of the FBH700-10, FBH760-10 and FBH820-10 bandpass filters	173
B.5	Spectral radiant sensitivity of the PM1002.	173
B.6	Spectral emission profile of the SOLIS 3-C white LED	174
C.1	Spectral response of the R, G and B matrices of the color camera. The efficiency of the Triple Band Filter (TBF) is also displayed.	175
C.2	Schematics of the experiments performed to determine the spectral overlap coefficients.	176
C.3	Schematic of the 'low temperature' blackbody calibration setup.	177
C.4	Schematic of the 'high temperature' blackbody calibration setup.	178
C.5	R/G ratio for various pixel gray value P_g and blackbody temperature. . . .	178
C.6	R/B ratio for various pixel gray value P_g and blackbody temperature. . . .	179
C.7	G/B ratio for various pixel gray value P_g and blackbody temperature. . . .	179
C.8	Experimental R/G values (dots) and R/G values obtained using equation C.6 (solid lines).	180
C.9	Relative error between calibration data and the calibration law from equation C.6.	180
C.10	Experimental R/B (dots) and corrected R/B using equation C.7 (triangles). .	181
C.11	Experimental G/B (dots) and corrected G/B using equation C.8 (triangles). .	182
D.1	Temperature profiles surrounding an Al droplet burning in air at various pressures	183
D.2	Temperature profiles surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures	184
D.3	Temperature profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures	184
D.4	Temperature profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar	185
D.5	Temperature profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures	185
D.6	Temperature profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars	186
D.7	Alumina volume fraction profiles surrounding an Al droplet burning in air at various pressures	186
D.8	Alumina volume fraction profiles surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures	187
D.9	Alumina volume fraction profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures	187
D.10	Alumina volume fraction profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar	188
D.11	Alumina volume fraction profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures	188
D.12	Alumina volume fraction profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars	189

D.13 Alumina particle size profiles surrounding an Al droplet burning in air at various pressures	189
D.14 Alumina particle size profiles surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures	190
D.15 Alumina particle size profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures	190
D.16 Alumina particle size profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar	191
D.17 Alumina particle size profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures	191
D.18 Alumina particle size profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars	192
D.19 Gaseous emissions profiles measured on the R matrix of the camera surrounding an Al droplet burning in air at various pressures	192
D.20 Gaseous emissions profiles measured on the R matrix of the camera surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .	193
D.21 Gaseous emissions profiles measured on the R matrix of the camera profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures	193
D.22 Gaseous emissions profiles measured on the R matrix of the camera profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .	194
D.23 Gaseous emissions profiles measured on the R matrix of the camera profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures	194
D.24 Gaseous emissions profiles measured on the R matrix of the camera profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars	195
D.25 Gaseous emissions profiles measured on the G matrix of the camera surrounding an Al droplet burning in air at various pressures	195
D.26 Gaseous emissions profiles measured on the G matrix of the camera surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .	196
D.27 Gaseous emissions profiles measured on the G matrix of the camera profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures	196
D.28 Gaseous emissions profiles measured on the G matrix of the camera profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .	197
D.29 Gaseous emissions profiles measured on the G matrix of the camera profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures	197
D.30 Gaseous emissions profiles measured on the G matrix of the camera profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars	198
D.31 Gaseous emissions profiles measured on the B matrix of the camera surrounding an Al droplet burning in air at various pressures	198

D.32 Gaseous emissions profiles measured on the B matrix of the camera surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .	199
D.33 Gaseous emissions profiles measured on the B matrix of the camera profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures .	199
D.34 Gaseous emissions profiles measured on the B matrix of the camera profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .	200
D.35 Gaseous emissions profiles measured on the B matrix of the camera profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures .	200
D.36 Gaseous emissions profiles measured on the B matrix of the camera profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars .	201

List of Tables

2.1	Specific impulse and thrust for various technologies	6
3.1	Chemical equilibrium temperatures from the CEA code for various pressures, in the case of liquid aluminum in air, both being initially and arbitrarily set at 1500 K	31
3.2	Chemical equilibrium temperatures from the CEA code for various diluents with 21 % vol. O_2 , in the case of liquid aluminum and oxidizer, both at 1500 K and 1 bar.	41
5.1	Temperature assessment methods and their efficiencies.	66
5.2	Emission wavelengths of Al containing gases	88
5.3	Summary of the optimization constraint errors for separate cases and averaged errors.	90
6.1	Summary of the processed experimental cases.	96
6.2	Summary of useful parameters for the theoretical evaluation of the SOR. . .	114
6.3	Summary of the experimental results.	129
7.1	Simulation parameters to assess the influence of the size of alumina droplets. .	137
7.2	Simulation parameters to assess the influence of oxide smoke diffusivity. . .	138
7.3	Simulation parameters to assess the influence of the pre-exponential factor of the condensation reaction.	140
7.4	Conditions for simulations performed using the CNT model.	146

Chapter 1

Introduction & Overview

In 1971, the first European project, 'Europa', was launched to make space more accessible. However, it failed to meet its objectives, leading to the birth of the Ariane program in 1973, initiated by the CNES (Centre National d'Études Spatiales). This program aims at providing a way for France to launch its own satellites into orbit, independently of the United States of America and of the URSS, which were at the time the leading countries regarding the access to space. The first successful launch of Ariane 1, in 1979, led to the rapid development of its successors, Ariane 2 launched in 1986, Ariane 3 launched in 1984, and Ariane 4 launched in 1988. Later on, in 1996, Ariane 5 is launched, equipped with new technologies improving performances and payload. In 2015, Airbus and Safran jointly formed ArianeGroup (formerly Airbus Safran Launchers) with in mind the development of the Ariane 6 launcher, which recently performed its first successful launch on the 9th of July 2024, as illustrated in Fig. 1.1.



Figure 1.1: Ariane 6 inaugural launch .

The increase in performances and cost reduction brought by Ariane 5 and Ariane 6 is correlated to the use of solid rocket motors, in which solid propellant is used, in opposition to the utilization of liquid propellants in previous versions of this launcher. This technology

provides a significant thrust at the early stages of the flight, along with a decrease in the costs of development, due to its ease of use, production and storage. With an increasing number of satellites in Earth’s orbit, as pictured by Fig. 1.2, decreasing the cost of the launches, along with improving the rocket’s performances, is at the core of many studies led by ArianeGroup.

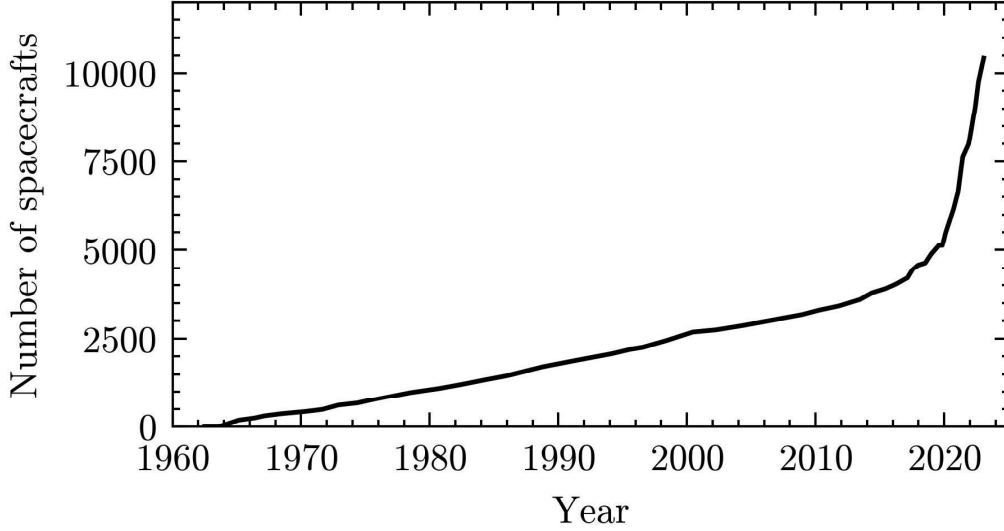


Figure 1.2: Historical increase of the cataloged spacecrafts in Earth’s orbit in February 2023, adapted from [1].

The work of this PhD thesis is motivated by the need for improving the performance of ArianeGoup’s motors, by enhancing our general understanding of solid propellant’s combustion, and more precisely, of aluminum combustion. The accurate combustion processes of aluminum particles, used as energetic additives in solid propellants, are yet to be fully understood and characterized. A state of the art of the current knowledge on aluminum combustion and its application is provided, and highlights the interest in the work of this PhD thesis. This work aims at deepening our knowledge on single aluminum particle combustion, with the use of a unique experimental setup coupled with newly developed optical diagnostics. The application of these diagnostics to the combustion of a single aluminum particle in various conditions enables crucial phenomena and mechanisms to be highlighted, and subsequently provides an unrivaled database for numerical models. In addition, the comparison of the obtained experimental data to ArianeGroup’s numerical model of aluminum combustion pinpoints areas of improvements for the model.

Chapter 2

Applications of aluminum combustion

Although being one of Earth's most common element, making up about 8% of the mass of its crust [2], aluminum has both a high energy density and specific energy. That is to say that if a given mass (related to specific energy) or a given volume (related to energy density) of aluminum was to ignite, it would produce a significant amount of energy. Figure 2.1 shows that aluminum has an energy density of about 23 kWh/l , greater than all gaseous and liquid fuels and than most solid fuels, while having a specific energy of about 9 kWh/kg , remaining competitive with other fuels.

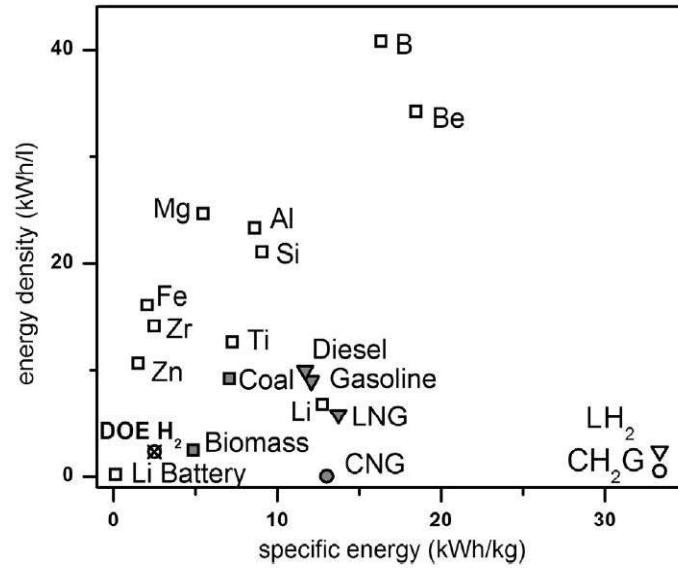


Figure 2.1: Energy density and specific energy for various gaseous fuels (○), liquid fuels (▽) and solid fuels (□), from [3].

Indeed, the combustion process for aluminum described later in this manuscript relies on the reaction of aluminum atoms Al with oxygen atoms O to produce alumina Al_2O_3 . The standard heat of formation at 298 K of alumina being of 1676 kJ/mol [4] compared to 393 kJ/mol for CO_2 and 242 kJ/mol for H_2O , the main combustion products of hydrocarbonated fuels, the formation of this aluminum oxide is a highly exothermic reaction, leading to high

flame temperatures and thus high energy production. The availability and thus the low cost of aluminum as well as its energetic potential, makes it a material of choice in the industry, whether it is for space conquest or for reducing our carbon emissions.

In this chapter, the application of aluminum combustion to the space propulsion field is introduced, followed by a short introduction to the use of aluminum as a clean energy carrier.

2.1 Aluminum in space propulsion

2.1.1 Introduction to rocket motors

Propulsive motors are based on the principle of ejecting gases at high velocity to thrust the system on the direction opposite to gas ejection. While some aircraft engines use the ambient air to burn the fuel, rocket motors use a fuel and an oxidizer that are stored within the engine. Ignoring the fuel and oxidizer storage, a propulsive system is generally composed of a combustion chamber, in which the fuel reacts with the oxidizer, and of a nozzle. In the combustion chamber, the chemical energy stored in the fuel is transformed into thermal energy to the gases through an exothermic reaction with the oxidizer. While the fuel burns, the gases in the combustion chamber are thus heated to high temperatures T_1 and reach high pressures p_1 . The repartition of the pressure forces in the chamber are provided in Fig. 2.2. The heated and pressurized gases with a velocity v_1 are transported towards a converging-diverging nozzle. As the mass flow $\dot{m}$ is conserved in the system, the decrease of the chamber's section at the nozzle results in high gas velocities v_t . Based on Newton's third law of movement, the ejection of the gases at temperature T_2 , pressure p_2 and overall, high velocity v_2 from the system provides a considerable amount of thrust to the engine. The chemical energy initially stored in the fuel is thus transformed to kinetic energy upon ejection of the gas phase.

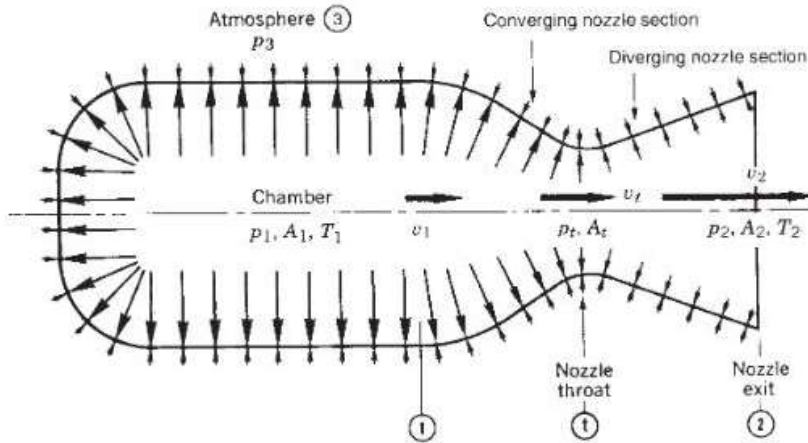


Figure 2.2: Schematic of a propulsive system, from [5].

The thrust F developed by the system from Fig. 2.2 can be expressed in terms of the mass flow $\dot{m}$ of the gases, the ejection velocity of the gases v_2 , the pressure of the gases at ejection p_2 , the ambient pressure p_3 and the section of the nozzle exit A_2 [5]:

$$F = \dot{m}v_2 + (p_2 - p_3) \cdot A_2 \quad (2.1)$$

In the case of rocket nozzles, the velocity profile of the ejected gases isn't uniform and is complex to assess. As a result, a mean effective velocity v_e is introduced [5] and we write:

$$v_e = F/\dot{m} \quad (2.2)$$

This mean effective velocity can be characterized by studying the variation of enthalpy of the gases. Assuming that during the combustion process, the gases are heated isentropically, that the thermal losses to the walls are negligible, and that the gases can be considered ideal, then the variation in enthalpy Δh of a mass m of gas of specific heat capacity c_p during its residence time in the combustion chamber can be described by Equation (2.3).

$$\Delta h = c_p m (T_1 - T_2) \quad (2.3)$$

The variation of enthalpy of the gas being equal to the kinetic energy transferred to the ejected gases $\frac{mv_e^2}{2}$, the squared mean effective velocity can be expressed as:

$$v_e^2 = 2c_p (T_1 - T_2) \quad (2.4)$$

Recalling the previous assumption of an isentropic transformation, and writing γ as the adiabatic index of the gas, R the ideal gas constant and M the molar mass of the gas, we write:

$$v_e^2 = \frac{2\gamma}{\gamma - 1} \frac{RT_1}{M} \left[1 - \left(\frac{p_2}{p_1} \right)^{\frac{\gamma-1}{\gamma}} \right] \quad (2.5)$$

The development of this mean effective ejection velocity is important as it enables the introduction of the specific impulse I_{sp} that characterizes the performances of a system. This specific impulse represents the time during which one kilogram of fuel can produce the required thrust to lift a mass of one kilogram in Earth's gravitational field. The specific impulse is defined as the ratio of the mean effective ejection velocity to the gravitational acceleration g .

$$I_{sp} = \frac{v_e}{g} = \frac{F}{\dot{m}g} \quad (2.6)$$

Equations (2.4), (2.5) and (2.6) demonstrate that the performances of the system are highly dependent on the properties of the propellant gas. The gas temperature reached in the combustion chamber T_1 , the molar mass of the gas M , or the heat capacity of the gas c_p are first order parameters when it comes to thrust and specific impulse. The choice of the fuel and of the oxidizer is thus crucial and needs to be made to account for the specifics of the mission. Some orders of magnitude of specific impulses and their associated thrusts for various technologies are presented in Table 2.1 below.

As highlighted by Table 2.1, a wide range of specific impulses can be obtained depending on the system used. It is important to note that although the specific impulse of the system helps characterizing its performances, getting the highest specific impulse does not always

Table 2.1: Specific impulse and thrust for various technologies

Technology	$I_{sp}(s)$	$F(kN)$
Turbojet	3000	50-500
Solid-propellant rocket	250	10000
Ariane 6 booster	278.5	4500
Liquid-propellant rocket	450	2000 - 3000
Ion thruster	1000-4000	0.0001
Nuclear	900	250
Magnetoplasma rocket	3000-12000	0.0005-0.005

match the requirements of the mission. Indeed, an ion thruster has a high specific impulse, but the thrust associated to this system is extremely low. As a result, ion thrusters are used in space where the systems do not require important thrust to generate the required movement, in opposition to first stages rocket motors for which the thrust needs to be sufficient to compensate the gravitational acceleration and to reach a sufficient velocity to exit the atmosphere. For rocket lift-off, liquid and solid propellant rockets are widely used as these systems have the greatest thrusts while maintaining relatively good specific impulses. Liquid-propellant rockets have a higher specific impulse than solid-propellant rockets and their operating regime can be adapted, but their design is much more complex and the storage of the liquid fuel and oxidizer, under pressure, is also a challenge. Solid-propellant rockets are thus a technology of choice due to the easier maintenance, design and fuel storage, but also because of its lesser cost compared to the thrust that it can develop. Solid-propellant rockets are thus widely spread in the space exploration field, and can be found for instance in the Ariane 6 launch system. In the case of Ariane 6, the solid propellants used in the boosters contains aluminum as an energetic additive to enhance the thrust and specific impulse of the system. In the context of this work, the solid-propellant rockets will be further described.

2.1.2 Solid-propellant rocket motors (SRM)

Solid-propellant rocket motors are a type of propulsive systems for which the fuel/oxidizer mixture is solid, and attached to the wall of the combustion chamber. SRM are the counterpart of the liquid-propellant rocket engines (LRE) for which the liquid fuel and the liquid oxidizer are stored under pressure, upstream of the combustion chamber as illustrated by the Figure 2.3.

Ariane booster's solid propellant, also called grain, is a mixture of 68 % of ammonium perchlorate NH_4ClO_4 (AP) acting as the oxidizer, 14 % of Hydroxyl-terminated polybutadiene (HTPB) that acts as a binder and a fuel, and 18 % of aluminum particles acting as a fuel and energetic additive [7]. The composition of the grain as well as its combustion process is described in Figure 2.4. The composition of the solid propellant is crucial as the use of a mixture made of 90 % AP and 10 % HTPB provides a combustion temperature of about 3000K while the addition of 16% of aluminum can boost the combustion temperature up to 3600K, hence the denomination of energetic additive for aluminum [6].

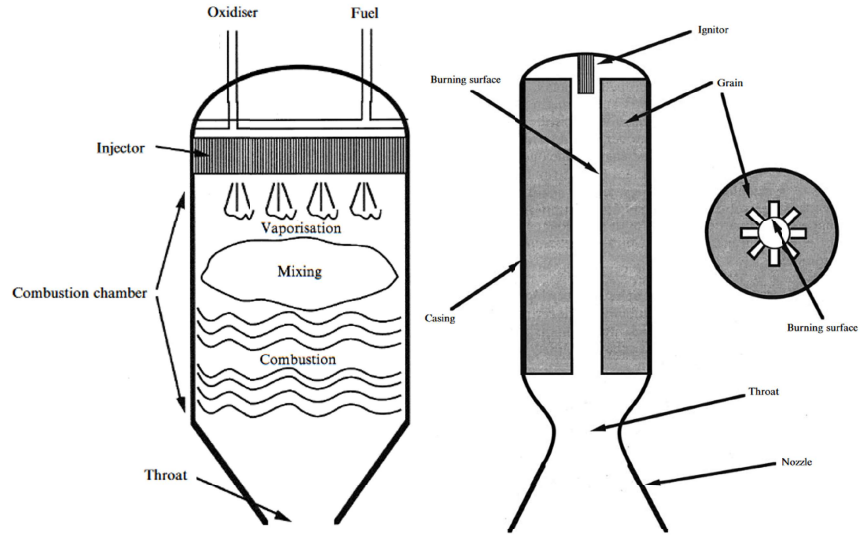


Figure 2.3: Schematic of a LRE (left) and a SRM (right), from [6] .

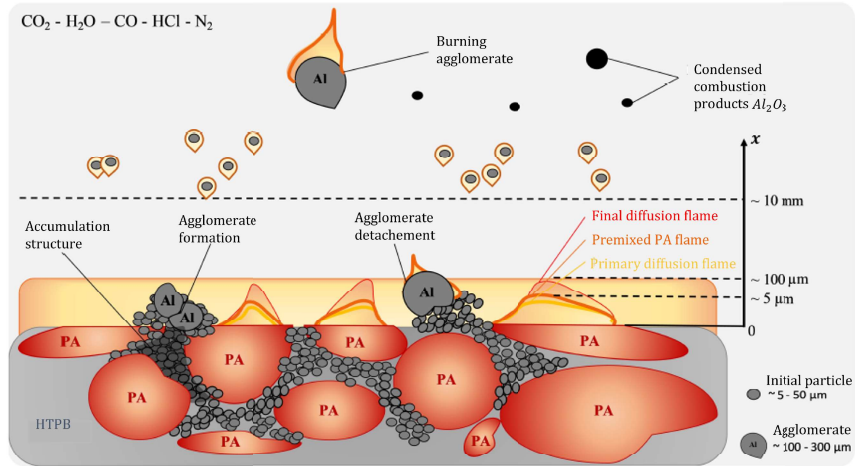


Figure 2.4: Schematic of the combustion process of an aluminized HTPB/AP solid fuel from [8].

The ignitor, represented at the top of the combustion chamber in Figure 2.3, enables the initiation of the combustion process. The HTPB reacts with the AP to form premixed and diffusion flames at the surface of the solid mixture, while aluminum particles form agglomerates that ignite and detach from the solid mixture to burn away from this surface as schematized in Figure 2.4 and experimentally observed in Figure 2.5 by Zarko et. al. [9]. The reaction of AP with HTPB produces the gaseous combustion products while aluminum combustion yields condensed alumina particles as a combustion product. At the nozzle exit, the exhaust composition is of about 32 % aluminum oxide Al_2O_3 , 20 % carbon monoxide CO ,

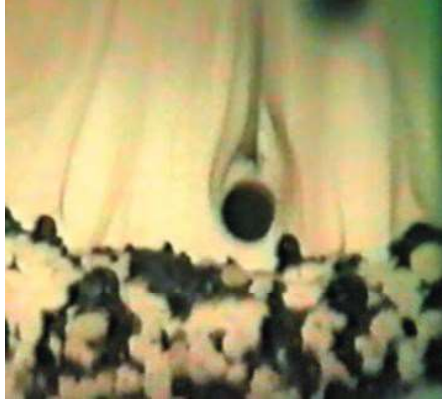


Figure 2.5: Image of a burning aluminum agglomerate, detached from the propellant surface, from [9].

16% water H_2O , 12% hydrogen chloride HCl , 10% nitrogen N_2 , 7 % carbon dioxide CO_2 and 3% chlorine Cl_2 and hydrogen H_2 . During this combustion process, the solid propellant is consumed and the surface regresses at a regression rate r_r such as:

$$\dot{m} = A_p r_r \rho_p \quad (2.7)$$

The mass flow of the gas is denoted as $\dot{m}$, the area of the solid propellant in contact with the gas is denoted as A_p and the density of the propellant is written ρ_p . An empirical relation enables to express the regression rate as a function of the pressure within the combustion chamber p :

$$r_r = ap^n \quad (2.8)$$

The factor a is an empirical constant and the term n is the combustion index. A high combustion index suggests that an important change in pressure will cause an important change in the propellant consumption rate while a low combustion index comes down to a regression rate almost independent of the pressure. The combustion index needs to be smaller than unity to avoid an exponential amplification of the pressure perturbations in the chamber. In the case of aluminized propellants, gas pressure and temperature are expected to be higher than in the case of non-aluminized propellants, increasing in turn the regression rate of the propellant for a non-zero combustion index, highlighting the need for a characterization of the impact of aluminum on the performances of a SRM.

2.1.3 The impact of aluminum

As mentioned previously, the aluminum particles within the solid propellant are considered as a fuel but also as an energetic additive, enabling a significant increase in the combustion temperatures and thus improving the overall performances of the motor. Once the combustion is initiated by the ignitor, not only the reaction of the HTPB with the AP leads to a regression of the propellant, enabling the diffusion of aluminum particles in the gas, but also the heat released from this reaction heats the gas at a temperature that is sufficient to initiate the

combustion of the aluminum particles. As illustrated by Fig. 2.4 and observed in Refs. [9, 10], the combustion of aluminum particles unfolds in a region detached from the flame zone of the propellant. Upon burning in the gas phase, the aluminum particles yield energy to the gas, reaching higher temperatures than with the use of non-aluminized propellants. From equations 2.5 and 2.2, it is obvious that the higher the gas temperature, the higher the thrust provided to the system. Han [11] investigated the influence of aluminum particles concentration on propellant surface temperature and gas phase temperature under various pressures. Upon increasing the concentration of aluminum particles from 1 % to 18 % in the propellant, they found that under a pressure of 18 MPa, the surface temperature of the propellant increases from 773 K to 1104 K while the maximum assessed gas phase temperature increases from less than 2000 K to about 3000K. Equations 2.4 and 2.2 showing that the thrust of the system is proportional to the square root of the gas temperature in the combustion chamber, and that a decrease in the molar mass of the gases results in an increase in the mean effective gas ejection velocity, the beneficial impact of aluminum in the solid propellant is evidenced.

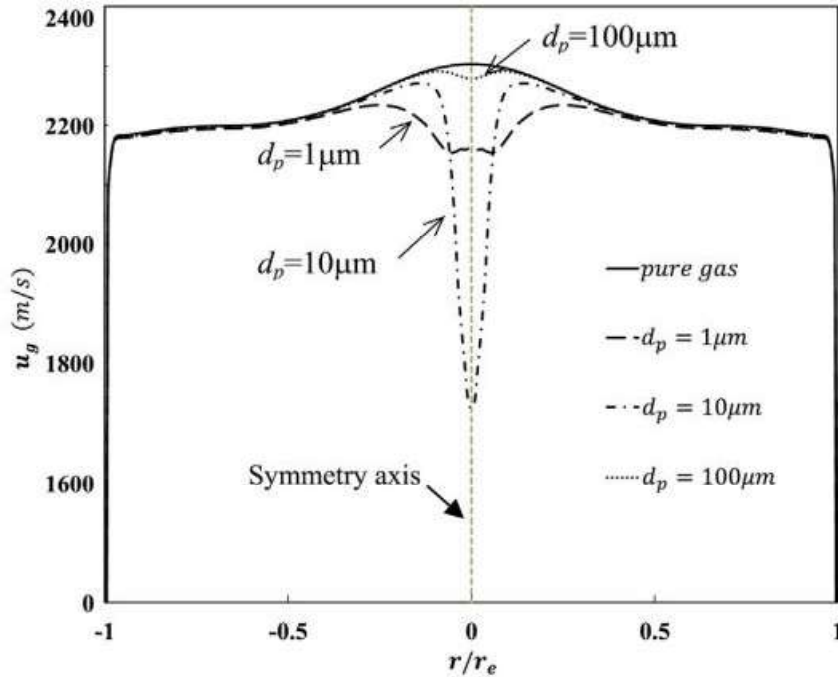


Figure 2.6: Gas phase velocity profiles at the nozzle exit for pure gas flow and two-phase flow with various alumina particle sizes, from [12].

Although aluminum enables better motor performances through combustion temperature, it can also be the source of performance losses due to the combustion products of aluminum. The reaction of aluminum with the available oxygen produces, at the final stages of the reaction, a stable phase of aluminum oxide, alumina Al_2O_3 , that condenses upon formation. The size of these condensed combustion products (CCPs) is not well known as it is highly dependent on the operating conditions of the system, such as the pressure or the concentration of aluminum particles [13]. Thus, in the context of aluminized rocket propellants, a wide range

of CCPs size is available in the literature such as $0.318\mu m - 45\mu m$ [14] or $0.1\mu m - 300\mu m$ [15]. The energy released from the formation of this condensed phase is highly valuable for motor performances, but the presence of this condensed phase within the flow can restrain this gain of performance because of two-phase flow losses. In opposition to a pure gas flow, a portion of the combustion mass in the two-phase flow cannot be compressed nor expanded and thus cannot be accelerated through the nozzle. Similarly, condensed particles have a significantly higher density than the surrounding gases and thus cannot accelerate as fast as the gas. This leads to alumina particles being ejected at the nozzle at lower velocities than gases due to momentum exchange between gases and condensed phase, resulting in a loss of performance as the mean effective ejection velocity is decreased from two-phase ejection. Figure 2.6 illustrates the decrease in the mean gas phase velocity at the nozzle exit in the case of addition of particles to the pure gas flow, leading to a decrease in the overall performances of the motor. Note however that Li et al. [12] simulate that the velocity lag magnitude in the case of larger particles ($100\mu m$) becomes negligible as for identical concentration of alumina, the effective area that drives momentum exchange decreases [16]. Furthermore, condensed matter shows a greater thermal inertia than gases and alumina particles can be ejected from the system before transferring their heat to the surrounding medium. Finally, condensed phases undergo radiative thermal losses in opposition to the gas phase, whose thermal energy is usually lost through conduction, diffusion or convection. The heat is poorly transferred to the non-condensed phases through radiation resulting in a share of the energy transported by the condensed phase transmitting to the walls or to the nozzle [17] and the efficiency of the energy conversion from chemical to kinetic is decreased.

Additionally, the regression of the propellant or the quick changes in altitude of the system leading to changes in thrust due to Equation (2.1) are parameters that are carefully accounted for during the development of the motor geometry, and the existence of condensed phase renders this task all the more complex. As alumina particles are produced within the combustion chamber, this condensed matter can, under certain conditions, be deposited at the nozzle throat, changing the geometry of the system and thus severely reducing the performances [18]. Similarly, the design of some systems can enable strong recirculation zones for the alumina particles, especially in the case of submerged nozzles [19]. In such recirculation regions, alumina accumulates and acts as an inert mass overloading the system, drastically decreasing the performances.

With appropriate geometries and fuel compositions, the energy that is gained from aluminum combustion is greater than the energy lost through two-phase flow losses and aluminum remains a competitive energetic additive for rocket propellants. However, another consequence of aluminum combustion in a motor needs to be accounted for as it does not only affect the performances, but it can also alter the integrity of the motor. Indeed, the use of a solid propellant can be the source of combustion instabilities that can result in more definitive consequences ranging from performances losses to motor failure. In rocket motors, combustion instabilities can occur due to thermo-acoustic instabilities described in this section or vortex shedding [5] that will not be discussed in the context of this work. Thermo-acoustic instabilities usually result from the coupling between pressure and heat release rate. In a system where such instabilities can occur, the initial pressure variations induce heat release rate variations that, in turn, impact the acoustic field within the combustion chamber, leading to greater pressure oscillations and thus greater heat release rate oscillations. A schematized

representation of thermo-acoustic instabilities is presented in Figure 2.7. Thermo-acoustic instabilities are predominant in LRE but were recently evidenced in SRM [20].

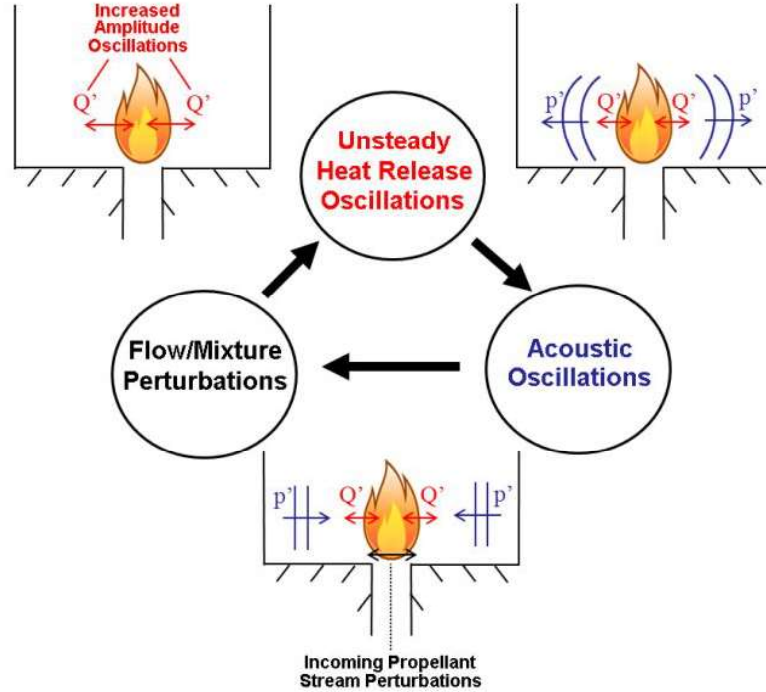


Figure 2.7: Schematized representations of thermo-acoustic instabilities, from [21].

The self-amplification of this phenomenon can lead to exponential increases of pressure in the combustion chamber. In the same way as geometry characterizes a flute, the onset of thermo-acoustic oscillations is strongly dependent on the geometry of the system. While the geometry design of the combustion chamber is crucial, in the case of solid propellant motors, the geometry of the combustion chamber changes with time as the solid propellant regresses during combustion and the effective volume of the combustion chamber increases, putting aside the potential alumina accumulation on the motor's walls. The coupling phenomenon responsible for thermo-acoustic instabilities is only problematic in motors when it meets a specific criterion on the heat release rate phase compared to the pressure oscillation phase. This criterion is called the Rayleigh criterion.

The work of Sutton [5] identifies phenomena that contribute to amplifications of vibrations and acoustic energy increase such as propellant formulation, combustion's dynamic response to the flow, flow interactions or vortices. Phenomena contributing to vibrations decrease and damping of the acoustic energy are identified as the viscous damping in the boundary layers at the walls, condensed phase damping (droplets), the presence of the nozzle or acoustic energy absorption by the propellant or the motor case. In opposition to Sutton's identification of the condensed phase as a damping factor of acoustic induced instabilities, Orlandi [22] identifies the combustion of aluminum droplets as a driving mechanism of the instabilities. Figure 2.8 illustrates the absence of pressure oscillations in the chamber for a monophasic flow in opposition to pressure oscillations reaching up to 7% of the average pressure in the

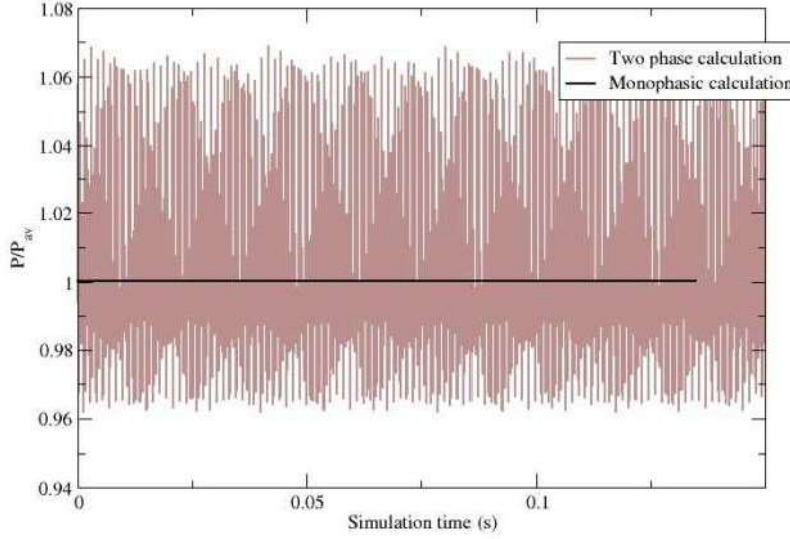


Figure 2.8: Simulations of pressure oscillations in monophasic and two-phase flow, from [22].

case of a two-phase flow.

It has to be noted that in addition to two-phase performance loss and combustion instabilities, the combustion time of aluminum particles is a crucial parameter to consider. As mentioned, high flow velocities are sought after in SRM designs and residence time of particles within the combustion chamber are consequently short. If the combustion time of an aluminum particle is longer than its residence time, then incomplete combustion of the aluminum is observed and the efficiency of the system decreases.

Using aluminum particles as an energetic additive to the solid-propellant enables a significant enhancement of the performances of the motor. Although two-phase flow losses and combustion instabilities can occur, a deep characterization of aluminum combustion, an adapted geometry or an appropriate fuel composition generally leads to the benefits of aluminum combustion outweighing the drawbacks.

2.2 Aluminum as a clean energy carrier

In the last seventy years, the unrestrained use of fossil fuels, hydrocarbons, in the industry has led to a dramatic increase in annual carbon dioxide emissions as displayed in Figure 2.9. In 2023, the annual emissions of this greenhouse gas reached more than 35 billion tons, significantly impacting climate change. Recent challenges to curtail carbon dioxide emissions involved replacing hydrocarbons by hydrogen but resulted in difficulties due to its low energetic density illustrated in Fig. 2.1 and issues related to usage and storage safety. In opposition to hydrogen, the generation of electricity from renewable sources has been multiplied by four over the last twenty-five years [23].

The main challenge of electricity generation through renewable energies is the storage as electricity cannot be stored nor shipped. The intermittency of renewable energies coupled to the incapacity of current technologies to store the produced electricity makes renewable

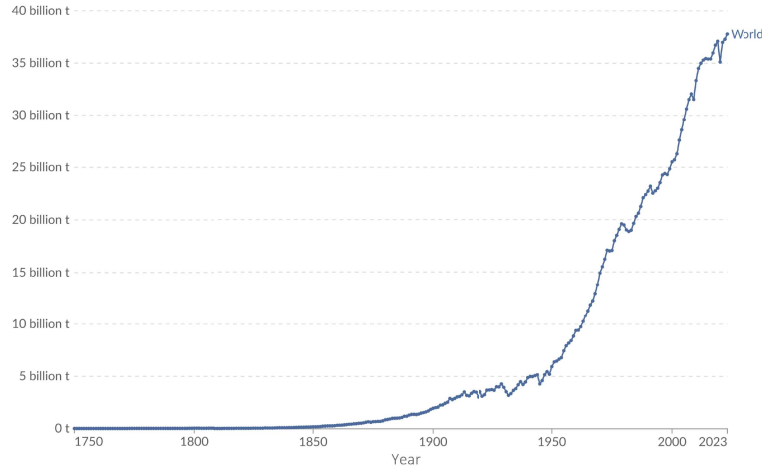


Figure 2.9: Annual CO_2 emissions from fossil fuels and industry, from [23].

energies complex to use on a daily basis. Recent studies have proposed aluminum combustion as a potential solution to store this renewable energy, making aluminum a clean energy carrier [3, 24]. Aluminum offers a high energy density, a high specific energy, can be easily and safely stored, and has the capacity to release its energy through a clean combustion process. Indeed the combustion process for aluminum can be simplified to the one step reaction $Al + O \rightarrow Al_2O_3$. In the hypothesis of aluminum droplets burning in oxygen without nitrogen, the combustion energy is released without production of carbonated species nor nitrogen oxides. Furthermore, the combustion products being condensed, they can be easily captured and the energy release thus involves no gaseous emissions. With the use of a reduction reaction that requires an energy income, the captured alumina can be reduced to form the aluminum fuel again along with a production of oxygen. The ease of storage and transportation of alumina enables its shipment towards locations where renewable energy is available and can be used as the energy income to produce the aluminum fuel from alumina reduction through a clean process. The formed aluminum fuel can be in turn transported and burned where the energy needs to be supplied. This clean energy cycle enabled by aluminum combustion is summarized in Fig. 2.10.

This described energy cycle is not ready to be implemented at large scales yet. While transportation of aluminum or of its oxide is fairly simple, there lacks systems to efficiently and quickly convert the energy stored within aluminum particles to electricity. Recent works are however studying the development of burners enabling this energy conversion, using aluminum [25] or iron particle [26].

Trowell et al. [24] compared the efficiency of the aluminum clean energy cycle to other carbon-free energy cycles and assessed an efficiency ranging from 20 to 26 % for the hydrogen cycle, from 15 to 18 % for the ammonia cycle and from 23 to 26 % for the aluminum cycle. Although aluminum shows good promises as a clean energy carrier and seems to compete relatively well with hydrogen, the enhancement of the cycle's efficiency is still being studied. Additionally, there are underlying complex challenges that are yet to be tackled. More specifically, for a perfectly clean energy cycle, the transportation of aluminum and aluminum

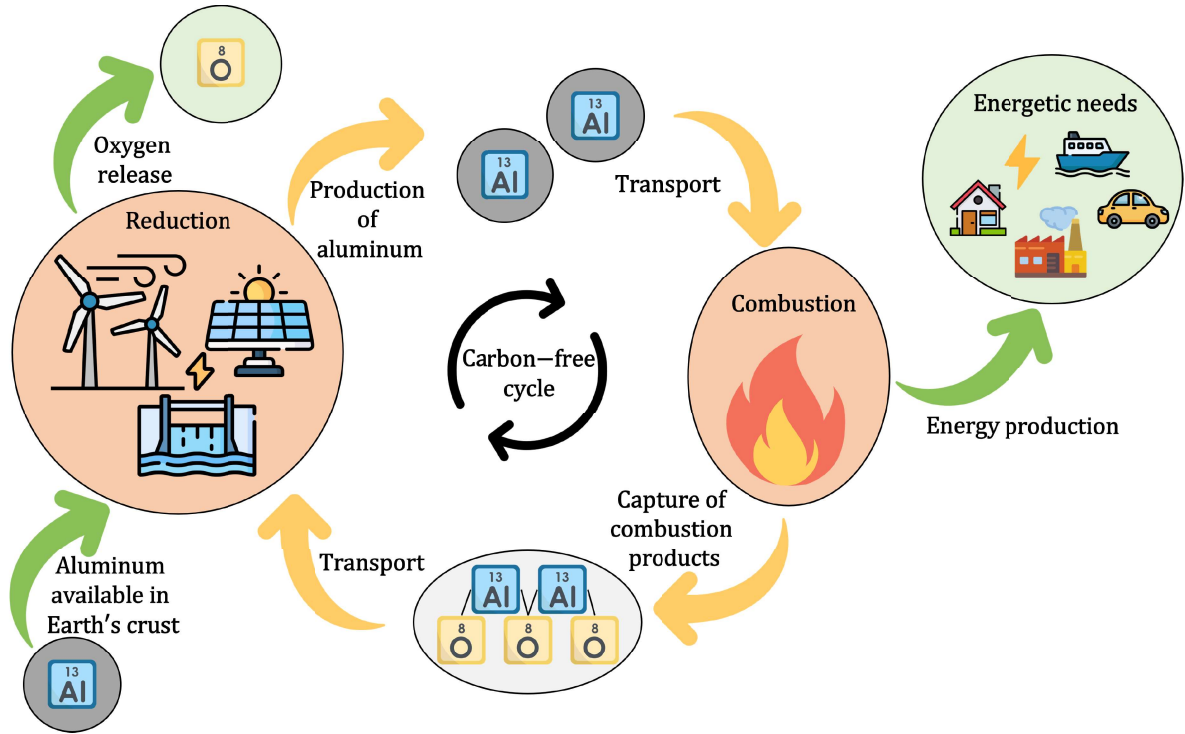


Figure 2.10: Aluminum-enabled carbon-free energy cycle.

oxide needs to be carbon-free and thus engines using aluminum or renewable energies have to be used. Also, the reduction process of alumina to aluminum needs to be clean and the 'classical' electrolysis using a sacrificial carbon electrode to reduce alumina cannot be used as this reaction produces carbon dioxide [27].

2.3 Summary

In propulsive systems, and mainly in the field of space exploration, solid-propellant motors are used because of design simplicity and high performances, i.e significant thrust for long durations. Most solid-propellants contain aluminum particles acting as an energetic additive. Upon propellant consumption, aluminum particles ignite and burn at higher temperatures than HTPB-AP flame temperatures, highly enhancing the performances of the motor. The combustion of aluminum forms aluminum oxide that condenses upon generation, yielding condensed phase aluminum oxide, alumina. The existence of condensed phase in rocket motors leads to performance losses from two-phase flow, combustion instabilities and incomplete combustion that are usually both compensated by performances enhancement. Aluminum is also actively studied as a clean energy carrier as alumina can be captured, following the highly exothermic combustion process, and can be easily transported to a location where renewable energies enable the reduction of alumina to aluminum. The reduced aluminum can be transported again to places of energetic needs, where it can be burned again to produced the required energy. Difficulties regarding cycle efficiencies or full carbon-free processes are the focus of current studies. Both of the applications of aluminum combustion in the

industry have shown how important aluminum is, and how crucial the future developments of aluminum combustion will be. For both applications, the high exothermicity of aluminum combustion is of utmost interest while the existence of condensed combustion products has appeared to be either beneficial or detrimental. With this introduction to the applications of aluminum combustion in the industrial field, it is essential to fully understand the mechanisms of aluminum combustion in order to maximize the efficiency of its utilization. In the context of this work, the following chapter will provide a full description of the combustion process of a single aluminum particle, from its ignition to its quenching.

Chapter 3

Single aluminum particle combustion

The mode of operation of the aluminized solid propellant rocket motors used on the Ariane 6 launchers, for instance, was introduced in the previous chapter. It has been highlighted that during the combustion of the solid propellant, aluminum particles or aggregates detach from the propellant and burn, as single particles, in the combustion chamber. It has been evidenced that aluminum combustion significantly enhances the performance of the motor. However, the energy provided by the combustion of aluminum to the system, as well as the occurrence of a two-phase flow problem, are the results of highly complex phenomena. In order to improve our overall understanding of the processes unfolding in the combustion chamber of solid propellant rocket motors, it is of prime importance to understand, at first, the combustion process of a single aluminum particle. In this chapter, the complete combustion process of an aluminum particle will be described, from ignition to quenching.

3.1 Ignition phase

The ignition phase of an aluminum particle is defined as the period during which external energy is provided to the particle until the chemical reactions of aluminum with oxidizers are initiated. These reactions are expected to produce enough energy for the particle to enter a self-sustained combustion regime. In theory, the particle is ignited when the oxidizers and the aluminum are in contact, at a temperature that is sufficiently high so that the activation energy of the oxidation reaction is exceeded. In its standard state, an aluminum particle is composed of an aluminum core, coated by an alumina layer that results from natural passivation. The thickness of this alumina layer is a few nanometers, generally measured between 2 and 8 nanometers [28, 29]. Note that for an aluminum particle of 70 microns in diameter with an oxide shell of 4 nanometers in thickness, the mass of alumina represents about 0.05 % of the mass of the aluminum core, at 300 K. Although insignificant in mass, the presence of this oxide shell entirely drives the ignition process of the aluminum particle, as this protective layer prevents the ambient oxygen from diffusing towards the underlying aluminum core. In most cases, the oxide layer must then be removed to enable particle ignition.

Let us consider an aluminum particle and its oxide layer in an oxygenated environment, such as represented by Figure 3.1 in which the oxide layer thickness is deliberately overstated.

The first structural change during the ignition phase happens upon reaching the melting temperature of aluminum, at 933 K. At this temperature, the oxide layer is still in its solid phase, while the aluminum core is liquid. With this phase change, the density of aluminum decreases and the liquid core expands, exerting pressure on the oxide shell. If the particle is small enough (bottom of Fig. 3.1), with a diameter below the micrometric scale, increasing the temperature leads to an increase in internal pressure, and the stress exerted by the liquid core enables the cracking of the oxide layer, while it remains solid [30]. Once the alumina shell is cracked, the ambient oxygen can find its way and diffuse to the aluminum liquid core and the first oxidation reactions of aluminum can occur, leading to thermal runaway. The particle is ignited.

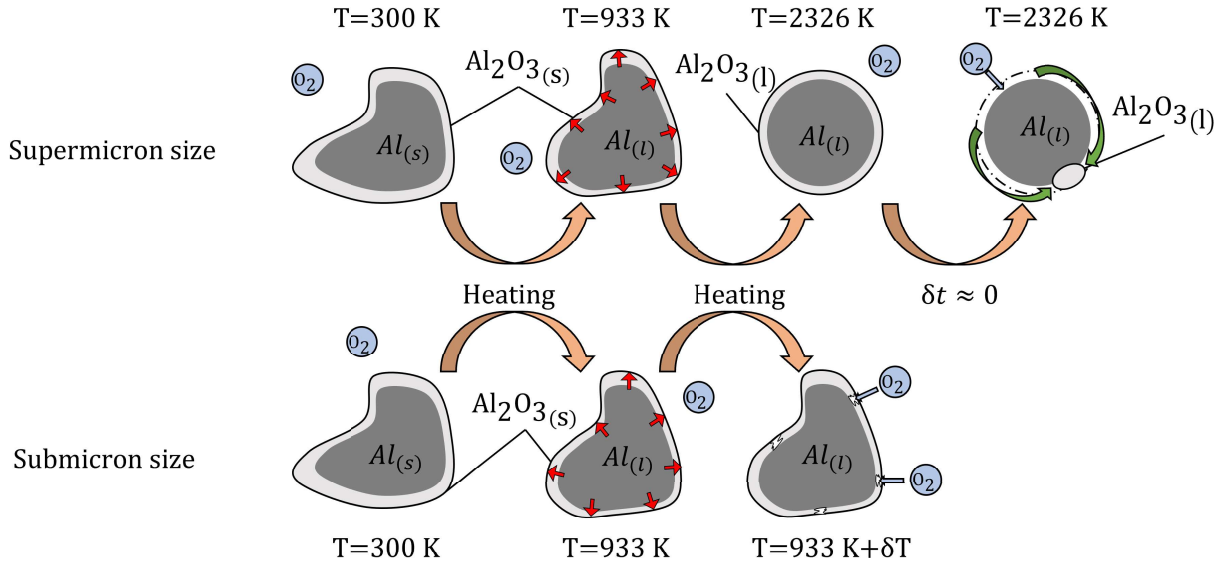


Figure 3.1: Schematized ignition process of an aluminum particle at high and low heating rates. Pressure forces are represented by red arrows.

For larger particles, with diameters larger than a micron (top of Fig. 3.1), the pressure exerted by the aluminum core on the oxide shell appears to not be sufficient to cause the cracking of the layer, or the existing cracks do not allow sufficient reactions to cause a thermal runaway. As a result, additional heating of the particle is necessary. Once the melting temperature of the oxide layer of 2326 K is reached, the oxide shell becomes liquid. The effects of surface tension cause the particle to become spherical. The time period for which the liquid oxide layer remains in the shape of a shell surrounding the aluminum core is very short. The non-miscibility of aluminum with alumina, the thinness of the shell and the pressure from the liquid core cause the alumina shell to retract and form a unique oxide cap on the surface of the aluminum core that has now become an aluminum droplet. Upon retraction of the oxide shell, the aluminum is in obvious contact with the ambient oxygen and the droplet ignites.

The work of Huang et al. [31] summarized the various ignition temperatures existing in the literature, as a function of the aluminum particle diameter and the results are presented in Figure 3.2. The low ignition temperatures for small particles can be attributed to the cracking

of the oxide layer, as described above [30], but other works attribute this phenomenon to the polymorphic phase transformations unfolding in the alumina layer [32]. The existence of a diameter of transition of around one micron is attributed to the increase in volumetric heat capacity, as the diameter of the particle increases [33].

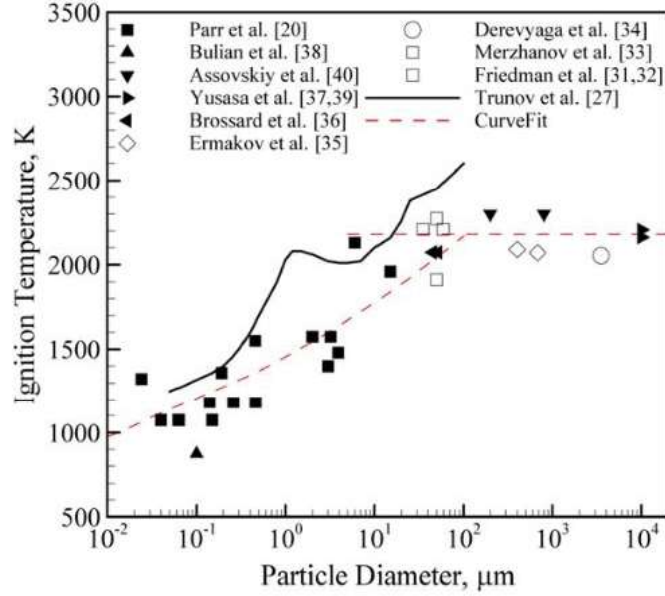


Figure 3.2: Ignition temperature as a function of aluminum particle size, from [31].

Along with the particle size, the heating rate of the particle is an additional parameter that can control the ignition process of the particle. It has been suggested that for low heating rates, the diffusion of oxygen atoms through the oxide shell is given enough time to operate. Inversely, fast heating rates generally lead to a fast-melting process of the oxide layer that enables the dispersion of the oxide layer when combined with pressure forces from the aluminum liquid core [34].

Finally, it has to be considered that the system used to ignite the particle can also impact the ignition process. In the literature, various ignition procedures can be found, such as hydrocarbon flame ignition [35], laser ignition [36, 37], ambience heating [38] or shock ignition [39]. Note that in the case of multiple particle systems, aluminum dust flames, the first aluminum particle needs to be ignited with a supply of external energy. However, when the first ignition is reached, the energy developed by aluminum particle combustion is generally sufficient to ignite the surrounding particles. Burning aluminum droplets thus supply the required ignition energy to non-ignited aluminum particles [40].

Once the oxygen reaches the liquid aluminum, the combustion reaction is initiated, thermal runaway occurs and no additional external energy is required to maintain the process. The aluminum droplet enters a self-sustained combustion phase that is described in the following section.

3.2 Combustion phase

This section aims at providing a complete overview of the combustion phase, following ignition, of a single aluminum droplet. The system considered is composed of the aluminum liquid droplet and its associated oxide cap. The aluminum is in contact with the ambiance, enabling combustion. The global phenomenology is introduced, prior to discussing temperatures and temporal aspects.

3.2.1 Phenomenology - A simplified approach

The previous section of this chapter identifies a maximal ignition temperature for aluminum particle that is the melting temperature of the oxide layer $T_m^{ox} = 2326K$. In this section, it is thus considered an aluminum droplet at an initial temperature of about 2300 K. The aluminum liquid droplet being in an oxygenated environment, it is assumed that aluminum reacts with oxygen to produce alumina along the following reaction:



Let Q_r be the heat of reaction, the amount of energy developed by reaction of Eq.3.1. Q_r can also be considered as the negative heat of formation of alumina. A share of this heat of reaction will heat the aluminum particle to higher temperatures, and the other share of this energy will be lost to the ambiance. If the heat from the combustion is not sufficient to bring the aluminum droplet to its boiling point, the aluminum remains in its liquid state and no evaporation can occur. In this case, the combustion is driven by the diffusion of oxygen to the surface, and limited by the kinetics of the surface reactions. This combustion regime is called a kinetically-limited regime, as the driving process of the oxidation reactions occur through heterogeneous reactions on the droplet's surface. This combustion regime is illustrated by Figure 3.3. This regime has been observed for particles with a diameter smaller than a critical diameter of a few microns. Bazyn et al. [41] observed a decrease in combustion temperature when the particle size decreases, to a point where the combustion of nanometric aluminum particles involves flame temperatures lower than the boiling point of aluminum. In this regime, the combustion process is complex, as alumina forms at the surface of the droplet, reducing the amount of aluminum surface that can react with oxygen, to a point where an oxide layer can surround the liquid aluminum core, in the same manner as during the ignition phase. In the kinetically-limited regime, three possible combustion modes are identified [33]. The first mode, involves diffusion of oxygen through cracks of the oxide layer, enabling reactions of oxygen with the liquid core. A second interfacial diffusion burning mode for which oxygen diffuses through the oxide layer to react at the core-shell interface, and a surface diffusion burning mode where aluminum can diffuse through the oxide layer to react with oxygen at the surface of the droplet. For all modes, the combustion products are generated attached to the fuel droplet. The initial particle thus gets larger and heavier during combustion, and aluminum is more difficult to consume, as alumina is produced.

In the context of this work, and in agreement with the composition of Ariane 6 solid propellants, particles with initial diameters larger than the critical diameter will be studied, introducing a second combustion regime for aluminum droplets. For large enough particles,

Kinetically–limited regime

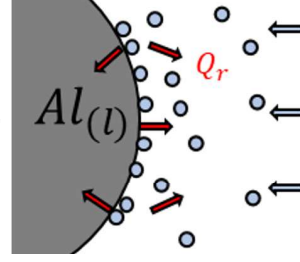


Figure 3.3: Combustion of an aluminum particle in a kinetically-limited regime. Blue circles: oxygen, blue arrows: oxygen flux, red arrows: heat release.

the energy released from the first reactions of aluminum with oxygen enables the heating of the droplet to its boiling temperature. Aluminum thus evaporates from the droplet and can react in a zone detached from the droplet’s surface. This regime is not controlled by the kinetics of the surface reactions, as opposed to the kinetically-limited regime, but is controlled by the diffusion of the reacting species to the flame region, leading to a diffusion-limited regime, also called the vapor phase regime. In order to maintain this vapor phase combustion, the droplet’s temperature needs to be at the boiling temperature and thus, the flame temperature needs to be greater than this boiling temperature. Additionally, Glassman and Yetter [4] state that physically, the flame temperature of a burning metal particle is limited by the volatilization-dissociation temperature of the produced oxide. If H_{vol} is the enthalpy required to heat the oxide from its reference temperature to its volatilization-dissociation temperature, then the heat that is available after combustion and oxide heating ΔH_{avail} is expressed as $\Delta H_{avail} = Q_r - H_{vol}$. Then, if ΔH_{vap} is the enthalpy required to vaporize the oxide, the following limitation for metal particle combustion arises [4]:

$$\Delta H_{avail} < \Delta H_{vap} \quad (3.2)$$

The flame thus does not develop sufficient energy to enable the existence of gaseous alumina in the system. These considerations put together help determine a maximum and a minimum value for the flame temperature T_f of an aluminum droplet burning in the vapor phase, respectively, the volatilization temperature of the produced oxide T_{vol}^{ox} and the boiling temperature of the metal T_b^{Al} . This is defined as Glassman’s criterion for vapor phase combustion:

$$T_{vol}^{ox} > T_f > T_b^{Al} \quad (3.3)$$

Note that for some metals such as boron, the volatilization temperature of its oxide is lower than the boiling temperature of boron, leading to the incapacity of boron to burn in the vapor phase, according to Glassman’s criterion.

Back to the vapor phase combustion of an aluminum droplet, if the assumption of infinitely fast kinetics is made, a flame occurs detached from the droplet and behaves in the way of a counterflow flame. Aluminum diffuses away from the droplet in a flux called the Stefan flow, to the infinitely thin reaction zone, while oxygen diffuses to the reaction zone via Fick’s diffusion. A schematic of an aluminum droplet burning in a diffusion-limited regime, with infinitely fast chemistry, is presented in Figure 3.4. In this model, a share of the heat produced

in the reaction zone is transferred to the droplet to maintain its temperature at the boiling temperature of aluminum, fueling the Stefan flow of aluminum vapor to the reaction zone.

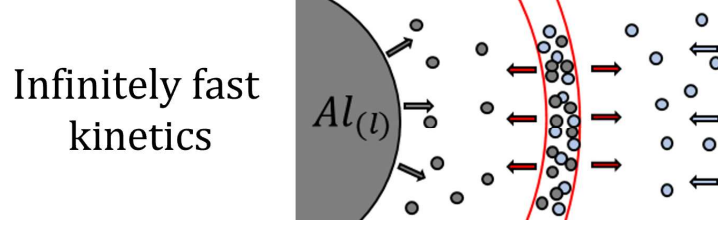


Figure 3.4: Combustion of an aluminum particle in a diffusion-limited regime, under the assumption of infinitely fast kinetics. The red circle segments delimit the reaction zone. Blue circles: oxygen, blue arrows: oxygen flux, gray arrows: aluminum Stefan flow, gray circles: aluminum vapor, red arrows: heat release.

With the introduction of the kinetically-limited regime and the diffusion-limited regime, a competition between kinetics and diffusion arises to determine the combustion regime of a burning aluminum droplet. Glassman and Yetter [4] derived the equations enabling the assessment of such competition. In the case of a particle burning in the vapor phase regime, the burning rate of the droplet k_{dif} can be expressed in terms of the density of the ambient gases ρ , of the diffusivity of the gases D , the droplet radius r_d , the stoichiometric coefficient of the reaction i and the oxygen concentration in the ambient gases $m_{o\infty}$:

$$k_{dif} = \frac{\rho D \ln(1 + im_{o\infty})}{r_d} \quad (3.4)$$

Upon integrating this burning rate, the combustion time of the droplet in a diffusion-limited regime $t_{b,dif}$ is obtained as a function of the droplet density ρ_d and droplet initial diameter d_0 :

$$t_{b,dif} = \frac{\rho_d d_0^2}{8\rho D \ln(1 + im_{o\infty})} \quad (3.5)$$

In the case of a particle burning in a kinetically-limited regime, the burning rate k_{kin} is a function of the reaction rate of the heterogeneous reactions k_s :

$$k_{kin} = i\rho m_{o\infty} k_s \quad (3.6)$$

The burnout time $t_{b,kin}$ is similarly assessed:

$$t_{b,kin} = \frac{\rho_d d_0}{2i\rho m_{o\infty} k_s} \quad (3.7)$$

It appears from Equations (3.5) and (3.7) that the burnout time in the diffusion-limited regime is proportional to the squared droplet diameter, whereas it is linearly proportional to the droplet diameter in the kinetically-limited regime, showing that a transition between these two regimes results in significant discrepancies in burning phenomenology. The Damkhohler number Da is introduced to characterize the combustion regime through comparison of the burnout times:

$$Da = \frac{t_{b,diff}}{t_{b,kin}} = \frac{d_0 im_{o\infty} k_s}{4D \ln(1 + im_{o\infty})} \quad (3.8)$$

In the literature, a reduced Damkholer number is introduced as $Da^* = \frac{Da}{Da+1}$ so that a reduced Damkholer close to unity means that the Damkholer is high and thus that the combustion regime is diffusion-limited, while a low reduced Damkholer is the result of a kinetically-limited regime. It is common to find in the literature that the combustion regime is determined by this reduced Damkholer number and that large aluminum particles burn in the vapor phase regime shown in Figure 3.4 [41, 42].

Transitional regimes are also introduced with reduced Damkholfers close to 0.5 [43], during which the vapor phase regime and the kinetically-limited regime can coexist. A flame is generated close to the droplet's surface, while heterogeneous surface reactions occur. Although this transitional regime is often considered in the literature for particles of sizes close to the critical size of a few micrometers [43], evidenced at 10 microns at 8.5 atm in the work of Bazyn et al. [41], recent works mention the existence of the transitional regime for a much larger range of particle sizes, from 100 nm to 100 microns [44].

While kinetically-limited and diffusion-limited combustion regimes enable a simplified characterization of a burning aluminum droplet, in the context of this work, these combustion regimes are set aside and a more detailed description of the combustion phenomenology is presented.

3.2.2 Phenomenology - A more detailed approach

Similarly to aluminum particles used in solid propellants, the diameter of the particles studied in this work is greater than 10 microns. For most conditions, particles of such size are not expected to burn in a kinetically-limited surface regime, at least with the use of oxygen as an oxidizer. A flame will develop, detached from the droplet's surface, while surface reactions can yet occur. A detailed description of this burning regime is introduced. What makes difficult the characterization of a burning aluminum droplet is the lack of spatially resolved experimental data. Although Bucher et al. introduced groundbreaking experimental spatially resolved temperature and species concentration profiles more than twenty years ago [45], there has been very few to none other spatially resolved experimental work since. However, various simulation studies tried to replicate Bucher's results [46, 47].

Gaseous suboxides

Let us consider an aluminum droplet that has been ignited. Gaseous aluminum thus evaporates from the droplet to reach the available oxygen for the combustion to occur. Interestingly, upon burning a 220 μ m-diameter aluminum droplet in an O_2/Ar 21/79%vol. mixture, Bucher [45] observes, using a planar laser induced fluorescence (PLIF) technique, that aluminum suboxides are formed close to the droplet's surface. From this observation, the simplistic assumption of a one step reaction assuming $Al + O \rightarrow Al_2O_3$ in an infinitely thin region does not hold, as species such as AlO observed by Bucher seem to be the first reaction intermediate, formed before any further oxidation of aluminum. Other suboxides such as Al_2O or AlO_2 are also expected to be important intermediates produced before the formation of alumina.

Bucher observes high relative concentrations of AlO at about 1 droplet radius from the droplet's surface. At this same location, Bucher measures relatively low concentrations of alumina, which are detected in higher concentrations at 3 to 4 droplet radii from the droplet's surface. Such measurements indicate that the reaction zone of aluminum with oxygen is wide and the assumption of infinitely fast kinetics does not hold. King [48] proposes a model of aluminum droplet combustion introducing a primary sheet flame and a condensation sheet. This structure, which can be found in Fig. 3.5, assumes that aluminum evaporates from the droplet towards the primary flame sheet, where it reacts to form AlO and Al_2O_2 . King assumes that AlO diffuses back to the droplet's surface and that between the primary flame sheet and the droplet's surface, AlO reacts with itself to produce a gaseous suboxide Al_2O and liquid alumina Al_2O_3 . The liquid alumina is assumed to be deposited on the oxide cap, which exists from ignition while Al_2O diffuses towards the primary flame sheet. The gaseous oxides Al_2O , Al_2O_2 and the share of AlO that is not diffusing to the droplet's surface, are diffusing towards the condensation sheet, where additional oxidation occurs and liquid alumina Al_2O_3 is formed.

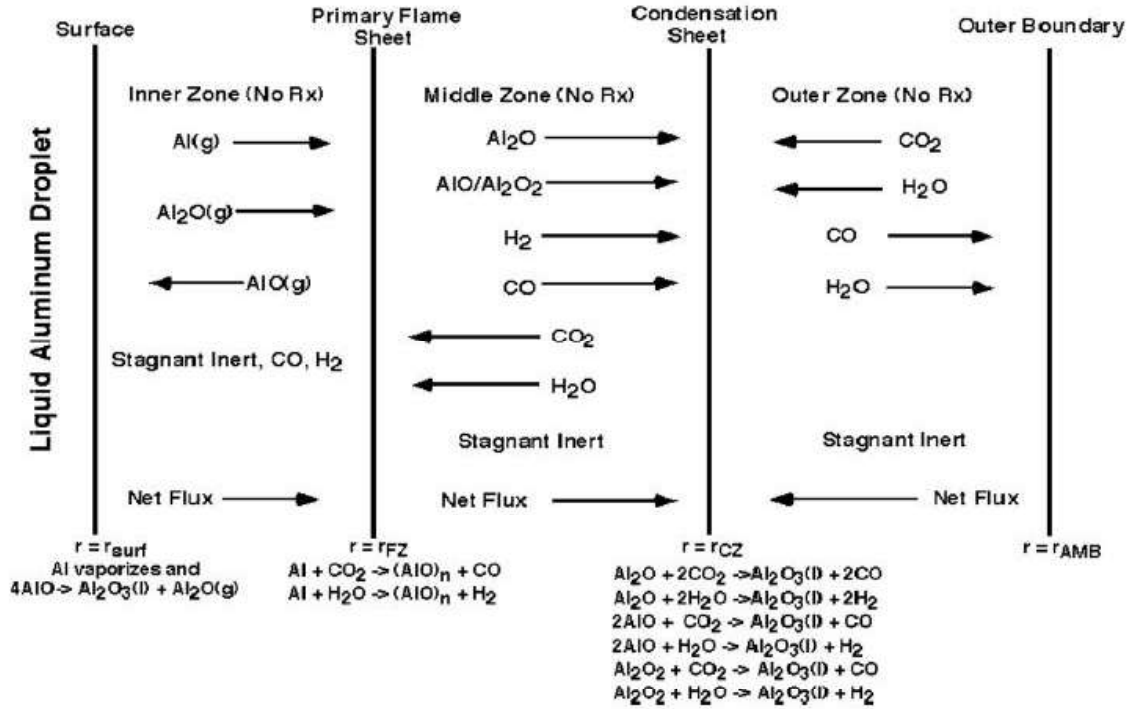


Figure 3.5: Structure of a burning aluminum droplet, according to Ref. [48].

King's work illustrates a more complex structure of aluminum droplet combustion, but relies on heavy assumptions such as the directions of the flows, or the absence of reactions between the various defined reactive sheets. Alternatively, the oxidation mechanisms of aluminum have been developed [49, 50], introducing numerous pathways for the generation of aluminum suboxides and alumina. Prevailing pathways are identified, but the presence of other suboxides than AlO has rarely been observed experimentally. Ultimately, accurate knowledge on the behavior of gaseous suboxides is lacking. It has, however, been evidenced

that gaseous suboxides are formed prior to alumina formation that results from an additional oxidation process. It thus comes that although alumina forms towards the outer edges of the system, the oxidizer diffuses through the alumina formation region to react with aluminum vapor, producing the gaseous aluminum suboxides.

Alumina - formation and transport

Alumina formation and condensation are the most exothermic reaction process and provide enough energy to maintain aluminum evaporation and thus, vapor phase combustion. Various mechanisms for aluminum combustion can be found in the literature [49, 50] and they attribute the formation of aluminum oxide to the oxidation of an aluminum suboxide, e.g. $Al_2O_2 + O \rightarrow Al_2O_3$ or to the combination of these suboxides, e.g. $AlO + AlO_2 \rightarrow Al_2O_3$. In most simulation procedures, the formed alumina is at first considered as a gaseous species $Al_2O_{3(g)}$ that is then condensed to form liquid alumina $Al_2O_{3(l)}$ along an empirical reaction based on an Arrhenius reaction with an activation energy set to 0 (barrierless reaction), and a high pre-exponential factor, $1 \cdot 10^{15}$ in Ref. [49]. Recent simulation efforts have improved this assumption of condensation along an Arrhenius law by applying the classical nucleation theory (CNT) corrected for alumina generation [51]. With such considerations, gaseous alumina is produced, but condenses through a physical process of nucleation. If alumina gaseous monomers exist at a sufficient concentration, the supersaturation of alumina S reaches values greater than unity. The supersaturation is described as the ratio of alumina partial pressure to the equilibrium vapor pressure of alumina over a flat surface. If supersaturation is greater than unity, alumina monomers regroup to form nuclei as the starting point of condensation. These nuclei, of a few alumina molecules, can then undergo growth and coagulation until the nanometric liquid alumina droplets are formed. The characterization of the produced alumina droplets has been the topic of various studies, as the importance of this condensed phase in the industry has already been mentioned. Mainly, the size of the alumina droplets as well as the concentration of such droplets in the surrounding of the Al particles are parameters of interest. Experimental techniques based on droplet quenching enable a characterization of the surroundings of the Al burning droplets. Bucher [52] and Dreizin [53] used this methodology to evaluate relative concentration profiles of alumina droplets in the cloud. Bucher observed that alumina droplets composing the oxide smoke are typically smaller than 5 microns, and observed droplets diameters in the range of 200 nm to 400 nm in the case of combustion in a CO environment. Glotov [54] used a sampling technique to study the condensed combustion products (CCPs) from solid propellant combustion. Glotov observed a multimodal mass distribution of the collected CCPs, with a first mode in the nanometric scale, corresponding to alumina nucleation. A second mode ranging between 4 and 8 microns is attributed to the condensation of alumina on the burning aluminum droplet as well as to coagulation phenomena, while the third mode of particles from 30 to 100 microns refers to agglomerates of aluminum droplets and alumina droplets. Later studies from Glotov [55] using a similar method with increased accuracy observed the dependence of the CCPs on the size of the burning aluminum droplet, and observed alumina particles with a diameter of a few tens of nanometers for a 4-micron aluminum droplet up to alumina particles with a diameter of about 60 nanometers in the case of a 340-micron aluminum droplet. The structure of the CCPs collected by Glotov form aggregates that are displayed in Figure 3.6.

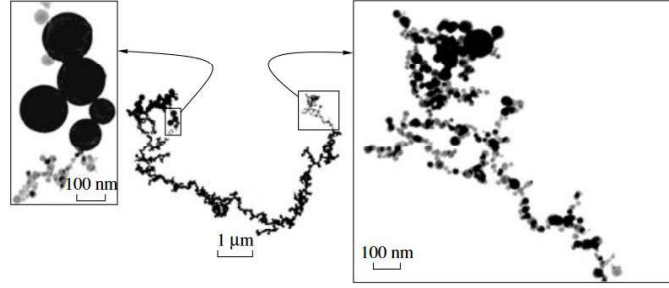


Figure 3.6: Structure of alumina aggregates collected from solid propellant combustion, from [55].

Similarly, Babuk et al. [56] collected the combustion products of aluminized propellants and observed that the CCPs grow due to the deposition of gaseous suboxides on their surface, similar to what is described in the CNT. They also observed particles ranging from 10 nm in diameter to about $1\ \mu\text{m}$. Similar work from similar groups generally observed the same trends and detailed the mechanisms of particle agglomeration [9, 57]. Although these studies provide crucial data to better characterize the combustion of aluminum and the formation process of alumina, the experimental procedures are either intrusive (quenching) or lack in situ spatial resolution (post combustion collection). Also note that most of the introduced studies using a collection method are based on the combustion of a carbonized solid propellant. In the presence of soot, it is possible that the combustion process of aluminum or that the condensation of alumina is altered.

The fact that the aluminum combustion process results in a multiplicity of steps, that can all occur at different locations makes the analysis of the complete system highly complex. The location of alumina production and condensation is usually hard to predict, although recent studies are getting closer to simulating the results obtained by Bucher [46]. Once the alumina is formed as a condensed phase in the vicinity of the burning droplet, it can be transported outside of the main condensation region. Alumina condensation being the most exothermic reaction, it generally occurs at the region of the highest temperature, as the surrounding gases are significantly heated by the reaction. From the aluminum droplet, a flow of gaseous aluminum and gaseous suboxides directed towards the condensation region is pushing the alumina particles away from the droplet. On the other side, a flow of oxidizer towards the reaction zone is retaining the alumina droplets within the cloud. Dreizin [58] assumed that the Stefan flow due to the droplet's evaporation prevails and thus that alumina particles are driven away from the droplet. However, recent work from Gallier et al. [59] proved that, in addition to the Stefan flow and diffusion of the oxidizer, phoretic motions, thermophoresis especially, can impact the transport of alumina particles. Alumina particles are being produced at the highest temperature in the system and are facing strong thermal gradients. Although combustion temperatures will be discussed later, the maximum combustion temperature is often close to 3700 K, while the droplet's surface temperature is often estimated close to 2600 K. This leads to a gradient of more than a thousand kelvins through a few tens of microns. In such thermal gradients, the momentum transfer from the gas molecules to the particle can show a similar gradient, leading to a displacement of the particles towards the cold gases.

Particles can thus undergo retrodiffusion towards the droplet's surface due to thermophoretic motions. Another temperature gradient exists between the condensation region and the ambiance, but it appears to be weaker and is thus less prone to thermophoresis. In the conditions simulated by Ref. [59], the Stefan flow is computed to be of about 12 m/s, while the thermophoretic velocity is calculated to reach up to 30 m/s. The Figure 3.7 schematizes, in a simplified way, the driving mechanisms of alumina particle transport.

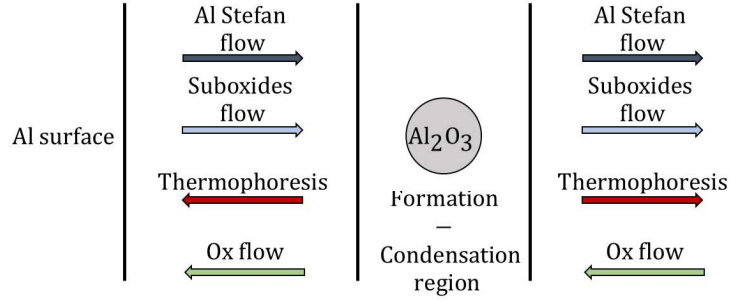


Figure 3.7: Driving mechanisms of alumina droplet transport.

To this day, the accurate behavior of the condensed alumina droplets is not well known. Bucher's experimental results from particle quenching [52] display a wide region in which condensed alumina is observed, from near the surface to distances up to 10 times the droplet's diameter. Similarly, microgravity experiments from Dreizin [53] show the presence of condensed alumina close to the droplet's surface. While most simulations predict the concentration of alumina at the droplet's surface to be zero [47, 60] experimental results are lacking to draw definitive conclusions on the behavior of condensed alumina, evidencing the complexity of the process.

Heterogeneous surface reactions

In order to accurately describe the combustion process of an aluminum droplet, the occurrence of heterogeneous reactions at the droplet's surface must be considered. Although the simplified approach of an aluminum droplet burning in the vapor phase regime neglects surface reactions, Glorian et al. [49] demonstrated that surface reactions can play an important role in the combustion process, as they assessed that at 1 bar in an O_2/Ar 21/79%vol. mixture, about 20% of the aluminum is consumed through heterogeneous reactions in the case of a 100-micron diameter particle. Figure 3.8 displays the importance of surface reactions assessed by Glorian et al. [49].

The occurrence of surface reactions is enabled due to the presence of aluminum suboxides close to the droplet's surface or to the potential diffusion of the oxidizer from the ambiance to the droplet's surface. The existence of these oxygen-containing species very near the droplet's surface enables the liquid aluminum from the surface to react heterogeneously to form alumina, or aluminum suboxides, at the droplet's surface. Glorian et al. [49] mention that the occurrence of surface reactions reduces the burnout time of the aluminum droplet. This can be an indicator that if alumina is produced on the droplet's surface, it may not accumulate on the surface, as this would drastically increase the burnout time, as aluminum

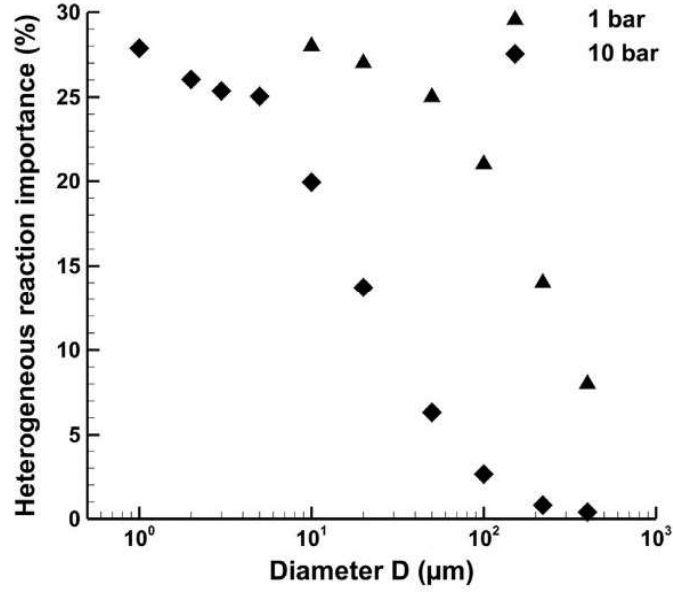


Figure 3.8: Importance of heterogeneous surface reactions, as a function of pressure and droplet size, from [49].

vaporization would be hindered. It is thus possible that surface reactions are a source for gaseous aluminum suboxides or alumina droplet production. From this, it comes that the simplified approach to the burning droplet with a vapor phase regime and a kinetically-limited regime is not accurate enough as Figure 3.8 evidences that, in almost every case, the droplet burns in a transition regime where heterogeneous surface reactions play a role as crucial as the gaseous phase reactions, hence the need for a detailed characterization of the burning droplet. Note, however, that aluminum combustion at high pressures can be expected to approach a perfect vapor phase combustion regime.

Alumina cap

As mentioned during the description of the ignition process of an aluminum droplet, the initial alumina shell gathers to form a liquid oxide cap on the surface of the burning droplet. During the combustion process, it has been observed that the size of the initial oxide cap changes. The definitive evolution of the oxide cap is not well known, as various conclusions can be found in the literature, but the driving mechanisms appear to be highly dependent on the composition of the ambient gases. Some studies observed a constant regression of the oxide cap during the combustion [61], assuming that the liquid aluminum droplet reacts with the liquid alumina cap to form gaseous aluminum oxides. Other studies simulate the growth of the oxide cap during the combustion, observing that alumina droplets can be transported through phoretic motions to the oxide cap [59, 48]. In both cases, the presence of the oxide cap on the surface of the droplet locally prevents aluminum evaporation, leading to a region depleted in aluminum vapor and thus, an asymmetric combustion regime is observed [58]. In a region where the aluminum flow from the surface is lacking, the retrodiffusing motions of

alumina meet no opposite forces and alumina deposition on the existing oxide cap is favored due to phoretic motions, supporting the hypothesis of oxide cap growth. Liu et al. [62] recently simulated the impact of the presence of the oxide cap on the combustion process as well as its growth mechanism. They found that the condensation of gaseous alumina on the oxide cap is the driving mechanism for the oxide cap growth. Its existence results in larger burnout times but barely impacts the aluminum evaporation rate as well as the droplet's temperature. It can also be hypothesized that heterogeneous surface reactions may produce alumina on the droplet's surface, which then migrates towards the oxide cap, participating in its growth. Note that the oxide cap plays a crucial role at the end of the combustion process, described later, due to an increase in the asymmetry of the combustion. The current proposed mechanism of oxide cap growth is illustrated in Figure 3.9.

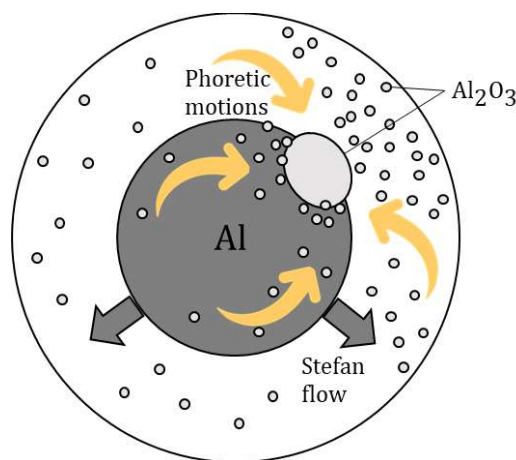


Figure 3.9: Commonly accepted growth mechanism of the oxide cap during aluminum droplet combustion.

With the introduction of the phenomenology of the oxide cap growth, the complete phenomenology of the burning aluminum droplet has been described, starting from the formation of the gaseous suboxides, to the formation of the condensed alumina, its transport, and its role on the alumina cap, while also introducing the role of heterogeneous surface reactions. The following section will describe the temperatures that are at play during the combustion of an aluminum droplet.

3.2.3 Temperatures

The additional oxidation of the aluminum suboxides to yield alumina that condenses upon formation generates a significant amount of energy. A share of this energy is lost heating the surrounding environment, while the remaining energy is transferred to the burning aluminum droplet, maintaining the evaporation process and thus, self-sustaining reaction. It has to be noted that surface reactions and formation of gaseous suboxides are, for the majority, exothermic processes that provide, in the same way, energy to the droplet, to the reaction zone, and to the environment. In order to accurately assess the amount of energy that can be produced by the system, it seems crucial to know the temperatures of the different

components of this system. As a first indicator, King [63] expresses the droplet's surface temperature T_d as a function of the partial pressure of aluminum vapor at the droplet's surface P_s^{Al} :

$$T_d = \frac{34860}{12.537 - \ln(P_s^{Al})} \quad (3.9)$$

The surface temperature of the droplet is displayed as a function of aluminum vapor partial pressure in Figure 3.10.

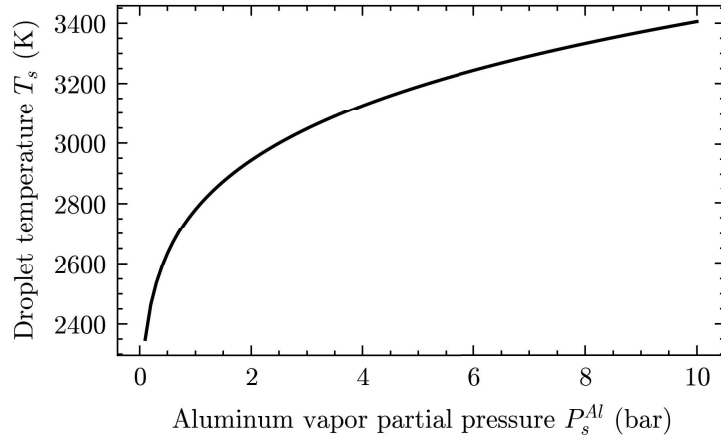


Figure 3.10: Aluminum droplet surface temperature as a function of aluminum vapor pressure on the surface, using Eq. 3.9.

In the case of an aluminum droplet burning in atmospheric air, the aluminum vapor partial pressure is often simulated to be of about 0.3 [64], yielding surface temperatures close to 2500 K. Note that upon increasing the ambient pressure, aluminum vapor pressure will consequently increase and at high pressures, surface temperatures higher than 3000 K can theoretically be obtained.

In the reaction zone, the temperatures are more complex to predict due to the complexity of the aluminum oxidation mechanism and to the spatial occurrence of various reactions. Nonetheless, an estimate of the maximum adiabatic temperature can be obtained through a chemical equilibrium calculation. The CEA (Chemical Equilibrium with Applications) code developed by the NASA can perform such calculations. Assuming liquid aluminum at 1500 K as a fuel and air at 1500 K as an oxidizer, at stoichiometry, the temperature of the chemical equilibrium can be obtained using the CEA code for various pressures and the results are presented in table 3.1

At atmospheric pressure, the adiabatic temperature during aluminum combustion is expected to be close to 3623 K, that is about 1100 K higher than the droplet's surface temperature. In the literature, various methods exist to assess the temperatures at play during aluminum droplet combustion although measuring combustion temperatures is challenging due to the complexity of the system: the aluminum droplet is a condensed phase at a given emissivity, at about 2500 K while alumina is another condensed phase, at a different emissivity, and at a temperature of about 3600 K. That set aside, the gaseous phase has a changing

Table 3.1: Chemical equilibrium temperatures from the CEA code for various pressures, in the case of liquid aluminum in air, both being initially and arbitrarily set at 1500 K

P (bar)	1	2	3	5	10
T (K)	3623	3743	3815	3910	4045

composition as a function of the distance from the droplet’s surface and doesn’t have a constant temperature. The use of integrated methods to measure the temperature of the system is relatively common in the literature, but the results are complex to analyze. Glasziou et al. [65] assessed the average temperature of the system composed of the burning droplet and its surrounding cloud in atmospheric air using a multi-spectral pyrometry method. They measured an average temperature of about 3250 K throughout the entire combustion process. Dreizin [53] used a similar multi-spectral pyrometry method to assess a mean temperature varying from about 3500 K to about 2300 K during the combustion of a 250-micron diameter aluminum droplet in microgravity, in air. The use of integrated methods thus relies on the assumptions that the sum of the light emissions behaves as a light-emitting system at the averaged temperature. Different methods relying on image processing enables the assessment of the temperature of specific objects during the combustion process. Chen et al. [66] used an imaging pyrometry method to assess particle and flame temperatures during combustion in solid propellant plumes. They measured an average particle temperature of 2640 K and flame temperatures ranging between 2300 K and 3000 K. Although droplet temperature is different from the gas phase temperature, the accurate temperature distribution surrounding the droplet is not known. Once again, the groundbreaking work from Bucher [64] offers the first spatial distribution of the temperature surrounding the aluminum droplet burning at atmospheric pressure, displayed in Figure 3.11. This temperature profile has been obtained using an *AlO* PLIF method, and is thus limited to region where *AlO* exists.

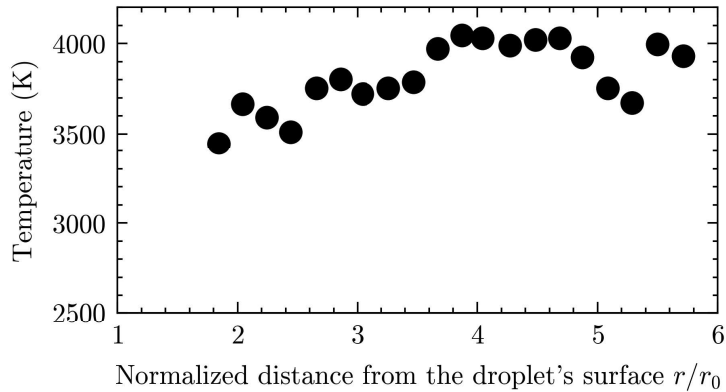


Figure 3.11: Temperature profile around a burning droplet of radius $r_0 = 110\mu m$ in a O_2/Ar (21/79) % vol. mixture, from Ref [64].

The temperature is observed to increase from 3500 K at 2 droplet radii until about 4000 K at 4 droplet radii before decreasing further away from the droplet’s surface. As alumina

condensation is the most exothermic reaction, it is assumed that the hottest region measured by Bucher is also the region where alumina condenses. The experimental apparatus did not allow for assessment of the temperature very near the droplet's surface, but the temperature is expected to reach the theoretical droplet surface temperature, close to 2500 K. This profile provided a first crucial target for simulations and since then, numerous work simulated temperature profiles surrounding a burning aluminum droplet to replicate this profile, such as the simulations from Beckstead et al. showed in Figure 3.12.

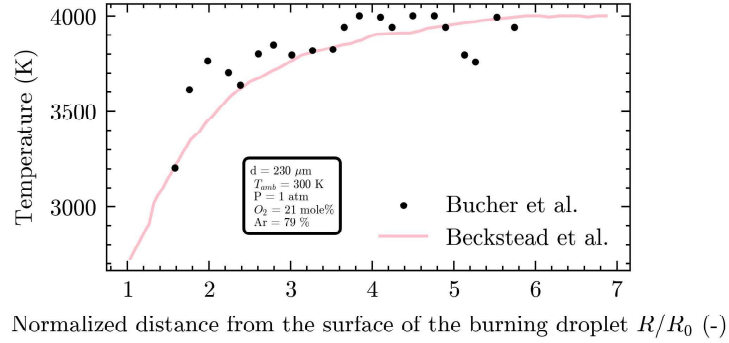


Figure 3.12: Temperature profile simulated by Beckstead to match Bucher's data, from Ref. [47].

Although many simulations efforts from various groups have been made to replicate Bucher's data or similar [60, 63, 46, 67], additional spatially resolved temperature profiles obtained from experiments are very scarce since Bucher's work. Qiu et al. [68] nonetheless recently managed to assess a particle temperature as well as a temperature profile surrounding the droplet, in the case of an aluminum droplet burning in steam, using a RGB pyrometry method.

3.2.4 Temporal aspects

To provide a complete characterization of a burning aluminum droplet, introducing timescales that have been ignored so far seems relevant. It is highly common to find studies about the burn times of aluminum particles. Indeed, for systems based on aluminum combustion, the burn time provides information on the duration for which energy is gained from aluminum combustion, but also it provides information on the completeness of the combustion, in the case of aluminum droplets burning in high velocity flows. In the case of hydrocarbon fuels, the burn time of a droplet corresponds to the duration between ignition and the time at which the totality of the droplet has evaporated and thus reacted. The very well-known d^2 law for hydrocarbon fuels [69] enables the prediction of the complete burn time when the initial size of the droplet is known. The droplet burning rate constant K is expressed as a function of the droplet's diameter d_d :

$$K = -\frac{d(d_d^2)}{dt} \quad (3.10)$$

From Eq. 3.10, the burnout time τ of a hydrocarbon droplet is expressed as a function of the initial droplet diameter $d_{d,0}$ and the burning rate constant:

$$\tau = \frac{d_{d,0}^2}{K} \quad (3.11)$$

Many studies have tried to verify the validity of the d^2 law in the case of aluminum droplet combustion. The work from Gill et al. [70] enables the assessment of micron-sized aluminum particles burn times for various particle sizes in various environments. In their studies, they used particles with diameters ranging between 1 and 20 microns and measured burn times ranging from about 0.5 ms to about 20 ms. Most importantly, they found that upon burning particles in air, the burn time could not be predicted with the d^2 law but observed the following correlation between the burn time and the initial droplet diameter: $\tau = 0.34 \cdot d_{d,0}^{0.95}$. They also compared their experimental results to correlations that have been developed in the literature, as illustrated by Fig. 3.13.

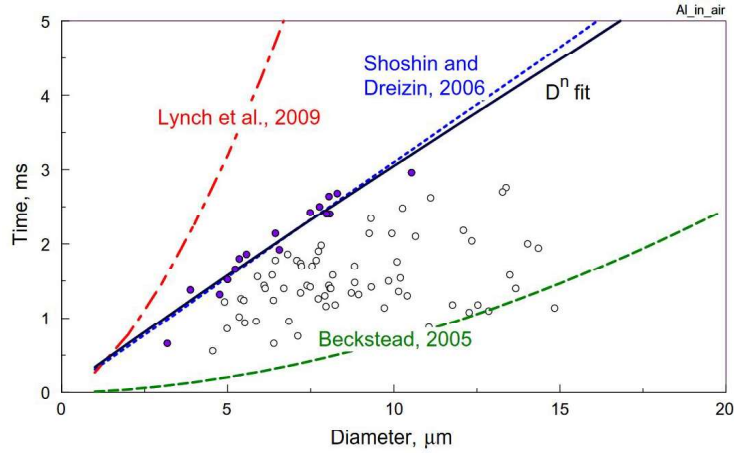


Figure 3.13: Combustion time for aluminum particles burning in air as a function of initial particle size. Comparisons with correlations from the literature are added, from Ref. [70].

Lynch's correlation displayed in Fig. 3.13 from Ref. [71] expresses the burn time as a function of the initial droplet diameter to a power n , but also as a function of pressure P compared to ambient pressure P_0 , oxidizer concentration X_{ox} and correlation constants a_0, a_1, a_2 : $\tau = a_0 X_{ox}^{a_1} \left(\frac{P}{P_0} \right)^{a_2} d_{d,0}^n$. Soshin's correlation from Ref. [72] defines a linear correlation between the burn time and the particle's diameter: $\tau = a \cdot d_{d,0}$. Finally, Beckstead's correlation from Ref. [73] observes a similar correlation to Lynch's correlation, introducing an effective oxidizer concentration X_{eff} , as well as the impact of ambient temperature T_0 : $\tau = \frac{a d_{d,0}^{1.8}}{X_{eff} P^{0.1} T_0^{0.2}}$. From these observations, the combustion of aluminum hardly follows the d^2 law that is defined for hydrocarbon droplets. Echoing the previous sections, the occurrence of surface reactions that are not generally expected for hydrocarbon fuels could be a potential reason for such discrepancies. Indeed, the Equation (3.7) predicts a burn time linearly proportional to the droplet diameter in the kinetically-limited regime. Correlating the burn time to droplet diameters to the power n can be hypothesized to account for surface reactions.

From Figure 3.8, it is expected that the burnout time for small particles approaches some linearity with particle diameter. Inversely, it is expected that for larger particles, the burnout time is proportional to the particle diameter to a power n , close to 2. Furthermore, the various transport mechanisms of the condensed alumina as well as the variety of suboxides make the prediction of the aluminum burn time more complex than in the case of hydrocarbons. In addition, the presence of the oxide cap on the droplet not only affects combustion, but it also impacts the final combustion phenomenology of the droplet, as it will be described further. For all these reasons, predicting the combustion time of an aluminum droplet through a theoretical relation is complex. As a result, most works rely on correlations with external parameters that are fitted to experimental results. Many correlations can be added to the one displayed in the Fig. 3.13, such as previous studies from our group that also intended to develop such correlations for aluminum droplet burn times [8, 74]. The burning rate constant, or evaporation coefficient, that is represented by K in equation Eq. (3.10) is another temporal parameter that can be studied. It has to be noted that this parameter is, however, obtained by considering a d^2 law and its validity can thus be questioned, as the law itself is not generally used. In his PhD work, Braconnier [8] assessed the burning rate constants of aluminum droplets burning in air, as a function of the droplet's initial size and the results are displayed in the Fig. 3.14.

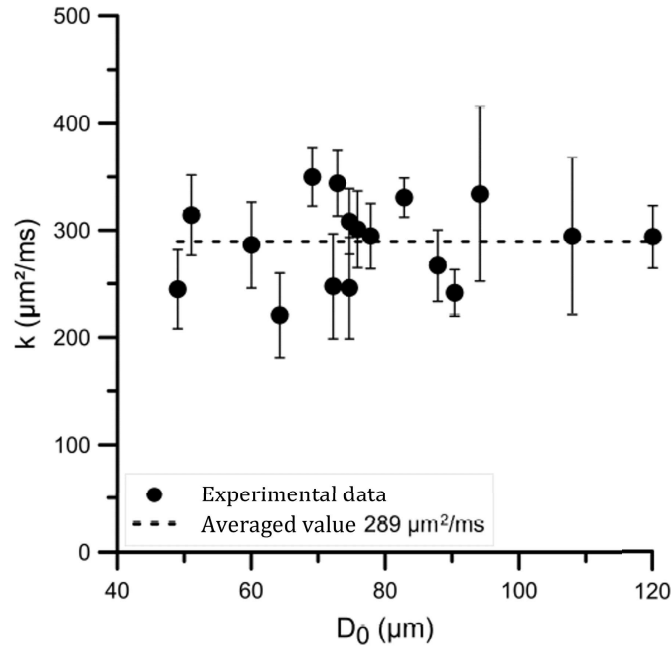


Figure 3.14: Aluminum droplet burning rate constants in air as a function of the initial droplet diameter, from Ref. [8].

Interestingly, the burning rate constant displays low variations to the initial droplet diameter. The burning rate constant shows an averaged value of $289\mu\text{m}^2/\text{ms}$. Although less intuitive than a combustion time and being issued from a law that doesn't appear to be suited to aluminum combustion, burning rate constants enable the characterization of

aluminum consumption from the droplets and thus can give approximations concerning the burnout time. The main conclusion of this section is that the complexity of aluminum droplet combustion leads to the incapacity to theoretically predict the burnout time of a droplet, resulting in the development of many correlations available in the literature. The variety of correlations indicates the high level of complexity of the phenomena occurring during aluminum combustion, as well as the potential influence of the burning conditions. In a simpler way, micrometric aluminum droplet burnout time ranges from below the millisecond to tens of milliseconds, depending on droplet size and burning conditions. Additionally, droplet burning rate constants show fewer variations with initial droplet size and are additional indicators of aluminum consumption rates, although questionable as, based on the d^2 law that doesn't appear to strictly apply to aluminum combustion.

3.3 Extinction phase

The extinction phase is defined as the time at which oxidation reactions stop occurring. Following this instant, the system does not produce energy, and cools down. The description of the cooling phase of the system is not of interest, as it is considered relatively easy to characterize. In this section, the few mechanisms that lead to the extinction phase are described. As detailed in the ignition and combustion phases, an oxide cap, containing the initial alumina shell as well as alumina potentially deposited during combustion, is present on the surface of the burning aluminum droplet. As combustion unfolds, the size of the aluminum droplet decreases due to evaporation, while the growth mechanisms of the oxide cap have been evidenced. The qualitative evolution of the aluminum droplet with its oxide cap during combustion is represented in Figure 3.15. As previously mentioned, the oxide cap locally prevents aluminum evaporation leading to an asymmetric combustion process. The growth of the alumina cap generally leads to an increase in the asymmetry. As observed by Dreizin [58], the asymmetric regime can lead to spinning or jetting of the burning aluminum droplet, as the aluminum flow is uneven and cannot maintain a symmetrical combustion. Figure 3.16 from [75] illustrates the occurrence of the jetting and spinning phenomenon after a symmetric burning phase. Such phenomena can result in high droplet velocities, up to 0.41 m/s, according to Wang et al. [76]. In the event of complete combustion, the totality of the aluminum evaporates during this jetting/spinning phase and a large CCP is generated, as upon total evaporation, the initial oxide cap, grown by condensation and deposition, remains. However, in other cases, as suggested by Braconnier [75], complete combustion does not occur. During jetting and spinning of the droplet, the oxide cap can potentially engulf the totality of the droplet's surface, hindering evaporation and thus combustion, leading to extinction with incomplete combustion. In such cases, the large remaining CCP contains a higher concentration of aluminum when compared to pure alumina. Liu et al. [14] observed such CCPs composed of alumina and unburned aluminum.

Although the oxide cap is a driving parameter of the extinction of burning aluminum droplet, in some conditions, a regression of this oxide cap can be observed. Sarou-Kanian et al. [61] observed this regression and referenced some other experimental evidences of this regression, usually in particular atmospheres. If regression of this oxide cap occurs, resulting in its disappearance from the droplet's surface, aluminum combustion remains symmetric for

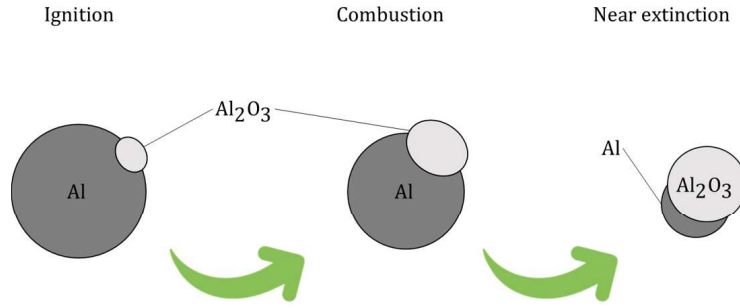


Figure 3.15: Qualitative size evolution of the aluminum droplet with its associated oxide cap during combustion.

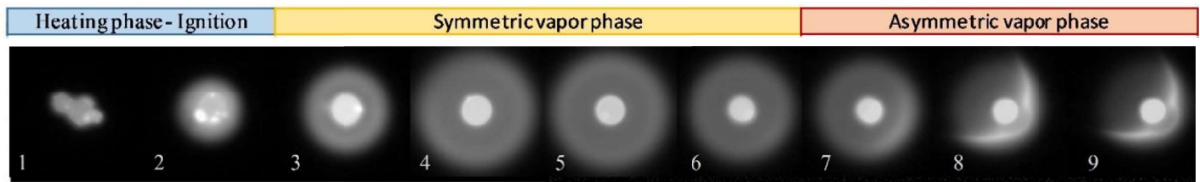


Figure 3.16: Occurrence of a jetting/spinning phenomenon, from [75].

significantly longer times. It is assumed in such cases that the aluminum droplet can either fully evaporate, similar to hydrocarbon droplet combustion, or that upon reaching a critical radius, the reaction region becomes critically close to the droplet and a chaotic combustion regime can be reached.

Finally, in an atmosphere containing nitrogen, another mechanism of droplet extinction can be observed. In the same work, Sarou-Kanian [61] observed the appearance of a solid phase, distinct from the oxide cap, on the surface of the burning droplet, growing until it covers the total surface of the burning droplet, leading to extinction. The sequence observed by Sarou-Kanian evidencing such a process is shown in Figure 3.17. This solid phase, identified as aluminum nitride AlN , quenches the droplet and usually results in incomplete consumption of the aluminum droplet.

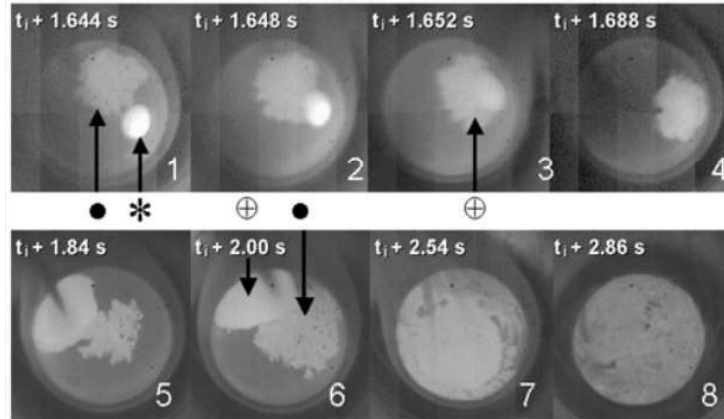


Figure 3.17: Sequence of aluminum droplet combustion in H_2O/N_2 (50/50) % vol. mixture, illustrating the coverage of the droplet by a solid phase, from [61].

With this, a global characterization of the combustion process of a single aluminum droplet has been given. Although briefly mentioned in the previous sections, the external parameters of the combustion, such as ambient pressure or composition, generally have a significant impact on the previously described processes. Thus, the next section highlights the key points of the influence of the external parameters on the combustion processes of a single aluminum droplet.

3.4 Influence of the ambiance

In this section, the influence of the ambient parameters on the combustion phase of a single aluminum droplet is described. The impact of the pressure, the diluent and the oxidizer will be characterized and a brief additional section will describe the impact of an electric field on the combustion.

3.4.1 Pressure

Upon increasing the pressure of the ambiance, the properties of the mixture change, as gas density, viscosity or diffusivity are influenced. First of all, the impact of the pressure on the theoretical droplet and flame temperatures is illustrated by Fig. 3.18 using equation 3.9 for the surface temperature, and the CEA code for the adiabatic flame temperature in air, setting aluminum and air both at 1500 K initially.

When pressure increases, the theoretical adiabatic flame temperature increases, mainly due to a decrease in the specific heat capacity of the equilibrium combustion products, from 21.9 kJ/kg/K at 1 bar to 13.9 kJ/kg/K at 10 bar, from the CEA code results. In conjunction, the droplet temperature increases with pressure. Note that the pressure indicated here acts as the partial pressure of aluminum on the droplet surface. With an increase in the theoretical flame temperature with pressure, the thermal feedback from the flame to the aluminum droplet is expected to increase, leading up to an expected increase in droplet temperature. As mentioned, the droplet temperature being related to aluminum partial pressure at the

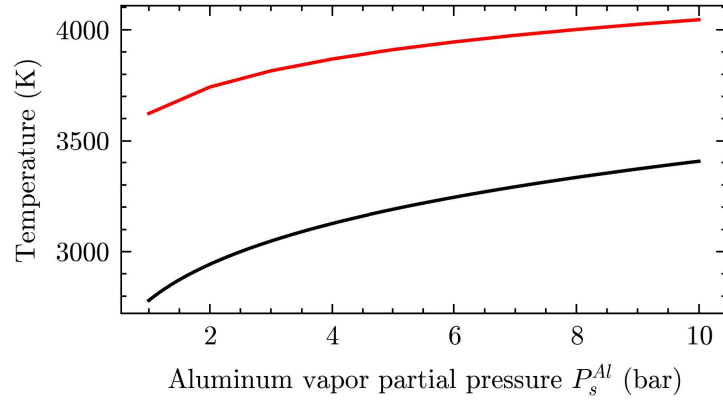


Figure 3.18: Theoretical droplet temperature (black) and adiabatic flame temperature (red) as a function of pressure in air.

droplet's surface, an increase in pressure thus results in an expected increase in aluminum partial pressure, and thus in the burning rate of the droplet, as the vaporization of aluminum is favored. Marion et al [77] studied the impact of air pressure on the burning rate of laser ignited aluminum particles. The resulting burning rate constants are available in Fig. 3.19. When no additional energy is provided to the droplet (laser off), the burning rate constant appears to slightly increase with the pressure, rising from $0.4 \text{ mm}^2/\text{s}$ at atmospheric pressure to about $0.5 \text{ mm}^2/\text{s}$ at 40 bars. This trend is also predicted by Bucher, Beckstead and Braconnier [8, 64, 73].

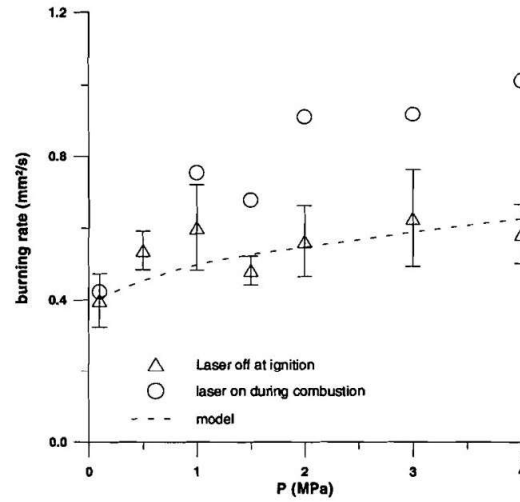


Figure 3.19: Aluminum droplet burning rate constants as a function of air pressure, from [77].

With an increase in the burning rate, a decrease in the combustion time is expected. However, in opposition to the correlation he identified in his PhD work that summarizes the 32 studied ambient conditions, Braconnier [78] identified a slight increase in the combustion time when air pressure increases. The results obtained in his work are presented in Figure 3.20.

The divergence from other correlations identifying that combustion time should decrease with increasing pressure is attributed to the consideration of many experimental data, thus lacking the accurate characterization of specific cases. The recent additional experimental data from Glasziou [74] confirm Braconnier’s observations and determine a correlation predicting a burn time slightly increasing with increasing pressure in the case of combustion with dioxygen as an oxidizer.

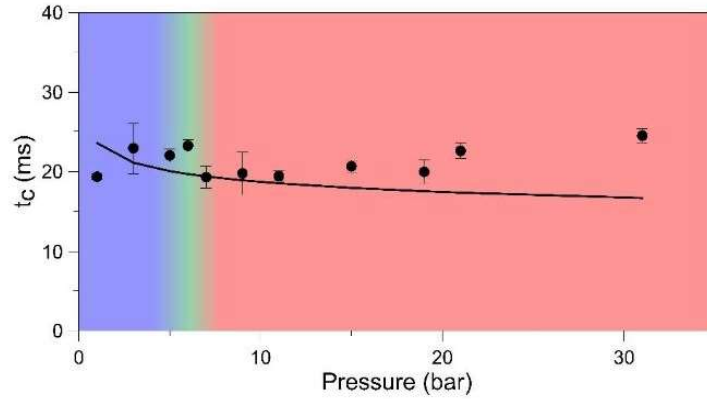


Figure 3.20: Aluminum droplet combustion time as a function of air pressure, from [78].

Note that, quite interestingly, Lynch [71] observes an increase in combustion time with pressure, only for CO_2 and H_2O as oxidizers, while he observes a decrease in combustion time with pressure when O_2 is the oxidizer. The impact of pressure on the combustion time or burning rate seems to be complex to characterize, as evidenced by the discrepancies in experimental results and in the variety in correlations that can be found in the literature. It is highly possible that the experimental conditions (flow, gravity, ambient temperature, ignition process ...) could impact the results, as well as the determination of the burn time. Although the flame temperature rises, the assessment of this impact on combustion time is hardly understood. However, changes in the global structure of the system are easily observed. Braconnier [8] observed that upon increasing the pressure of the medium, the reaction zone, as well as the oxide smoke, move closer to the droplet. The Stand-Off Ratio (SOR) defined as the flame radius r_f normalized by the droplet’s radius r_d thus decreases when pressure increases. Indeed, with low variations in combustion times, the Stefan flow velocity appears to decrease with pressure. Assuming mass flow is conserved, that droplet density ρ_d and droplet regression rate v_d are pressure-independent—since combustion time also seems unaffected by pressure—and that the density of gaseous aluminum ρ_g^{Al} varies linearly with pressure, it follows that the Stefan flow velocity of aluminum, v_g^{Al} , is inversely proportional to pressure:

$$\rho_d \cdot v_d = \rho_g^{Al} \cdot v_g^{Al} \quad (3.12)$$

In opposition, the increase in pressure leads to a greater availability of the oxidizer and the reaction thus settles at shorter distances from the source of aluminum vapor. With sufficiently high pressures (>15 bar), the oxide smoke becomes very dense and settles very near the droplet’s surface, usually leading to chaotic combustion behaviors [78].

When pressure increases, the decrease in Stefan flow velocity results in alumina particles being transported away slower, as pictured in Figure 3.7. Additionally, the decrease in SOR leads to the formation of alumina in a decreasing volume. Alumina thus tends to accumulate in the oxide cloud surrounding the droplet in increasing concentration. This results in denser smoke that can be opaque if the pressure is sufficiently high as illustrated by Figure 3.21. This illustrates that an increase in pressure results in an increase in the concentration of alumina in the droplet's vicinity. Both phenomena of SOR decrease and alumina concentration increase have recently been simulated by Finke et al. [60].

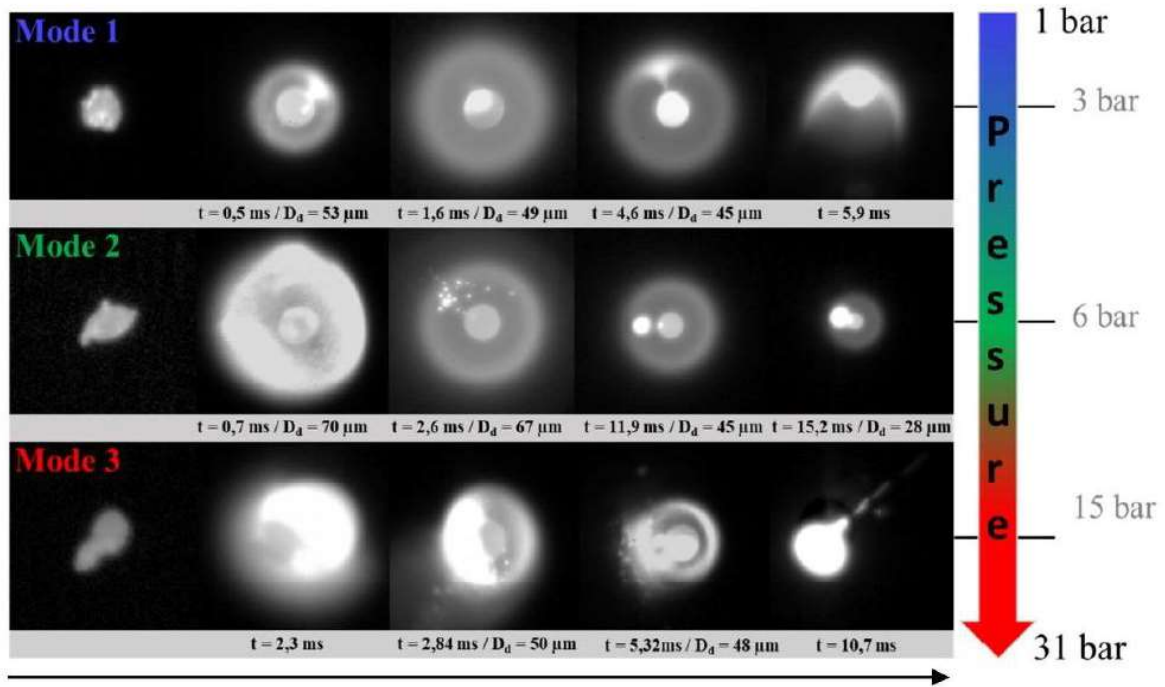


Figure 3.21: Aluminum droplet combustion sequences with increasing air pressure, from [78].

The work from Glotov [54] tackles the impact of pressure on the size of the collected CCPs from aluminized propellant combustion. The increase in pressure appears to favor the growth of the alumina droplets formed during single droplet combustion. Inversely, larger CCPs usually attributed to incomplete combustion, with diameters larger than 60 microns, are smaller when the pressure increases. This is due to the decrease in size of the agglomerates formed during the combustion of the propellant when pressure increases. A similar trend of alumina droplet particle growth with increasing pressure is observed by Babuk et al. [56].

An increase in temperature, alumina droplet concentration and an increase in alumina droplet size that can result in a change in emissivity [79] generally results, according to Planck's law, in a brighter system, as light emission is increased and thus to greater radiative heat losses from the system.

Table 3.2: Chemical equilibrium temperatures from the CEA code for various diluents with 21 % vol. O_2 , in the case of liquid aluminum and oxidizer, both at 1500 K and 1 bar.

Diluent	N_2	Ar	He
T (K)	3623	3666	3666

3.4.2 Diluent

It has been evidenced that the ambient pressure can have a drastic influence on the combustion process of an aluminum droplet. Nonetheless, the composition of the mixture also has to be accounted for. Indeed, various diluents with various thermodynamic properties can be used such as helium, nitrogen, or argon. The influence of these gases as well as their concentration on the combustion of a single aluminum droplet is briefly described.

The influence of nitrogen on aluminum combustion is generally significant, as it is not an inert diluent. Indeed, it is well known that in high temperature combustion processes, nitrogen oxides can be produced. Additionally, the formation of aluminum nitrides and aluminum oxynitrides has been detected [45, 58] and is hypothesized to cause the transition to the asymmetric combustion regime. The formation of aluminum nitride evidenced after quenching has also been described earlier. Bucher [52] also captures light intensity oscillations in the presence of nitrogen that are not observed in argon experiments, evidencing the asymmetric combustion regime. Other work from Bucher [45] measures greater AlO concentrations and higher temperatures in an O_2/Ar (21/79) % vol. mixture than in air. Bucher [45] attributes the decrease in temperature in the case of air to the formation of AlN that is endothermic and thus creates a heat sink in the reaction zone. The concentration of condensed species containing aluminum is, however, measured to be greater in the case of air. A similar trend is observed for the temperature using the CEA code, predicting an adiabatic temperature of 3623 K for air compared to 3666 K for argon, at atmospheric pressure. The results from the CEA code are summarized in Table 3.2. The decrease in AlO concentration in air can be similarly attributed to the decrease in aluminum and oxygen availability due to AlN and $AlNO$ formation.

A very thorough study by Braconnier [75] references the influences of the diluents on the combustion parameters. It is observed that at similar oxygen concentrations and similar droplet sizes, the combustion time is hardly impacted by the nature of the diluent, but an increase in oxygen concentration significantly decreases the combustion time and increases the droplet burning rate constants. Note, however, that the relation between combustion time and droplet diameter in helium and argon differs from nitrogen. This is attributed to the oxide cap growth mechanism. It is hypothesized that argon and helium hinder the diffusion of the formed oxide towards the oxide cap. These results are summarized in Figure 3.22.

Given the low differences in the predicted adiabatic flame temperatures from Table 3.2, similar burning rates are expected, as the thermal feedback from the flame drives the evaporation of the burning droplet. However, it is interesting to note that although combustion time and temperatures are relatively close, the use of helium as a diluent causes a significant change in the structure of the system due to its light molecular mass. Indeed, using helium as a diluent favors the diffusion of species and the oxidizer is more efficiently

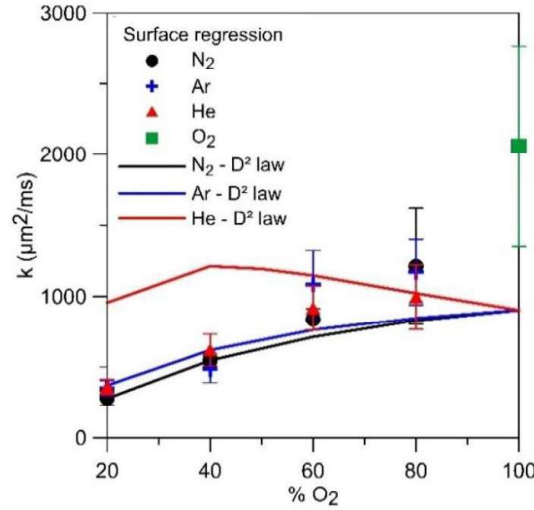


Figure 3.22: Aluminum droplet burning rate constants as a function of diluent nature and concentration, from [75].

transported to the reaction zone. In the same way as when pressure increases, the use of helium results in a decrease in the SOR, from about 4 with argon or nitrogen, to about 2. Similarly, the oxide cloud appears less dense in the case of helium than in argon and nitrogen, suggesting a potential fast diffusion of the alumina in the ambiance if the CCPs production rate is considered identical to all diluents. Note, however, that the impact of the diluent on the CCPs characteristics has not been fully studied yet.

3.4.3 Oxidizer

Although aluminum combustion in air is the most fundamental case to study, in the solid propulsion field, aluminum generally burns within a gaseous mixture of complex composition. In opposition to air, in solid propellants, dioxygen is absent and species such as CO_2 , H_2O or CO are the main oxidizers that can entertain a combustion process. The influence of a few oxidizers will be briefly characterized in this section.

In its burn time correlation, Beckstead [73] defines relative oxidizing efficiencies. Dioxygen is considered as the oxidizer of reference as it is more prone to react with aluminum than CO_2 for example, for which bonds between carbon and oxygen atoms must be broken to enable oxidation of the aluminum. Beckstead measured an efficiency of 0.6 for H_2O and an efficiency of 0.22 for CO_2 . This means that, upon using similar conditions, replacing dioxygen with carbon dioxide should multiply the burn time of the droplet by a factor of about 5. Braconnier observes the same trend in his PhD work [8] adding the efficiency of carbon monoxide assessed at 0.028 and Glasziou observes identical trends but with different efficiencies [74]. However, the same problem encountered when introducing burn time correlations arises. The discrepancies between correlations make the characterization of the influence of the oxidizer on the burn time complex.

To provide phenomenological insights, Bucher [52] quenched aluminum droplets burning in O_2 , N_2O , CO_2 , H_2O and CO atmospheres. He observes that in a CO atmosphere, the droplet

burns in a kinetically limited regime in opposition to all other atmospheres where vapor phase combustion prevails. The temperature developed by the combustion of aluminum in carbon monoxide is thus believed to be below the vaporization temperature of the aluminum droplet. This behavior is confirmed by Braconnier's PhD work. In N_2O atmospheres, the brightest combustion is observed, with the greatest amount of condensed phase measured after quenching. This relates to the previous section, as the presence of nitrogen possibly enables the formation of aluminum nitrides and aluminum oxynitrides as condensed species. The flame developed is, however, similar to the flame observed with air, with the addition of larger CCPs in the flame. In CO_2 atmospheres, a flame develops very near the droplet's surface with a SOR of about 2, favoring the mass transfer between the oxide cap and the oxide droplets located in the cloud. A decrease in the SOR is theoretically expected, as Beckstead [47] simulates a flame temperature of less than 3000 K in a CO_2/Ar 21/79 % vol. mixture. An asymmetric combustion regime is thus reached at the early stages of the combustion. A higher concentration of CCPs than in air or argon is observed by Bucher, but this can be attributed, similarly as for nitrogen, to the possible presence of aluminum carbides as a condensed phase.

Finally, aluminum combustion in water vapor is at the core of various studies, as a peculiar combustion regime is usually observed. This regime is described by Halter et al. [80] and illustrated by Figure 3.23

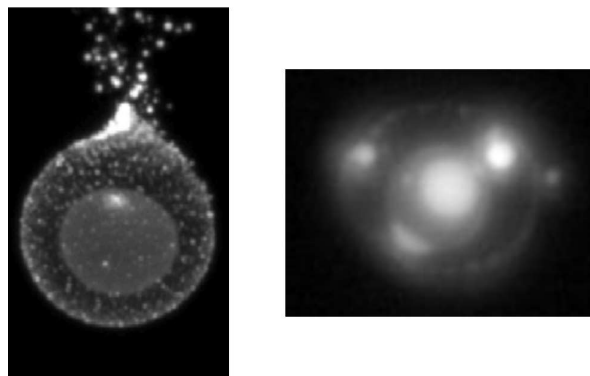


Figure 3.23: Aluminum droplet combustion images in pure water vapor, extracted from [81] (left) and [80] (right).

In steam, the alumina produced during combustion forms a 'crown' surrounding the burning aluminum droplets, and is composed of alumina droplets much larger, as they can be distinctly visualized, than the expected nanometric droplets observed in other atmospheres. Particles located in this crown can regroup to form even larger droplets called 'satellites' that remain in the vicinity of the burning droplet instead of diffusing away as expected. Although curious, this peculiar regime will not be further described, as it is not of prime interest in the context of this work, and articles available in the literature provide more detailed descriptions [81, 80, 68]. In the same way as for diluents, the accurate characterization of the CCPs for various oxidizers has yet to be worked on.

3.4.4 Electric field

While the influence of the most evident parameters that define an ambiance has been characterized, the influence of an electric field on the combustion process is less obvious but needs to be questioned. Indeed, as described in the following chapter, aluminum droplet combustion is studied in the present work by levitating a particle using electrical fields. A brief review of the known influences of the electrical field on combustion processes is thus necessary.

In order to levitate an aluminum droplet, an initial charge is given to it. In 1991, Bar-Ziv and Saforim provided an extensive study of electrodynamic levitators as a tool to study materials at high temperatures [82]. In their work, they mention that the initial charges given to the material are lost in short times when it is heated due to thermoionic emissions, as summarized in Figure 3.24. Indeed, this figure shows that the characteristic time for charge losses decreases exponentially when temperature increases, with increasing time for materials presenting elevated work functions. Note that the work function of aluminum is assessed at 4.08 eV [83]. Given the characteristic times of catastrophic charge losses, the aluminum particle is expected to become neutrally charged during its heating phase, prior to ignition.

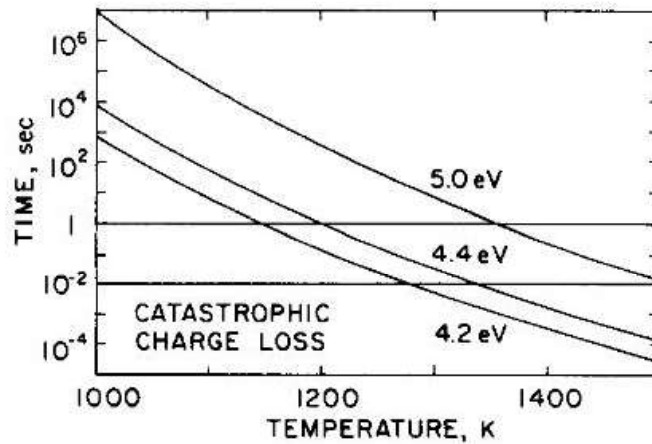


Figure 3.24: Characteristic charge loss times as a function of temperature for different material work functions, from [82] .

During the combustion of a fuel, in the reaction zone, ions and free electrons are generated [84]. It is well known that under the influence of an electric field that can basically be considered as an electronic wind, charged species can undergo a behavioral change due to external forces. It thus comes out that applying an electric field to a flame can impact the characteristics of the combustion. Let us assume that the reaction region of a flame is placed between a negatively charged and a positively charged electrode. The positive species produced in the reaction zone are thus expected to move towards the electrode of negative charge, while electrons are displaced towards the positive electrode, potentially impacting the flame's shape and thus heat release rates. Figure 3.25 schematizes the expected influence of an electric field on a flame.

In the field of gaseous fuel combustion, it is known that the presence of an electric field

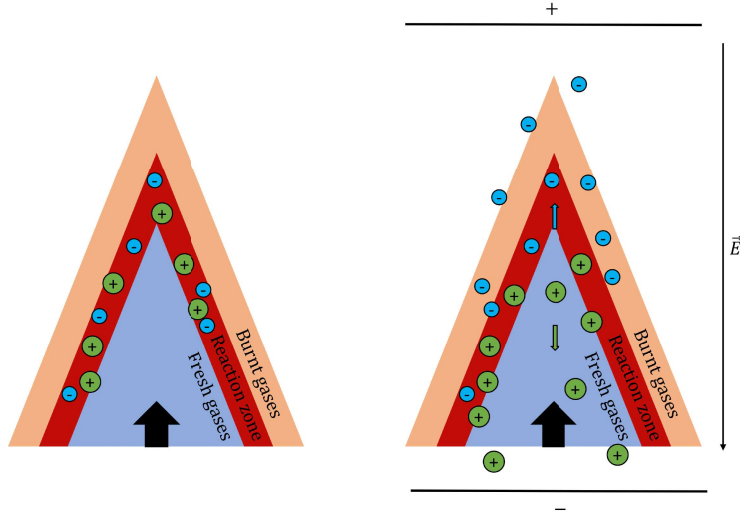


Figure 3.25: Schematics of the influence of an electric field on the charged species produced in the reaction zone of a flame.

can help in flame stabilization, alter soot production, or change the flame shape. If this transposes to metal combustion, it is crucial to assess in what ways. Dreizin [85] studied the influence of alternative and constant electric fields on single aluminum droplet combustion. His first observation is that in the presence of a constant electric field (500 to 2000 kV/cm), the burning droplets were attracted to the negatively charged electrode, indicating that the droplets are positively charged. Then, with the application of alternative electric fields with similar amplitudes, a decrease in the combustion time is observed with a decrease in droplet temperature. Later work from Yagodnikov et al [86] observed the influence of the electric field on the combustion of aluminum dust. They compared results obtained in classical conditions to results obtained applying a 0.9 kV negative potential to the Bunsen burner's nozzle. They observe a change in the shape of the flame upon applying the electric field, attributed to the influenced motion of aluminum and aluminum oxide particles. They hypothesized that during combustion in an electric field, ions and electrons are chaotically transported to the produced alumina particles, and attached to their surface, leading to the formation of charged oxide droplets.

More recent works from Altman et al. [87] and Tran et al. [88] significantly helped improve the fundamental understanding of metal combustion, and associated the generation of nano-oxide with an electron-emission process. Altman observes that the electronic emission from nano-oxides is a driving parameter of the energy accommodation coefficient (EAC), thus driving the heat transfer between the gas phase and nano-oxides. Such phenomena can impact the growth mechanisms of the oxides. Indeed, he observes a change in the morphology of the collected CCPs of magnesium combustion when an ion source is directed towards the flame. As an additional consequence of electronic emissions, Tran observes that the excited electrons can emit light and radiate, thus playing a role in the overall energy balance of the combustion process.

Although the recent studies from Altman and Tran provide crucial insights on the

potential influence of an electric field on metal combustion, the consequences of an electric field generated by the levitator on aluminum combustion cannot be accurately predicted. A short set of experiments available in Appendix A nonetheless provides some insights on the influence of the electrodes on the combustion process.

3.5 Conclusion

This chapter provides a wide characterization of the combustion process of a single aluminum particle, from ignition to extinction, and the main conclusions are summarized in the following list.

- Large aluminum particles, typically with a diameter greater than 10 microns, ignite upon melting of the oxide shell, at 2326 K. Lower ignition temperatures are reached for smaller particles due to cracks in the oxide shell that enable diffusion of the oxidizer to the Al core.
- A simplistic approach to Al combustion consists of assuming that small particles (<10 microns) burn in a kinetically-limited regime, with the reactions occurring at the surface of the droplet. Larger particles burn in a diffusion-limited regime, with a flame establishing detached from the droplet's surface. The occurrence of a transition regime is observed and a more detailed characterization is required.
- Aluminum suboxides such as AlO , Al_2O or AlO_2 are identified as crucial reaction intermediates that are formed prior to the alumina.
- Upon formation, alumina immediately condenses to form CCPs of sizes typically ranging from 10 nm to 400 nm according to the burning conditions. These CCPs can either diffuse away from the system, mainly due to the Stefan flow, or diffuse towards the droplet, mainly due to thermophoresis. The latter can result in the accumulation of the oxide on the oxide cap, leading to its growth.
- Heterogeneous surface reactions are evidenced to play a crucial role during aluminum particle combustion, even for particles with diameters up to 100 microns.
- In atmospheric air, the droplet's surface temperature is expected at about 2500 K, while the flame temperature is expected close to 3600 K.
- Al particles don't strictly follow the d^2 law. Many studies evaluated particle diameter, oxidizer composition, pressure and temperature as first-order parameters to predict the combustion time of an aluminum particle.
- The growth of the oxide cap generally leads to the jetting and spinning of the aluminum droplet, usually leading to extinction. In the presence of nitrogen in the diluent gas, the formation of AlN as a condensed phase on the droplet's surface was observed, resulting in quenching.

- An increase in pressure results in a decrease in SOR, in an increase in alumina concentration, and in higher flame temperatures. Pressure has a weak influence on combustion times.
- Adiabatic flame temperatures and combustion times are expected to undergo weak variations when the diluent varies between N_2 , Ar or He , although oxide cap growth is favored by the presence of N_2 , and the SOR decreases when He is used.
- Combustion is significantly impacted by the nature of the oxidizer. Oxidizing efficiencies are measured lower for steam and carbon dioxide. In addition, the presence of nitrogen or carbon is hypothesized to result in AlN , $AlNO$ or AlC formation as CCPs.
- Placing a burning aluminum particle within an electric field is expected to slightly influence the CCPs formation process.

Although the global understanding of aluminum particle combustion has significantly improved in recent years, what stands out from this introduction, hopefully, is that, apart from Bucher's results, very scarce spatially resolved experimental data is available in the literature. There are few, if any, temperature or species concentration profiles that surround the burning aluminum droplet can be found, let alone for various burning conditions. The lack of spatially resolved data hinders the validation of the numerous simulations that are available in the literature. This PhD work thus aims at providing such data. To meet this need, an experimental device previously used by Braconnier and Glasziou [8, 74] has been improved and will be fully described in the next chapter.

Chapter 4

Experimental Setup

In this chapter, the experimental setup that has been used throughout this work is entirely introduced. Firstly, the experimental devices available in the literature that enable the study of single aluminum particle combustion are introduced, followed by a general overview of the experimental setup developed at ICARE-CNRS. Then, a detailed description of the electrodynamic levitator is provided, followed by a description of the associated optical diagnostics. The ignition system is detailed prior to concluding this chapter.

4.1 Single particle experiments in the literature

In the literature, the experimental study of aluminum combustion is divided into two areas. The first one, not described in this work, relies on aluminum dust flame studies. The combustion process of a single aluminum droplet is set aside and aluminized flames are studied instead, providing less fundamental data, but more generalized and applied results of aluminum combustion. The second area, which is the core of this work is single aluminum droplet combustion. In this domain, the combustion of a unique droplet is studied to collect fundamental data, improving our understanding of aluminum combustion at the smallest scale. In this section, a few experimental devices that supported studies available in the literature are presented.

The most straightforward experimental device to study single droplet combustion is to set a single particle on a plate and proceed to its ignition. Xu et al. [89] used this device to study single particle ignition. A single particle is isolated on the bottom's surface of a combustor, and a laser ignites the particle while a high-speed camera and a three-color pyrometer monitor the combustion process. Although straightforward, the combustion takes place in contact with a surface, which significantly alters the combustion process when compared to aluminum droplets combustion in solid-propellant rocket motors. To prevent contact between a surface and the particle, various systems have been developed. Wilson et al. [37] attached an aluminum droplet to the tip of a glass fiber by adhesive electrostatic forces. A laser is used to heat the particle to its ignition temperature. Upon ignition, the droplet detaches from the fiber that melts at laser impact, and burns in a free-fall regime while a high speed camera observes the process. Instead of using a support for the particle, Macek et al. [90] injected single particles into the hot gas stream generated by a burner. The

particle ignites due to the hot environment, and aluminum combustion unfolds as the droplet follows the gas stream.

Some other studies are based on the principle of heating an aluminum wire until its melting point, generating an aluminum droplet that ignites when the required temperature is reached. Generally, the formed droplet gains momentum in the early stages of its free-fall, prior to ignition, and thus has a non-zero velocity when combustion is initiated. Bucher [64] uses a CO_2 laser to heat the aluminum wire, located at the top of a combustion chamber. The particle thus detaches from the wire and ignites. The free-fall of the burning droplet is observed with a CCD camera. Similarly, Dreizin [58] uses a micro-arc electrode to melt the aluminum wire and observes the free-fall of the droplet until its quenching on a plate. A pyrometer and a camera were used to record the combustion process. Finally, Wu et al. [81] use a hydrogen flame to heat the wire. The detached droplet thus follows the gaseous stream from the flame, instead of falling, and burns in the combustion products of the hydrogen flame, mainly steam. Two cameras coupled with a blue laser enable simultaneous observation of emission and laser absorption from the system. These experimental devices enable highly repeatable experiments in the desired gas compositions, but result in aluminum droplets being observed in a forced flow or in free-fall. Although aluminum droplets burn in a low velocity flow in SRM, in the vicinity of the propellant's surface, the presence of this flow complicates the study of the fundamental combustion characteristics due to enhanced convective effects. To avoid flow or free-fall effects, Liang et. al. [91] use an ultrasonic levitator to stabilize a nanometric aluminum droplet in a controlled environment, without contact with any surface. A coil is used to ignite the particle, which is then observed by a high-speed camera, a spectrometer and an infrared thermometer. It is, however, known that acoustic waves can interact with the heat release rate, as it occurs during thermo-acoustic instabilities. A similar experimental setup, using an aerodynamic levitator, was also used by Sarou-Kanian [61].

There is a wide variety of experimental devices available in the literature that enable the study of single aluminum particle combustion. However, most of these devices rely on a physical contact between a particle and a surface, on the presence of a flow, or on the observation of a droplet during its free fall, making the access to fundamental data a challenge. Furthermore, the assessment of locally resolved data is crucial to help improve current simulation models, which lacks in current experiments because droplet motion hampers the use of significant magnifications. To provide an order of magnitude of current resolutions, data from Wu et al [81] are extracted with a resolution of about 8 microns per pixel. The use of an electrodynamic levitator that enables the ignition and combustion of a stabilized aluminum droplet, without flow effects, acoustic perturbations, and free-falling effects, along with high-resolution observations (less than 2 microns per pixels) thus appears highly beneficial. The next section provides a general overview of this experimental device before going into more details in further sections.

4.2 General overview

The results that will be presented in the present work have been acquired using an electrodynamic levitator. The levitator itself, detailed in the next section, is composed of three

electrodes generating a stable position for a charged aluminum particle. The levitator is located in a combustion chamber that enables the use of a controlled atmosphere, both in composition and pressure. This cylindrical chamber is made of aluminum alloy AlCu_4MgSi , and is hermetically sealed at both ends. The chamber is designed to operate at pressures of up to 120 bar, although no experiments have ever been performed at pressures beyond 50 bars. A vacuum pump, connected at the bottom of the chamber, is used to remove the gas from the chamber, enabling a precise filling of the desired mixture. At the same location, three independent high-pressure gas lines are used to fill the chamber. The use of a numerical pressure gauge (0-4 bar, 1 mbar $\pm 0.1\%$; 0-31 bar, 10 mbar $\pm 0.1\%$) enables a control of the pressure within the chamber. With the partial pressure law and micrometric valves, the composition of the mixture of the chamber can be accurately controlled during the filling of the chamber. The power supply of the three electrodes is also connected at the bottom of the combustion chamber.

The top electrode of the levitator is drilled to allow the injection of aluminum particles into the levitator. A compensated piston is mounted on the top cover, connected to a PEEK injector that contains aluminum particles, aligned with the electrode's drilled injection port. This system enables the injection of aluminum particles while maintaining the chamber hermetic.

The combustion chamber is equipped with four optical accesses, equally distributed on its perimeter, to enable particle ignition and the use of optical diagnostics. The two optical accesses on the camera's axis feature windows in barium fluoride (BaF_2) that provide a complete transmission in the visible spectrum. These two optical ports enable the monitoring of the combustion process with the use of a high-speed camera coupled to a backlight system as well as a multi-wavelength pyrometer. The two other accesses, aligned with the laser's trajectory, feature windows made of zinc selenide (ZnSe) that favor the transmission of infrared emissions. These latter windows are coupled to focal lenses $f = 50 \text{ mm}$ that enable the focusing of the laser on the levitating aluminum particle. The experimental setup is presented in Figure 4.1.

4.3 The electrodynamic levitator

In this section, the principle of the electrodynamic levitator is briefly introduced. Initially developed as a balance [92, 93], the electrodynamic levitator is a device that enables the stabilization of a charged particle using a set of three electrodes [82]. The principle of levitation relies on applying a contactless force to an object in order to compensate for its weight. In the case of a charged particle, an electric field generated by two planar electrodes enables for the compensation of its weight. For the current experimental setup, aluminum particles are charged using a plastic syringe, via the triboelectric effect. The triboelectric effect is complex to characterize, as it depends on the materials or their interaction, but it appears from our experiments that aluminum particles become negatively charged upon friction with the syringe. The generation of a constant electric field, with the positive electrode located above the negative electrode, would cause the particle located in its center to levitate if this electric field is well tuned. In the event of a weak electric field, the weight of the particle will prevail and the particle will fall. On the opposite, an excessively strong electric field

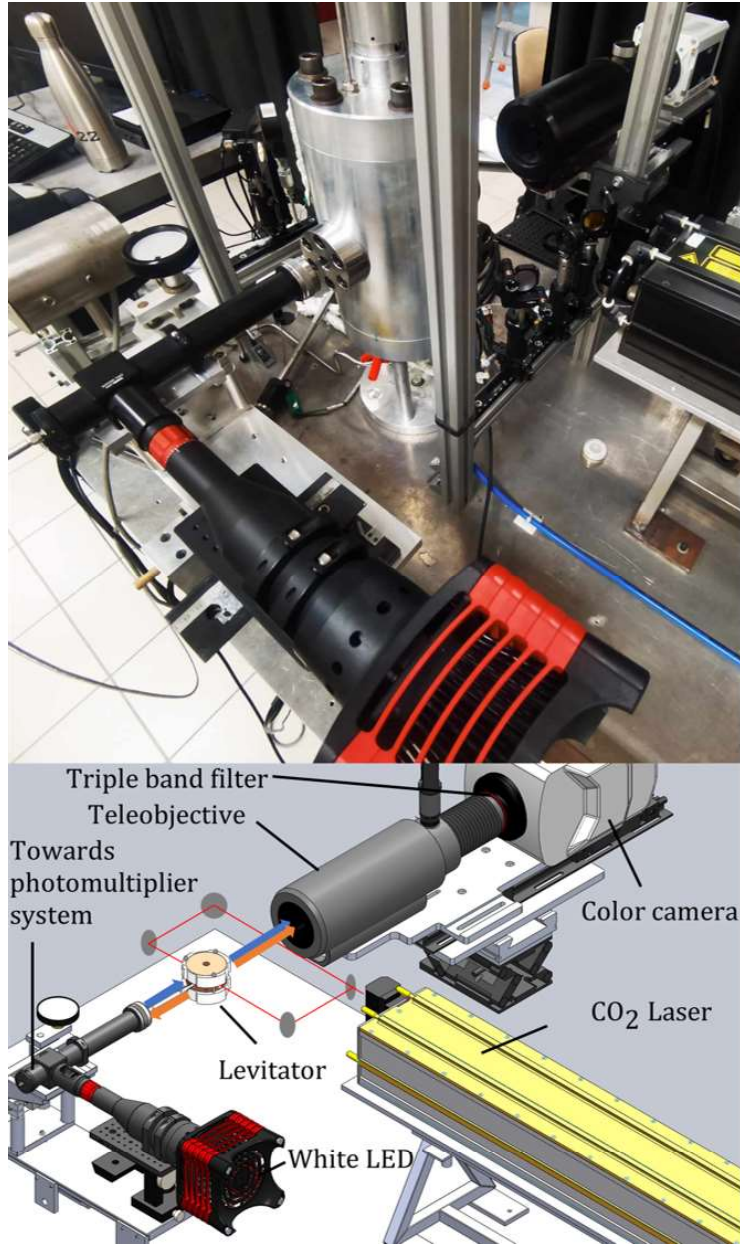


Figure 4.1: Photography of the experimental setup (Top) and simplified drawing of the experimental setup (Bottom). Red lines illustrate the laser path. Blue arrows represent LED emissions. Orange arrows represent combustion emissions.

will overcompensate the particle's weight and the particle will rise to stick to the positive electrode, undergo a discharge and then fall due to its newly acquired neutral state. The levitation of the particles thus needs a fine-tuning of these two planar electrodes.

With a fine-tuning of the electrodes, the weight of the particle is well compensated, but the particle generally isn't stable, as horizontal motion is still allowed and the location of the particle isn't fully constrained. To ensure a stable location for the particle, a third circular

electrode is required to force the particle into a defined location. Note that generally, stability cannot be obtained with the sole use of constant fields; an alternating electric field is required and is ensured by this ring electrode. Figure 4.2 summarizes the structure of the levitator. The two planar electrodes are designated as weight compensated electrodes, while the circular electrode is designated as the centering electrode. The aluminum droplet is represented, not at scale, along with the injection and optical accesses that enable data acquisition.

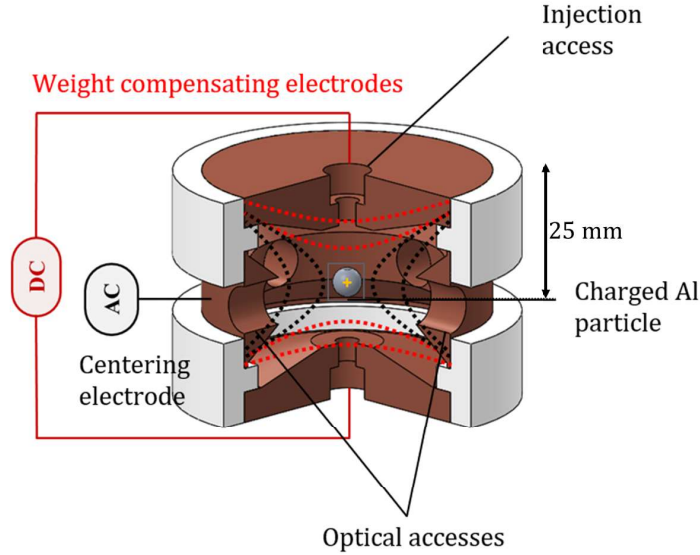


Figure 4.2: Schematized representation of the electrodynamic levitator.

In order to levitate an aluminum particle of about 70 microns in diameter, generally, an electric potential of about -400 V is applied to the negative bottom electrode, 400 V is applied to the positive top electrode, and a potential alternating between 1600 V and 0 V is applied to the circular electrode, usually at frequencies of about 15 Hz. A derivation, largely inspired by the work of Frickel [94], of the resulting electric field generated by this set of three electrodes is available in appendix A. The figure 4.3 represents the two shapes between which the electric potential generated by the circular electrode alternates. In the figure on the left, the particle is located at the center of the chamber, at a radial coordinate $R = 0$ and height $Z = 0$. The particle is negatively charged. It is attracted to the maximum potentials and thus, towards greater absolute heights while remaining at the same radial coordinate $R = 0$. If this field was to be constant, the potential on the left figure would cause the particle to slide away, as the location of the particle is not stable. The potential thus alternates with the shape of the right figure. In this figure, the height now becomes stable, as the maximum potential is obtained at a height $Z = 0$ but the radial location of the particle is unstable. Upon alternating at sufficiently high frequencies, the changes in shape of the potential don't provide enough time for the particle to fall due to the unstable location and is thus locked at the center of the levitator. Stable levitation is thus reached by alternating sufficiently fast between unstable levitation states.

A study of the stability of the system is available in the work of Frickel et al. [94]. Note also that Maxwell's laws of electromagnetism predict the presence of a magnetic field induced

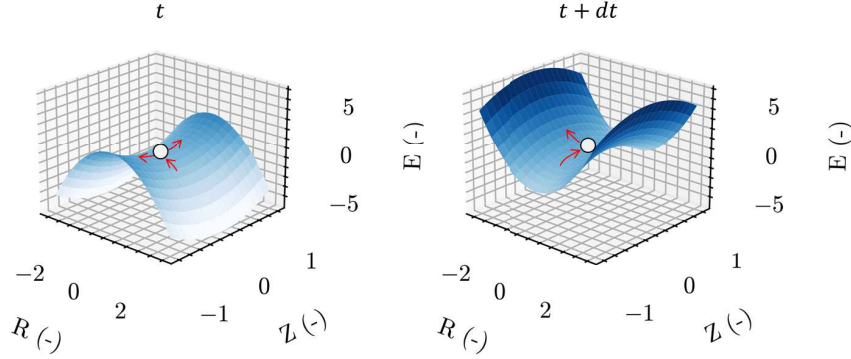


Figure 4.3: Time variation of the electric potential induced by the circular electrode within the levitator. The location of the particle is represented by the grey circle. The preferred movement of the particle due to the potential is represented with red arrows.

by the time varying electric field. Aluminum is paramagnetic, meaning that it follows Curie's law and thus has very low magnetism at high temperatures. The impact of the induced magnetic field on aluminum can be neglected during combustion. Alumina, however, is a diamagnetic material and thus preserves its magnetism at high temperatures. A short study available in appendix A evidences that the impact of the magnetic field can also be neglected for alumina and, thus, can be ignored at all times.

4.4 Optical diagnostics

4.4.1 Photomultipliers

During the combustion of an aluminum particle, the light emission from the droplet passes through the optical ports of the levitator and the combustion chamber, reaching a light collecting tube. At the end of the tube, a THORLABS dichroic filter *D1* DMLP650R deflects all emissions at wavelengths lower than 650 nm. As a result, the emission spectrum above 650 nm is focused and transmitted through an iris to modulate the overall intensity and then via an optic fiber towards the photomultipliers system. Light first passes through a second dichroic filter *D2* DMLP735R that deflects emissions at wavelengths lower than 735 nm towards the first photomultiplier PM_1 . In front of PM_1 , a THORLABS bandpass filter *BP1* FBH700-10 centered at 700 nm with a full width at half maximum (FWHM) of 10 nm is used to select the wavelength of the emissions reaching the photomultiplier. PM_1 thus measures the emission intensity at 700 nm. The light that is not deflected by *D2* reaches the third dichroic *D3* DMLPR805R that reflects emissions below 805 nm towards PM_2 . PM_2 is coupled to a bandpass filter *BP2* FBH760-10 centered at 760 nm with FWHM of 10 nm and thus measures the emission intensity at 760 nm. Finally, the emissions transmitted through *D3* reach PM_3 coupled to the bandpass filter *BP3* FBH820-10 centered at 820 nm with a

FWHM of 10 nm and thus captures light intensity at 820 nm. This system is summarized in Figure B.5.

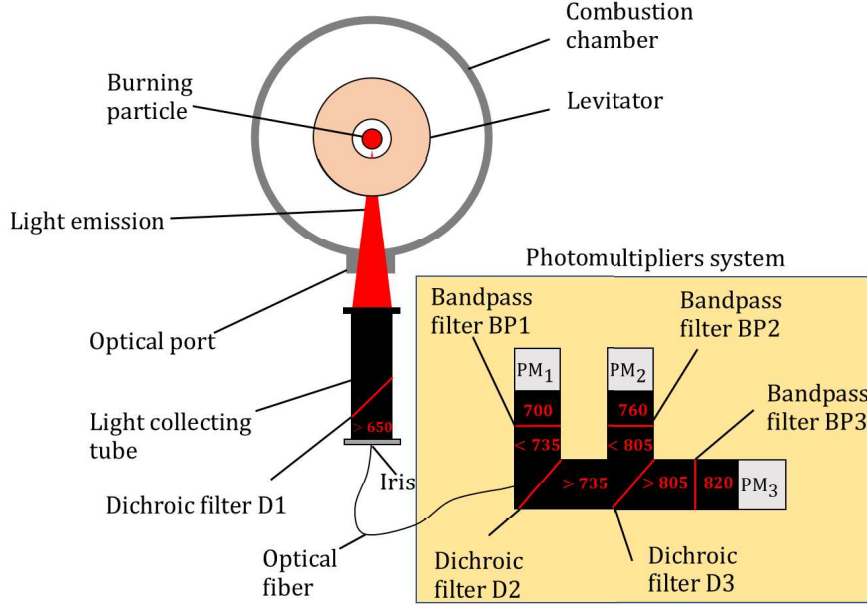


Figure 4.4: Schematics of the photomultipliers (PM) system. Information of locally available wavelengths is displayed in red.

The photomultipliers used are PMT1002 from THORLABS. The spectral responses of the dichroic filters, bandpass filters and photomultipliers are available in appendix B.

This photomultipliers system enables the measure of the integrated, i.e., total light emission intensity of the burning droplet, at three different wavelengths, from ignition of the droplet, to its quenching or until the particle exits the field of view of the light collecting tube. Note that depending on the opening of the iris, the field of view of the photomultipliers system ranges from a circle of 10 mm in diameter to a circle of 5 mm in diameter, at the center of the levitator. The field of view of the photomultipliers is greater than the camera's.

As an example, and without going into the details of the results, Figure 4.5 presents an example of signals from the photomultipliers system.

4.4.2 Camera

The work presented in this manuscript relies on the acquisition of spatially resolved data, making the camera system a crucial element of the experimental setup. On the opposite side of the photomultipliers system, light emission from the particle exits the levitator and the combustion chamber and enters a QUESTAR QM100 teleobjective. This short focal teleobjective enables a significant magnification of the observed system. The working distance, between the teleobjective and the particle, is about 270 mm. The light exiting the teleobjective is transmitted to the camera, passing through a triple band filter from EDMUND OPTICS. The camera-teleobjective system is summarized in Figure 4.6. The bandwidths of the filter are centered at 457 nm, 530 nm, and 628 nm, with respective bandwidths of 22 nm, 20 nm,

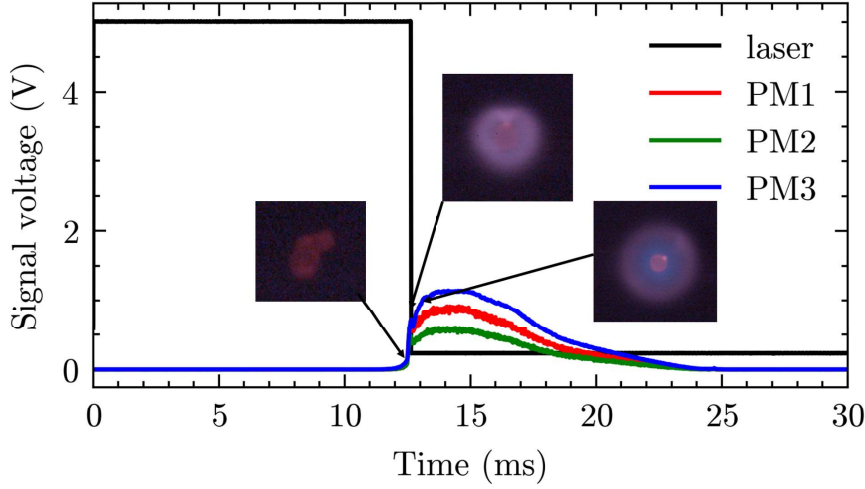


Figure 4.5: Example of signals from photomultipliers during aluminum combustion. Images of the particle at various times are also added.

and 28 nm. As light emission from a condensed phase follows the Planck's law, the use of a triple band filter enables us to observe emissions with the camera at precise wavelengths, instead of emissions integrated over large wavelength bands. As an example, from Figure 4.7 it comes that the R matrix of the camera contains emissions ranging from 400 nm to 700 nm. With the use of the triple band filter, it is considered that the R matrix only observes emissions at 630 nm, along with emissions at 450 nm and 530 nm from spectral overlap issues that can be accounted for. This is detailed further in the next chapter.

A high-speed color PHANTOM T-2410 camera is used in the experimental setup. This camera is equipped with RGB matrices and their associated spectral responses are presented in Figure 4.7, along with the relative spectral response of the triple band filter. The maximal frequency of the color camera is 25 000 fps in full scale, and the spatial resolution obtained from the teleobjective is about $1.3 \mu\text{m}/\text{px}$. From this camera-teleobjective coupling, excellent spatial and temporal resolutions are achieved while observing light emissions at three different wavelengths.

4.4.3 Backlight

To conclude about the optical diagnostics, the backlight system is detailed. In order to proceed to absorption measurements from the CCPs generated during aluminum combustion, a backlight is required. A SOLIS-3C THORLABS LED is used for its high power (4.6 W) and white light (5700 K). Indeed, the methodology requires the measurement of absorption from CCPs at various wavelengths enabled by a white LED. As the emission is spread across the visible spectrum, the triple band filter coupled to the camera cuts off the majority of the light intensity, hence the need for a high-power LED. The emission spectrum of the LED can be found in Appendix B.

This LED is mounted on the same light collecting tube as the one used for the photomultipliers system. A low-pass filter *LP1* FESH0650, at a cut-off wavelength of 650 nm,

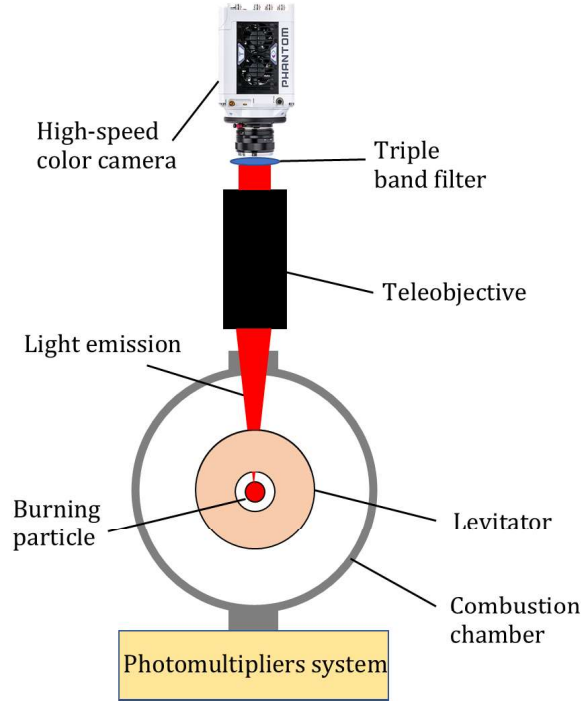


Figure 4.6: Schematics of the camera-teleobjective system.

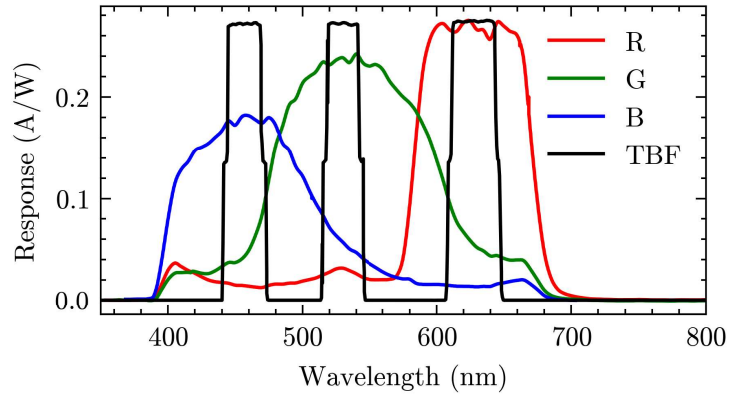


Figure 4.7: Spectral response of RGB matrices of the high-speed color PHANTOM T-2410 camera. The relative spectral response of the triple band filter (TBF) has been added (black).

ensures that no LED emission can go to the photomultipliers. Thus, the dichroic filter $D1$ reflects the LED emissions towards the center of the levitator, then reaching the teleobjective and camera system. In addition, this LED can be externally controlled and pulsed at high frequencies up to 100 kHz.

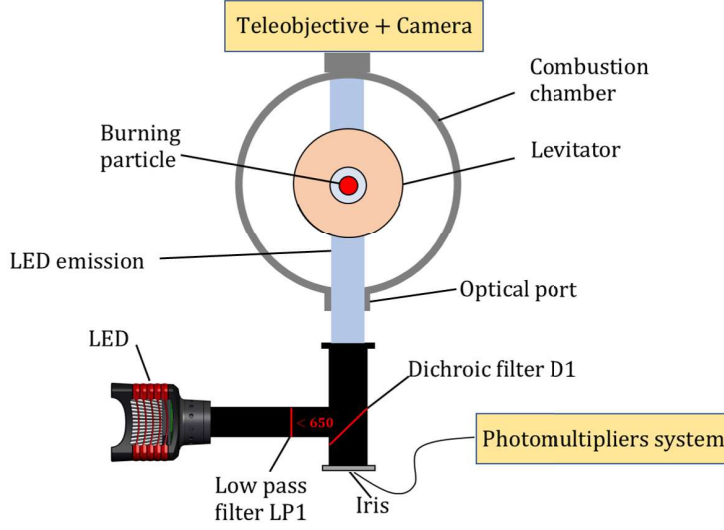


Figure 4.8: Schematics of the LED system.

4.5 Ignition device

As discussed in the previous chapter, upon heating, the initially charged particle loses its charge rapidly, decreasing the efficiency of the levitating device as the particles begins to fall. A rapid and contactless device is required to provide the ignition energy to the particle. A SYNRAD 48-5 CO_2 laser, at a wavelength of 10.6 microns, and output power of 50 W, serves as the ignition device. The laser initially provides a Gaussian beam of diameter $D_0 = 3.5$ mm, which is then split into two perpendicular beams using a 50/50 $ZnSe$ beam splitter. The first beam is reflected with the use of one silicon mirror towards the combustion chamber, while the other beam is reflected by two silicon mirrors to reach the combustion chamber. The optical ports on the chamber are equipped with converging lenses ($f = 50$ mm) to focus the laser beams at the center of the levitator. The diameter of the laser beam at this location is crucial, as it characterizes the energy density that can be provided to the particle. Let us define the Rayleigh distance as the distance from the location of beam generation, at which the cross-section area doubles, or the beam radius is multiplied by a factor $\sqrt{2}$. This Rayleigh distance Z_R can be expressed as a function of the initial beam diameter and the beam's wavelength λ_0 .

$$Z_R = \frac{\pi D_0^2}{4\lambda_0} \quad (4.1)$$

With the current laser, the Rayleigh distance is evaluated at $Z_R = 0.91$ m. The longest beam path (the beam reflecting on two mirrors), before reaching the converging lens, is about 0.83 m. It is thus approximated that the beam diameter at the combustion chamber is identical to the initial beam diameter. In Gauss conditions, the beam diameter at the center of the levitator D_l is expressed as a function of the focal length of the lens, the beam diameter at the converging lens, and the wavelength of the laser.

$$D_l = \frac{4f\lambda_0}{\pi D_0} \quad (4.2)$$

From this, it comes that the beam diameter at the location of the particle is $D_l = 193 \mu m$. As a result, an aluminum particle of 70 microns in diameter can be entirely impacted by the laser beam, from two opposite sides. The use of two beams allows the radiative pressure effects on the particle to cancel out, keeping it in the field of view of the optical devices. Additionally, it provides a homogeneous heating of the particle. The laser is used for a single pulse, with a controlled duration and controlled power. A power of 50 W can be delivered in total by the sum of the power from both beams. The global transmission efficiency of the optics (splitter, lenses, mirrors) being of about 0.84, a power density of $1.44 \cdot 10^9 W/m^2$ is developed by the laser at the center of the levitator. Wilson [37] defines a flux of $10^{11} W/m^2$ as excessive, leading to jetting effects pushing the particle away, and recommends a flux of $10^{10} W/m^2$ to ignite an aluminum particle in 200 μs . The slightly lower power delivered by our laser results in slightly longer ignition delays, usually of a few milliseconds depending on the location of the particle in the laser paths. The use of the laser to ignite the Al particle requires a precise alignment of the two laser beams. A careful adjustment of the laser, the splitter, and the mirrors is necessary. This procedure must be frequently done and can require up to two days of calibration, prior to commencing experiments.

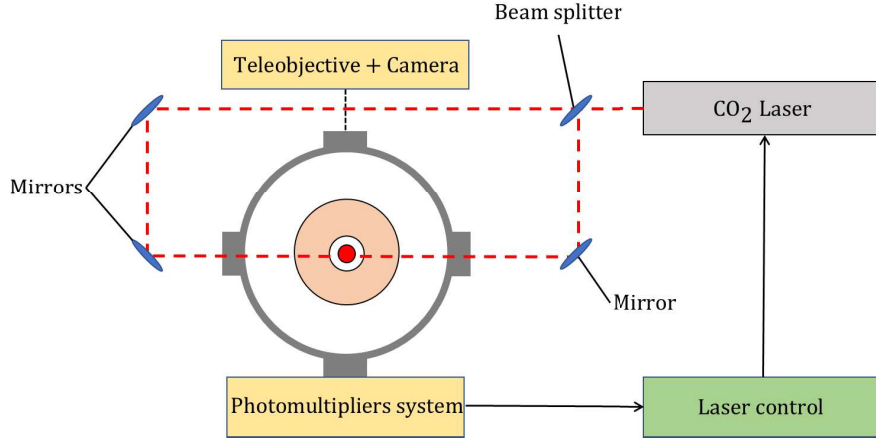


Figure 4.9: Schematics of the ignition system. The red dashed line is the laser path.

Note that to observe a self-sustained combustion of the particle, the laser needs to be cut-off when ignition of the particle is observed. The significant increase in the particle's light emission during ignition is used to cut the laser off. Indeed, one of the photomultipliers is connected to the laser control system. While the signal from a selected PM is lower than an arbitrarily defined threshold, the laser pulse is maintained. As the PM signal rises from an increase in particle light emission, it reaches the threshold value, and the laser control panel shuts the laser off. Self-sustained combustion is thus observed. The laser shutoff is illustrated in Figure 4.5. In this figure, the laser heats the particle for about 12.5 ms. As PM3 reaches about 0.5 V, the laser shuts off and combustion unfolds. Figure 4.9 summarizes the structure of the ignition system.

4.6 Summary

In this chapter, the experimental setup used throughout this work has been introduced. It contains a discussion about the experimental setups that can be found in the literature, as well as a general introduction to the setup used in this work. The electrodynamic levitator is further detailed, followed by an introduction to the optical diagnostics that equip the system. The ignition device is then introduced.

The equipment that is detailed in this chapter enables the application of a newly developed method to characterize aluminum combustion described in the following chapter.

Chapter 5

Development and performance assessment of an original characterization method

Aluminum particles with sufficiently large diameters are expected to burn in a vapor phase regime in most atmospheres. The occurrence of a flame established away from the droplet's surface results in a highly complex system, consisting of the droplet, reaction zones, gaseous suboxides, condensed combustion products (CCPs), high temperature gradients, etc. To provide a characterization of this system, a new method, based on optical diagnostics, has been developed. This method enables the measurement of spatially resolved alumina concentration, size, and temperature profiles surrounding the burning aluminum droplet. This chapter aims at providing a complete description of this method.

At first, the optical methods existing in the literature are briefly discussed. This discussion is followed by the hypotheses that are required for the application of the method. Then the Multispectral Light Extinction Method (M-LEM) is introduced, followed by the description of the enhanced Modulated Absorption/Emission (MAE) method. To ease the detailing of the analysis, the most fundamental case of an aluminum particle burning in atmospheric air is used as supporting data. The results are not discussed in terms of combustion phenomenology in this chapter. The limitations of the method are discussed before concluding.

5.1 Optical diagnostics & Al combustion - state of the art

In this section, the optical diagnostics that are used in the literature to characterize the combustion of aluminum particles are briefly discussed.

5.1.1 Alumina characterization

Various studies have characterized the CCPs formed during aluminum combustion, but rarely using in situ methods. Some studies are based on collecting the CCPs after the combustion process. This enables the measurement of alumina particle sizes with the use of a Scanning Electron Microscope (SEM). However, local information is usually lost, as particles are generally sampled on the walls of the combustor or using an aerosol filter coupled with a set of sieves [54]. To obtain local information, the quenching methods, already mentioned in the

previous chapters and used by Bucher [52] or Dreizin [53] are used. These methods consist in catching burning aluminum droplets on plates, quenching the combustion. The Al particle and its surrounding alumina smoke is thus captured and observable. Relative concentration profiles of the oxide can be obtained, as well as alumina particle size using a SEM. Although these methods provided groundbreaking results, it has to be considered that they are not in situ, and that the quenching can influence the measurements. An example of data obtained by Bucher [52] is presented in Fig. 5.1. From the quenched particle on the left figure, relative concentration profiles of condensed combustion products are obtained as a function of the distance to the droplet's surface.

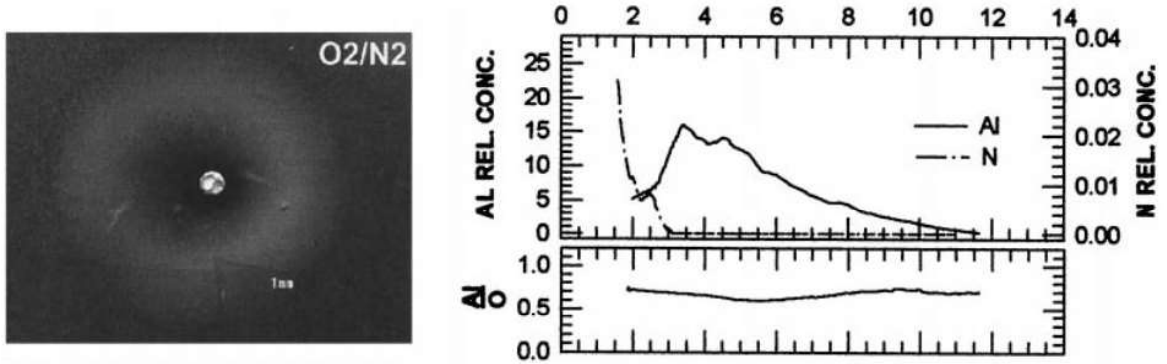


Figure 5.1: SEM image of a quenched particle (left) and Abel recovered CCPs relative concentration profiles (right) in the case of atmospheric air, from Ref.[52].

Additionally, very recent work from Jungst et al. [95], used a light extinction method coupled with a light scattering method applied to an aluminum dust flame to observe, in situ, the nanoparticles sizes and volumes generated during the jetting phase of aluminum burning droplets. Indeed, the efficiency of alumina droplets to scatter and absorb light provides information about their size and concentration. This work is amongst the first ones to provide in situ and spatially resolved data about the CCPs generation during Al combustion, significantly improving the general understanding of alumina condensation.

5.1.2 Temperature measurements

The measurement of the various temperature levels at play during aluminum combustion is a challenge. Although the system is complex, the assessment of an averaged temperature for the whole system is widespread in the literature. A large share of these studies is based on the multi-wavelength pyrometry method, briefly introduced here.

Multi-wavelength pyrometry

Let us consider a system, made of a hot condensed phase, with an emissivity ϵ constant over the full spectrum (grey body assumption). This system thus emits light, according to Planck's law, at any wavelength. The spectral radiance B_λ , function of the wavelength of emission λ and the system's temperature T , is the amount of power emitted by 1 m^2 of the system, in a unit solid angle, at a given wavelength, in $W \cdot m^{-3} \cdot sr^{-1}$. The Planck's law

gives the expression of this spectral radiance as a function of Planck's constant h , the speed of light c , and Boltzmann's constant k_b :

$$B_\lambda(\lambda, T) = \epsilon \frac{2hc^2}{\lambda^5} \frac{1}{e^{\frac{hc}{\lambda k_b T}} - 1} \quad (5.1)$$

Note that, for a wavelength of 800 nm, $\frac{hc}{\lambda k_b T} \approx \frac{18000}{T}$. For the temperature ranges expected in the case of aluminum combustion, well below 5000 K, and while using wavelengths close to the visible range, the following approximation can be made:

$$B_\lambda(\lambda, T) \approx \epsilon \frac{2hc^2}{\lambda^5} e^{-\frac{hc}{\lambda k_b T}} \quad (5.2)$$

Figure 5.2 illustrates the evolution over the spectrum of the spectral radiance of a black body (unit emissivity), for various temperatures, using Eq. (5.2). Note that at 3000 K, the maximum spectral intensity is reached in the infrared region of the spectrum. With increasing temperature, the maximum spectral radiance is observed at lower wavelengths.

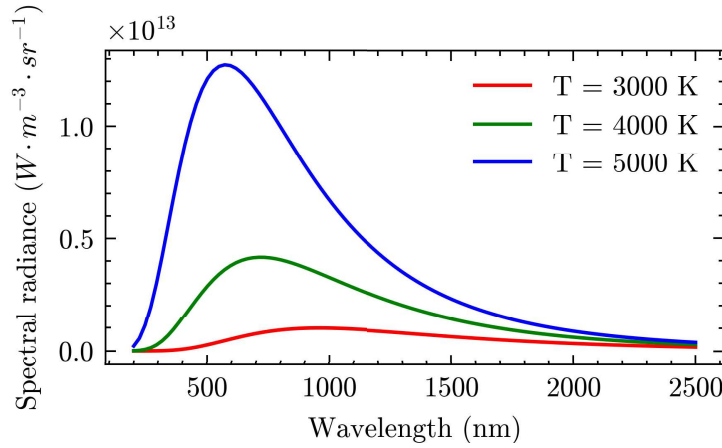


Figure 5.2: Spectral radiance of a black body, as a function of wavelength at 3000 K (red), 4000 K (green), and 5000 K (blue).

The multi-wavelength pyrometry uses Planck's law, in conjunction with a grey-body assumption, to assess the temperature of the system. Indeed, given a set of two photomultipliers that collect the grey-body emissions in the same geometrical conditions (identical solid angle Ω , identical observed area A) at two different wavelengths λ_1 , λ_2 , then two signals S_{λ_1} and S_{λ_2} are measured. Assuming that the two photomultipliers have respective specific spectral responses R_{λ_1} and R_{λ_2} then the measured signal can be written as follows:

$$S_{\lambda_1} = R_{\lambda_1} \cdot \Omega \cdot \epsilon \cdot A \cdot \frac{2hc^2}{\lambda_1^5} e^{-\frac{hc}{\lambda_1 k_b T}} \quad (5.3)$$

Thus, when the ratio of the signals is calculated, the geometrical coefficients cancel out, as well as the emissivities and it comes:

$$\frac{S_{\lambda_1}}{S_{\lambda_2}} = \frac{R_{\lambda_1}}{R_{\lambda_2}} \cdot \frac{\lambda_2^5}{\lambda_1^5} \cdot e^{-\frac{hc}{k_b T} \left(\frac{1}{\lambda_1} - \frac{1}{\lambda_2} \right)} \quad (5.4)$$

If the photomultipliers are accurately calibrated, the spectral responses are known, as well as the two wavelengths of interest λ_1 and λ_2 and the remaining unknown of Eq. (5.4) is the temperature T of the system. This method has been widely used in the literature with various numbers of photomultipliers, improving the accuracy of the method [65, 96, 97].

The main asset of this method is the ability to obtain pyrometry signals at high temporal resolution. The same method can be used with a spectrometer that enables the computation of significantly more signal ratios, but requires longer exposure times, and the temperature is usually not assessed at all times of the combustion process of a single aluminum droplet. Spectrometers are thus generally used in stable combustion systems such as aluminum dust flames [40]. Although time resolution cannot be obtained, in return, the emission spectrum of *AlO* can be measured and can be used as an additional tracker of the temperatures at play by fitting a theoretical emission spectrum that is a function of *AlO* temperature.

Even though the multiwavelength pyrometry method has a good temporal resolution, and the spectrometry method has improved accuracy regarding temperature evaluation, both methods show no spatially resolved data and are thus designated as integrated methods. In the event of a particle burning in a kinetically limited regime, integrated temperature measurement methods are relevant, as the observed system is fairly simple and shows almost no spatial emission variation. In the case of an aluminum droplet burning in the vapor phase, the system is much more complex:

- The aluminum droplet, at about 2500 K in atmospheric air, has a given emissivity and emitting surface.
- The alumina droplets, CCPs, are expected to display a wide range of temperatures, according to their location in the cloud. The CCPs have an emissivity and emitting surface (function of their concentration) that is different from the Al droplet.
- The gaseous aluminum and gaseous aluminum suboxides show emissions in the visible range, summarized by Dabos et al. [98]. For instance, *AlO** shows a continuous emission spectrum due to chemiluminescence between 500 nm and 640 nm
- On a more complex level, De Iuliis et al. [99] recently suggested light emission due to the presence of electrons during the formation of nanoparticles can perturb the pyrometric measurements

From this, it comes that the signal observed by a photomultiplier is the integrated sum of multiple emissions and is thus highly complex to analyze, making the direct use of Eq. (5.4) irrelevant. The use of pyrometers suffers from the lack of characterization of light emitters. With these considerations, the need for a spatially resolved temperature measurement method rises. Using a camera to measure the temperature enables the identification of the light emitters and thus greatly facilitates the analysis. Spatial temperature measurement is based on the same principle as multi-wavelength pyrometry. A first method consists in simply replacing photomultipliers with cameras coupled with bandpass filters. If two cameras measure

the same image of the system at two different wavelengths, then each pixel can be treated as a photomultiplier and Eq. (5.4) can be used on each pixel to provide a temperature field [66, 100]. This method, however, is challenging as the two cameras need to be perfectly aligned to observe exactly the same image, thus enabling the image processing to retrieve the temperature field. Note that to improve the accuracy of the measured temperature using a third signal ratio, a third camera is required and the design of the experimental setup becomes a real challenge. Another method, introduced in an in-depth study by McNesby et al. [101] consists in using a single color camera coupled with a triple-band filter. The main idea is to use the RGB matrices of the camera as three cameras to assess the temperature field of the system. Although the RGB matrices usually present wide spectral responses, placing a triple-band filter in front of the camera results in the R matrix measuring emissions at a precise wavelength in the red region of the spectrum, and similarly with the G and B matrices respectively measuring emissions at precise wavelengths in the green and blue regions of the spectrum. The ratio of the intensities measured by these three matrices R/G, R/B and G/B thus provides the temperature field. This method greatly facilitates image processing, as the three matrices come from the same sensor. Nonetheless, this method requires a careful calibration process that is described in Appendix C. With such a method, in theory, a single image is sufficient to assess a temperature field resulting in correct temporal resolutions with the use of high-speed cameras.

LEM & MAE

Note that so far, the methods to characterize the CCPs and the temperature are not compatible and usually cannot be done simultaneously. In addition, the temperature measurement relies on the grey body assumption. To overcome these two limitations, a Light Extinction Method (LEM) and a Modulated Absorption/Emission (MAE) technique can be used. Initially developed for soot characterization [102, 103, 104, 105], the evaluation of light extinction by a medium provides crucial characteristics of this medium. The LEM usually results in the evaluation of the concentration of the attenuating species, as well as their emissivities. With knowledge of such data, the light emission from the CCPs can be measured, accounting for self-absorption and emissivity to provide more accurate temperature profiles. The combination of these two methods is rather complex, as it requires some hypotheses to be made as well as a careful calibration process. However, this method provides an accurate identification of the emitters with good spatial and temporal resolution, and mostly, a simultaneous characterization of the CCPs concentration and temperature. The method will be entirely introduced in the following sections.

PLIF

Another method used in the literature to assess temperatures at play during aluminum combustion is a Planar Laser Induced Fluorescence (PLIF) technique. Although briefly described here, it is widely detailed in the literature and enables spatially resolved temperature measurements. Bucher [64] used an *AlO* PLIF method to assess the temperature of the *AlO* species. With the use of two lasers, two specific transitions of *AlO* molecules are excited. The laser emission is absorbed by the gas and then re-emitted at a different wavelength. Bucher

Table 5.1: Temperature assessment methods and their efficiencies.

Method	Spatial resolution	Time resolution	Characterization of emitters
Multi-wavelength pyrometry	None	Best	Bad
Spectrometer based pyrometry	None	None	Intermediate
Two-cameras pyrometry	Best	Correct	Good
Single-camera pyrometry	Best	Correct	Good
PLIF	Best	Correct	Good (Limited to AlO)
M-LEM + MAE	Best	Correct	Best

uses two different cameras coupled with filters to observe the re-emission from AlO at specific wavelengths. A careful calibration of the system enables the assessment of the spatially resolved AlO temperature by assessing the ratio of the two PLIF images from the two cameras. Bucher, with the use of this method, provided the first temperature profile surrounding a burning Al droplet. Not only this method is complex and requires accounting for many calibration laws, as well as accounting for condensed phase emissions, but this method is only limited to assessing temperature in regions where AlO concentration is sufficient.

Summary

For the majority of the existing studies, the grey-body assumption is made when alumina temperature is assessed. This can be avoided with the knowledge of the optical properties of oxide smoke that can be obtained using Light Extinction Methods. With such information, the spatially resolved pyrometry method can be improved to account for emissivity and absorption, resulting in a method called Modulated Absorption Emission (MAE) method. Although the PLIF method can provide a spatially resolved temperature profile, it only provides information about AlO and lacks a more complete characterization of the system, especially of alumina. The table (5.1) presents a summary of the various temperature measurement methods, their assets and their drawbacks. These considerations motivated the development of the multispectral LEM and MAE method coupling to provide an accurate characterization of aluminum combustion, under hypotheses detailed in the next section.

5.2 Hypotheses

In this section, the hypotheses that are made to enable the application of the method introduced in the following section are listed.

- In the system, the gaseous phase is optically thin. Condensed phase species are the only species considered as light-attenuating.
- Alumina is the only condensed phase, apart from the Al droplet.

- The presence of the triple-band filter enables the measurement of monochromatic values on the R G and B matrices of the camera.
- The system is spherically symmetric during the stable phase of the combustion.
- The oxide cap has no effect on the symmetry.
- The oxide cloud is sufficiently diluted to neglect multiple scattering.
- At a given location in the cloud, all alumina particles have the same size.
- Alumina particles within the cloud are spherical and follow the Mie theory.
- A local thermal equilibrium is considered at each location in the cloud.

5.3 Multispectral Light Extinction Method (M-LEM)

In this section, the developed M-LEM method is introduced, as well as the results it provides. First, the general principle of the LEM is introduced, followed by a description of the deconvolution process using the onion-peeling method. Then, the evaluation of the extinction cross section of alumina particles is discussed, prior to introducing the M-LEM.

5.3.1 LEM: general principle

Firstly, let us consider a light beam that contains multiple wavelengths λ , of initial spectral intensity $I_0(\lambda)$, traveling in a medium along a path of length l . If this medium is composed of light-attenuating species, at the end of the path, the beam has a spectral intensity $I(\lambda)$ such as $I(\lambda) < I_0(\lambda)$. The optical depth of the medium $\tau(\lambda)$ is defined as:

$$\tau(\lambda) = -\ln \left(\frac{I(\lambda)}{I_0(\lambda)} \right) \quad (5.5)$$

Note that if the light is not attenuated, the optical depth of the medium is 0, while in the case of a fully attenuated beam intensity, the optical depth of the medium is infinite.

The Beer-Lambert's law relates the optical depth of a medium, to the concentration of the attenuating species and to their attenuating efficiencies. Let us denote the number density of species i at location z of the beam's path as $n_i(z)$. The attenuating efficiency of a species i , generally called extinction cross section, at a location z of the beam's path and wavelength λ is denoted as $\sigma_i(z, \lambda)$. Beer-Lambert's law gives:

$$\tau(\lambda) = \int_0^l \sum_i^s \sigma_i(z, \lambda) \cdot n_i(z) dz \quad (5.6)$$

A beam attenuated by s species on a path of length l is depicted in Fig. 5.3.

Let us now transpose Beer-Lambert's law to the system of interest: the burning Al droplet and its surrounding oxide smoke. The same beam now passes through the oxide smoke generated by the combustion of the Al droplet. The smoke is composed of a gaseous phase

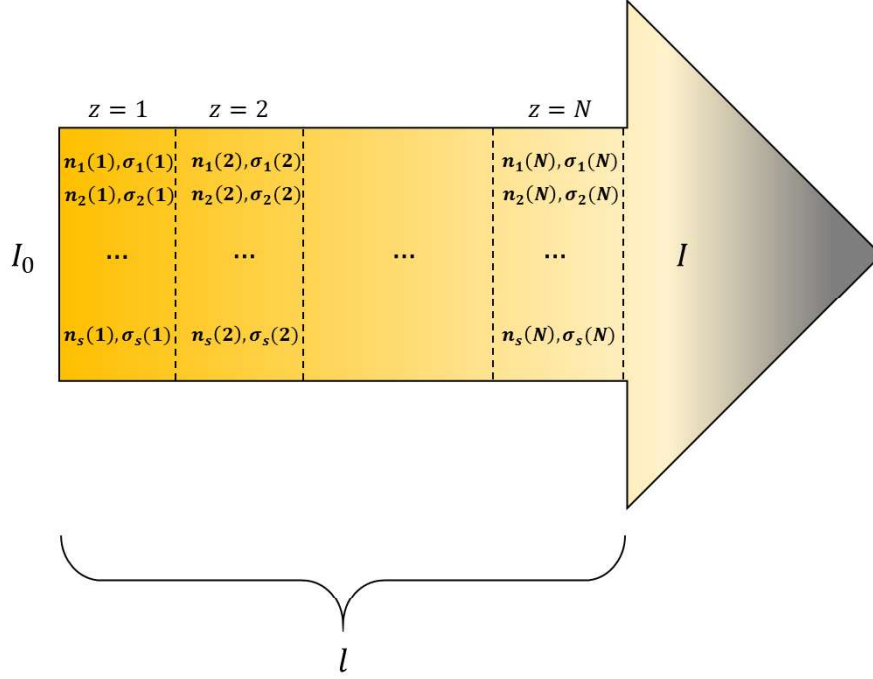


Figure 5.3: Representation of a beam of initial intensity I_0 passing through a medium of length l discretized in N slabs, composed of s attenuating species. The beam's final intensity is I .

and alumina droplets. It is assumed that the gaseous phase is optically thin and does not participate in attenuating the beam. Note that the attenuation cross section of an alumina particle is a function of its radius r_p . Thus, only alumina attenuates the beam intensity and Beer-Lambert's law becomes:

$$\tau(\lambda) = \int_0^l \sigma_{Al_2O_3}(r_p(z), \lambda) \cdot n_{Al_2O_3}(z) dz \quad (5.7)$$

The number density of alumina $n_{Al_2O_3}$ is defined as the ratio of the number of alumina droplets $N_{Al_2O_3}$ to volume V . It comes $n_{Al_2O_3} = \frac{N_{Al_2O_3}}{V}$. Introducing $V_{Al_2O_3}$ as the total volume of alumina in the considered volume, then $N_{Al_2O_3} = \frac{V_{Al_2O_3}}{\frac{4}{3}\pi r_p^3}$, with r_p the radius of an alumina particle. From this, the number density of alumina at a location z can be expressed as a function of the alumina volume fraction at this location $f_v(z)$ and the alumina particle radius at this location (z):

$$n_{Al_2O_3}(z) = \frac{V_{Al_2O_3}(z)}{\frac{4}{3}\pi r_p^3(z) \cdot V(z)} = \frac{f_v(z)}{\frac{4}{3}\pi r_p^3(z)} \quad (5.8)$$

The Equation (5.7) can thus be rearranged to give the following equation:

$$\tau(\lambda) = \int_0^l \sigma_{Al_2O_3}(r_p(z), \lambda) \cdot \frac{f_v(z)}{\frac{4}{3}\pi r_p^3(z)} dz \quad (5.9)$$

Equation (5.9) is the general principle of the LEM, relating light extinction, or optical depth, to the characteristics of the medium. The left-hand side of this equation, the optical depth, is a fairly simple term, function of the initial and final beam intensity. Experimentally, the initial intensity is easily assessed and the attenuated intensity can be measured with a particular process discussed further. The right-hand side of the equation is crucial as it contains data of high interest. Information about the local concentration and size of alumina particles are comprised in the integral. The left-hand side thus consists of an integrated measurement of local values, and the use of an Abel inverse transform, described later, enables the evaluation of the local values. Assuming that the extinction cross section of an alumina particle can be theoretically computed, as described further, this equation remains undetermined, as it contains two unknowns: the alumina particle radius and the alumina volume fraction. The multispectral nature of the method makes it possible to overcome this limit.

5.3.2 Optical depth measurement

In this section, combustion images are introduced. An image is a matrix of pixels that measure light intensity and it is assumed, at first, that all of the pixels measure the same intensity. This assumption is only made to facilitate the detailing of the method.

The optical depth of a medium can be measured using Equation (5.5), with knowledge of the initial intensity of a beam and of its intensity at the exit of the attenuating medium. The measurement of the optical depth of a non-emitting medium is straightforward. A LED is placed in front of a camera and the initial intensity is measured. Each pixel of the camera measures a light intensity I_0 . If the attenuating medium is placed between the LED and the camera, each pixel now measures an intensity I . The comparison of a pixel's value without the medium to a pixel's value with the medium enables the evaluation of the optical depth. In the case of a light-emitting medium, additional information is required. Indeed, the initial intensity is measured similarly to a non-emitting medium, but when the medium is added, the pixels measure an intensity that consists of the attenuated intensity of the LED, summed to the intensity of the medium's emissions. Figure 5.4 represents this situation in the case of a burning Al droplet and its light-emitting surroundings. To assess the attenuated LED intensity I , the intensity of the combustion emission, I_c in Fig. 5.4, must be subtracted to the intensity measured by the camera in the presence of the LED $I + I_c$.

In the case of aluminum combustion, Eq. (5.5) is rather expressed as:

$$\tau(\lambda) = -\ln \left(\frac{(I(\lambda) + I_c(\lambda)) - I_c(\lambda)}{I_0(\lambda) - I_d(\lambda)} \right) \quad (5.10)$$

The term $(I + I_c)$ is obtained in a backlit image of a burning Al droplet. The term I_c is obtained in a non-backlit image of a burning Al droplet. The term I_0 is obtained in an image of the backlight in the absence of the Al droplet. Finally, the term I_d accounts for the dark noise of the camera, which is already canceled out in the numerator due to the difference of two images, but that would be missing at the denominator if I_0 was the sole term. This term is obtained without LED emission and in the absence of the burning Al droplet. These four images as well as the conditions at which they are acquired are represented in Fig. 5.5.

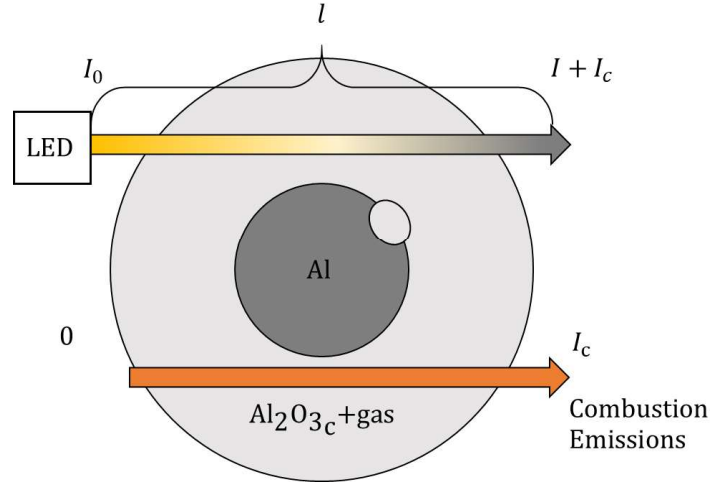


Figure 5.4: Representation of a beam of initial intensity I_0 passing through the oxide smoke of a burning Al droplet, along a path of length l , with final intensity $I + I_c$ due to combustion emissions.

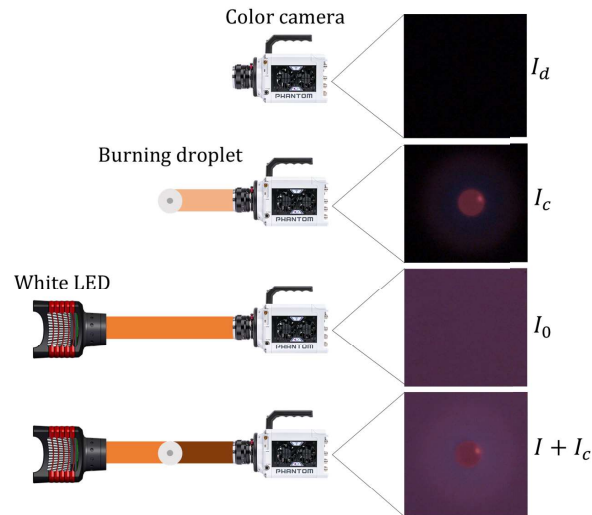


Figure 5.5: Specific images and the conditions in which they are acquired. From top to bottom: Dark noise image; Combustion image; LED image; Backlit combustion image.

In order to obtain the best possible accuracy, the two terms at the numerator of Eq. (5.10) should be measured, in theory, at exactly the same time. As this is not possible, the chosen solution is to observe two successive images of the burning droplet, one backlit, and the subsequent one without the LED. To enable such measurements, the LED should be pulsed at half the frequency of the camera. However, the LED doesn't show a perfect square signal when pulsed. The time the LED takes to reach maximum intensity from 0 intensity and vice versa is not zero. Thus upon synchronizing the LED at half the camera's frequency, all images appear backlit. To provide enough time for the LED to respond to the pulse, the LED is pulsed at a third of the camera's frequency. The stability of the LED was verified

for these operating conditions. This results in observing successively, a fully backlit image (LED emission decreasing), a non-backlit image (LED emission is zero) and a dimmer backlit image (LED emission increasing). A sequence obtained in such conditions for an Al particle burning in atmospheric air is presented in Fig. 5.6. The first six successive images of this sequence depict the ignition phase of the particle. The last three successive images illustrate the aluminum droplet burning in a stable vapor phase. Images of the LED alone (without Al combustion) and of the dark noise are not shown here but are easily obtained.

An optical depth image can thus be computed, and each pixel of this image has an intensity that is calculated with Eq. (5.10), using the intensities of the acquired images. Note that the oxide cloud induces a low attenuation of the backlight. Thus, computing an optical depth image from a simple pair of images {backlit ; non-backlit} results in a noisy optical depth image. In order to be able to measure the optical depth of the oxide cloud surrounding the aluminum droplet, a set of a few images, usually from 5 to 15 pairs of images depending on the stability of the combustion, is used. This results in the calculation of 5 to 15 optical depth images. To further enhance the accuracy, from the perfect symmetry of the system during the combustion, as observed in Fig. 5.6, an averaged optical depth profile can be evaluated. To do so, each pixel equally distant from the center of the droplet, in each optical depth image, is averaged to yield a single value corresponding to this distance, enabling the reconstruction of a radial optical depth profile.

An example of the resulting optical depth is depicted in Fig. 5.7. Three optical depth profiles are obtained as a color camera is used and Eq. (5.10) can be applied to the R, G, and B matrices of the camera. It is important to recall that the camera is coupled with a triple band filter, and thus the optical depths measured on the R,G and B matrices of the camera correspond to optical depths measured at three distinct wavelengths λ_R , λ_G and λ_B . The profiles were obtained by processing a set of 6 pairs of images. Note that an offset is observed for the three profiles. This offset is also measured when comparing two LED images with no attenuating medium, and is thus attributed to noise and can be easily accounted for. An increase in the optical depth is observed from 4 droplet radii r_0 down to lower distances to the droplet's surface.

The optical depth of the oxide smoke surrounding a burning Al droplet can thus be obtained with the current experimental setup. The left hand term of Eq. (5.9) is thus determined. However, this measured optical depth is integrated along the optical path. In order to retrieve local information for alumina, local information about optical depth is required. Indeed, Eq. (5.9) can be rewritten as:

$$\tau(\lambda) = \int_0^l \tau_{loc}(z, \lambda) dz \quad (5.11)$$

And thus it comes that:

$$\tau_{loc}(z, \lambda) = \sigma_{Al_2O_3}(r_p(z), \lambda) \cdot \frac{f_v(z)}{\frac{4}{3}\pi r_p^3(z)} \quad (5.12)$$

The evaluation of τ_{loc} at all locations of the cloud is thus crucial to assess local alumina characteristics. An onion-peeling Abel transform method is used to enable the calculation of the local optical depths.

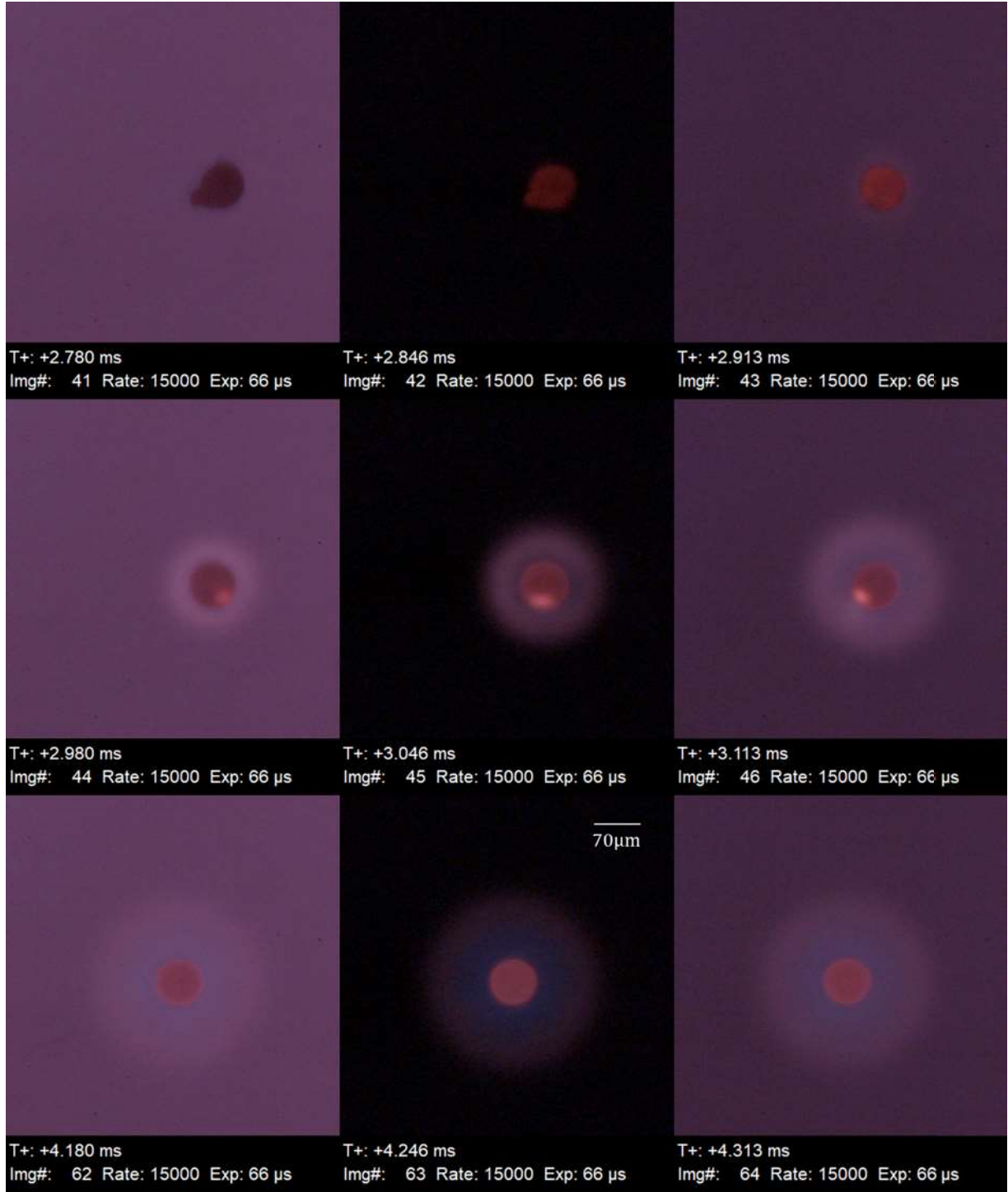


Figure 5.6: Sequence of an aluminum particle burning in atmospheric air. The resolution is $1.22 \mu m/px$.

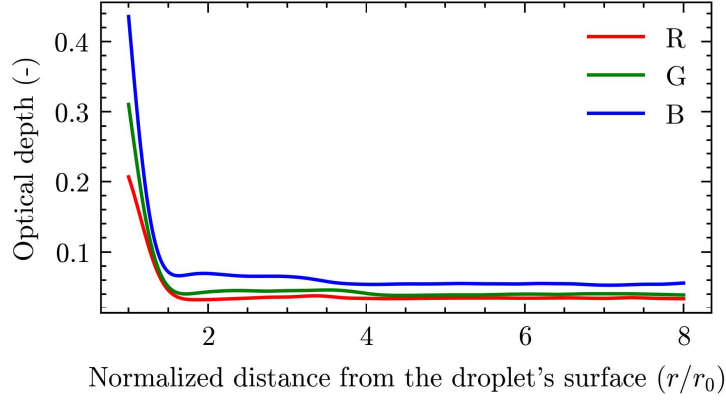


Figure 5.7: Optical depth profile surrounding an Al particle burning in atmospheric air, computed on the R,G, and B matrices of the camera.

5.3.3 Abel transform: Onion-peeling

The oxide smoke surrounding the Al droplet can be discretized into N layers so that Eq. (5.11) can be discretized. A representation of the shell discretization is given in Fig. 5.8

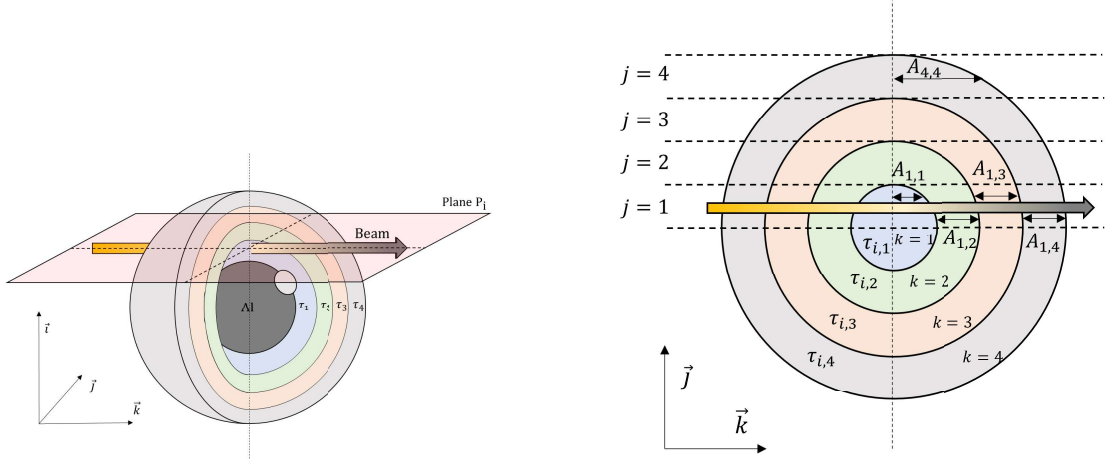


Figure 5.8: Schematics of the discretized oxide smoke (left). Top view of the plane P_i from the left figure (right).

Assuming that the normal to the plane encompassing the camera sensor is $\vec{k}$, the depicted beam reaches the sensor on the (i, j) pixel and thus measures an integrated optical depth $\tau_{i,j}^{integ}$. From the left figure, it is made obvious that this integrated optical depth is the sum of local optical depths weighed by geometric terms. As an example, from Fig. 5.8, for the beam located at a height i and impinging the $j^{th} = 1$ pixel, the measured integrated optical depth is expressed as:

$$\tau_{i,1}^{integ} = 2A_{1,4} \cdot \tau_{i,4}^{loc} + 2A_{1,3} \cdot \tau_{i,3}^{loc} + 2A_{1,2} \cdot \tau_{i,2}^{loc} + 2A_{1,1} \cdot \tau_{i,1}^{loc} \quad (5.13)$$

Please note that, if a ray hits the pixel at $j = 4$ at the same height, the integrated optical

depth would be directly proportional to the local optical depth. This is because the beam intersects a single layer of the system, $\tau_{i,4}^{integ} = 2A_{4,1} \cdot \tau_{i,4}^{loc}$. Moving down to $j = 3$, the integrated optical depth is a function of $\tau_{i,4}^{loc}$ and $\tau_{i,3}^{loc}$. As $\tau_{i,4}^{loc}$ has already been measured, $\tau_{i,3}^{loc}$ can also be determined. By proceeding through all shells, we can obtain all the local optical depths. This is the principle used in the onion-peeling deconvolution procedure. The equations that summarize the procedure, taken from Legros et al. [105], are presented. From Eq. (5.13), it comes for any height i :

$$\tau_{i,j}^{integ} = 2 \sum_{k=1}^N A_{jk} \cdot \tau_{ik}^{loc} \quad (5.14)$$

With A_{jk} the geometrical coefficients of the system. At any height i , the square matrix A^{OP} of size $(N \times N)$ is function of the camera's resolution δ_R in meters per pixel, and is expressed as:

$$A^{OP} = \begin{cases} 0 & \text{if } j > k \\ \delta_R \cdot [k^2 - (j - 0.5)^2]^{0.5} & \text{if } j = k \\ \delta_R \cdot ([k^2 - (j - 0.5)^2]^{0.5} - [(k - 1)^2 - (j - 0.5)^2]^{0.5}) & \text{else} \end{cases} \quad (5.15)$$

Let us consider the pixel line at the specific height i_s represented by the green line in Fig. 5.9. The integrated optical depth measured at the pixel $(i_s, j = n)$ is thus considered to be only proportional to the local optical depth of the shell n at height i_s : $\tau_{i_s,n}^{loc}$. The integrated optical depth measured on the next pixel $(i_s, j = n - 1)$ is thus function of the local optical depths $\tau_{i_s,n}^{loc}$ and $\tau_{i_s,n-1}^{loc}$. As we progress towards the center of the image, all local optical depths are obtained, allowing us to evaluate the local characteristics of the cloud at any given location.

Let us denote $X = (\tau_{i_s,k}^{loc})_{k=1,\dots,N}$ as the vector containing the local optical depths at height i_s and of the layers from 1 to N , and $B = (\tau_{i_s,j}^{integ})_{j=1,\dots,N}$ the vector containing the integrated optical depths computed on the pixels at height i_s and horizontal coordinate $j = 1, \dots, N$. The local optical depths can be calculated by solving the equation:

$$2A^{OP} \cdot X = B \quad (5.16)$$

This equation is ill-conditioned. It cannot be solved with a simple inversion of the matrix A^{OP} as a significant noise amplification would occur. To overcome this, a Tikhonov regularization is used. This process, described by Kashif [106] in his PhD work, is only briefly described here. A Tikhonov matrix L_0 of size $(N - 1 \times N)$ is introduced such as:

$$\begin{bmatrix} 1 & -1 & 0 & \dots & 0 \\ 0 & 1 & -1 & \ddots & \vdots \\ \vdots & \ddots & \ddots & \ddots & 0 \\ 0 & \dots & 0 & 1 & -1 \end{bmatrix} \quad (5.17)$$

Let us define a regularization parameter λ_{reg} and a matrix A_{reg} as:

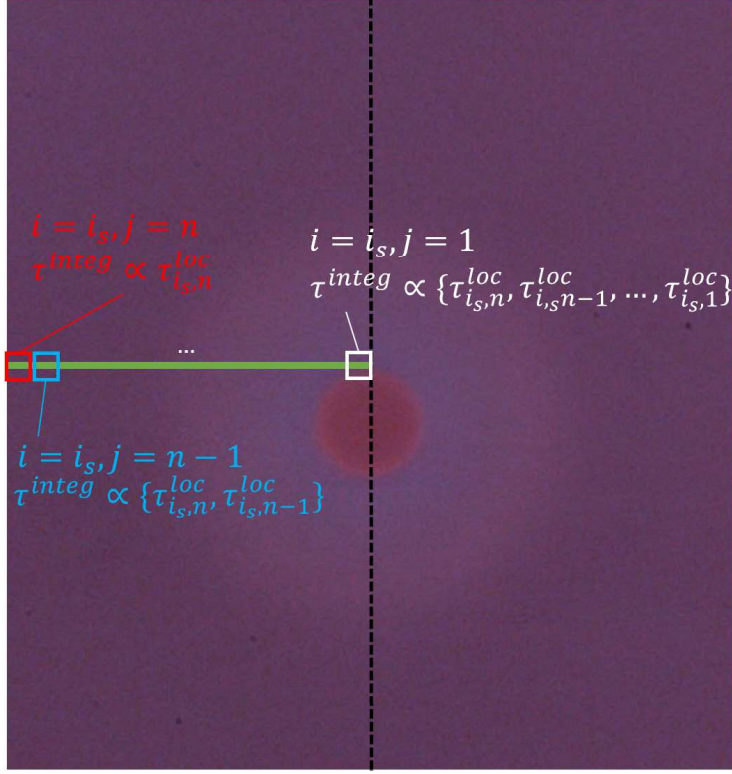


Figure 5.9: Image of a burning Al droplet and the line selected to apply the onion-peeling procedure.

$$A_{reg} = \left((A^{OP})^T \cdot A^{OP} + \lambda_{reg} \cdot A^{OP} \cdot L_0^T \cdot L_0 \right) \quad (5.18)$$

The selection of the regularization parameter is not addressed in this work but is thoroughly discussed in the literature [107]. The choice of this parameter is the result of a compromise between a desired noise attenuation, best at low values of λ and a non-desired attenuation of the gradient, best at large values of λ . A matrix B_{reg} is also defined as:

$$B_{reg} = (A^{OP})^T \cdot B \quad (5.19)$$

By applying the Tikhonov regularization technique, the solution vector X is obtained as the solution of the regularized equation $2A_{reg} \cdot X = B_{reg}$. This equation is no longer ill-posed, so it can thus be solved using linear algebra.

Knowledge of the vector X provides knowledge of the local optical depth at any distance from the droplet's surface. The left-hand side of Eq. (5.12) is known. An example of local optical depth profiles, obtained at λ_R , λ_G and λ_B , is presented in Fig. 5.10.

5.3.4 Extinction cross section

Let us consider a spherical particle, of diameter d_s and complex refractive index m . Let us consider light radiation at a wavelength λ , impacting the spherical particle, as depicted by

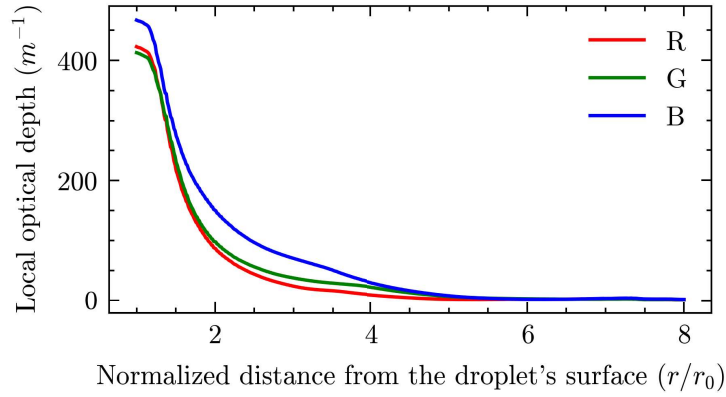


Figure 5.10: Local optical depth profile surrounding an Al particle burning in atmospheric air, at the three wavelengths of interest, obtained from the deconvolution of the integrated optical depth.

Fig. 5.11.

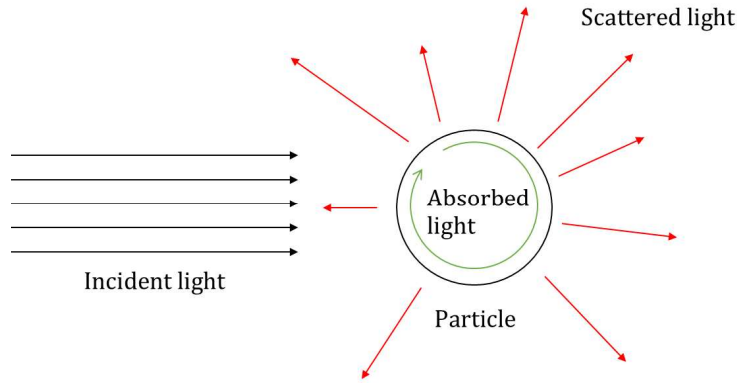


Figure 5.11: Absorbed and scattered light due to incident light impacting a particle.

A portion of the incoming light impinging on the particle is scattered in various directions with varying intensities. Another part of the light is absorbed by the particle without being re-emitted. This combination of absorption and scattering is called extinction. The efficiency with which a single particle scatters, absorbs and extinguishes radiation is precisely described by Mie theory. The following derivation, based on the work of van de Hulst, Bohren and Huffman [108, 79], allows us to characterize the optical efficiencies of a spherical particle.

As a first step, the Bessel function of the first kind, of order n , J_n is introduced and defined as:

$$J_n(x) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m! \Gamma(m+n+1)} \left(\frac{x}{2}\right)^{2m+n} \quad (5.20)$$

Where Γ is the gamma function defined as $\Gamma(x) = \int_0^{\infty} t^{x-1} e^{-t} dt$. Then, the spherical Bessel functions of order n , j_n and y_n are introduced as functions of the Bessel function:

$$j_n(x) = \sqrt{\frac{\pi}{2x}} J_{n+\frac{1}{2}}(x) \quad (5.21)$$

$$y_n(x) = \sqrt{\frac{\pi}{2x}} J_{-n-\frac{1}{2}}(x) \quad (5.22)$$

Finally, the Ricatti-Bessel functions are introduced:

$$\psi_n(x) = x j_n(x) \quad (5.23)$$

$$\chi_n(x) = -x y_n(x) \quad (5.24)$$

$$\xi_n(x) = \psi_n(x) - i\chi_n(x) \quad (5.25)$$

Then, the scattering coefficients of order n , a_n and b_n can be introduced as a function of the size factor of the particle $x = \frac{2\pi r_p}{\lambda}$ and its complex refractive index m :

$$a_n = \frac{m\psi_n(mx) \psi'_n(x) - \psi_n(x) \psi'_n(mx)}{m\psi_n(mx) \xi'_n(x) - \xi_n(x) \psi'_n(mx)} \quad (5.26)$$

$$b_n = \frac{\psi_n(mx) \psi'_n(x) - m\psi_n(x) \psi'_n(mx)}{\psi_n(mx) \xi'_n(x) - m\xi_n(x) \psi'_n(mx)} \quad (5.27)$$

Using the properties of the Ricatti-Bessel functions, Matzler [109] introduces a function $D_n(x)$ such as:

$$D_n(x) = \frac{\psi'_n(mx)}{\psi_n(mx)} \quad (5.28)$$

This function can be determined by recurrence with the following relation:

$$D_{n-1}(x) = \frac{n}{x} - \frac{1}{D_n(x) + \frac{n}{x}} \quad (5.29)$$

This enables to avoid the computation of derivatives of Bessel functions and save computation times, and thus, the scattering coefficients can be rearranged to give:

$$a_n = \frac{\left(\frac{D_n(mx)}{m} + \frac{n}{x}\right) \psi_n(x) - \psi_{n-1}(x)}{\left(\frac{D_n(mx)}{m} + \frac{n}{x}\right) \xi_n(x) - \xi_{n-1}(x)} \quad (5.30)$$

$$b_n = \frac{\left(D_n(mx)m + \frac{n}{x}\right) \psi_n(x) - \psi_{n-1}(x)}{\left(D_n(mx)m + \frac{n}{x}\right) \xi_n(x) - \xi_{n-1}(x)} \quad (5.31)$$

Let us denote $\Re(x)$ as the real part of the complex number x and $\Im(x)$ its imaginary part. The extinction efficiency Q_{ext} and scattering efficiencies Q_{sca} of a spherical particle are determined as:

$$Q_{ext} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \Re(a_n + b_n) \quad (5.32)$$

$$Q_{sca} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) (|a_n|^2 + |b_n|^2) \quad (5.33)$$

As the extinction is the result of absorption and scattering, the absorption efficiency Q_{abs} can be computed from the extinction and scattering efficiencies:

$$Q_{ext} = Q_{sca} + Q_{abs} \quad (5.34)$$

Finally, the extinction cross section of a spherical particle is calculated from its extinction efficiency as:

$$\sigma_{ext} = \pi r_p^2 \cdot Q_{ext} \quad (5.35)$$

Note that Bohren and Huffman [79] recommend using summations up to a maximal integer $n_{max} = x + 4x^{1/3} + 2$ to evaluate Eq. (5.32, 5.33). Using the Eq. (5.20) - (5.35), the extinction cross section of an alumina droplet at a given wavelength can be computed with the knowledge of its radius and its complex refractive index. Dombrovsky [110] developed a function to express the complex refractive index of liquid alumina as a function of wavelength and temperature T . The refractive index for alumina is thus expressed as:

$$m_{Al_2O_3}(\lambda, T) = n_0(\lambda) \cdot (1 + \xi_1(T - T_1)) + ik_0(\lambda) e^{\xi_2(T - T_*)} \quad (5.36)$$

With $\xi_1 = 2.02 \cdot 10^{-5} K^{-1}$, $T_1 = 473K$, $\xi_2 = 1.847 \cdot 10^{-3} K^{-1}$, $T_* = 2950K$ and the intermediate functions n_0 and k_0 defined, with λ in microns, as:

$$n_0(\lambda) = \sqrt{1 + \frac{1.024\lambda^2}{\lambda^2 - 0.003776} + \frac{1.058\lambda^2}{\lambda^2 - 0.01225} + \frac{5.281\lambda^2}{\lambda^2 - 321.4}} \quad (5.37)$$

$$k_0(\lambda) = 0.002 (1 + 0.007\lambda + 0.06\lambda^2) \quad (5.38)$$

The temperature T is arbitrarily set at 3550 K. It is highly common, in the literature, to consider that the particles are much smaller than the wavelength. In other words, a very common assumption is to say that the size factor is much smaller than unity $x \ll 1$. This assumption is the Rayleigh approximation. Although this hypothesis can be relevant in some applications, it is mainly used because it greatly simplifies Eq. (5.9). If the Rayleigh approximation is made, the Ricatti-Bessel functions of orders greater than 2 can be neglected and the extinction efficiency Q_{ext}^{Ray} is expressed as:

$$Q_{ext}^{Ray} = \frac{2}{x^2} (3\Re(a_1 + b_1) + 5\Re(a_2 + b_2)) \quad (5.39)$$

Using series expansion if required, the four terms a_1 , a_2 , b_1 and b_2 using the Rayleigh approximation and limited to the sixth order, are expressed as:

$$a_1^{Ray} = -\frac{2ix^3}{3} \frac{m^2 - 1}{m^2 + 2} - \frac{2ix^5}{5} \frac{(m^2 - 2)(m^2 - 1)}{(m^2 + 2)} + \frac{4x^6}{9} \frac{(m^2 - 1)^2}{(m^2 + 2)} \quad (5.40)$$

$$b_1^{Ray} = -\frac{ix^5}{45} (m^2 - 1) \quad (5.41)$$

$$a_2^{Ray} = -\frac{ix^5}{15} \frac{m^2 - 1}{2m^2 + 3} \quad (5.42)$$

$$b_2^{Ray} = 0 \quad (5.43)$$

It thus comes from a mathematical development:

$$Q_{ext}^{Ray} = 4x \Im \left(\frac{m^2 - 1}{m^2 + 2} \right) + \frac{8}{3} x^4 \Re \left(\left(\frac{m^2 - 1}{m^2 + 2} \right)^2 \right) \quad (5.44)$$

And once again, if x is smaller than unity:

$$Q_{ext}^{Ray} \approx 4x \Im \left(\frac{m^2 - 1}{m^2 + 2} \right) \quad (5.45)$$

And thus:

$$\sigma_{ext}^{Ray} \approx \frac{8\pi^2 r_p^3}{\lambda} \Im \left(\frac{m^2 - 1}{m^2 + 2} \right) \quad (5.46)$$

With the Rayleigh approximation, the particle radius r_p that appears from the number density of particles cancels out with the particle radius that appears from the extinction cross section. The Equation (5.9) is now determined, as the alumina volume fraction remains as the sole unknown. The question of the validity of Rayleigh's approximation thus arises. Figure 5.12 illustrates the behavior of the Rayleigh approximation compared to the classical Mie theory. For a wavelength of 400 nm, the Rayleigh approximation induces a relative error in the extinction efficiency greater than 10 % for particles larger than 11 nm in diameter. At 800 nm, a relative error greater than 10 % is obtained for particles of diameters larger than 26 nm.

Similarly, assuming a medium of constant composition (identical optical depth τ at all locations of the medium) and unit length l , it can be approximated that $f_v = \frac{\tau \cdot 4/3 \cdot r_p}{l \cdot Q_{ext}}$. Assuming an optical depth of 0.1, the volume fraction of alumina in this medium can be computed with the Mie theory and with the Rayleigh approximation. The results presented in Fig. 5.13 evidence that a 10 % error on the volume fraction is reached in similar conditions as for the extinction efficiency. It is also observed that for larger particles, the error significantly increases, reaching up to 400 % at a particle diameter of 40 nm at a wavelength of 400 nm, and more than 40% at a particle diameter of 40 nm at a wavelength of 800 nm.

Figures 5.12 and 5.13 highlight that the Rayleigh approximation is only valid, in the entire visible range, for alumina particles with diameters well below 10 nm. Given the alumina droplet sizes observed in the literature mentioned in the previous chapter, in this work, the choice is made not to use the Rayleigh approximation, in order to develop a method with greater accuracy based on the multispectral character of the measurements.

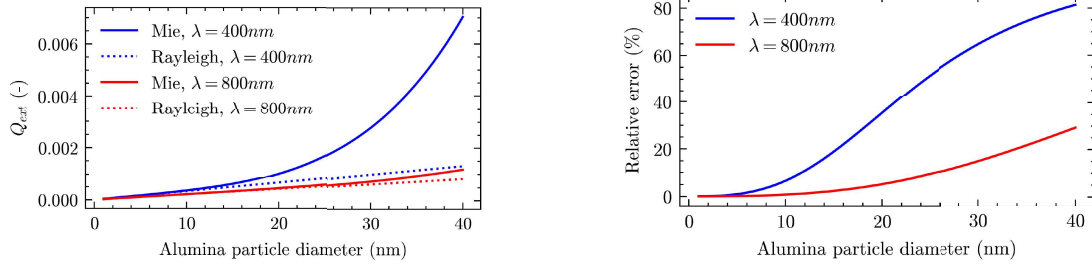


Figure 5.12: Extinction efficiency of an alumina particle as a function of its size, evaluated with the classical Mie theory, and with the Rayleigh approximation, for two different wavelengths (left). Relative error between extinction efficiencies computed with Mie and Rayleigh methods for two different wavelengths (right).

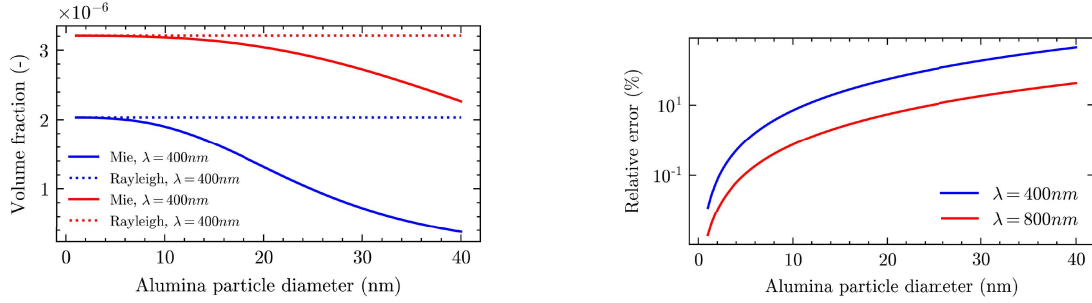


Figure 5.13: Volume fraction computed under the hypothesis of a medium of unit length, constant composition and optical depth of 0.1, evaluated with the classical Mie theory, and with the Rayleigh approximation, for two different wavelengths (left). Relative error between volume fractions computed with Mie and Rayleigh methods for two different wavelengths (right).

5.3.5 Multispectral LEM

The use of a color camera coupled with a triple band filter enables the measurement of the local optical depths, using the LEM and an onion-peeling deconvolution method, at three distinct wavelengths λ_R , λ_G and λ_B , respectively on the R,G and B matrices of the camera. The three profiles of local optical depths have been introduced in Fig. 5.10. Equation (5.12) can be rewritten, for any wavelength, as:

$$\frac{\tau_{loc}(z, \lambda)}{\sigma_{Al_2O_3}(r_p(z), \lambda)} = \frac{f_v(z)}{\frac{4}{3}\pi r_p^3(z)} \quad (5.47)$$

Although this equation cannot be solved due to f_v and r_p being unknown with only τ_{loc} as a measured value, it is crucial to notice that the right-hand side of this equation is independent on the wavelength. Thus, it comes out:

$$\frac{\tau_{loc}(z, \lambda_R)}{\sigma_{Al_2O_3}(r_p(z), \lambda_R)} = \frac{\tau_{loc}(z, \lambda_G)}{\sigma_{Al_2O_3}(r_p(z), \lambda_G)} = \frac{\tau_{loc}(z, \lambda_B)}{\sigma_{Al_2O_3}(r_p(z), \lambda_B)} \quad (5.48)$$

τ_{loc} is measured at the three wavelengths of interest, and $\sigma_{Al_2O_3}$ can be computed at any

wavelength for a given particle radius. Thus, in the set of three equations contained in Eq. (5.48), the only unknown is the alumina particle radius. The Equation (5.48) is thus solved for r_p at any distance from the droplet's surface. To do so, an optimization procedure is based on minimizing the relative errors between the three ratios $\frac{\tau_{loc}}{\sigma_{Al_2O_3}}$. Denoting $A_i^{r_p,z}$ the value of this ratio for the wavelength $(\lambda_i)_{i=R,G,B}$ and for a particle size r_p at a location z , then a mean ratio $A_m^{r_p,z}$ is expressed as $A_m^{r_p,z} = 1/3 \cdot (A_R^{r_p,z} + A_G^{r_p,z} + A_B^{r_p,z})$ and at each location z of the cloud, the size of alumina particles is determined to minimize the error $e(r(z))$:

$$e(r_p(z)) = e_{abs}(r_p(z)) \cdot e_{rel}(r_p(z)) \quad (5.49)$$

With

$$e_{abs}(r(z)) = \left(\frac{A_m^{r_p,z} - A_R^{r_p,z}}{A_m^{r_p,z}} \right)^2 + \left(\frac{A_m^{r_p,z} - A_G^{r_p,z}}{A_m^{r_p,z}} \right)^2 + \left(\frac{A_m^{r_p,z} - A_B^{r_p,z}}{A_m^{r_p,z}} \right)^2 \quad (5.50)$$

$$e_{rel}(r(z)) = (A_m^{r_p,z} - A_R^{r_p,z})^2 + (A_m^{r_p,z} - A_G^{r_p,z})^2 + (A_m^{r_p,z} - A_B^{r_p,z})^2 \quad (5.51)$$

This optimization procedure enables the evaluation of an alumina particle radius at any location in the oxide smoke. The resulting alumina particle radius profile is presented in Fig. 5.14.

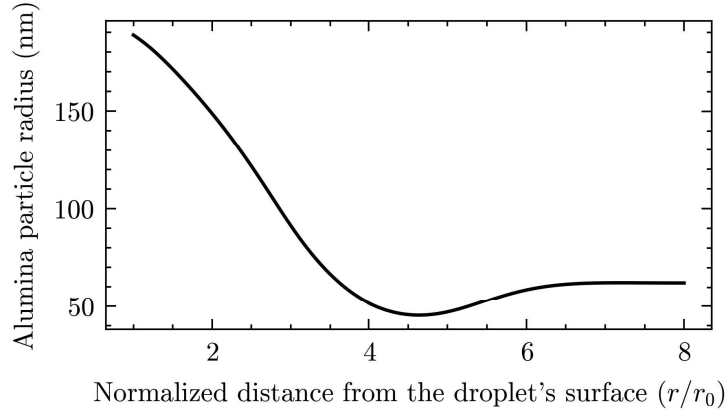


Figure 5.14: Alumina particle radius profile surrounding the burning Al droplet.

Equation (5.47) can be rearranged:

$$f_v(z) = \frac{\frac{4}{3}\pi r_p^3(z) \cdot \tau_{loc}(z, \lambda)}{\sigma_{Al_2O_3}(r_p(z), \lambda)} \quad (5.52)$$

From this, it comes out that the evaluation of an optimized particle size profile leads to the assessment of an optimized alumina volume fraction profile. In order to assess the efficiency of the method, let us compare the alumina volume fraction profiles evaluated under the assumption of a constant alumina particle size of 75 nm in radius, to the alumina volume fraction profile evaluated with the optimized alumina particle size profile. The results are displayed in Fig. 5.15.

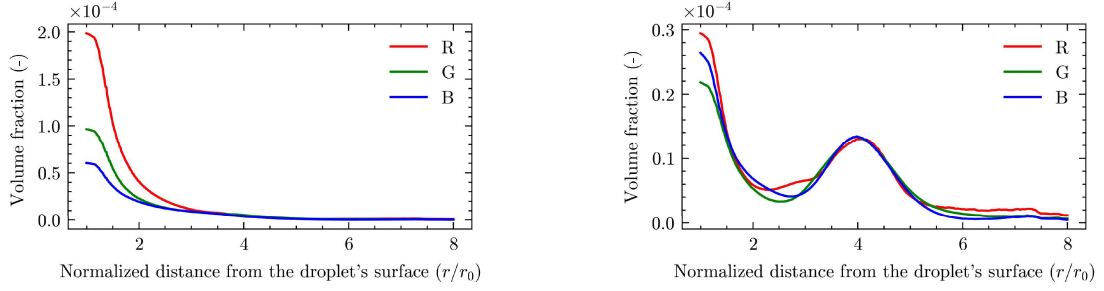


Figure 5.15: Alumina volume fraction profile computed under the assumption of constant alumina particle size $r_p = 75nm$ (left). Alumina volume fraction profile computed with the optimized alumina particle size (right).

The left figure evidences the need for the optimization procedure as, considering a constant alumina particle radius leads to significant discrepancies between the volume fraction profiles computed at the three wavelengths of interest. It is obvious that the volume fraction must be constant, whatever the wavelength at which it is measured. The right figure shows that the optimization procedure is efficient, as the three volume fractions are close to identical.

5.3.6 Summary

A pulsed LED is coupled with a color camera in order to evaluate the integrated optical depth of the oxide cloud, at three distinct wavelengths, using Eq. (5.10). The local optical depth profile is evaluated with the use of an onion-peeling deconvolution procedure. The Rayleigh approximation is discarded due to its non-validity, and the extinction cross section of an alumina particle is calculated using the Mie theory. Then, an optimization procedure is applied to the local optical depth profiles at the three wavelengths of interest in order to assess the alumina particle radius. From this, Eq. (5.52) enables the evaluation of the alumina volume fraction profile. The procedure is summarized in Fig. 5.16.

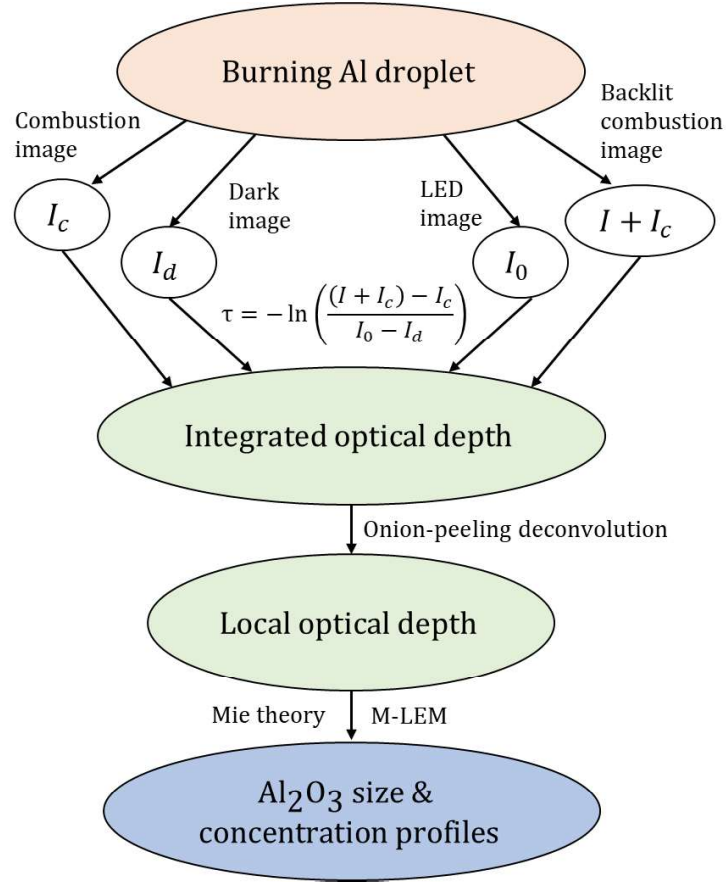


Figure 5.16: Summary of the M-LEM.

5.4 Modulation Absorption Emission (MAE)

In the previous section, the alumina size and concentration profiles are assessed using the M-LEM, but the alumina temperature profile remains unknown. The use of a Modulated Absorption/Emission method uses the previously obtained information about extinction and combustion emissions to assess the temperature profile surrounding the burning Al droplet.

5.4.1 Local black body emissions

Let us recall Eq. (5.11):

$$\tau(\lambda) = \int_0^l \tau^{loc}(z, \lambda) dz \quad (5.53)$$

Let us introduce a local spectral absorption coefficient $\kappa^{loc}(z, \lambda)$ and a local spectral scattering coefficient $\beta^{loc}(z, \lambda)$ such as:

$$\tau(\lambda) = \int_0^l (\kappa^{loc}(z, \lambda) + \beta^{loc}(z, \lambda)) dz \quad (5.54)$$

Thus, $\tau^{loc}(z, \lambda)$ appears as a local spectral extinction coefficient. Modest [111] mentions that at the local thermal equilibrium, the local spectral absorption coefficient $\kappa^{loc}(z, \lambda)$ is equal to the local spectral emission coefficient, denoted as $\epsilon^{loc}(z, \lambda)$. In the previous section, it was shown that scattering, absorption and extinction efficiencies $Q_{sca}(r_p(z), \lambda)$, $Q_{abs}(r_p(z), \lambda)$ and $Q_{ext}(r_p(z), \lambda)$ can be computed using the Mie theory. Thus, the importance of absorption in relation to extinction, $P_{abs}(r_p(z), \lambda)$ can be assessed as $P_{abs}(r_p(z), \lambda) = \frac{Q_{abs}(r_p(z), \lambda)}{Q_{ext}(r_p(z), \lambda)}$. From this, the local spectral emission coefficient can be computed from the previously assessed local optical depth profile, also called the local extinction coefficient profile:

$$\epsilon^{loc}(z, \lambda) = P_{abs}(r_p(z), \lambda) \cdot \tau^{loc}(z, \lambda) \quad (5.55)$$

From Eq. (5.1), let us introduce the spectral radiance of a blackbody $B_\lambda^{bb}(\lambda, T)$

$$B_\lambda^{bb}(\lambda, T) = \frac{2hc^2}{\lambda^5} e^{-\frac{hc}{\lambda k_b T}} \quad (5.56)$$

For clarity, $B_\lambda^{bb}(\lambda, T)$ is simply denoted as B_λ^{bb} . Thus, each location of the oxide smoke emits light with an intensity that is proportional to the product of the local spectral emission coefficient with the spectral radiance of a blackbody. This light emission travels the oxide smoke before reaching a sensor and is thus attenuated, with an efficiency proportional to the local extinction coefficient τ^{loc} . Let us consider the combustion emission intensity $I_{i,j=3}^c$ measured by the camera on the pixel $(i, j = 3)$. The light beam issued from the combustion is represented in Fig. 5.17. The measured combustion intensity is thus a sum of the emissions from each layer, attenuated by each layer located between the emitting layer and the sensor, through an exponential law [111].

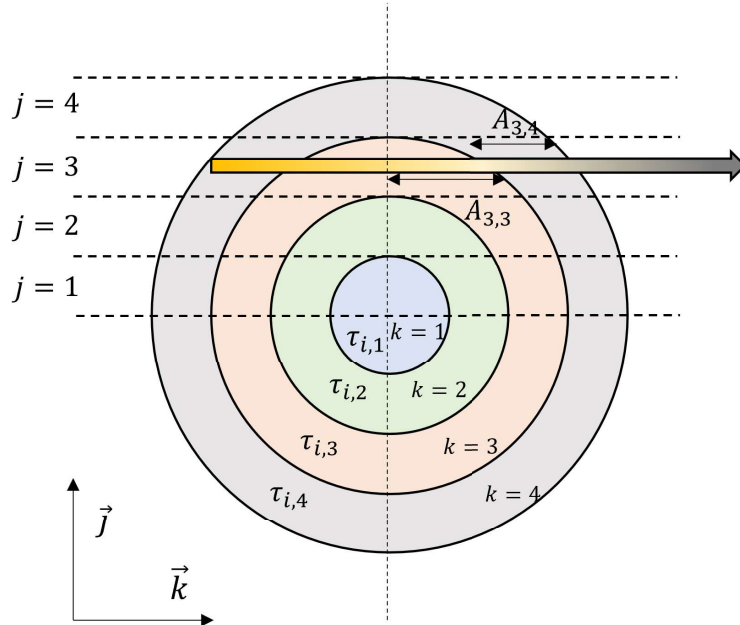


Figure 5.17: Schematics of combustion emission and self absorption.

Thus, the following proportionality can be written as follows, with the terms starting from the end of the beam's path:

$$\begin{aligned}
I_{i,j=3}^c &\propto A_{3,4}\epsilon_{i,4}^{loc} B_{\lambda,i,4}^{bb} + A_{3,3}\epsilon_{i,3}^{loc} B_{\lambda,i,3}^{bb} e^{-A_{3,4}\tau_{i,4}^{loc}} \\
&\quad + A_{3,3}\epsilon_{i,3}^{loc} B_{\lambda,i,3}^{bb} e^{-A_{3,4}\tau_{i,4}^{loc} - A_{3,3}\tau_{i,3}^{loc}} \\
&\quad + A_{3,4}\epsilon_{i,4}^{loc} B_{\lambda,i,4}^{bb} e^{-A_{3,4}\tau_{i,4}^{loc} - A_{3,3}\tau_{i,3}^{loc} - A_{3,3}\tau_{i,3}^{loc}}
\end{aligned} \tag{5.57}$$

Note than proportionality is written instead of equality. Indeed, the term on the right hand side expresses the light intensity emitted by the system. It has to be accounted for that the generated beam is transmitted to the camera via collection optics, with given efficiencies. The beam then reaches a pixel that collects beams from a given solid angle, and during a given exposure time. The beam intensity is then converted to pixel intensity with a given efficiency. The need to introduce optical constants that account for the overall described phenomena arises. From now on, the terms K_R , K_G and K_B denote constants that relate theoretical cloud emissions, the right-hand side of Eq. (5.57), to pixel intensity, the left hand side of this same equation, for the wavelengths of interest λ_R , λ_G and λ_B . Let us introduce the vector $I^c = (I_{i,j}^c)_{j=1,\dots,N}$ as the vector of integrated combustion intensities measured at pixels located at height i and with horizontal coordinates j . The vector is chosen at the height i_s that matches the green line previously introduced in Fig. 5.9. Let us introduce a vector $X = (\epsilon_{i,k}^{loc} \cdot B_{\lambda,i,k}^{bb})_{k=1,\dots,N}$ as the vector containing the local emission terms at the height i of each layer k . Then, at the height i_s , for any of the three wavelengths of interest, it comes out:

$$C \cdot (K_l)_{l=R,G,B} \cdot X = I^c \tag{5.58}$$

With the matrix C defined, for any height i , as:

$$C_{jk} = \begin{cases} 0 & \text{if } k < j \\ A_{j,k} \cdot e^{-\sum_{m=k+1}^N A_{j,m}\tau_{i,m}^{loc}} \left(1 + e^{-A_{j,k}\tau_{i,k}^{loc}}\right) & \text{if } j = k < N \\ A_{j,k} \cdot e^{-\sum_{m=k+1}^N A_{j,m}\tau_{i,m}^{loc}} \left(1 + e^{-2\sum_{m=j}^{k-1} A_{j,m}\tau_{i,m}^{loc} - A_{j,k}\tau_{i,k}^{loc}}\right) & \text{if } j < k < N \\ A_{j,k} \cdot \left(1 + e^{-2\sum_{m=j}^{k-1} A_{j,m}\tau_{i,m}^{loc} - A_{j,k}\tau_{i,k}^{loc}}\right) & \text{if } j < k = N \\ A_{j,k} \cdot \left(1 + e^{-A_{j,k}\tau_{i,k}^{loc}}\right) & \text{if } j = k = N \end{cases} \tag{5.59}$$

Similar to the M-LEM, a Tikhonov regularization is used to solve Eq. (5.58), and the three vectors $(K_l)_{l=R,G,B} \cdot X$ are evaluated at the three wavelengths of interest. Thus, locally, the product of the optical constant $(K_l)_{l=R,G,B}$, with the emission term X is known. The emission term is the product of the local emission coefficient $\epsilon_{i,k}^{loc}$ and the local spectral black body radiance $B_{\lambda,i,k}^{bb}$. As mentioned, the emission coefficient $\epsilon_{i,k}^{loc}$ is known from the extinction coefficient $\tau_{i,k}^{loc}$ evaluated via the M-LEM. The product of the local optical constant and the black body emission can thus be evaluated at the specific height i_s :

$$(K_l)_{l=R,G,B} \cdot B_{\lambda,i_s,k}^{bb} = \frac{X_{i_s,k}}{\epsilon_{i_s,k}^{loc}} = \frac{X_{i_s,k}}{P_{abs} \left((r_p)_{i_s,k}, \lambda \right) \cdot \tau_{i_s,k}^{loc}} \quad (5.60)$$

With $(r_p)_{i_s,k}$ the alumina particle radius measured in layer k at the specific height i_s . For simplicity, we write R as the radial profile built from the vector $(K_l)_R \cdot B_{\lambda_R,i_s,k}^{bb}$, G as the radial profile built from the vector $(K_l)_G \cdot B_{\lambda_G,i_s,k}^{bb}$ and B as the radial profile built from the vector $(K_l)_B \cdot B_{\lambda_B,i_s,k}^{bb}$. R , G and B are thus radial profiles of local blackbody emissions, multiplied by an optical constant. An example of such profiles is displayed in Fig. 5.18.

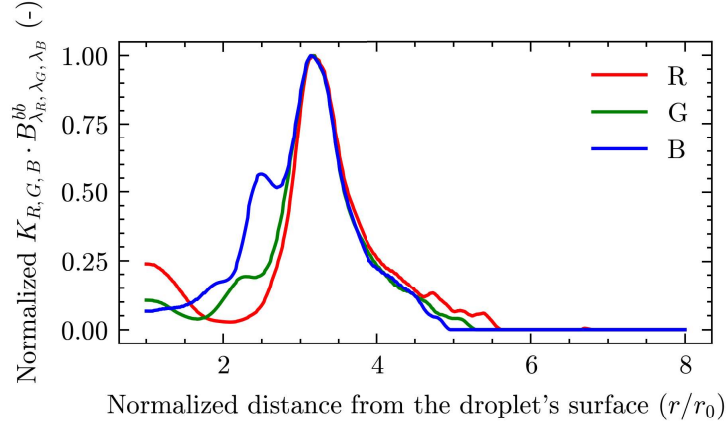


Figure 5.18: Normalized profiles of local black body spectral radiance multiplied by the optical coefficient for the three wavelengths of interest.

Although the local spectral radiance is a function of the temperature, the optical constant prevents from a direct assessment of the local temperature.

5.4.2 Temperature evaluation

Blackbody calibration

The MAE method enables the measurement of local black body emissions at the three wavelengths of interest R , G and B , which consist of the product of the Planck's black body radiance, with an optical constant. In order to obtain the temperature profile, a similar procedure to the one used for the multi-wavelength pyrometers is used. As an example, the R profile is related to the temperature along the following equation:

$$R = K_R \cdot \frac{2hc^2}{\lambda^5} e^{-\frac{hc}{\lambda_R k_b T}} \quad (5.61)$$

As previously introduced, the constant K_R accounts for geometrical efficiencies and spectral efficiencies. In order to account for these coefficients, a black body calibration, available in Appendix C, is performed. In this calibration procedure, the constants K_R are not determined, as it would result in a highly complex calibration process that would barely transpose to the experiments. Instead, the ratios of the local black body emissions

are computed: R/G , R/B and G/B . Upon doing so, the geometrical share of the optical coefficient cancels out, leaving the spectral share of this coefficient, facilitating the calibration procedure. A calibration function f is thus determined. Denoting R^{cal} , G^{cal} and B^{cal} as the pixel intensity obtained observing a blackbody at the three wavelengths of interest, the temperature T of the blackbody is obtained from the calibration function as:

$$f(R^{cal}/G^{cal}) = f(R^{cal}/B^{cal}) = f(G^{cal}/B^{cal}) = T \quad (5.62)$$

An example of the local blackbody emission ratios experimentally obtained from Fig. 5.18 is displayed in Fig. 5.19.

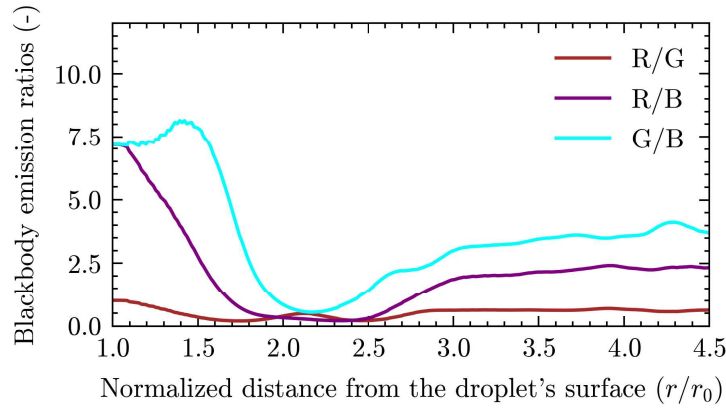


Figure 5.19: Profiles of the ratios of local black body emission (radiance multiplied by optical coefficient).

Using the blackbody calibration function, Fig. 5.19 should provide the temperature profile surrounding the burning Al droplet. However, a direct use of the calibration function on such profiles results in temperatures greater than 8000K, well above the expected temperatures. This is attributed to gaseous phase emissions, which perturbs the blackbody nature of the emission.

The influence of gaseous phase emissions

Generally, when the temperature is measured during aluminum combustion, the wavelengths at which the combustion is recorded are carefully chosen. The set of wavelengths is generally chosen in regions where gaseous phase emissions are non-existent to preserve the condensed phase nature of the observed system [65]. In recent work, Dabos [98] identified and summarized the wavelengths that are impacted by gaseous phase emissions. The wavelengths at which aluminized gases can emit are summarized in Table 5.2.

The work from Parigger et al. [112] also predicts via simulation an emission spectrum of AlO that spans from about 450 nm to about 550 nm at 3000 K. Recalling that the triple band filter enables emissions from 446 nm to 468 nm to impinge the B matrix of the camera, emissions from 520 nm to 540 nm to impinge the G matrix of the camera and emissions from 614 nm to 648 nm to impinge the R matrix of the camera, it appears that gaseous emissions must be accounted for when using the calibration law to determine the temperature profile.

Table 5.2: Emission wavelengths of Al containing gases

Wavelengths (μm)	Origin
{0.500-0.640} continuum	Chemiluminescence of AlO^*
{0.400-0.800} continuum	
0.3092 - 0.3944 - 0.3961 peaks	Al emission
0.464 - 0.486 - 0.507 peaks	AlO emission
0.429 peak	Al_2O emission
0.500 peak	AlO_2 emission

It has to be noted that this phenomenon is observable in Fig. 5.18. Indeed, in regions near $2r/r_0$, the G and B emissions are significantly high, while the R emissions remain low. In this region, the blackbody emission ratios are thus abnormally low, as observed in Fig. 5.19. It has to be noted that in the combustion sequence in Fig. 5.6, a blueish halo is observable in this same region. It is thus evidenced that gaseous phase emissions are measured, especially by the G and B matrices, and thus, by the G and B local blackbody emission profiles. The measurement of the gaseous emissions thus results in R/G and R/B ratios much lower than in the case of a condensed phase at about 4000 K. As the Planck's law attributes lower wavelengths to greater temperatures, it thus explains the incapacity of the classical method to evaluate the temperature profile surrounding the burning Al droplet. The development of a method accounting for gaseous emissions is thus required.

Numerical method to account for gaseous emissions

A direct application of the calibration function to the local blackbody emissions results in incorrect temperatures. Moreover, as the three ratios are perturbed by gaseous emissions, the emissions do not obey Planck's law. The temperatures assessed from the three ratios are thus different, in addition to being incorrect.

$$f(R/G) = T_{RG} \neq f(R/B) = T_{RB} \neq f(G/B) = T_{GB} \quad (5.63)$$

The previously called local blackbody emissions R , G and B are now simply called local emissions. These terms are assumed to be composed of a share of blackbody emission, and of a share of gaseous phase emission. Denoting the local emission from blackbody radiation at λ_R as R_{bb} , and the local emission from gaseous emissions at λ_R as R_g and similar to G and B, it can be written:

$$\begin{cases} R = R_{bb} + R_g \\ G = G_{bb} + G_g \\ B = B_{bb} + B_g \end{cases} \quad (5.64)$$

Three parameters α , β and γ , with values between 0 and 1, can be introduced to represent the share of local radiation that is attributed to local emission from blackbody radiation:

$$\begin{cases} R_{bb} = \alpha R \\ G_{bb} = \beta G \\ B_{bb} = \gamma B \end{cases} \quad (5.65)$$

These three parameters are unknown, but can be evaluated with the use of an optimization procedure at every location of the local emission profile. For each triplet (α, β, γ) , a triplet of corrected temperature is obtained when the calibration function is applied to each corrected ratio $(T_{RG}^c, T_{RB}^c, T_{GB}^c) = \left(f\left(\frac{\alpha R}{\beta G}\right), f\left(\frac{\alpha R}{\gamma B}\right), f\left(\frac{\beta G}{\gamma B}\right)\right)$. The optimization procedure relies on three constraints on these temperatures. The first constraint ensures that the blackbody nature of radiation is respected. The second one is a self-consistency constraint. Indeed at any location of the profile, if the corrected temperature is accurately evaluated, then Eq. (5.65) gives:

$$R \propto \frac{1}{\alpha} \frac{2hc^2}{\lambda_R^5} e^{-\frac{hc}{\lambda_R T^c}} = R_{rec} \quad (5.66)$$

This equation is written to illustrate a proportionality between the measured local emission R and the reconstructed local emission R_{rec} from gaseous emissions and the corrected temperature. The second optimization constraint thus ensures that the ratios (R/R_{rec}) , (G/G_{rec}) and (B/B_{rec}) remain constant throughout the entire profile. This ensures that the measured gaseous emissions and corrected temperature profile are consistent with the camera's observations. Finally the third constraint is a simple continuity constraint that prevents physically inaccurate temperature jumps from occurring in the profile. The optimization constraints as well as the related errors are summarized in the following list.

- As the temperature is measured from blackbody radiations $(\alpha R, \beta G, \gamma B)$, the three temperatures assessed via the calibration law must be equal. $T_{RG}^c = T_{RB}^c = T_{GB}^c = T^c$. If the temperatures are not equal, the system doesn't radiate according to Planck's law and gaseous emissions are not correctly accounted for. This constraint is called the blackbody constraint. The minimized associated error is based on the computation of a mean temperature $T_m = \frac{1}{3}(T_{RG}^c + T_{RB}^c + T_{GB}^c)$. The computed error is the following:

$$e_{bb} = \left(\frac{1}{3} [(T_m - T_{RG}^c)^2 + (T_m - T_{RB}^c)^2 + (T_m - T_{GB}^c)^2] \right)^{0.5} \quad (5.67)$$

- The measured gaseous emissions and corrected temperature must be consistent with camera observations. The ratios of the measured profiles to the reconstructed profiles are, thus, to remain constant throughout the entire profile. $(R/R_{rec}) = cst_R$, $(G/G_{rec}) = cst_G$ and $(B/B_{rec}) = cst_B$, at any location of the profile. The associated minimized error is based on the relative error between the normalized measured profiles X^{norm} and the normalized reconstructed profiles X_{rec}^{norm} :

$$e_{sc} = \frac{100}{3} \cdot \left(\frac{|R^{norm} - R_{rec}^{norm}|}{R^{norm}} + \frac{|G^{norm} - G_{rec}^{norm}|}{G^{norm}} + \frac{|B^{norm} - B_{rec}^{norm}|}{B^{norm}} \right) \quad (5.68)$$

Table 5.3: Summary of the optimization constraint errors for separate cases and averaged errors.

Experimental case	$e_{bb} (K)$	$e_{sc} (\%)$	$\sigma_{bb} (K)$	$\sigma_{sc} (\%)$	$e_{bb}^{mean} (K)$	$e_{sc}^{mean} (\%)$
1	20.2	6.9	21.4	6.4		
2	10.6	4.6	11.9	1.9		
3	18.7	3.5	15.6	2.3	14.8	4.8
4	12.0	4.6	12.0	4.3		
5	12.5	4.4	15.8	4.5		

- The temperature profile must be continuous throughout the entire profile. This constraint ensures that the temperature at location z is close to the temperature measured at location $z + 1$: $T(z) \approx T(z + 1)$. The associated error is computed as follows:

$$e_c = (T^c(z - 1) - T^c(z))^2 \quad (5.69)$$

The triplet of coefficients (α, β, γ) is thus optimized at each location of the profile, using a genetic algorithm, minimizing the product of the three introduced errors. Table 5.3 summarizes the measured errors along the blackbody and the self-consistency constraints over five experimental cases.

An example of the three temperatures profiles obtained after the optimization procedure is presented in Fig. 5.20. This figure illustrates the respect of the blackbody and the continuity constraints.

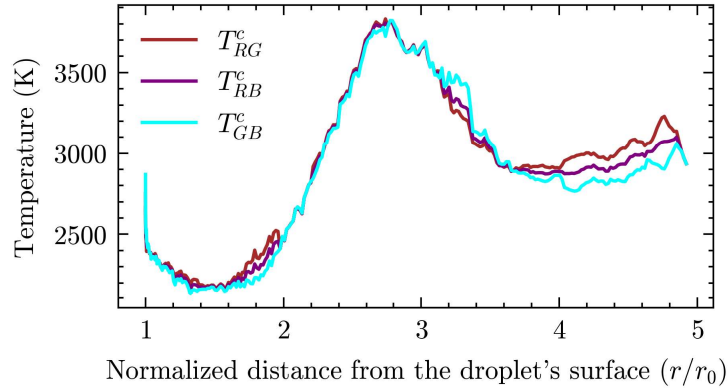


Figure 5.20: Temperature profiles from the corrected local emission ratios $\left(\frac{\alpha R}{\beta G}, \frac{\alpha R}{\gamma B}, \frac{\beta G}{\gamma B}\right)$.

The respect for the self-consistency constraints is evidenced by Fig. 5.21. The normalized reconstructed local emission profiles display an almost perfect fit to the measured local emission profiles.

With the success of the optimization procedure, the alumina temperature profile surrounding the burning Al droplet is evaluated. Additionally, the coefficients α , β and γ are evaluated

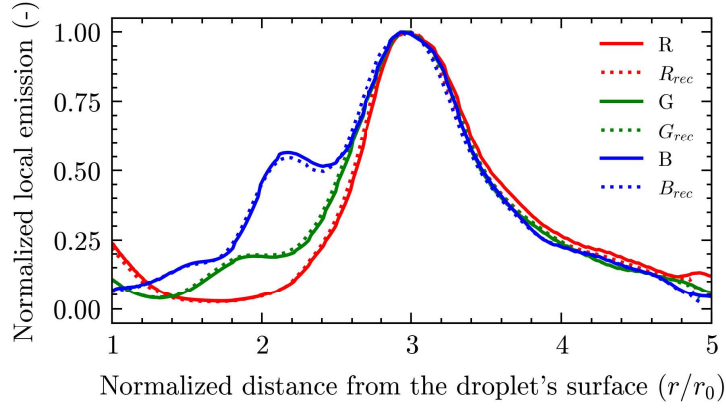


Figure 5.21: Normalized measured local emission profiles compared to normalized reconstructed local emission profiles from Eq. (5.66).

as a result of the optimization procedure. From Eq. (5.64) and (5.65), the emission profile of gaseous emissions can be assessed:

$$\begin{cases} R_g = (1 - \alpha) R \\ G_g = (1 - \beta) G \\ B_g = (1 - \gamma) B \end{cases} \quad (5.70)$$

Figure 5.22 displays the three gaseous profiles obtained from this equation.

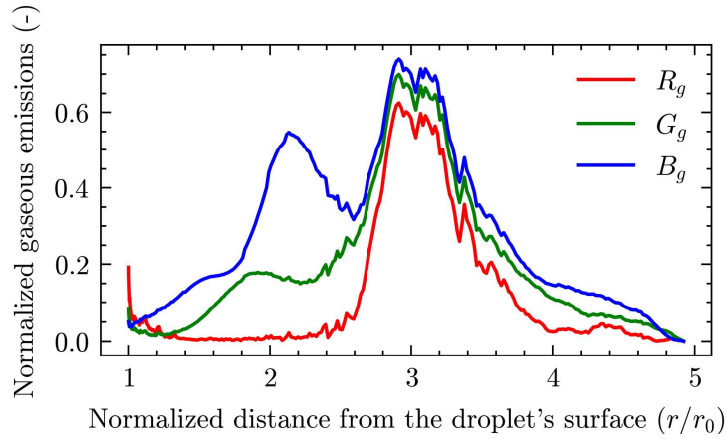


Figure 5.22: Gaseous emission profiles normalized by the respective maximum local emissions, evaluated by Eq. (5.70).

Note that gaseous emissions in the blue and green regions of the spectrum are measured at a region of about 2 droplet radii, as expected earlier. Moreover, significant gaseous phase emissions are measured at about 3 droplet radii. These emissions are attributed to an electronic emission through a Bremsstrahlung process, discussed in the following chapter.

5.4.3 Summary

Images of combustion intensities, without the backlight, are measured by the color camera. Each pixel intensity is obtained from a sum of local contributions. The local intensities consist of a product of the blackbody radiance, function of the temperature, with an emission coefficient measured via the M-LEM. A deconvolution method accounting for self-extinction is performed to retrieve the local intensities at any locations in the cloud. With knowledge of the emission coefficient, the product of an optical constant with the blackbody radiance is known. The use of a blackbody calibration enables the assessment of the temperature profile using the ratio of local blackbody emissions. However, gaseous emissions are evidenced to perturb the measurements, preventing a direct assessment of the temperature. An optimization procedure is developed to split the gaseous emissions from blackbody emissions. As a result of this procedure, the temperature profile of alumina is assessed, and the normalized profiles of gaseous phase emissions are obtained. The entire process is summarized in Fig. 5.23.

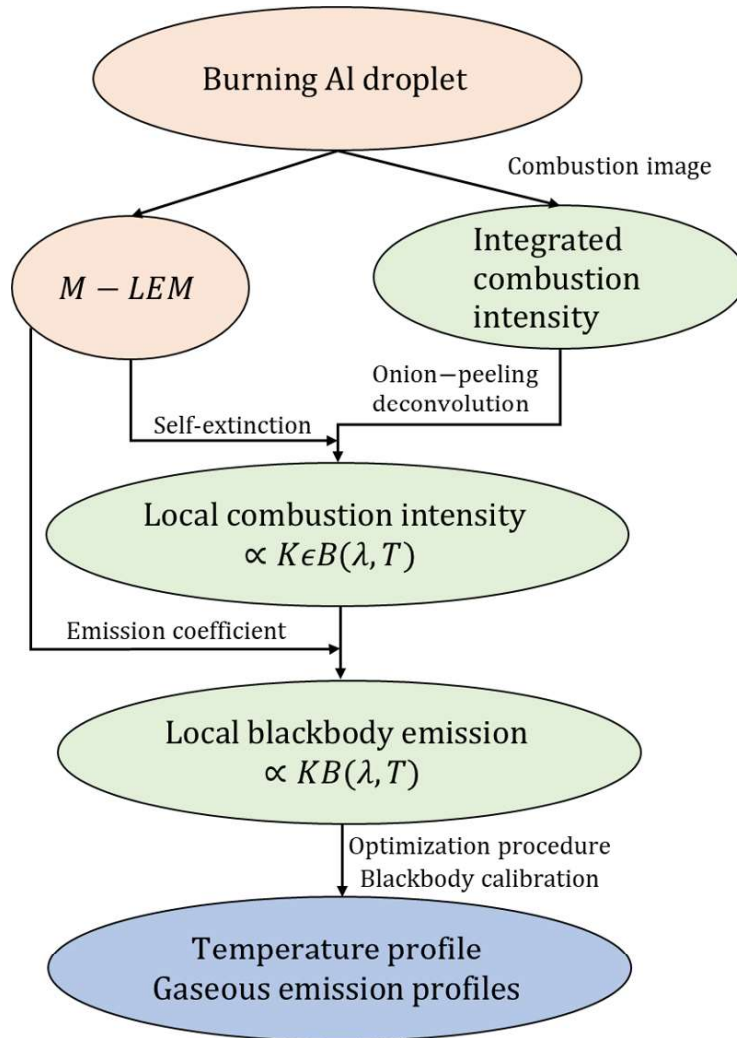


Figure 5.23: Summary of the MAE method.

5.5 Limitations

In this section, the limitations of the method are listed.

- In order to perform the Onion-peeling deconvolution procedures, the system must maintain a spherical or cylindrical symmetry. Thus, the often chaotic ignition phase and extinction phase of the Al droplet cannot be characterized. Similarly, burning conditions that result in chaotic regimes cannot be characterized (highly oxygenated environments or high pressure environments)
- For the M-LEM to be applied, the oxide cloud must be sufficiently attenuating, while not being opaque. Burning conditions for which the concentration of alumina is too low cannot be characterized. Similarly, at high pressure, the opacity of the cloud results in the inefficiency of the method.
- As a pair of images is required for the global process, using a high frame rate for the camera is encouraged, limiting the time difference between the two images. However, high acquisition frequencies result in low exposure times, which can then lead to the inefficiency of the method if the cloud's intensity is too low. Thus, at low cloud temperatures, and thus low condensed phase emissions, the camera's frame rate must be decreased, penalizing the method's accuracy.
- In order to use the M-LEM method, the alumina's refractive index must be assessed. As this parameter is a function of the temperature, an assumption must be made as temperature is not known at this time of the method. Although not resulting in significant changes, this could be improved.
- Given the complexity of the method, the assessment of an experimental uncertainty is of high complexity, as the error must be propagated through the entire procedure. Note, however, that a pixel value fluctuation of about 1% around the mean pixel value has been measured. This fluctuation can result in an 8% error on the optical depth. Nonetheless, this error nullifies upon applying Eq. (5.48) as the error on one pixel is the same for the three matrices. To overcome the limitation of uncertainty assessment, multiple tests are carried out in each condition in order to evaluate a 95% confidence interval.
- If a condensed species, different from alumina, was to be produced in the cloud, it would be inaccurately considered as alumina.

Chapter 6

Experimental Results

In this chapter, the experimental results obtained from the application of the M-LEM/MAE methods to the combustion of Al particles are introduced. The presentation of the acquired data aims at providing a complete characterization of the combustion of aluminum in various conditions. Thus, the experimental cases used throughout this study are introduced. Following this, the structure of the measured profiles is discussed. Since all experimental cases yield comparable results, the focus of our discourse is solely on combustion in ambient air. Instead of presenting a comprehensive list of profiles, we will highlight crucial combustion parameters and examine their sensitivity to burning conditions. This will allow us to elucidate the role of the medium's properties in influencing aluminum combustion.

6.1 Experimental cases

Al particles have been burned in mixtures of dioxygen O_2 and various diluents: nitrogen N_2 , argon Ar and helium He . Oxygen concentration and pressure have been varied. A test in a mixture containing carbon dioxide CO_2 as the oxidizer has also been performed. In each condition, about 10 experiments were performed, and about half of these experiments were selected to be processed. The other half was not apt for processing, due to asymmetric combustion regimes, particles moving out of focus, the prevailing oxide cap, etc. The summary of the selected experimental cases is presented in Table 6.1.

Air has been selected as a reference case due to its ease of implementation, making this the most fundamental condition. Nitrogen is likely to react with oxygen and aluminum to form nitrogen oxides and aluminum oxynitrides. In order to assess the influence of such mechanisms, nitrogen has been replaced with argon due to its similarity in its physical properties. In the case of argon, an inert gas, the oxygen concentration has been varied to observe the influence of such changes on the combustion. The helium replaced argon to assess the influence of a significant change in the physical properties of the diluent, switching from a relatively heavy diluent to the lightest one, significantly influencing the diffusivity of the medium. Finally, the influence of a carbonized species is studied with carbon dioxide selected as the oxidizer. In most cases, pressure was increased up to the symmetric combustion limit. That is to say that a chaotic combustion regime was met with pressures slightly above the studied pressures.

Table 6.1: Summary of the processed experimental cases.

Oxidizer	(%)	Diluent	(%)	Pressure (bar)	Number of processed tests
O_2	21	N_2	79	1	5
O_2	21	N_2	79	2	5
O_2	21	N_2	79	3	6
O_2	21	N_2	79	5	4
O_2	21	N_2	79	6	2
O_2	21	Ar	79	1	4
O_2	21	Ar	79	2	4
O_2	21	Ar	79	3	5
O_2	21	Ar	79	4	3
O_2	15	Ar	85	2	4
O_2	15	Ar	85	3	4
O_2	15	Ar	85	5	6
O_2	30	Ar	70	1	5
O_2	21	He	79	3	3
O_2	21	He	79	5	5
O_2	21	He	79	7	5
O_2	21	He	79	9	6
O_2	21	He	79	11	6
CO_2	60	N_2	40	2	5

In order to provide visual information prior to the analysis of the results, a combustion image, taken in the symmetric burning regime, is displayed for each case in Figs. 6.1, 6.2, 6.3 and 6.4.

6.2 Profiles - Atmospheric air

In this section, the profiles obtained in the case of atmospheric air are discussed. The profiles are obtained as the data averaged from the five tests performed in air. Each profile is thus presented along with its 95 % confidence interval, obtained via the calculation of the standard deviations among these five tests. This discussion will describe the knowledge that is provided by these profiles and will thus facilitate the evaluation of the influence of the burning conditions.

6.2.1 Alumina volume fraction

The alumina volume fraction profile surrounding the Al droplet burning in air is presented in Figure (6.5). A high concentration of alumina is measured at the surface of the droplet. Although never predicted by simulations [47, 46, 60], this high concentration near the droplet's

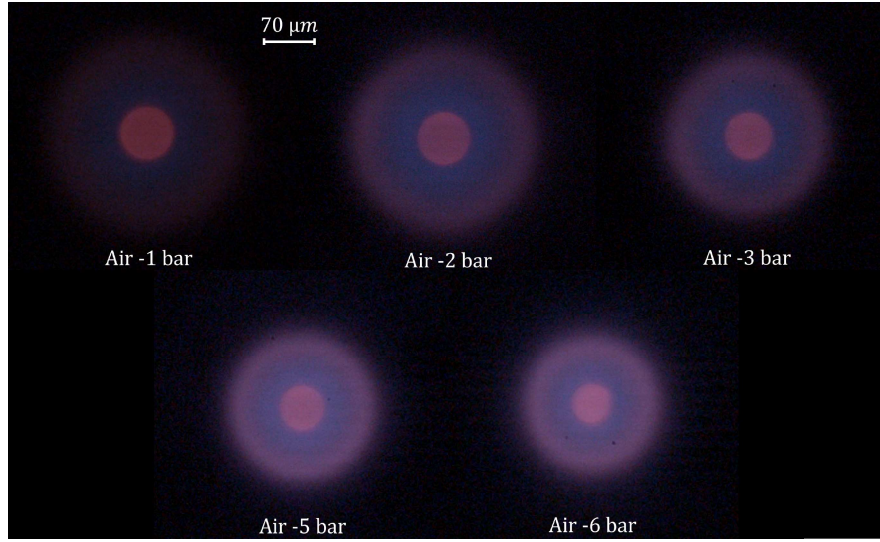


Figure 6.1: Images of Al combustion in pressurized air. Camera settings are kept identical.

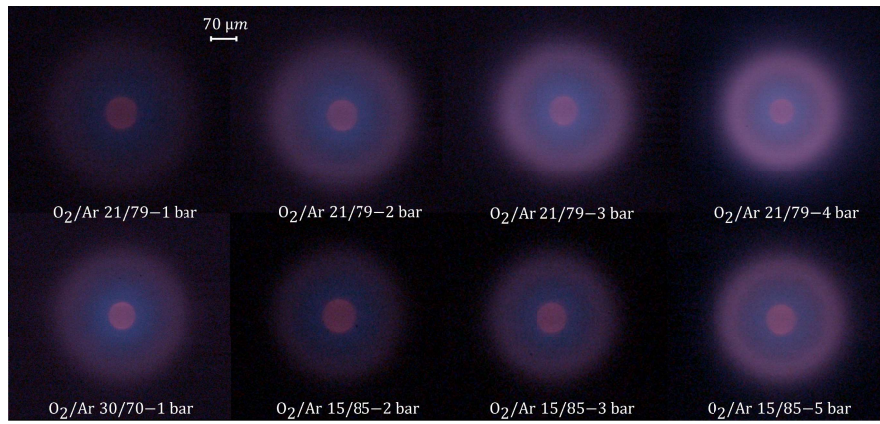


Figure 6.2: Images of Al combustion in O_2/Ar mixtures, at various O_2 concentrations and pressures. Camera settings are kept identical.

surface has recently been observed by Ruan et al. in the case of aluminum burning in a steam environment [113]. The current data isn't sufficient to bring definitive conclusions as to the origin of this alumina. Two hypotheses are, however, proposed. The first hypothesis consists in considering that the alumina, produced further away from the droplet, is transported to the droplet's surface by phoretic motions. The influence of the phoretic forces on aluminum combustion has already been studied to explain the dynamics of the oxide cap [59]. In this study, thermophoresis is shown to be an efficient mechanism to transport alumina towards the droplet, prevailing over the aluminum Stefan flow that is directed towards the outer edges of the system. The second hypothesis assumes that surface reactions, already proven to be of high importance [49], result in the production of alumina at the droplet's surface. As surface reactions can occur, near-surface reactions are also to be considered. This can result in the generation of alumina droplets in this region of the cloud. The combination of these two hypotheses could result in the observed high alumina concentration at the Al droplet's

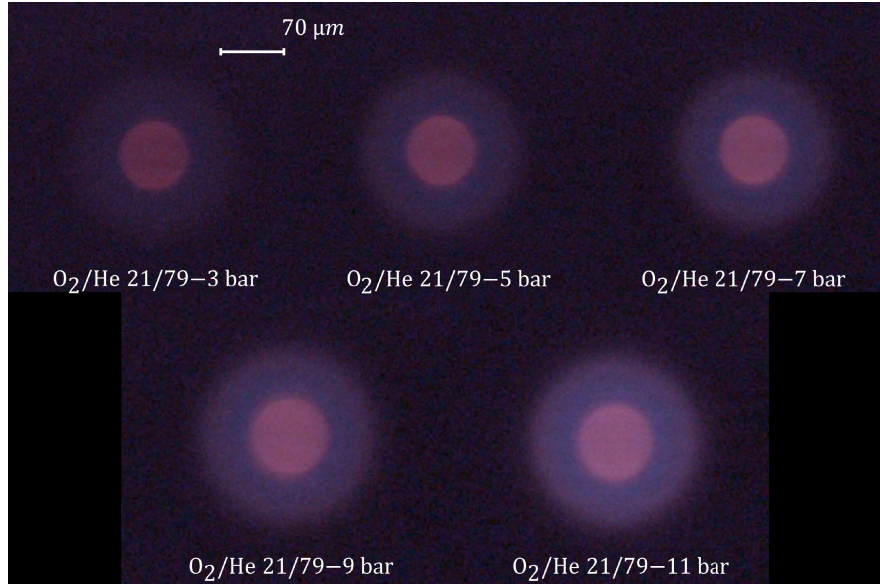


Figure 6.3: Images of Al combustion in an O_2/He (21/79) % vol. mixture, at various pressures. Camera settings are kept identical.

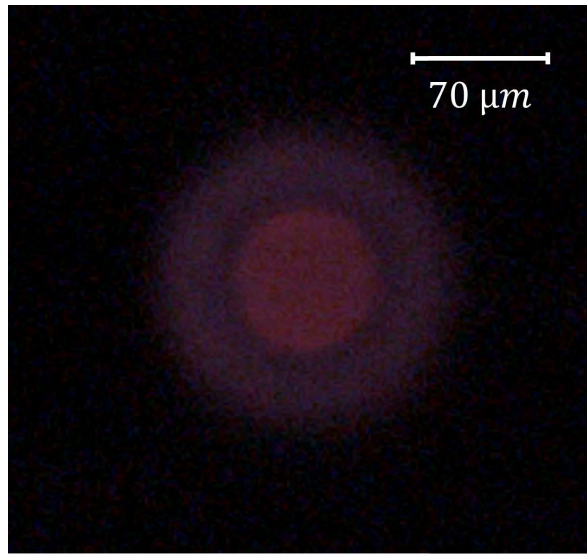


Figure 6.4: Image of Al combustion in a CO_2/N_2 (60/40) % vol. mixture, at 2 bar.

surface. From the droplet's surface to about 2.5 droplet radii, the concentration of alumina decreases. Then, from 2.5 droplet radii to about 3.5 droplet radii, alumina concentration increases and a local maximum is reached. The share of alumina produced in the cloud that is not transported to the droplet's surface is transported away from the droplet by the Stefan flow and thermophoresis. It is, however, assumed that the oxidizer flow to the reaction zone slows the outward transport of alumina particles, resulting in an accumulation region of alumina, depicted by this local maximum in concentration. At greater distances from the droplet's surface, alumina concentration decreases monotonically.

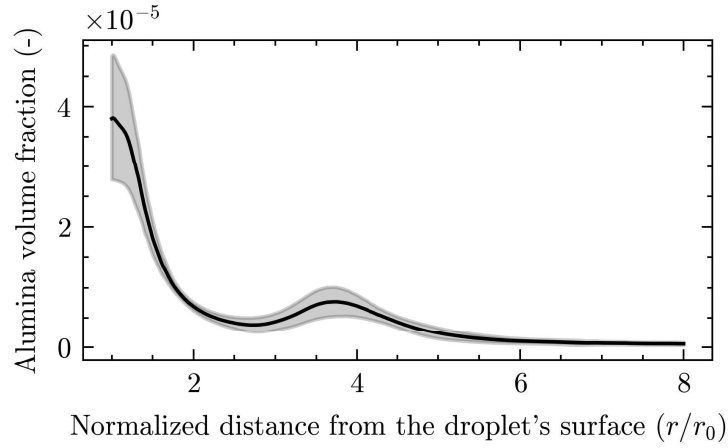


Figure 6.5: Alumina volume fraction profile and the associated 95 % confidence interval obtained in atmospheric air.

6.2.2 Alumina size

The alumina size profile measured surrounding an Al particle burning in atmospheric air is presented in Figure 6.6. The 95 % confidence interval is added and evidences some consistency in the measurements. The alumina particles are measured to be large near the droplet's surface, with a radius of about 200 nm. Further away from the droplet, the alumina particles reach a minimum radius of about 67 nm at 4 droplet radii. This radius remains relatively constant further from the droplet

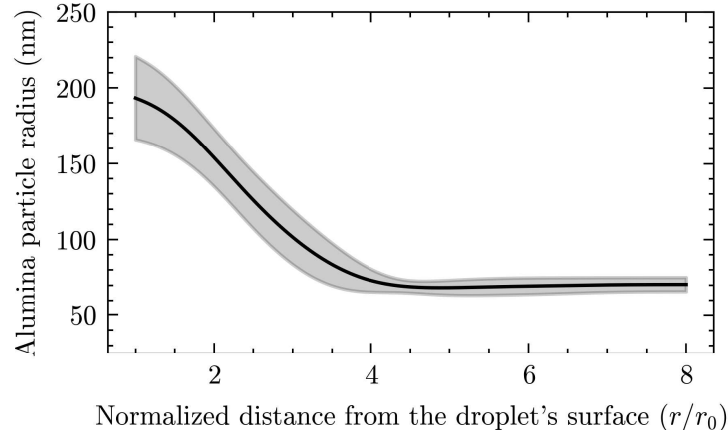


Figure 6.6: Alumina particle size profile and the associated 95 % confidence interval obtained in atmospheric air.

As alumina particles get transported towards the Al droplet, the interdistance between the particles decreases and this results in an increase in the collision frequency between the alumina droplets. The alumina produced via the potential surface reactions also plays in favor of an increase in collision frequency. Thus the coalescence between droplets is favored,

possibly resulting in the generation of larger droplets. In addition, lower temperatures favor the growth rate of alumina particles according to the Classical Nucleation Theory (CNT) [51]. As near the droplet, the temperature decreases from the flame temperature to the aluminum droplet's temperature, the growth of existing alumina particles is favored. In contrast, the alumina particles that are driven away from the droplet's surface undergo a cooling phase during which the collision frequency decreases due to an increasing distance between particles and thus, coalescence is curtailed. Similarly, the temperature gradient is less important for particles being driven away from the droplet's surface, leading to a lower growth rate than in the vicinity of the droplet. Also note that the density of alumina increases when the temperature drops. Thus, further from the Al droplet, alumina droplets will tend to shrink due to this increase in density.

6.2.3 Temperature

The temperature profile surrounding an Al droplet burning in atmospheric air is displayed in Figure 6.7 and a discussion is proposed.

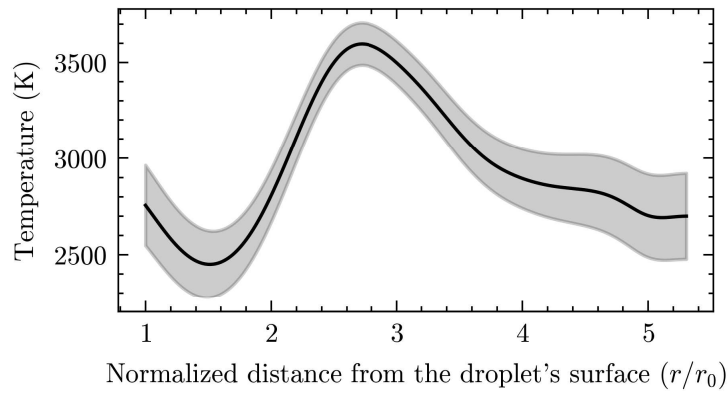


Figure 6.7: Temperature profile and the associated 95 % confidence interval obtained in atmospheric air.

Maximum temperature

At about 2.7 droplet radii from the center of the droplet, the temperature rises and reaches a maximum temperature of 3600 K, near the maximum temperature measured by Bucher in similar conditions [45], and close to, but below, the adiabatic flame temperature predicted by the CEA code of 3728 K for a stoichiometric condition with an initial droplet temperature of 2500 K. Further from the droplet, the temperature monotonically decreases, due to the thermal losses to the environment, and diffusion of the hot condensed species to colder regions of the cloud.

Surface temperature

At the Al droplet's surface, a temperature of 2755 K is evaluated, with a confidence interval ranging from 2550 K to 2960 K. Although the measured temperature is near the boiling

temperature of aluminum, the Al partial pressure is expected to be below 1 bar and generally, the droplet temperature is expected at about 2550K [46]. This temperature remains within the evaluated confidence interval. The discrepancies between the expected temperature and the measured temperature can either be attributed to measurement noise amplified by Abel transforms, or to the presence of surface reactions that locally increase the temperature above aluminum’s boiling point. It is also to be considered that the partial pressure of gaseous aluminum at the droplet’s surface could be underestimated in the numerical simulations, leading to numerical temperatures lower than the measured temperature.

Near surface temperature

Moving away from the droplet’s surface, the temperature decreases to reach a minimum of about 2500 K at about 1.5 droplet radii. This decrease in temperature can be attributed to the occurrence of local endothermic reactions, or to a local change in the alumina’s structure as the temperature approaches the melting point of alumina, resulting in changes in the optical properties of the droplet and thus, to a potentially inaccurate assessment of the temperature at this location of the cloud. Firstly identified by Bucher, with the $Al + N \rightarrow AlN$ reaction, endothermic reaction can occur within the oxide cloud [45]. Saba [50] et al. identified in recent work the main oxidation path for aluminum. He concludes that once AlO is formed, whether is it formed from Al or AlO_2 , it reacts with itself to produce Al_2O releasing an oxygen atom. Then, the main oxidation reaction of Al_2O is identified as $Al_2O + O_2 \rightarrow Al_2O_2 + O$. This reaction is endothermic and could locally lead to a thermal sink. Other non-prevailing reactions of Al_2O with dioxygen to produce $Al + AlO_3$ or $AlO + AlO_2$, and reactions of Al_2O with monoatomic oxygen to yield $AlO + AlO$ or $Al + AlO_2$ are also identified as endothermic [50]. Close to 1.5 droplet radii, such endothermic reactions could provide the thermal sink observed in Figure 6.7. An in depth study of the reaction path of aluminum oxidation and the local concentration of aluminum suboxides and oxygen would be required to draw a definitive conclusion on this matter.

6.2.4 Gaseous emissions

The gaseous emissions measured on the R, G and B matrices of the camera are presented in Figure 6.8, and a discussion is proposed.

Near droplet AlO emissions

At 2 droplet radii from the droplet’s center, green and blue gaseous emissions are measured, while red gaseous emissions are negligible. As highlighted by Dabos et al. [98] and Parigger et al. [112], the AlO shows a broad emission spectrum that ranges between 450 nm and 550 nm. As the triple band filter enables the wavelengths close to 450 nm and 530 nm to respectively impinge the B and G matrices of the camera, the AlO emissions are measured. As AlO shows no emissions in the red region of the spectrum, this corroborates the low measured red gaseous emissions at this location. It can be assumed that AlO concentration is important at 2 droplet radii. From 2 to 2.5 droplet radii, the green and blue gaseous emissions slightly decrease, evidencing a decrease in AlO concentration.

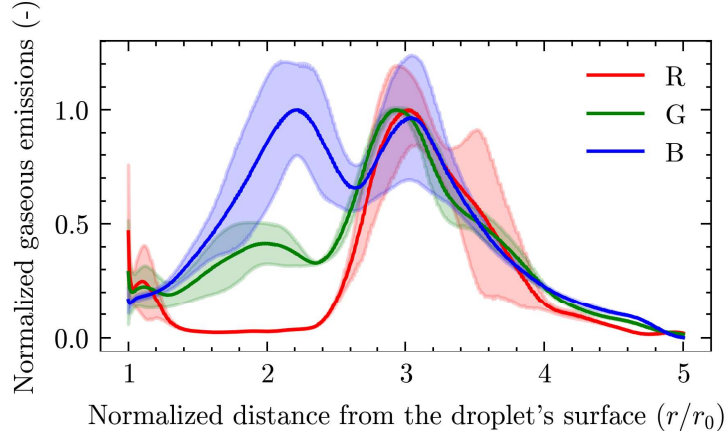


Figure 6.8: Gaseous emission profiles measured at λ_R , λ_G and λ_B and the associated 95 % confidence intervals obtained in atmospheric air.

Negligible surface emissions

Negligible gaseous emissions are measured at the surface of the burning Al droplet. This is expected, as in the vicinity of the droplet, the main species that are expected with the highest concentrations are gaseous aluminum, condensed alumina, and oxygen. These species are not predicted to demonstrate strong gaseous emissions in the spectral range of the triple band filter.

Multispectral emissions from the reaction zone

At 3 droplet radii, unexpected gaseous emission are measured. In addition, the measured emissions span over the entire visible spectrum as they are detected on the three matrices of the camera. Usually attributed to condensed phase emissions, this continuous spectrum does not originate from alumina emissions, as it differs from a conventional spectrum expected from the Planck's law. The gaseous emissions between 2.8 and 4 droplet radii can be averaged, and a normalized emission spectrum can be identified. This spectrum, built from the three wavelengths at which the gaseous spectrum is measured, is presented in Figure 6.9 and could be attributed to light emission from electrons. As observed by De Iuliis et al. [99] while studying titanium flames using a laser-induced incandescence method, the presence of electrons at elevated temperatures results in light emissions that perturb the temperature measurements with classical pyrometry. Further studied in various works [88, 87], this light emission results from the overheating of electrons during the condensation process of alumina. The presence of overheated electrons can result in a light emission process called Bremsstrahlung, due to the slowing of electrons in a medium. The Bremsstrahlung spectral emission $\epsilon^B(\lambda, T_e)$ is expressed as a function of the electronic temperature T_e [114]:

$$\epsilon^B(\lambda, T_e) = K \cdot T_e^{-1/2} e^{-\frac{hc}{\lambda k_b T_e}} \cdot g_f \quad (6.1)$$

K is a parameter that contains the influence of electron concentration and ion concentration and charges, not described here. h is the Planck constant, k_b is the Boltzmann constant and

c is the celerity of light. g_f is called the gaunt factor and can be approximated as:

$$g_f \propto \log \left(\frac{\lambda k_b T_e}{hc} \right) \quad (6.2)$$

At a given electronic temperature, a proportionality for the Bremsstrahlung emission can be written:

$$\epsilon^B(\lambda, T_e) \propto e^{-\frac{hc}{\lambda k_b T_e}} \cdot \log \left(\frac{\lambda k_b T_e}{hc} \right) \quad (6.3)$$

From this, at a given electronic temperature, the shape of the emission spectrum resulting from Bremsstrahlung can be computed. Note that, usually, the electron excitation energy can be several eV, resulting in a few tens of thousands of degrees in electronic temperature [88]. A Bremsstrahlung spectrum is thus fitted to the measured gaseous emission spectrum, in Figure 6.9, and an electronic temperature of about 21 000 K seems to result in a relatively good fit.

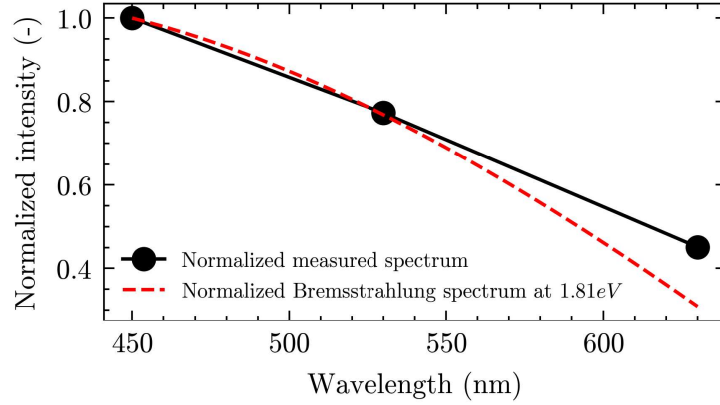


Figure 6.9: Gaseous emission spectrum compared to a Bremsstrahlung spectrum computed at 1.81 eV.

The work function of a material is the energy required to remove an electron from this material. In the literature, no available data can be found on the work function of alumina and thus, no comparison can be proposed. Note that the location of the peak in electronic emissions matches the location of the maximum temperature. As alumina condensation is the highest exothermic reaction, the formation of liquid alumina droplets results in a high temperature region, where the energy is sufficient for electrons to exit the condensed alumina. These produced electrons have a given energy that can be lost through Bremsstrahlung, emitting light.

6.2.5 Cloud structure

The knowledge of the temperature, alumina concentration, and gaseous emission profiles enable the identification of a flame structure, displayed in Figure 6.10. Between 1 and 2 droplet radii, the temperature remains close to the droplet's temperature. It is hypothesized

that the thermophoresis from the flame region, in conjunction with possible surface reactions, prevails compared to the Al Stefan flow. This results in an important concentration of alumina in this region. As alumina concentration is high, the coalescence of alumina particles is favored. In addition, as the temperature is low, the growth rate of alumina particles increases, as predicted by the classical nucleation theory, and highlighted by equations 52 and 53 of Ref. [51]. Both these mechanisms favor the formation of large alumina particles. At 2 droplet radii, aluminum begins its oxidation process, and AlO is detected. Upon reaching 3 droplet radii, AlO is oxidized to form alumina. Condensation of alumina generates high temperatures and emits light through Bremsstrahlung by generating electrons. A share of this produced alumina is transported to the droplet's surface by thermophoresis, while the other is transported outside of the system. At 4 droplet radii, it is hypothesized that the diffusion of the oxidizer to the flame region slows the transport of alumina towards the environment, generating a region of alumina accumulation. In this region, the temperature is greater than near the Al droplet and the distance between alumina particles is greater than near the droplet due to a lower volume fraction of alumina. Alumina coalescence isn't favored, and the growth rate of alumina droplets being low at high temperatures, this results in the observation of small alumina droplets.

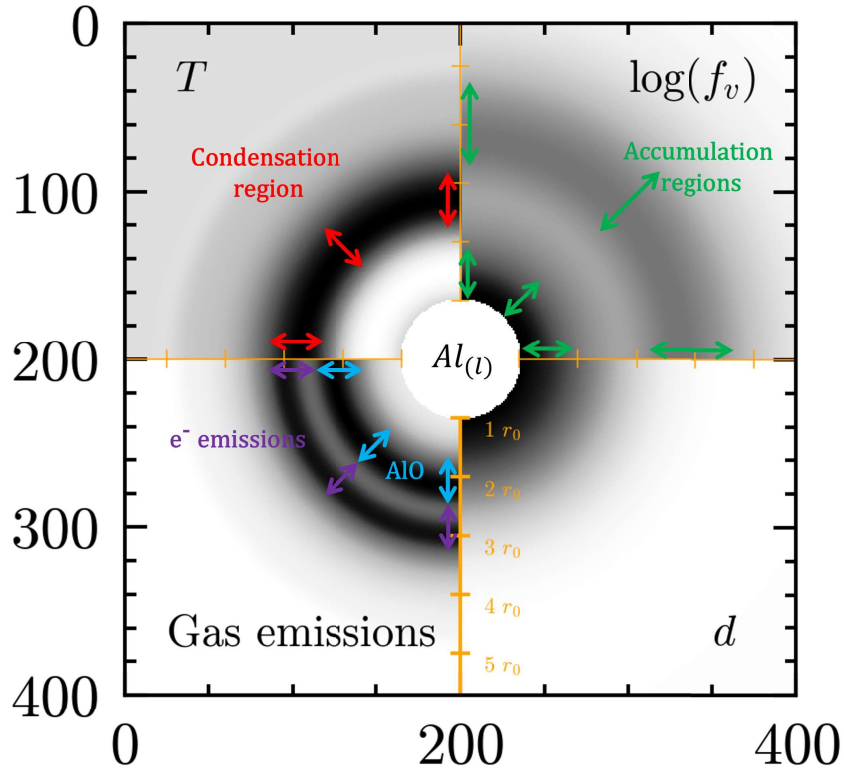


Figure 6.10: Identified cloud structure. All values are normalized with respect to their own maximum. Gaseous emissions are represented at λ_B .

With the complete description of the results in the case of atmospheric air, the influence

of the medium on key combustion parameters can be studied. These key parameters are first introduced.

6.3 Key combustion parameters (KCPs)

The previously introduced method enables the measurement of the alumina size, alumina concentration, alumina temperature, and normalized gaseous emissions profiles surrounding a burning Al droplet. These four profiles are crucial to enable the characterization of the influence of the medium on the combustion process. However, as 19 experimental conditions have been studied, Key Combustion Parameters (KCPs) are identified to avoid displaying too many profiles. These profiles are nonetheless available in Appendix D.

Temperature

A first set of KCPs is identified in the temperature profile. The temperature at the droplet's surface T_{surf} , the maximum temperature of the profile T_{max} and the radial location of this maximum $r/r_0(T_{max})$ are selected as KCPs regarding temperature. These KCPs are represented, in the case of atmospheric air, in Figure 6.11. Throughout this work, it will also be considered that the location of the maximum temperature is the Stand-Off Ratio (SOR) of the flame.

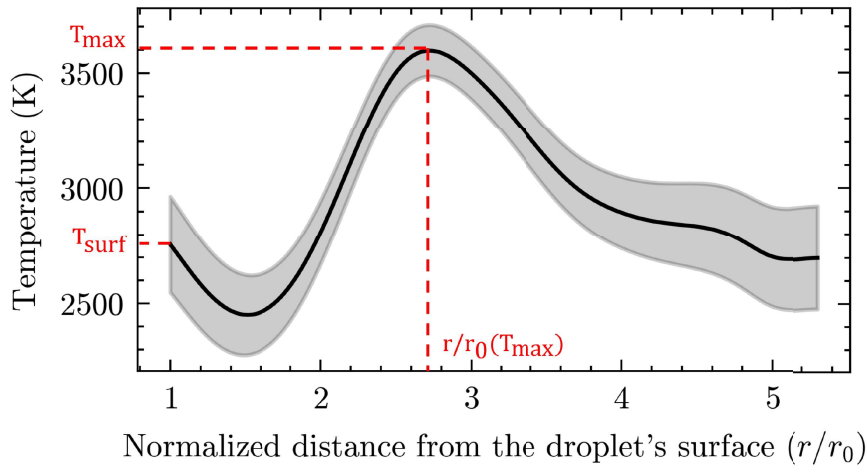


Figure 6.11: KCPs related to temperature.

Alumina concentration

The set of KCPs regarding volume fraction is rather similar to the KCPs selected for temperature. The local maximum in alumina volume fraction (excluding the surface peak) as well as the radial location of this maximum are selected. The volume fraction at the surface of the droplet is not considered as a KCP. The KCPs are represented, in the case of atmospheric air, in Figure 6.12.

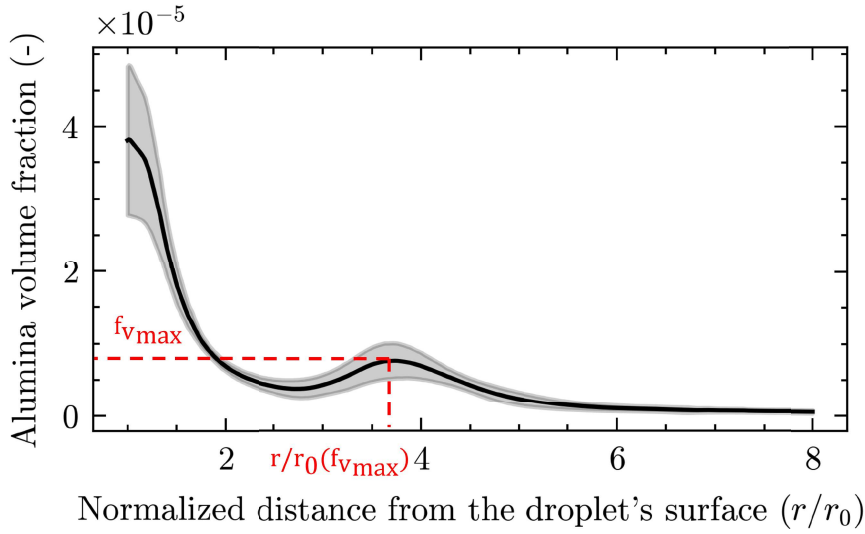


Figure 6.12: KCPs related to alumina concentration.

Alumina particle size

A single KCP is identified for the size of alumina particles. In this work, the influence of the conditions on alumina particle size will only be studied regarding the average alumina particle size. For each case, alumina particle size profiles are averaged to provide a single average value.

Gaseous emissions

Gaseous emissions contain a wide variety of information, as the profiles contain *AlO* emissions and electronic emissions. The first KCP enables the characterization of *AlO* formation, as the location of the *AlO* peak is studied. The last two KCPs characterize electronic emissions. The location of the peak of electronic emissions is studied, along with the associated electronic temperature T_e calculated by fitting a Bremsstrahlung spectrum to the measured spectrum. These KCPs are summarized in Figure 6.13.

6.4 Influence of the medium on the KCP

In this section, the influence of the medium on the key combustion parameters is studied. Every KCPs are presented for all the combustion cases presented in Table 6.1, and the properties of the medium that influence these KCPs are identified. When available, data from three references in the literature are added to the presented results. The chosen references are the numerical work from Beckstead et al. [47], the experimental work from Bucher et al. [45], and the numerical work from Muller et al. [46]. Note that these three studies are for Al particles of diameters greater than 200 microns, while the initial diameter of the particles in this work is near 70 microns.

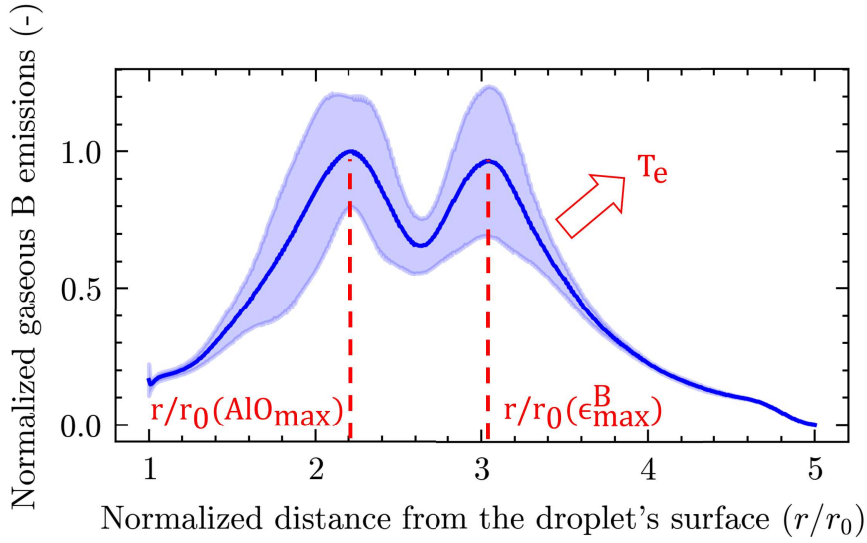


Figure 6.13: KCPs related to gaseous emissions.

6.4.1 Temperature

In this section, the influence of the burning conditions on the KCPs related to the temperature is studied.

Maximum temperature

The evolution of the maximum measured temperature for the various experimental cases is displayed in Figure 6.14.

The maximum temperature displays a low dependency on the pressure. Indeed, slight increases in maximum temperature are observed as the pressure increases. The nature of the diluent appears to have a stronger effect on the maximum temperature as, for similar oxygen concentrations, lower temperatures are measured in the case of helium than in the case of argon or nitrogen. Note, however, that the argon and nitrogen cases seem to display similar maximum temperatures. In O_2/He 21/79 % (vol.) mixtures, temperatures are measured ranging from 3560 K at 3 bars to 3690 K at 11 bars. In the case of air and O_2/Ar mixtures, the temperatures remain relatively close. In air, the maximum temperature varies from 3600 K at 1 bar to 3800 K at 3, 5 and 6 bars. In the case of O_2/Ar 21/79 % (vol.), the temperatures are greater than in air at low pressures, ranging from 3670 K at 1 bar to 3770 K at 3 bars, but then the temperature decreases with 3750 K at 4 bars.

Varying the oxygen concentration also influences the maximum flame temperature. Upon increasing the oxygen concentration at 1 bar, the temperature increases, as a temperature of 3700 K is measured for the O_2/Ar 30/70 % (vol.) mixture while a temperature of 3670 K is measured for the 21 % oxygen mixture. Similarly, with a decreasing oxygen concentration, the measured temperature is lowered. Indeed, in the O_2/Ar 15/85 % (vol.) mixture, the temperature is 3650 K at 2 bars while a temperature of about 3700 K is measured for the 21 % oxygen mixture at the same pressure.

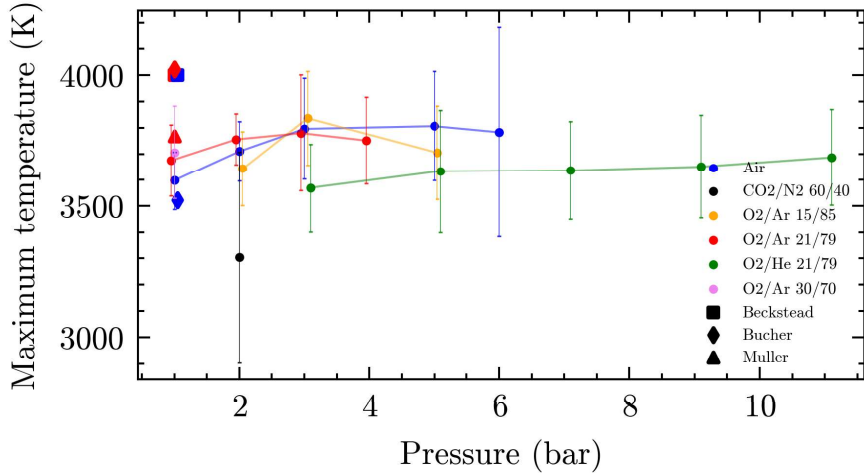


Figure 6.14: Maximum temperature measured for all experimental cases. Error bars represent the 95 % confidence interval. A negligible horizontal offset has been added to pressure values to facilitate the reading of the graph. The compositions are in % (vol.). Literature data has been added for comparison with similar colors to experimental data.

The nature of the oxidizer strongly influences the maximum flame temperature as using CO_2 as an oxidizer significantly decreases the flame temperature. Indeed, the lowest flame temperature of 3300 K is measured in the CO_2/N_2 60/40 % (vol.) case at 2 bars, with a large confidence interval due to the asymmetry of the combustion process.

The simulations from Muller in argon, as well as the experimental work from Bucher in air, are relatively close to experimental measurements. However, simulations from Beckstead as well as the experiments from Bucher in argon show high temperatures, near the dissociation temperature of alumina. To analyze these temperatures, theoretical adiabatic flame temperatures obtained with the CEA code are calculated and displayed in Figure 6.15. Note that CO_2 calculations cannot be performed with this code.

In the case of helium as a diluent, the correlation between the measured temperatures and the calculated flame temperatures is particularly good. The temperatures as well as the evolution with pressure are accurately predicted by the CEA code, indicating that Al combustion in an O_2/He mixture is near adiabatic. In the case of argon and nitrogen as diluents, the trend regarding oxygen concentration is correctly predicted. However, two main discrepancies arise:

- Predicted temperatures are a few hundred kelvins greater than measured temperatures
- Predicted temperatures increase monotonically with pressure, while measured temperatures reach a plateau or decrease when pressure increases.

Both discrepancies are attributed to the adiabatic character of the calculations. To assess the deviation of the combustion process from adiabaticity, it is proposed to evaluate the thermal losses of the flame region to the environment. Let us make the simple assumption of an infinitely thin reaction shell, located at a radius r_f . The chemical energy released into this

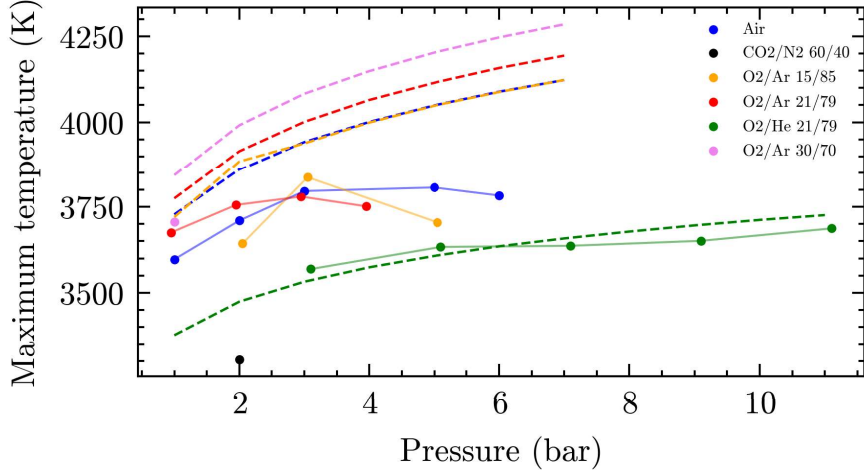


Figure 6.15: Theoretical adiabatic flame temperature (dashed lines) and maximum experimentally measured temperature (points and solid lines).

layer is written E_{chem} . In the assumption of an adiabatic process, the energy produced by the reaction is utilized to heat the products of the reaction. Let us denote the heat capacity of liquid alumina c_{p,Al_2O_3} , in $J.kg^{-1}.K^{-1}$. Let us also denote the mass of alumina in the layer as $m_{Al_2O_3}$. Finally, let us denote L_f the latent heat of fusion of alumina, in $J.kg^{-1}$. Then, if T_0 is the reference temperature of 300 K and T_{ad} is the adiabatic temperature of the reaction, as computed by the CEA-NASA code, it can be written in an adiabatic configuration:

$$E_{chem} = m_{Al_2O_3} C_{p,Al_2O_3} (T_{ad} - T_0) + m_{Al_2O_3} L_f \quad (6.4)$$

Now let us consider that thermal losses can occur, and heat is transferred from the reaction layer to the environment. The energy lost to the environment is denoted as Q_{loss} , and with this consideration, the mixture in the layer is heated to a flame temperature T_f that is lower than the adiabatic temperature due to these thermal losses:

$$E_{chem} = m_{Al_2O_3} C_{p,Al_2O_3} (T_f - T_0) + m_{Al_2O_3} L_f + Q_{loss} \quad (6.5)$$

Upon subtracting equation 6.5 to equation 6.4, it comes:

$$Q_{loss} = (T_{ad} - T_f) \cdot (m_{Al_2O_3} C_{p,Al_2O_3}) \quad (6.6)$$

Convective heat losses are neglected and thus, radiative and conductive energy losses only are considered. Let us denote Q_{rad} the radiative energy losses from the reaction layer and Q_{cond} the conductive heat losses from the reaction layer. The radiative heat losses are expressed using the Stefan-Boltzmann law and, in order to maximize the heat losses, let us consider that the entire layer is made of pure alumina with an emissivity of one. Then, the energy lost by radiation during a time dt is express:

$$Q_{rad} = 4\pi r_f^2 \sigma \cdot (T_{ad}^4 - T_0^4) \cdot dt \quad (6.7)$$

The radiative heat losses from eq. 6.7 come from the radiation from condensed species, or condensed alumina Al_2O_3 . The conductive heat losses are expressed using Fourier's law, and considering that the layer loses heat to the inner region, where the aluminum droplet at a temperature T_p is, and to the outer region of temperature T_0 :

$$Q_{cond} = dt \left(4\pi\lambda r_f (T_{ad} - T_0) + 4\pi\lambda \frac{T_{ad} - T_p}{\frac{1}{r_p} - \frac{1}{r_f}} \right) \quad (6.8)$$

The assumption is made that the thermal conductivity λ is identical in both regions and taken as the thermal conductivity of the gaseous mixture at the adiabatic temperature. It is chosen to consider the mass of alumina in the layer as proportional to the experimentally determined mass fraction of alumina Y computed from the volume fraction of alumina $m_{Al_2O_3} \propto Y$. The following equation arises:

$$Q_{loss} \propto (T_{ad} - T_f) \cdot (Y C_{p,Al_2O_3}) \quad (6.9)$$

Upon evaluating equations 6.7 and 6.8, it is observed that for the studied conditions, $Q_{cond} \approx Q_{rad}$. Although the radiative heat losses have been significantly overestimated, the conductive heat losses remain in a similar order of magnitude. For this reason, radiative heat losses are neglected. Thus, upon combining equation 6.9 and equation 6.6, it can be written:

$$(T_{ad} - T_f) \propto \frac{Q_{cond}}{Y C_{p,Al_2O_3}} = \frac{Q_{cond}}{\bar{C}_p} \quad (6.10)$$

Figure 6.16 displays the left-hand term of this equation as a function of its right-hand term. The observed linear trend directly supports the hypothesis of conductive heat losses as the main mechanism for the non-adiabaticity of the system. The non-perfect linearity can, however, be attributed to strong assumptions that were required to enable a fairly simple study of the adiabaticity. It is believed that a more accurate model could provide better linearity. Nonetheless, it can be observed that an increase in pressure leads to an increase in adiabatic temperature, but Fig. 6.16 illustrates that it also leads to an increase in conductive heat losses, due to an increase in the condensed phase concentration. Thermal losses in an O_2/He 21/79 % (vol.) mixture are observed to be negligible due to low concentrations of alumina in the cloud. On the other hand, combustion in nitrogen and argon results in greater adiabatic flame temperatures but also results in greater thermal losses. As more alumina is produced, it is hypothesized that heat loss mechanisms become more important and the adiabatic assumption cannot be made.

In order to simulate realistic flame temperatures, thermal losses and particularly conductive heat losses of the system must be accounted for. The assumption of an adiabatic system is proven to be incorrect for most mixtures. However, accounting for conductive heat losses in simulation means being able to locally assess the transport properties of all species, including the condensed phase species. Similarly, to account for radiative heat losses, the optical properties and geometrical properties of the alumina particles must also be locally tracked, which has rarely been done in the literature.

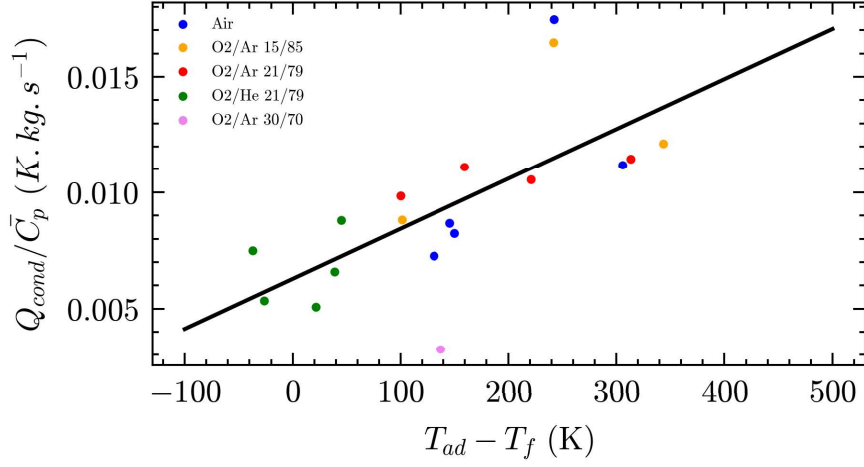


Figure 6.16: Theoretical conductive heat losses divided by the heat capacity of the mixture as a function of the difference between the adiabatic flame temperature and the measured flame temperature.

Aluminum surface temperature

The surface temperature measured in all experimental cases is displayed, along with the 95 % confidence intervals depicted as error bars, in Figure 6.17. No clear trends can be identified in this figure. In addition, error bars are relatively important, mainly due to the fact that at the droplet's surface, the deconvolution error is the greater as it accumulates along the profile. However, it is observed that the surface temperature tends to increase with pressure, which is theoretically expected from the Clausius-Clapeyron relation. The surface temperature shows no obvious variation with the composition of the mixture. In addition, both numerical work from Muller and Beckstead show temperatures near the temperature values in the case of air.

In the literature, a common relation developed by King [63] relates the droplet's surface temperature T_s to the aluminum partial pressure at the droplet's surface $P_{Al,s}$. This relation is presented in the Equation (6.11).

$$T_s = \frac{34680}{12.537 - \ln(P_{Al,s})} \quad (6.11)$$

Considering that at the droplet's surface, the mole fraction of gaseous aluminum is $x_{Al,s}$, Equation (6.11) can be written as a function of the ambient pressure P :

$$T_s = \frac{34680}{12.537 - \ln(x_{Al,s} \cdot P)} \quad (6.12)$$

From this equation, the gaseous aluminum mole fraction at the droplet's surface can be obtained from the droplet's surface temperature, and the results are presented in Figure 6.18.

Note that the gaseous aluminum mole fraction for the O_2/Ar 15/85 % (vol.) mixture at 3 bars is discarded, as the measured temperature is above the evaporation temperature of aluminum, leading to mole fractions greater than unity. Similarly as for surface temperature, no clear behavior is identified regarding the evolution with pressure. The mole fraction of

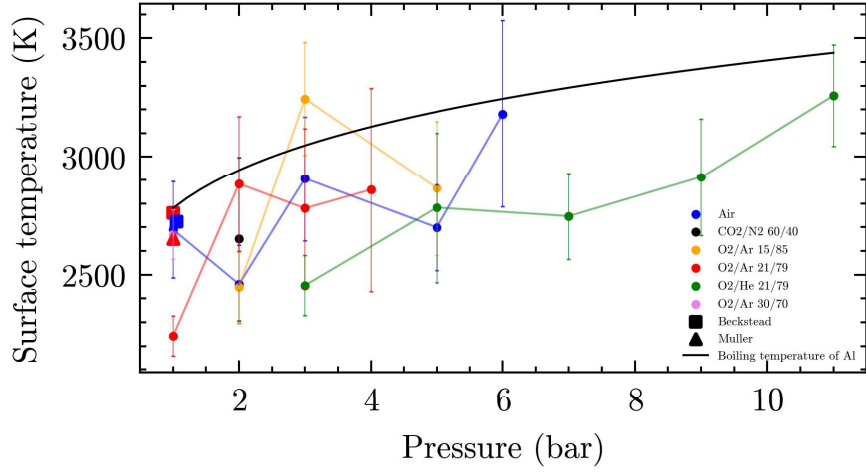


Figure 6.17: Aluminum droplet surface temperature measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).

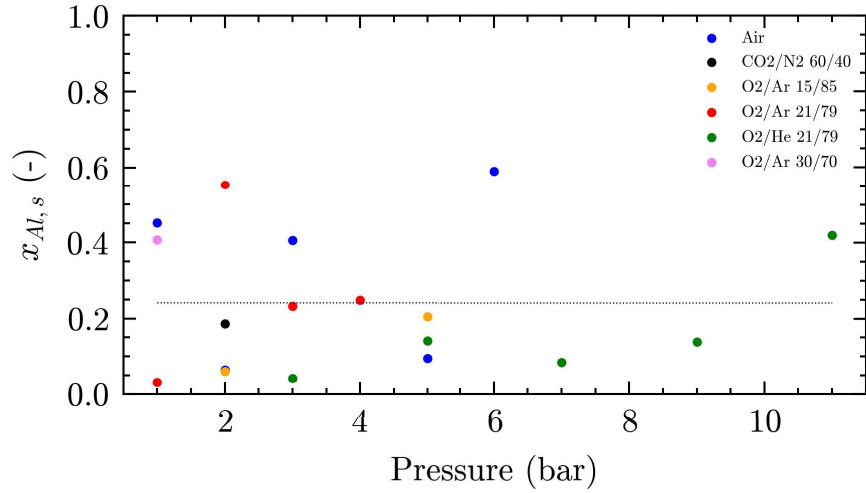


Figure 6.18: Gaseous aluminum partial pressure as a function of pressure. The average value of all mole fractions is also displayed (black dashed line).

aluminum on the droplet's surface appears lower in the case of helium, while argon and nitrogen show similar surface mole fractions. This could be explained by the high diffusion coefficient of helium compared to argon and nitrogen. All mole fractions remain close to an average value of 0.29. Since x_s results from a balance between evaporation and diffusion (both independent on pressure), this result may not be surprising.

Location of maximum temperature - SOR

The radial location of the maximum temperature, defined as the Stand-Off Ratio (SOR), is displayed, for all experimental cases in Figure 6.19

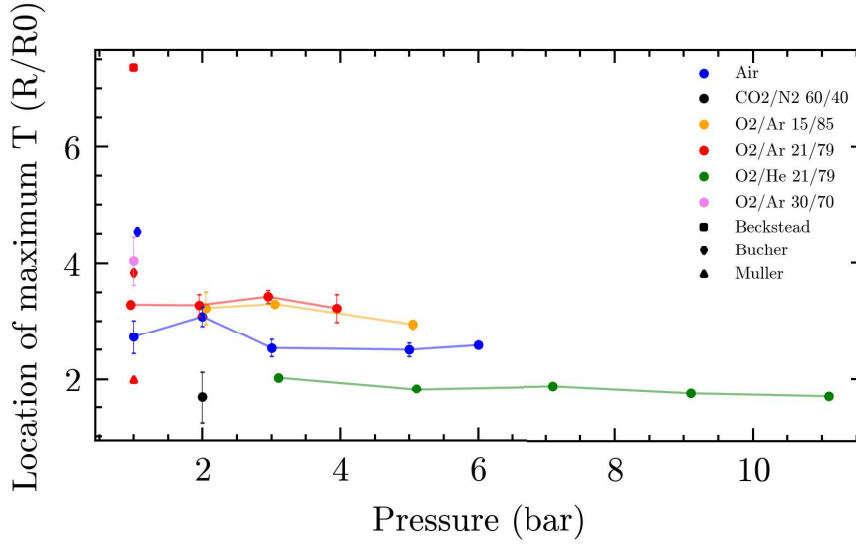


Figure 6.19: Radial position of the experimental maximum temperature. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).

At 21 % (vol.) of oxygen, using helium as a diluent results in the smallest SOR of about 2 droplet radii. A similar SOR is obtained in the CO_2/N_2 60/40 % (vol.) mixture. When nitrogen is used as the diluent (air), the standoff ratio increases from 2 to about 2.5 droplet radii. The largest SOR is obtained upon using argon as a diluent and reaches about 3.3 droplet radii. As oxygen concentration increases, the standoff ratio increases. Additionally, as pressure increases, the SOR is observed to slightly decrease. The values issued from the literature are significantly different from the current experimental results. Such discrepancies are however attributed to the difference in initial size of the Al droplet, as in the literature, particles of diameter near 230 microns are considered and observed, while in current work, the initial diameter of the particles is near 70 microns, possibly influencing the diffusion processes. The SOR results from a complex interplay between flame temperature and oxygen availability. As an example, when oxygen concentration is increased, the aluminum vapor oxidizes closer to the Al droplet due to the increase in oxygen availability, resulting in a theoretical decrease of the SOR. However, as the SOR decreases, the thermal feedback to the flame increases. Indeed, the flame temperature is greater as oxygen concentration increases, and the flame is closer to the droplet. As thermal feedback to the droplet increases, the Al Stefan flow increases, thus pushing away the reaction zone, and the SOR thus increases. This coupling is complex to analyze and predict. In order to provide some insights on these observations, theoretical formulas to compute the SOR are used. The parameters used for these formulas are summarized in table 6.2.

In the case of a hydrocarbon droplet, Liu et al. [115] extracted from Law et al. [116] the stand-off ratio SOR_{hc} expressed as:

Table 6.2: Summary of useful parameters for the theoretical evaluation of the SOR.

Symbol	Description	Units
M_{mix}	Molar mass of the mixture	$kg \cdot mol^{-1}$
C_p	Specific heat capacity at constant pressure of the mixture	$J \cdot kg^{-1} \cdot K^{-1}$
λ	Thermal conductivity of the mixture	$W \cdot m^{-1} \cdot K^{-1}$
ρ	Density of the mixture	$kg \cdot m^{-3}$
D_O	Mass diffusion coefficient of oxygen	$m^2 \cdot s^{-1}$
L	Molar latent heat of vaporization of the fuel	$m^2 \cdot s^{-1}$
q_F	Molar heat of combustion	$J \cdot kg^{-1} \cdot K^{-1}$
T_∞	Temperature of the ambience	K
T_s	Temperature of the surface of the fuel	K
$Y_{O,\infty}$	Oxygen mass fraction in the mixture	-
$\tilde{T} = \frac{Y_{O,\infty} \cdot C_p \cdot T}{q_F}$	Reduced temperature T	-
$\tilde{L} = \frac{L}{q_F}$	Reduced heat ratio	-
$\tilde{Y}_O = \frac{Y_O}{\sigma}$	Reduced oxygen mass fraction	-
ν_O	Stoichiometric coefficient of oxygen in the oxydation reaction $\nu_F F + \nu_O O \rightarrow P$	-
ν_F	Stoichiometric coefficient of the fuel in the oxidation reaction $\nu_F F + \nu_O O \rightarrow P$	-
W_O	Molecular weight of oxygen	kg
W_F	Molecular weight of the fuel	kg
$\sigma = \frac{\nu_O W_O}{\nu_F W_F}$	Stoichiometric oxidize to fuel mass ratio	-
$Le_O = \frac{\lambda}{C_p \cdot \rho \cdot D_O}$	Lewis number of oxygen	-

$$SOR_{hc} = \frac{Le_O \cdot m_{O,hc}}{\ln \left(1 + \tilde{Y}_{O,\infty} \right)} \quad (6.13)$$

With m_O expressed as:

$$m_{O,hc} = \ln \left(1 + \left[\left(1 + \tilde{Y}_{O,\infty} \right)^{1/Le_O} - \left(\tilde{T}_s - \tilde{T}_\infty + 1 \right) \right] / \tilde{L} \right) \quad (6.14)$$

This theoretical solution for the SOR is determined for combustion cases in which the combustion products are in the gaseous phase. Figure 6.20 represents the SOR computed with this correlation, as a function of the measured SOR. Although a certain linearity is observed, the SOR from the classical hydrocarbon law is generally overestimated.

In the same study, Liu et al. determine a new theoretical formulation of the standoff ratio, accounting for the presence of condensed combustion products (CCPs). With the use of an activation energy asymptotics, Liu discards the classical mass conservation equation by considering that the existence of CCPs reduces the mass of the gas, and thus, that the mass of the gas phase is not conserved. This study leads to a new correlation expressed as:

$$SOR_{ccp} = - \frac{\sigma Le_O \cdot m_{O,ccp}}{\ln (1 - Y_{O,\infty})} \quad (6.15)$$

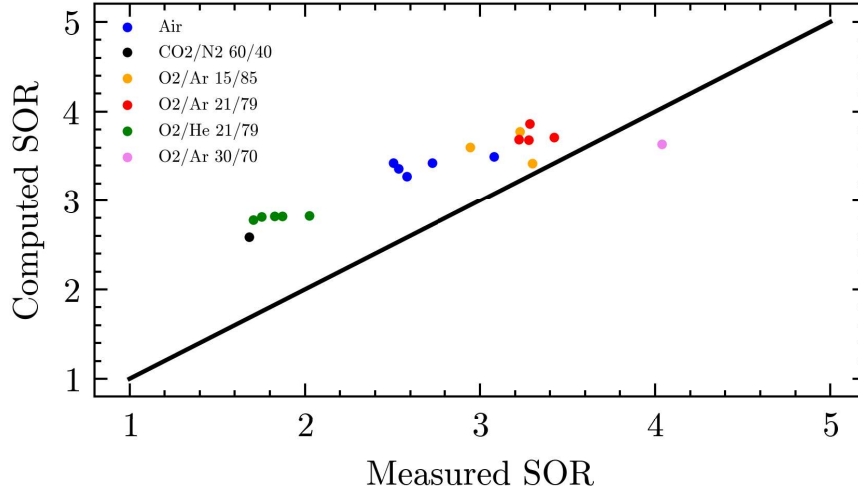


Figure 6.20: SOR computed according to the classical law for hydrocarbon droplets from [116], as a function of the measured SOR.

With $m_{O,ccp}$ defined as:

$$m_{O,ccp} = \ln \left(\frac{1 - (1 - Y_{O,\infty})^{1/Le_O} - \sigma (\tilde{T}_s - \tilde{T}_\infty - \tilde{L})}{\tilde{L} \left[(1 + \sigma) - (1 - Y_{O,\infty})^{1/Le_O} \right] \cdot (1 - Y_{O,\infty})^{1/(\sigma Le_O)}} \right) \quad (6.16)$$

Figure 6.21 represents the SOR computed with Liu's correlation that accounts for CCPs, as a function of the measured SOR.

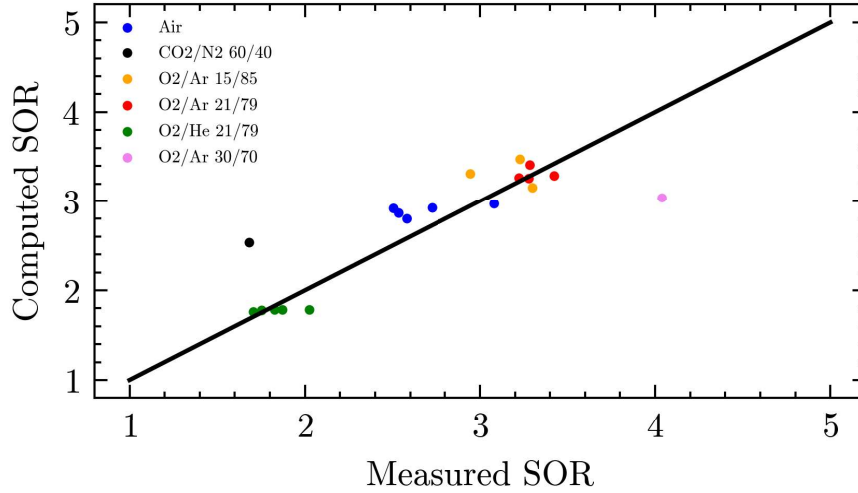


Figure 6.21: SOR computed according to the law developed by Liu et al. [115] accounting for CCPs, as a function of the measures SOR.

A much better agreement between the correlation and experiments is now observed. It has to be noted that, in the case of the CO_2/N_2 60/40 % (vol.) mixture, the computed SOR

is slightly overestimated. In addition, in the case of the O_2/Ar 30/70 % (vol.) mixture, the computed SOR is slightly underestimated. Although this correlation enables an accurate prediction of the SOR in the range of mixtures considered, it is believed that the correlation would fail for high oxygen concentrations. Indeed, the measurements in this work, as well as in the work of Braconnier [8], the SOR is observed to increase with O_2 concentration, while the current correlation predicts a decrease in the SOR with an increasing O_2 concentration. Given the complexity of such correlations, no conclusions can be drawn as to what assumptions result in incorrect predictions at high oxygen concentrations. Nonetheless, it can be hypothesized that the one step reaction assumption fails at higher oxygen concentration.

6.4.2 Alumina concentration

Alumina peak volume fraction

The local maximum volume fraction that establishes further from the droplet than the high alumina concentration at the droplet's surface, denoted as the alumina peak volume fraction, is measured for all experimental cases. The results are presented in Figure 6.22. In all mixtures, the peak volume fraction of alumina is observed to increase with the pressure. Disregarding pressure effects, it is observed that the minimum volume fraction of alumina is measured when helium is used as a diluent. At low pressures, argon and nitrogen show similar alumina peak concentrations, but at pressures greater than 3 bars, alumina peak concentrations measured in argon are greater than in nitrogen. In addition, increasing the oxygen concentration seems to result in an increase in the alumina peak volume fraction. Finally, at equivalent pressure, the largest alumina concentration is measured in the CO_2 mixture.

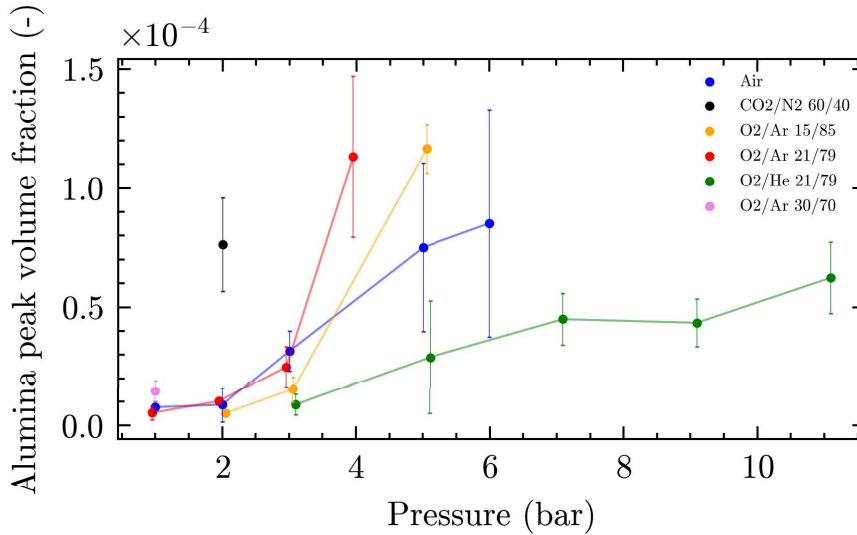


Figure 6.22: Alumina peak volume fraction measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).

This peak of alumina concentration is the result of a complex interplay of various phenomena. As a first approximation, this peak alumina concentration is influenced by the production rate of alumina, as well as the residence time of the particles within the cloud. At constant residence time, the more alumina is produced, the greater the peak volume fraction. However, if the production rate of alumina is constant, the peak volume fraction depends on the time taken by the particle to diffuse away from the cloud. The following list summarizes the mechanisms that can influence the production rate and residence time of alumina particles.

- The flame temperature. It dictates the thermal feedback to the aluminum droplet, thus drives the aluminum Stefan flow and the transport of the fuel to the reaction zone. As the flame temperature decreases, less aluminum is transported to the reaction zone and it is thus expected that less alumina is produced. The Stefan flow is also expected to drive the alumina particles to the outer edges of the oxide cloud. The flame temperature is also expected to influence the reaction rates and thus alumina production.
- The SOR. As alumina particles condense, a share of these particles can be transported towards the droplet's surface by thermophoresis. This mechanism transports the particles located in a temperature gradient towards the lowest temperatures and the amplitude of this mechanism depends on the temperature gradient, driven by the flame temperature, but also by the SOR. As the SOR increases, the temperature gradient decreases and thermophoresis is less important. The SOR is also strongly correlated to the flame temperature.
- The mass diffusivity of the particles in the environment. Alumina particles produced in the cloud are driven to the outer edges of the cloud by the Stefan flow, by thermophoresis, and also by the mass diffusivity of the particles. As the pressure increases and the composition of the mixture varies, these transport properties can significantly change.

To my knowledge, no correlations exist in the literature to predict the evolution of the alumina peak volume fraction. Given the complexity of the system, the determination of a theoretical correlation is not intended. Instead, an empirical correlation is determined, providing a first degree of understanding of the parameters at play regarding the alumina volume fraction in the surrounding of a burning Al droplet. It is proposed to use the very first order properties of the mixture to determine this correlation. The effective oxygen mole concentration in the mixture X_{O_2} , the pressure of the ambiance P , and the average molar mass of the initial gaseous mixture M are selected as the parameters of the correlation. Note that the diffusivity of the mixture is expected to influence the alumina peak volume fraction, but is not considered as it also contains the influence of the molar mass of the mixture and the pressure. The determined correlation is the following for the maximum local volume fraction f_v^{max} :

$$f_v^{max} = \left[1.87 \cdot 10^{-7} \cdot X_{O_2}^{0.21} \cdot P^{6.59} \cdot M^{-3.3} \cdot (2.1 \cdot 10^5)^{X_{O_2}} \cdot 0.51^P \cdot 1.29^M \right] + 8.57 \cdot 10^{-6} \quad (6.17)$$

Figure 6.23 displays the maximum volume fraction computed with the empirical correlation compared to the experimental maximum volume fractions. This correlation highlights that

increasing the oxygen concentration in the mixture leads to an increase in alumina volume fraction. The influence of the pressure is more complex as the product $P^{6.59} \cdot 0.51^P$ increases with P at low pressures, and then decreases when the pressure is sufficiently high. The influence of the molar mass of the mixture is as complex as the product $M^{-3.3} \cdot 1.29^M$ decreases with M at very low values of M and then increases with M . Note however that for the values studied in this work, the alumina volume fraction increases with the molar mass of the initial gaseous mixture.

In most cases, the evolution of the maximum alumina volume fraction is correctly described by the correlation. Further measurements would, however, enable a refinement of this correlation.

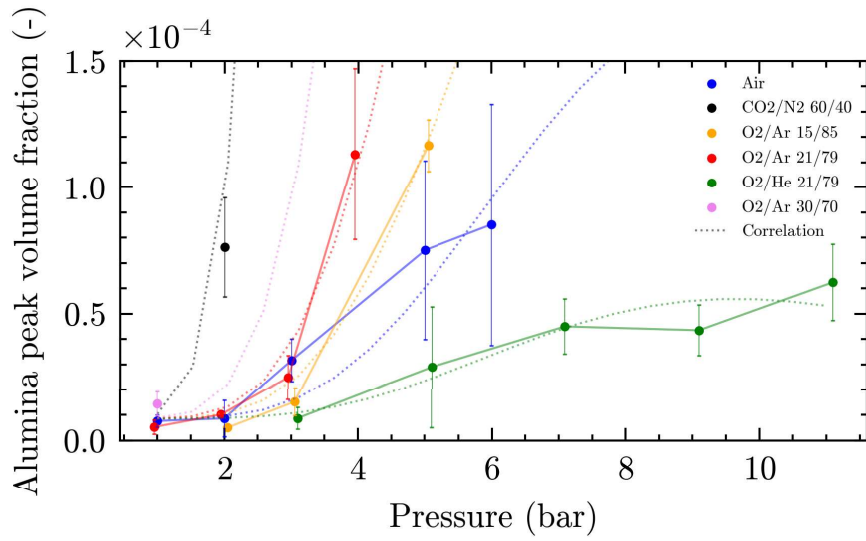


Figure 6.23: Determined correlation (dashed lines) compared to experimental results (points and solid lines).

Location of alumina peak volume fraction

The location of the local maximum in alumina volume fraction r_{f_v} is measured for each experimental case and displayed in Fig. 6.24. As previously mentioned, this alumina peak volume fraction results from the accumulation of alumina particles in a specific region. It is assumed that the location of this accumulation strongly depends on the forces that drive the motion of the alumina particles. The Stefan flow, thermophoretic motions and diffusivity of the particles, driving a Fickian motion of the condensation nuclei, are hypothesized to be first order parameters regarding this KCP.

As the SOR has already been introduced and a theoretical correlation for this KCP has already been mentioned, the location of the local maximum in alumina volume fraction r_{f_v} is correlated to the corresponding SOR. The following linear correlation is observed:

$$r_{f_v} = 1.41 \cdot SOR - 0.12 \quad (6.18)$$

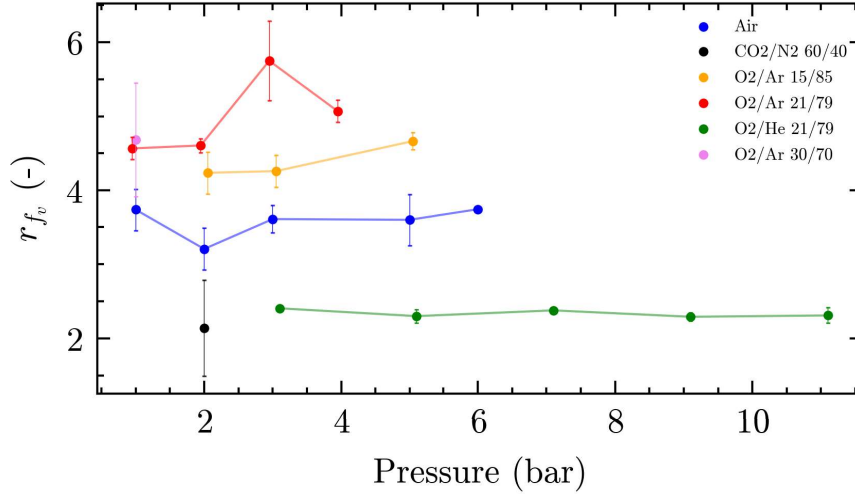


Figure 6.24: Location of the alumina peak volume fraction measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).

The measured location of the alumina peak volume fraction is compared to the predicted location using the measured SOR in Fig. 6.25.

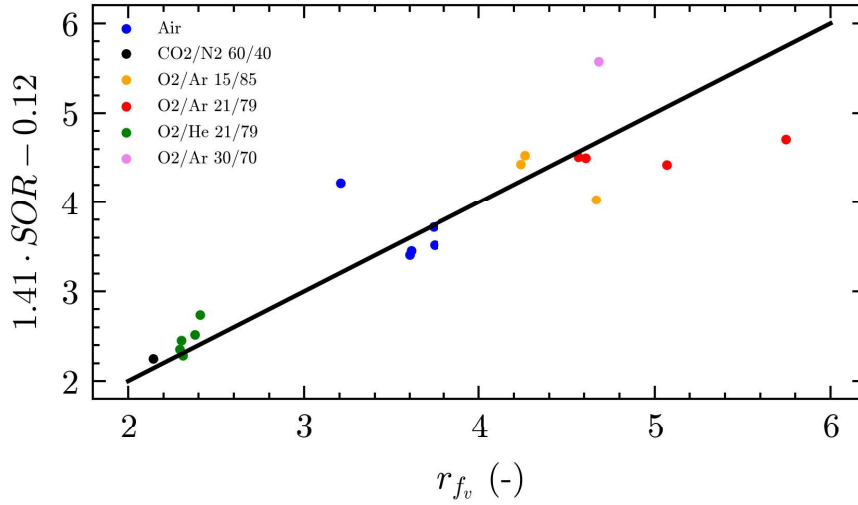


Figure 6.25: Correlation between Stand-Off Ratio (SOR) and location of alumina peak volume fraction r_{f_v} .

Although the correlation presents a slight constant offset of 0.12 between the SOR and r_{f_v} , the 1.41 proportionality factor illustrates that as the SOR increases, the location of the peak alumina volume fraction increases even further. The SOR being considered as the region of alumina condensation due to the significant exothermicity of this reaction, the accumulation of alumina is established further from the production region as the production region moves further from the droplet's surface. The displacement of the SOR further from the droplet

being attributed to an increase in the Stefan flow, the correlation shows that the Stefan flow has an even greater impact on the dynamics of alumina particles.

6.4.3 Average alumina particle size

The alumina particle sizes, averaged across the entire profile, are measured for each experimental case and displayed in Fig. 6.26

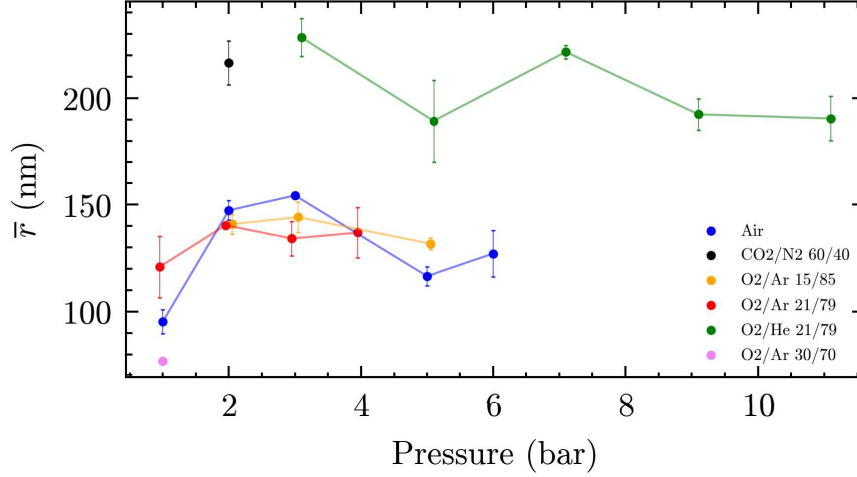


Figure 6.26: Average alumina particle size measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).

The alumina particle sizes are observed to be relatively constant, and span between 100 nm and 200 nm, all conditions considered. No significant variations are observed when the pressure is increased, and larger particles are measured in the case of helium as a diluent, and in the case of CO_2 as the oxidizer. The oxygen concentration seems to have a small influence on the alumina particle sizes. In order to understand the alumina particle sizes presented in Fig. 6.26, the Classical Nucleation Theory (CNT) must be introduced. This short introduction is based on the work of Finke et al. [51]. The CNT is a theoretical model that enables the characterization and description of the kinetics of nucleation. As aluminum reacts with oxygen to form alumina, although gaseous alumina is not expected to exist, as the flame temperature is limited by the vaporization of the alumina oxide [4], an alumina monomer is supposed to form. As multiple monomers are formed, alumina nuclei are produced at a rate called the nucleation rate, which will not be described in this work. Upon formation, the alumina nuclei can grow or shrink under specific conditions. Let us introduce S , the supersaturation for alumina monomers, defined as follows:

$$S = \frac{P_v}{P_e} \quad (6.19)$$

Where P_v is the partial pressure of alumina monomers (i.e., gaseous alumina) $P_v = X_{Al_2O_3} \cdot P$ and P_e is the equilibrium vapor pressure of gaseous alumina over a flat surface.

The vapor pressure of alumina P_v is expressed as a function of the flame temperature T , according to a function extracted from the work of Beckstead [47]. This equation is derived from a Clausius-Clapeyron relation with the ideal gas approximation for alumina monomers. Note, however, that it appears that Beckstead assumes this partial pressure to be the partial pressure of all the aluminum oxides, meaning alumina and the other suboxides of aluminum. Given the lack of other available data, the choice is made to accept this relation.

$$P_v = e^{\frac{0.000250501}{14.132 \cdot 10^{-6}} - \frac{1}{14.132 \cdot 10^{-6} \cdot T}} \quad (6.20)$$

The equilibrium vapor pressure can be expressed as a function of the thermodynamic properties of alumina:

$$P_e = P_0 \cdot e^{\left(-\frac{\mu_v^{pure}(p_0, T) - \mu_l^{pure}(p_0, T) - v_m(P - P_0)}{k_b \cdot T} \right)} \quad (6.21)$$

P_0 is the standard pressure of 1 atm, $\mu_v^{pure}(p_0, T)$ is the chemical potential of pure alumina vapor at the standard pressure and temperature T , $\mu_l^{pure}(p_0, T)$ is the chemical potential of pure liquid alumina at the standard pressure and temperature T , v_m is the molecular volume of an alumina monomer and k_b is the Boltzmann's constant. The chemical potentials are obtained using the data from Burcat et al. [117]. In the case of a spherical droplet of radius r , the equilibrium vapor pressure is expressed as:

$$P_e^r = P_e \cdot e^{\frac{2\sigma_l v_m}{r k_b T}} \quad (6.22)$$

Where σ_l is the surface tension of liquid alumina, expressed as $\sigma_l(T) = 0.65 - 3.9 \cdot 10^{-5} \cdot (T - 2500)$, from Ref. [51]. Let us introduce a critical droplet diameter r_0 expressed as $r_0 = \frac{2\sigma_l v_m}{k_b T \ln S}$ such as if a droplet's radius is larger than the critical radius, it will grow. In opposition, if the droplet's radius is smaller than the critical radius, the droplet will shrink.

In the case of droplet smaller than the mean free path of the gas, the growth of the alumina droplet is driven by the kinetics of the gases. The corresponding growth rate of the nucleus G_{kin} , in $m^3 s^{-1}$, is expressed as:

$$G_{kin} = \frac{v_m (P_v - P_e^r) \pi}{\sqrt{2\pi m_m k_b T}} \cdot \sqrt{\frac{v_m}{v} + 1} \cdot (d + d_v)^2 \quad (6.23)$$

Where m_m is the mass of an alumina monomer, v is the volume of the droplet of radius r and diameter d , and d_v is the Lennard-Jones collision diameter of an alumina monomer. In the case of a droplet larger than the mean free path of the gas, the diffusivity of the monomer is the driving mechanism for the droplet growth and the corresponding growth rate of the nucleus G_{dif} , in $m^3 s^{-1}$, is expressed as:

$$G_{dif} = \frac{2\pi d D v_m (P_v - P_e^r)}{k_b T} \quad (6.24)$$

Where D is the diffusivity of alumina monomers in the gas. The parameters to compute the diffusivity of alumina monomers can be found in Ref. [118]. Finke et al. mention that the harmonic average of the two growth rates gives a good approximation of a growth rate valid for all particle sizes. Thus, a total growth rate G can be computed as:

$$G = \frac{G_{kin}G_{dif}}{G_{kin} + G_{dif}} \quad (6.25)$$

The total growth rate describes the volume gained by an alumina particle per unit of time. If the initial size of a nucleus is known, then the final size, after growth, of this nucleus can be evaluated. In this work, we consider an initial alumina nucleus composed of n monomers such as:

$$n = \left(\frac{2\sigma_l (36\pi)^{1/3} v_m^{2/3}}{3k_b T \ln S} \right)^3 \quad (6.26)$$

The radius of the initial nucleus r_n is thus expressed as:

$$r_n = \left(\frac{3nv_m}{4\pi} \right)^{1/3} \quad (6.27)$$

Upon determining an arbitrary growth time $t_g = 1 \cdot 10^{-5} s$ during which the nucleus can grow, the final radius of the nucleus can be computed using the introduced set of equations. The results are presented in Fig. 6.27

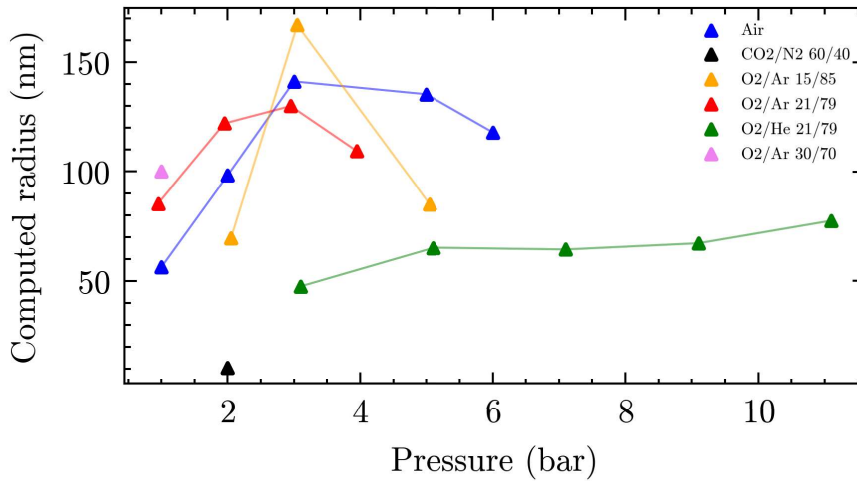


Figure 6.27: Final alumina particle size computed using the CNT.

A comparison between the measured alumina particle sizes from Fig. 6.26 and the computed alumina particle sizes from Fig. 6.27 shows that although the absolute particle sizes are not correctly predicted by calculations, the evolution of the diameter with the pressure is well captured by the calculation. The error in terms of values between the computed sizes and the measured sizes, presented in Fig. 6.28, can be attributed to two parameters. A first parameter is the alumina partial pressure used in Eq. 6.20. This equation is derived from a Clausius-Clapeyron relation, with two hypotheses. A first hypothesis is the consideration of a constant latent heat of vaporization. This hypothesis is made due to the absence of data concerning vapor alumina in the literature, as alumina is considered to decompose rather than vaporize. The second hypothesis is the consideration of alumina vapor as an ideal gas.

This hypothesis is required due to the complexity of the derivation of the Clausius-Clapeyron relation for non-ideal gases, as well as to the scarce data for alumina vapor. These hypotheses are not validated and are a potential source of error for the presented calculations. A second source of error is the arbitrarily chosen growth time. The growth time of an alumina particle is dependent on the residence time of the particle in a region of high supersaturation. Thus, the velocity of the particle must be known to exactly assess the size of the alumina particles. From Fig. 6.28, it can be seen that the residence time of alumina particles in the case of helium as a diluent, must be greater than in other mixtures to match the experiments. A similar behavior is observed for the CO_2 experiments. The flame temperature as well as the nature of the diluent appear to have an influence on growth time. In addition, there is a possibility that the homogeneous theory of nucleation and growth does not apply to this study. The consideration of a heterogeneous condensation process could potentially provide a more accurate prediction of the alumina particle sizes.

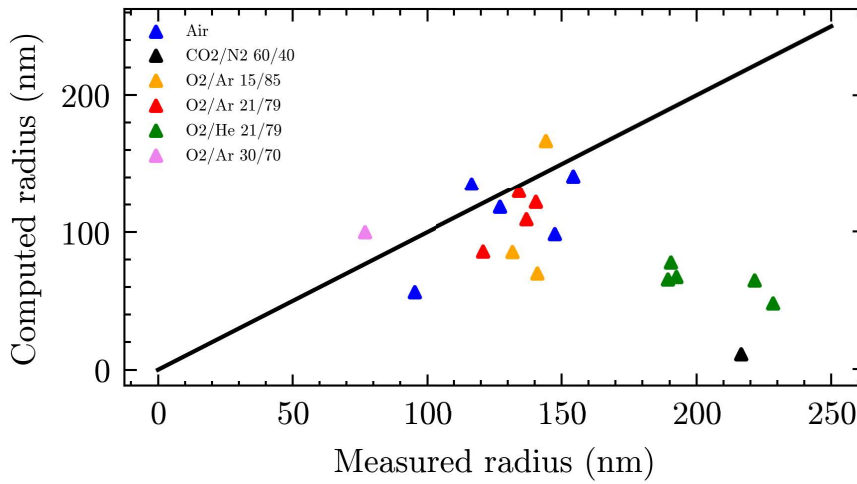


Figure 6.28: Computed alumina particle radii vs. measured alumina particle radii.

The growth time can be optimized for each experimental case in order to provide a linear correlation between the measured particle sizes and the calculated sizes using the CNT. The optimized growth times are presented as a function of the flame temperature in Fig. 6.29. The choice has been made to represent the residence time as a function of temperature to highlight a potential correlation between both, as no obvious trend is observed when the optimized growth time is compared to pressure.

Upon using the optimized growth times from Fig. 6.29, the computed particle sizes perfectly match the measured particle sizes. Although this does not confirm the use of this model, it provides more insights regarding some parameters of interest for the evaluation of the alumina particle sizes.

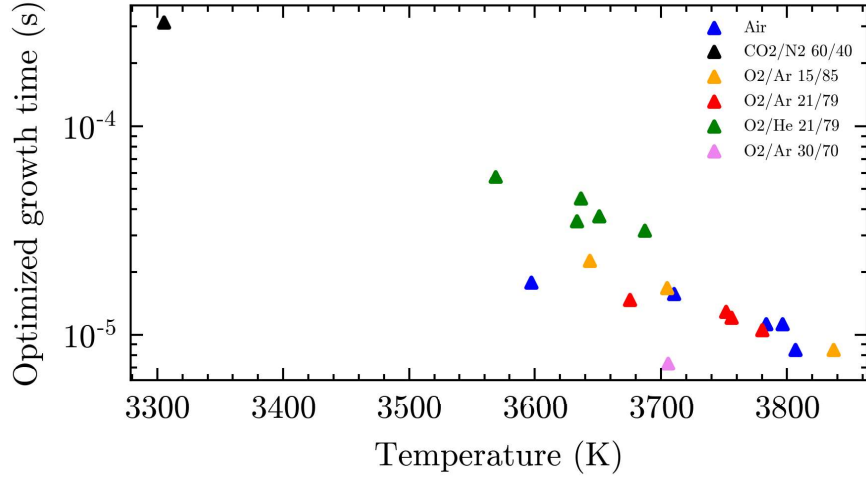


Figure 6.29: Optimized growth time vs. temperature for each experimental case.

6.4.4 Gaseous emissions

Location of AlO peak emissions

The locations of the maximum AlO emissions r_{AlO} are gathered for each experimental case and summarized in Fig. 6.30

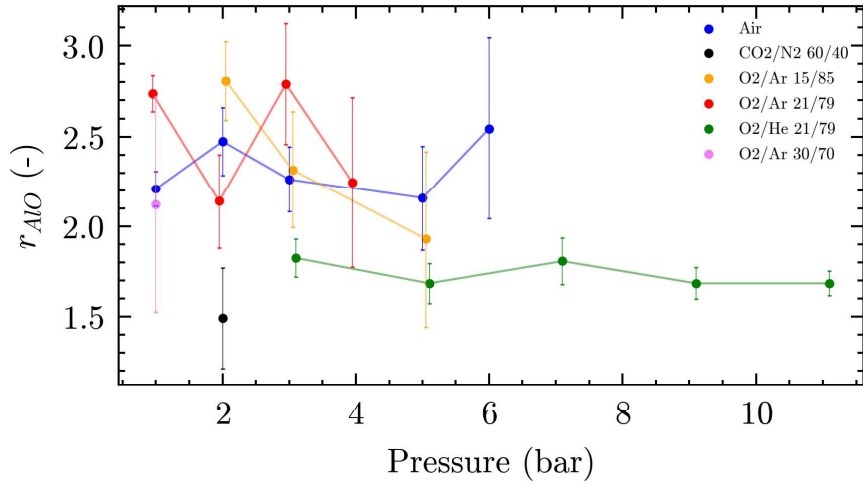


Figure 6.30: Location of the maximum AlO emissions (in a unit of droplet radius) measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).

It is hypothesized that the location of the maximum AlO emissions r_{AlO} is influenced by the same parameters as the SOR and the location of the alumina peak volume fraction. Although no significant variations are observed, r_{AlO} is observed to be mainly dependent on the composition of the gaseous environment. It is proposed to compare r_{AlO} to the respective

SOR for each experimental case. The figure 6.31 displays the evolution of the SOR as a function of the location of the maximum AlO emissions. In all cases, the SOR is located further from the droplet than the location of the maximum AlO emission, indicating that AlO is a first suboxide formed prior to the condensation region.

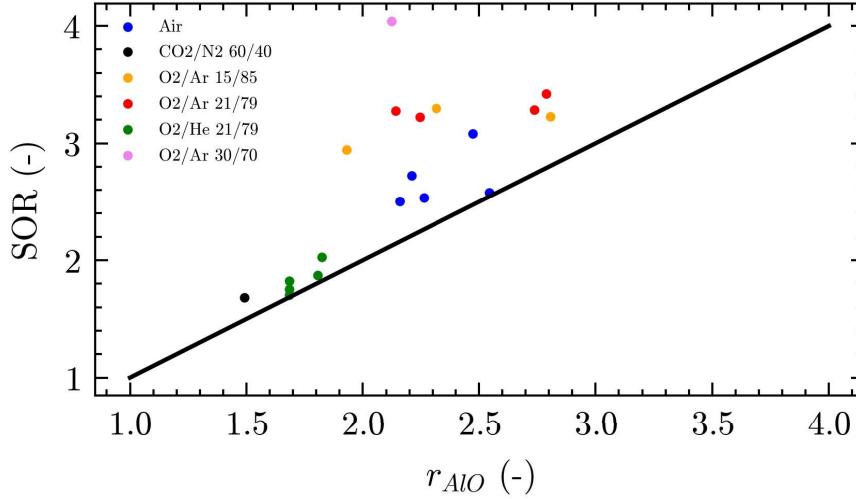


Figure 6.31: Experimental SOR as a function of the experimental r_{AlO} .

As a theoretical correlation has previously been introduced for the SOR, it is proposed to correlate r_{AlO} to its respective SOR. A linear correlation is proposed:

$$r_{AlO} = 0.42 \cdot SOR + 1 \quad (6.28)$$

The linear correlation is displayed in Fig. 6.32. In the case of helium and CO_2 , r_{AlO} is close to the SOR. This observation indicates that at low SOR, the formation of AlO occurs near the formation of Al_2O_3 . Upon considering that, in all cases, the oxidation time of aluminum to alumina is identical, then a potential explanation for this trend, is to consider that for low temperature flames, the diffusion speed of aluminum to the reaction zone is low. This is supported by a low SOR, and results in aluminum oxidizing to AlO and then to Al_2O_3 in near locations. At high flame temperatures, if the aluminum diffuses faster to the reaction region, the oxidation to AlO and then to Al_2O_3 will appear to be in remote regions, due to the diffusion speed.

6.4.5 Bremsstrahlung emissions

Location of maximum Bremsstrahlung emissions

The locations of the maximum Bremsstrahlung emissions r_{Brem} are measured for each experimental case and represented in Fig. 6.33

As Bremsstrahlung emissions are expected to occur upon condensation of alumina, r_{Brem} is compared to the SOR. Indeed, alumina condensation being the most exothermic reaction, Bremsstrahlung emissions are expected to be the most important at the location of the highest temperature. The SOR is compared to r_{Brem} in Fig. 6.34.

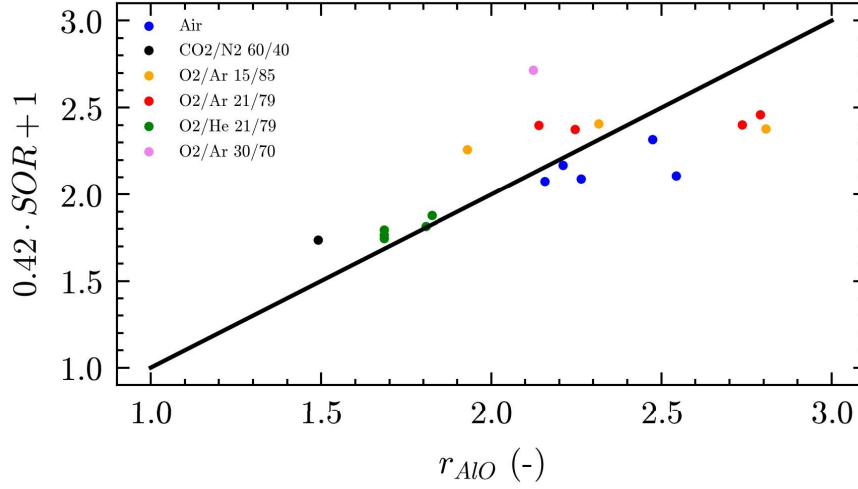


Figure 6.32: r_{AIO} computed using the correlation from Eq. 7.1 vs the experimental r_{AIO} .

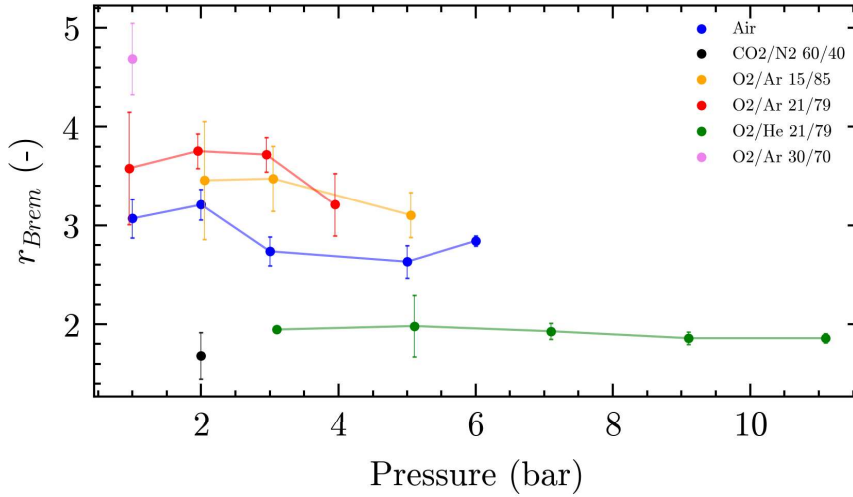


Figure 6.33: Location of the maximum Bremsstrahlung emissions (in a unit of droplet radius) measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The compositions are in % (vol.).

A quasi-perfect linearity is observed between the SOR and r_{Brem} indicating that Bremsstrahlung emissions prevail at the location of alumina condensation. It is proposed to consider $r_{Brem} = SOR$ as a correlation to predict the location of maximum Bremsstrahlung emissions.

Electronic temperature

The electronic temperatures measured from the Bremsstrahlung emissions, for each experimental case, are computed and summarized in Fig. 6.35

The measured electronic temperatures do not display any obvious trends when the pressure or the nature of the mixture is varied. As a result, all the electronic temperatures are measured

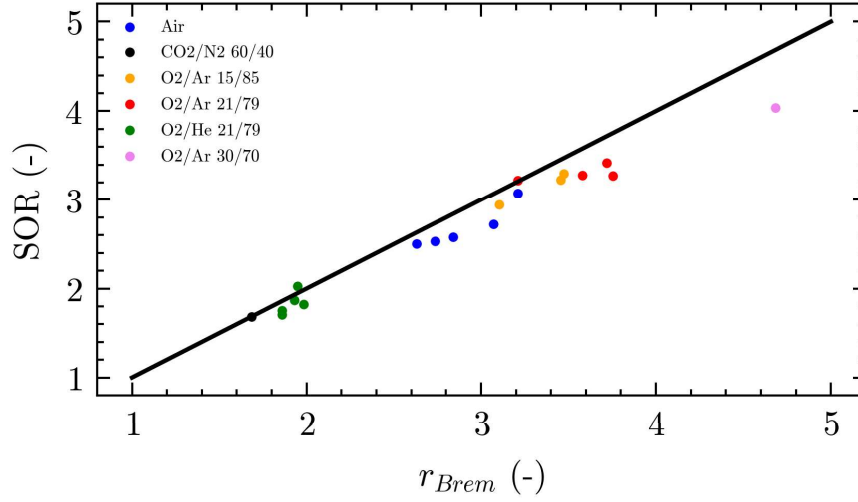


Figure 6.34: Experimental SOR as a function of the location of the maximum Bremsstrahlung emissions.

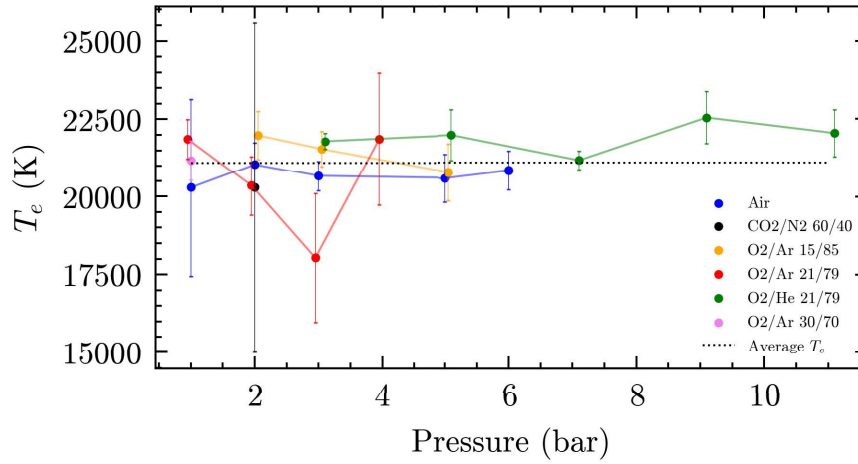


Figure 6.35: Electronic temperature measured for all experimental cases. Error bars represent the 95 % confidence interval. A horizontal offset has been added for clarity. The average electronic temperature is also displayed (black dotted line). The compositions are in % (vol.).

near an average electronic temperature of 21 096 K. Dushman [119] correlates the electronic temperature T_e to the electronic current I per unit surface and time, and the temperature of the emitter T :

$$T_e = -T \cdot \ln \frac{I}{A \cdot T^2} \quad (6.29)$$

With A a constant. In this work, Dushman observes an accurate fitting of experimentally measured current to the correlation from Eq. 6.29 for varying emitter temperatures T , while measuring constant electronic temperatures T_e . From Dushman's work, it is thus

assumed that the electronic temperature T_e is only dependent on the material. An electronic temperature $T_e = 21096K$ is measured. In the context of this work, this corresponds to a work function of alumina ϕ with $\phi = 1.82eV$. No values could be found in the literature to provide a comparison.

6.5 Summary

The following table briefly summarizes the influence of the medium on the studied KCPs and the correlations used to characterize the KCPs.

Table 6.3: Summary of the experimental results.

KCP	Influence of pressure	Influence of diluent	Influence of oxidizer (a%<b%)	Correlation
Maximum temperature	If P $\nearrow$, T $\nearrow$ up to a certain P. Then, T constant or T $\searrow$	$T(He) <$ $T(Ar) \approx T(N_2)$	$T(CO_2) <$ $T(a \cdot O_2) <$ $T(b \cdot O_2)$	-
Surface temperature	If P $\nearrow$, $T_s \nearrow$	$T_s(He) <$ $T_s(Ar) \approx$ $T_s(N_2)$	-	$T_s \approx \frac{34680}{12.537 - \ln(0.29 \cdot P)}$
SOR	-	$SOR(He) <$ $SOR(N_2) <$ $SOR(Ar)$	$SOR(CO_2) <$ $SOR(a \cdot O_2) <$ $SOR(b \cdot O_2)$	$SOR = \frac{\sigma_{LeO} \cdot m_{O,ccp}}{\ln(1 - Y_{O,\infty})}$
Alumina peak volume fraction	If P $\nearrow$, $f_v \nearrow$	$f_v(He) <$ $f_v(N_2) <$ $f_v(Ar)$	$f_v(CO_2) <$ $f_v(a \cdot O_2) <$ $f_v(b \cdot O_2)$	Eq. 6.17
Location of alumina peak volume fraction	-	$r_{f_v}(He) <$ $r_{f_v}(N_2) <$ $r_{f_v}(Ar)$	$r_{f_v}(CO_2) <$ $r_{f_v}(a \cdot O_2) <$ $r_{f_v}(b \cdot O_2)$	$r_{f_v} = 1.41 \cdot SOR - 0.12$
Alumina particle size	If P $\nearrow$, d $\nearrow$ up to a certain P. Then, d $\searrow$	$d(N_2) \approx$ $d(Ar) < d(He)$	$d(b \cdot O_2) <$ $d(a \cdot O_2) <$ $d(CO_2)$	Predicted by CNT and residence times
Location of AlO emissions	-	$r_{AlO}(He) <$ $r_{AlO}(Ar) \approx$ $r_{AlO}(N_2)$	$r_{AlO}(CO_2) <$ $r_{AlO}(a \cdot O_2) \approx$ $r_{AlO}(b \cdot O_2)$	$r_{AlO} = 0.42 \cdot SOR + 1$
Location of Bremsstrahlung emissions	-	$r_{Brem}(He) <$ $r_{Brem}(N_2) <$ $r_{Brem}(Ar)$	$r_{Brem}(CO_2) <$ $r_{Brem}(a \cdot O_2) <$ $r_{Brem}(b \cdot O_2)$	$r_{Brem} = SOR$
Electronic temperature	-	-	-	$T_e \approx 21000K$

Chapter 7

Simulations

This chapter discusses the numerical efforts to simulate the combustion of a single aluminum droplet. At first, a short state of the art of the existing numerical models and their results are presented. The numerical model used in this work is then introduced. A brief parametric study using this model is performed, and a comparison between experimental and numerical results is presented. Further simulation results obtained with the implementation of the Classical Nucleation Theory (CNT) are introduced with the aim of improving the performances of the simulation. Finally, conclusions and perspectives are discussed.

7.1 State of art

Although experimental data on single aluminum droplet combustion is scarce, numerical simulations are more widespread. In general, this lack of experimental data makes the development of numerical methods more complex, as only a few comparisons between experiments and models are available. This highlights the role of this work, providing various targets for numerical methods. A vast variety of simulations, using various numerical procedures, can be found in the literature. In this section, only a few numerical procedures and corresponding results are described.

In 1979, King proposed a model of a single Al particle burning in a CO_2/N_2 atmosphere [63]. At the droplet's surface, aluminum is consumed through evaporation, or through reaction with CO_2 and AlO to yield Al_2O . Similar to all the other models, the aluminum droplet is considered isothermal with a constant diameter and the interface between the liquid and gaseous phase is simply modeled with mass and energy conservation equations. In the inner region, between the droplet's surface and an infinitely thin condensation region, aluminum reacts with the CO_2 to produce AlO and CO . In this thin condensation zone, the aluminum suboxides are considered to react infinitely fast to form Al_2O_3 . In the outer region, further from the droplet than the condensation region, it is considered that no reactions can occur. With the use of conservation equations and boundary conditions, the location of the condensation region is guessed until a combustion temperature of 3500 K is reached at this location. With this model, King simulates temperature profiles, Al and Al suboxides concentration profiles, and CO and CO_2 concentration profiles. As a condensation region containing all the alumina is considered, no alumina concentration profiles are measured.

The results obtained by King for an aluminum droplet of 15 microns in radius burning in a CO_2/N_2 (50/50) (vol.) mixture at 1 bar are compared to the experimental results obtained for CO_2/N_2 (60/40) (vol.) mixture at 2 bar. The temperature profiles are presented in Fig. 7.1 and the normalized AlO concentration profile from King is compared to the normalized gaseous emission profile of the B matrix of the camera in Fig. 7.2.

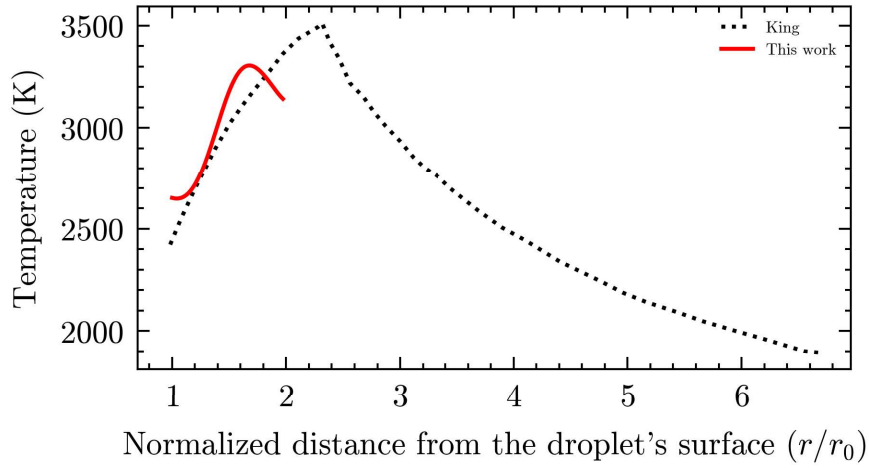


Figure 7.1: Temperature profiles from experiments (CO_2/N_2 (60/40) (vol.) 2 bar) and simulations from King (CO_2/N_2 (50/50) (vol.) 1 bar) .

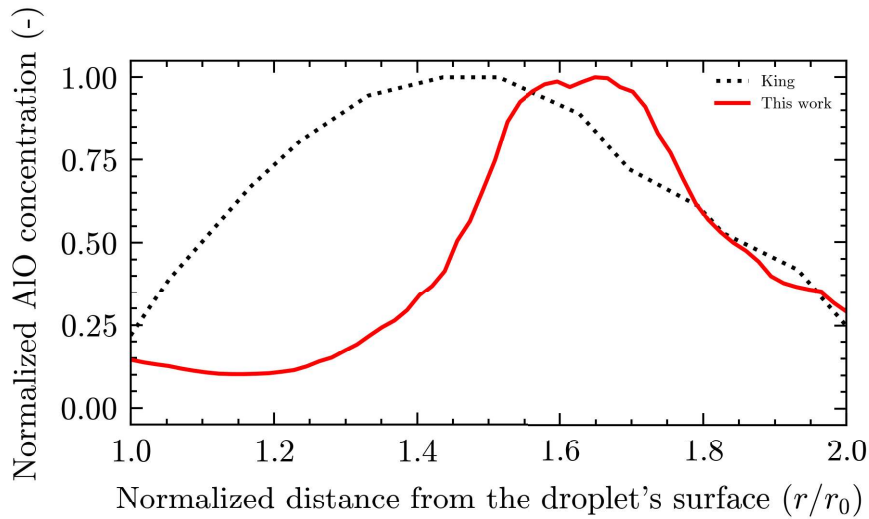


Figure 7.2: Normalized blue gaseous emission profile from experiments (CO_2/N_2 (60/40) (vol.) 2 bar) and simulated normalized AlO concentration profile from King (CO_2/N_2 (50/50) (vol.) 1 bar).

Although combustion conditions slightly differ and the model being overly simplistic, the simulation results remain relatively close to the experimental data.

In 2002, Beckstead proposed a "2-dimensional, unsteady state, evaporation-diffusion-kinetics controlled numerical model" [47]. This model is an improvement from King's model, as the infinitely thin condensation region assumption as well as the infinitely fast reactions assumption are discarded. In this model, a kinetic mechanism describes the surface reactions and the gas phase reactions. Apart from aluminum evaporation, a single surface reaction is considered: $Al_{(l)} + AlO_g \rightarrow Al_2O_{(g)}$. As aluminum suboxide is formed, it is considered that homogeneous reactions form Al_2O_3 in the gas phase, and then a homogeneous condensation reaction occurs to produce liquid alumina. Beckstead implemented the CNT to compute the rate of the homogeneous condensation reaction. However, the assumption is made that the vapor pressure of aluminum oxide is equal to the vapor pressure of gaseous aluminum and that the partial pressure of alumina is equal to the partial pressure of aluminum suboxides to evaluate the supersaturation term. Oxide deposition on the droplet's surface is accounted for. With the use of conservation equations and boundary conditions, temperature and species concentration profiles are simulated. The results obtained by Beckstead in the case of an aluminum droplet of 230 microns in radius in an O_2/Ar (21/79) (vol.) mixture at 1 bar are compared to experimental results in the same environment in Figures 7.3, 7.4 and 7.5. Note that the particle size in the simulations is about 6 times larger than the particles used in the experiments.

In 2017, Muller et al. [46] developed a more accurate model for an Al droplet burning in a steady state, in a 2D axisymmetric configuration, ignoring the presence of the oxide cap and the radiative heat losses, but considering a surface kinetic model in addition to the gas phase kinetic model. The droplet's combustion time τ , which drives the flow of gaseous aluminum to the reaction zone, is represented using a law that resembles the d^2 law, as a function of the initial diameter D_0 :

$$\tau = aD_0^n \quad (7.1)$$

It is proposed to compare our experimental results to the simulations from Muller et al. when all surface reactions and a reversible condensation reaction of alumina are considered, in the case of an Al droplet of 100 microns in diameter burning in an O_2/Ar (21/79) (vol.) mixture at 1 bar. The comparison of Beckstead's simulations and Muller's simulations are made in the same figures as similar conditions, except particle size, are modeled.

The simulated temperature profiles are compared to the experimental temperature profile in Fig. 7.3, and discrepancies in both the shape of the profiles and the maximum temperatures are observed. Similarly, the mass fractions of alumina significantly differ between simulations and experiments as observed in Fig. 7.4. Finally, a relatively correct correlation is observed in Fig. 7.5 between the AlO concentration profile simulated by Beckstead and Muller and the experimental gaseous blue emissions. Note, in addition, that the results from Beckstead's simulations show relatively strong discrepancies to the results from Muller's simulations. It is, however, important to note that Muller's numerical model displays temperatures that remain close to the experimental temperatures. Also, the temperature decreasing slope observed at the external edge of the oxide cloud due to thermal losses and diffusion to the environment matches relatively well the experiments.

Finally, in 2023, Finke and Sewerin [51] proposed a model that tracks the evolution of alumina droplets throughout the oxide cloud using the CNT. Compared to previous models

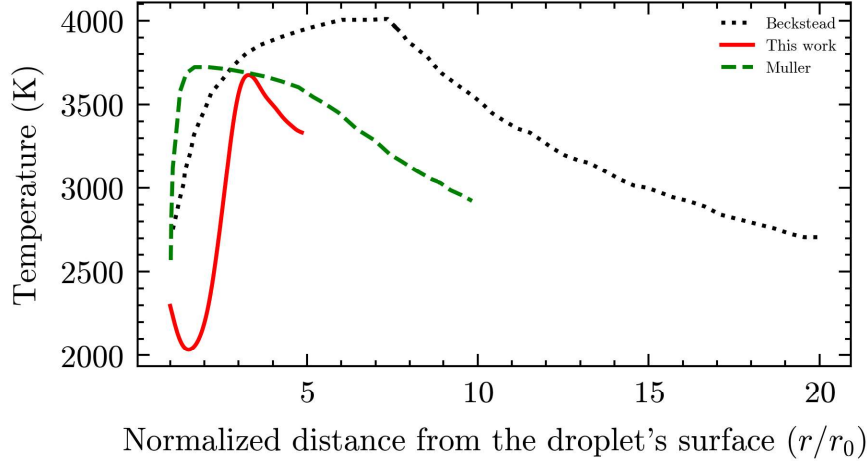


Figure 7.3: Temperature profiles from experiments and simulations from Muller and Beckstead in an O_2/Ar (21/79) (vol.) mixture at 1 bar.

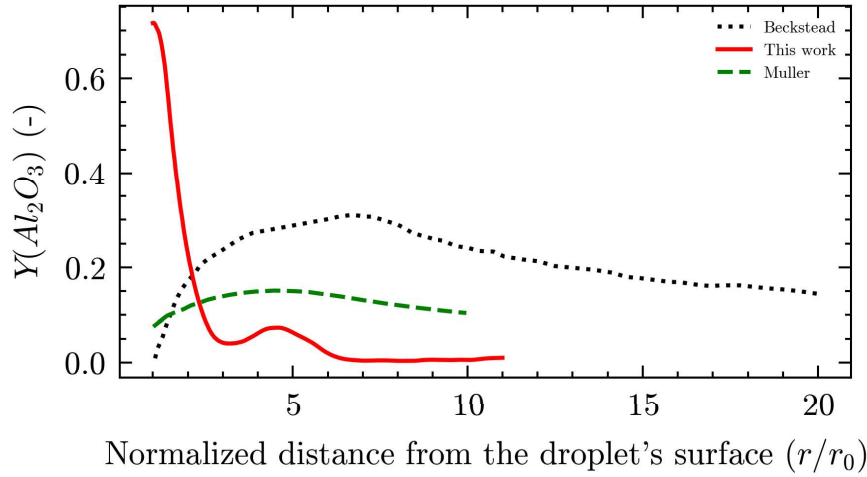


Figure 7.4: Alumina mass fraction profiles from experiments and simulations from Muller and Beckstead in an O_2/Ar (21/79) (vol.) mixture at 1 bar.

in which alumina is often considered as a gaseous species, this numerical method is the first that provides accurate tracking and consideration of the condensed phase produced by the combustion reaction. Nucleation rates, growth rates, and coagulation rates are all computed at each location in the oxide cloud, while maintaining detailed kinetic mechanisms. The results from this complex model cannot be directly compared to experimental results due to the evaluation of parameters of interest only shortly after ignition, and presented through colormaps instead of spatial profiles. Nonetheless, it is believed that this paper provides the most accurate simulation of a burning Al droplet, accounting for mechanisms so far ignored in most numerical models. Note that only a few models were introduced in this section, but many more are available in the literature, such as the works from Fabignon [120], DesJardin [121] or Washburn [122].

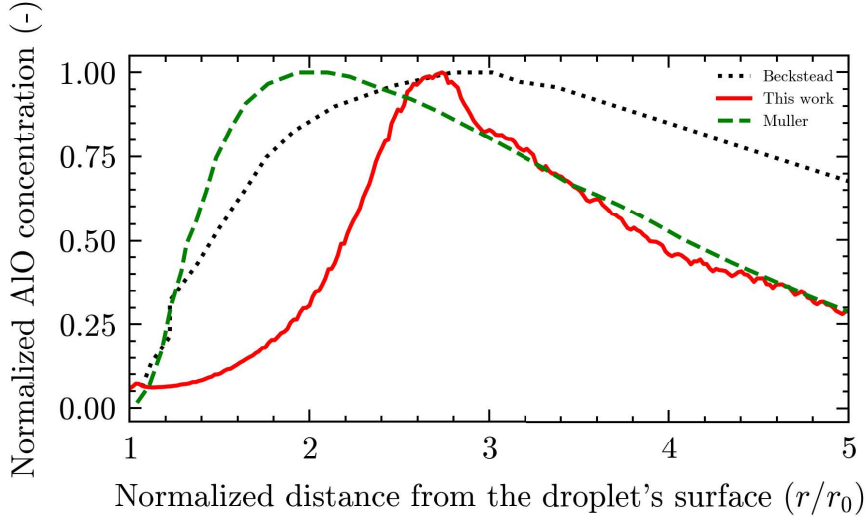


Figure 7.5: Normalized blue gaseous emission profile from experiments and simulated normalized AlO concentration profile from Muller and Beckstead in an O_2/Ar (21/79) (vol.) mixture at 1 bar.

In addition to these models, an in-house model of a single aluminum droplet in combustion was developed at ArianeGroup and used in this study. This model is briefly introduced in the next subsection.

7.2 Description of the numerical model

The description is deliberately kept very short, and details are provided in Refs. [49, 59]

7.2.1 System

The simulated system is an aluminum droplet of 70 microns in diameter, burning in a determined atmosphere. The Al droplet is considered to be ignited, isothermal, of constant diameter and without internal flow. The axisymmetric two-dimensional reactive Navier-Stokes equations are solved around the burning Al droplet. Conservation equations of mass, momentum, energy and species, thoroughly discussed in Ref. [49], are solved using a finite volume approach. The numerical domain extends up to $50 r_0$ with minimum grid spacing of $r_0/35$ at the droplet surface.

7.2.2 Physical model

The gas phase kinetic mechanism is based on the work of Catoire and Swihart, summarized in Ref. [49]. The gas phase mechanism contains 22 gaseous species and 51 associated reactions. In oxygen, the gas phase mechanism reduces to 16 reactions and 9 species. At the droplet's surface, an evaporation reaction of the liquid aluminum is considered, along with various surface reactions. The surface mechanism contains 19 surface species and 49 associated

surface reactions. In oxygen, the surface mechanism reduced to 23 reactions and 8 species. The transport properties of the gaseous species are computed by standard kinetic theory, and are summarized in Ref. [49]. As alumina is considered as a condensed phase within the system, the transport mechanisms differ from the gaseous phase transport. The transport of condensed alumina is driven by convection and phoretic motions, thermophoresis, and diffusiophoresis that can be enabled and disabled in the model. These mechanisms that result from gradients of temperature and concentration respectively drive the transport of alumina based on the size of the alumina particles considered. The local tracking of alumina size is not implemented in this model and thus, alumina particles are considered to have a constant diameter at all locations of the system. The diameter of alumina particles is set at 1 micron, but can be modified. In addition, a Fickian diffusion coefficient, by default set at $10^{-5} \text{m}^2 \cdot \text{s}^{-1}$, is added to the alumina smoke to prevent non-physical accumulations, and enables the modeling of diffusive effects such as Brownian diffusion. The value of this diffusion coefficient can also be modified.

7.2.3 Alumina formation

A simple approach is considered for alumina formation. It is assumed that alumina monomers (i.e., gaseous alumina) Al_2O_3 are produced by oxidation of aluminum and aluminum suboxide according to the gas-phase kinetic mechanism, although it is often stated that gas-phase alumina does not exist. It is then considered that the formed gaseous alumina condenses to form liquid alumina following a simple reaction, governed by an empirical Arrhenius law. The condensation reaction is the following:



The activation energy of reaction (7.2) is set to 0. The pre-exponential factor of this reaction is set to 10^{15} to model very fast condensation, but can be modified. By default, this reaction is considered to be irreversible, but this can be modified in the model.

7.3 Parametric study

In this section, a brief parametric study of the numerical model is proposed. The case of an Al droplet of 70 microns in diameter burning in an O_2/N_2 (20/80) (vol.) at 1 bar is simulated. The temperature of the environment is 300 K. Alumina particles size, the reversibility of the condensation reaction, the pre-exponential factor of the condensation reaction and the diffusivity of the oxide smoke are parameters that have been modified, and the influence of these changes on the simulation results is studied. The simulated results are compared to the experimental results obtained in identical conditions.

7.3.1 Alumina size and condensation reversibility

The influences of the alumina particle size and reversibility of the condensation reaction in the model are discussed here. Four simulation cases are compared. For each simulation, the size of the alumina droplets is considered constant at any location of the oxide cloud. For all

Table 7.1: Simulation parameters to assess the influence of the size of alumina droplets.

Case	$d_{Al_2O_3}$ (nm)	Condensation reversibility
A	200	Reversible
B	200	Non-reversible
C	1000	Reversible
D	1000	Non-reversible

four simulations, thermophoretic and diffusiophoretic motions of the oxide smoke are enabled. The diffusivity of the oxide smoke is set to $10^{-4}m^2s^{-1}$. The pre-exponential factor of the condensation reaction is set at 10^{15} . The variation of the alumina size only influences the phoretic motions. The simulation conditions are presented in Table 7.1.

The experimental and simulated temperature profiles are presented in Fig. 7.6. From these results, it is observed that changing the size of alumina particles does not significantly alter the temperature profile, whether the reaction is reversible or not.

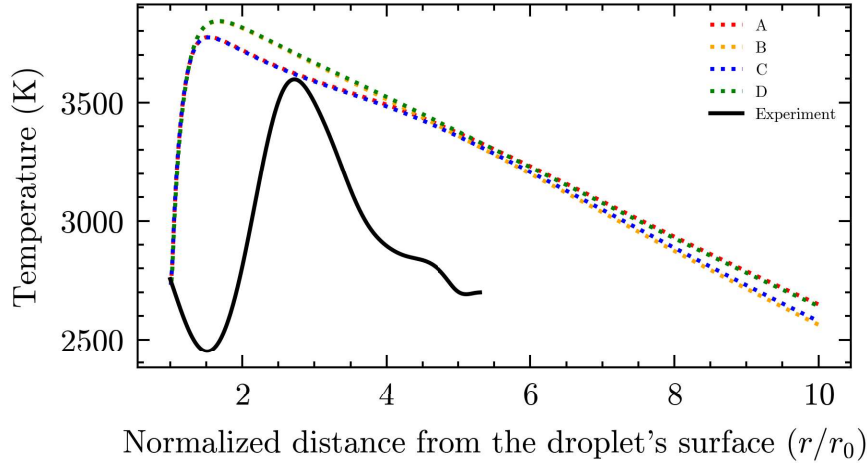


Figure 7.6: Experimental and simulated temperature profiles for atmospheric air.

The influence of the alumina particle size on the volume fraction of alumina is presented in Fig. 7.7. Similarly, the size of the considered alumina particle sizes does not have a significant impact on the concentration of alumina in the oxide cloud. In these conditions, the size is so small that the thermophoretic constant levels off to the constant value of the free molecular regime.

Upon considering a reversible condensation reaction (cases A and C), the temperature and alumina concentration are measured lower than in the case of a non-reversible condensation reaction. This trend is expected, as the inverse condensation, if allowed, consumes condensed alumina through an endothermic reaction. However, significant discrepancies are observed between simulations and experiments for the four simulation cases. Thus, for the following cases, the condensation reaction will be considered reversible and the alumina particle size is

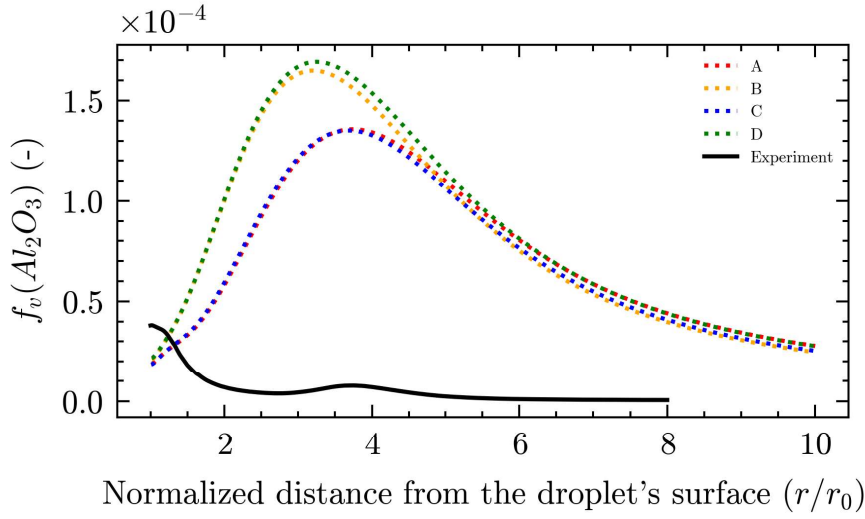


Figure 7.7: Experimental and simulated alumina volume fraction profiles for atmospheric air.

Table 7.2: Simulation parameters to assess the influence of oxide smoke diffusivity.

Case	Diffusion coefficient (m^2s^{-1})
E	10^{-4}
F	10^{-3}
G	10^{-2}

kept at 1 micron.

7.3.2 Diffusivity of the oxide smoke

In this subsection, the influence of smoke diffusivity on the simulations is observed. Three simulation cases are compared. For each simulation, the diameter of alumina droplets is 1 micron, phoretic motions are enabled, the pre-exponential factor of the condensation reaction is set at 10^{13} and the condensation reaction is considered reversible. The simulation conditions are presented in Table 7.2.

While strong differences are still observed between the simulated and experimental temperatures profiles in Fig. 7.8, the diffusion coefficient of alumina smoke appears to have a non-negligible influence on the temperature profiles.

Similarly, the simulated alumina volume fraction profile is strongly dependent on the diffusivity of the alumina smoke, as depicted by Fig. 7.9. Although the shape of the alumina volume fraction profile is not well predicted, relatively acceptable concentrations of alumina are predicted upon selecting a diffusion coefficient close to $10^{-3}m^2s^{-1}$. Although the diffusion coefficients tested here are unrealistic, this highlights that alumina transport properties are first order parameters regarding alumina concentration profiles. This supports

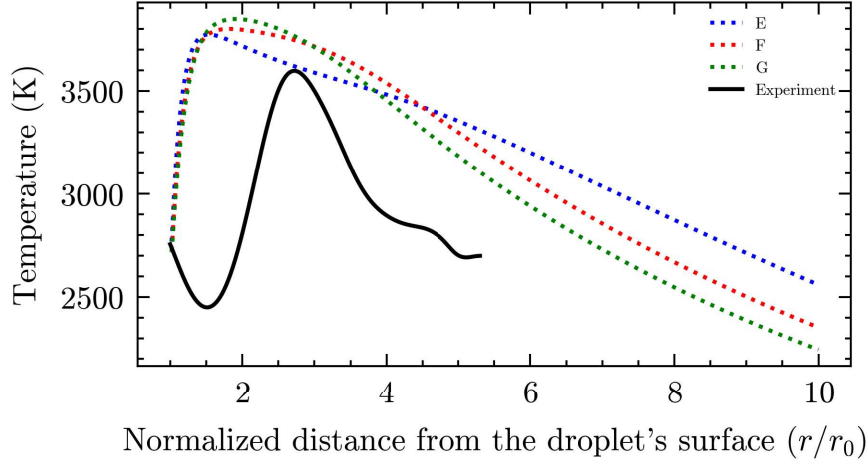


Figure 7.8: Experimental and simulated temperature profiles for atmospheric air.

the experimental results that show a complex relation to the alumina's residence time within the oxide cloud.

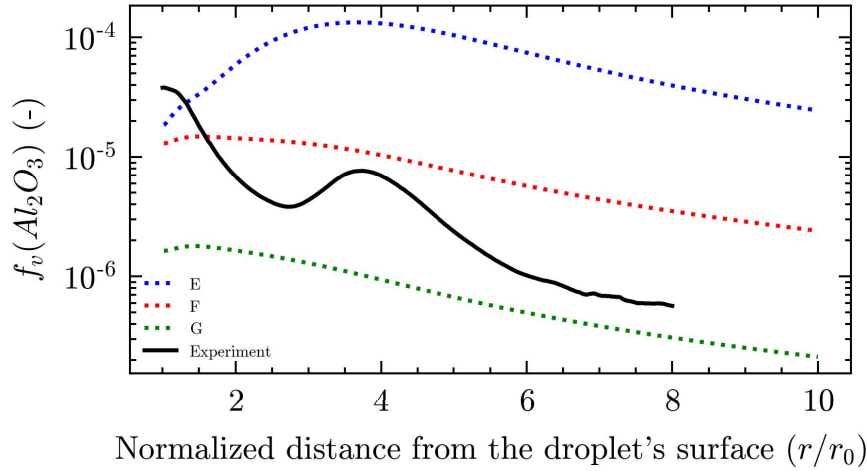


Figure 7.9: Experimental and simulated alumina volume fraction profiles for atmospheric air.

7.3.3 Pre-exponential factor of the condensation reaction

Lastly, the influence of the pre-exponential factor of the condensation reaction on the simulations is assessed. Three simulation cases are compared. For each simulation, the alumina droplet diameter is 1 micron, phoretic motions are enabled, the diffusion coefficient of the oxide smoke is set at $10^{-4} m^2 s^{-1}$ and the condensation reaction is considered reversible. The simulation conditions are presented in Table 7.3.

Figure 7.10 shows that the temperature decreases at every location of the oxide cloud upon decreasing the pre-exponential factor. This behavior is expected due to the decrease of

Table 7.3: Simulation parameters to assess the influence of the pre-exponential factor of the condensation reaction.

Case	Pre-exponential factor
H	10^8
I	10^{10}
J	10^{15}

the reaction rate of the highly exothermic condensation reaction. Similarly, the concentration of alumina within the oxide cloud decreases as the pre-exponential factor decreases. Although strong discrepancies are still observed between simulations and experimental data, it is made obvious that the condensation reaction rate can strongly influence the simulation results. It still has to be noted that although changes to the condensation are made, the temperature profiles are, in all cases, significantly different from the experiments, highlighting the need for a more accurate model.

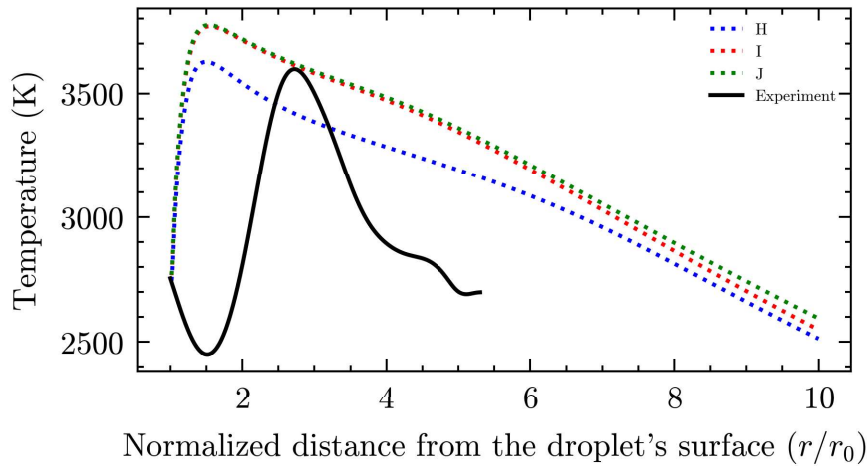


Figure 7.10: Experimental and simulated temperature profiles for atmospheric air.

7.3.4 Conclusion

In this section, various parameters of the simulations have been tested. It has been observed that changing the alumina particle size, the reversibility of the condensation reaction or enabling phoretic motions only slightly modifies the results, while maintaining strong discrepancies when compared to the experimental data. The pre-exponential factor of the condensation reaction or the diffusivity of the oxide smoke have a greater influence on the results, but none of the configurations presented a correct correlation to the experimental results. Although not presented here, the influence of phoretic motion on the results has also been assessed. It was observed that the phoretic motions of the oxide smoke barely influence the temperature profiles, and discrepancies with the experimental temperature

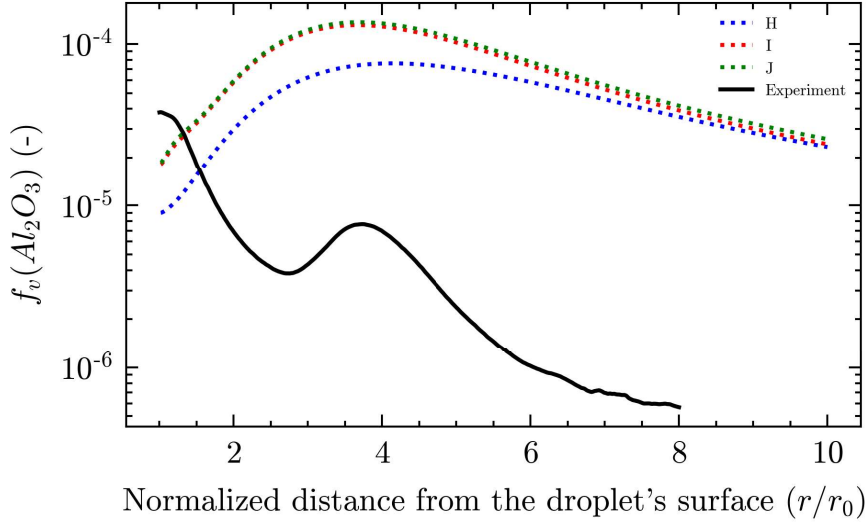


Figure 7.11: Experimental and simulated alumina volume fraction profiles for atmospheric air.

profile remain significant. The comparison of the simulated alumina volume fraction profiles to the experimental alumina volume fraction profile shows that, in the absence of phoretic motions, the alumina volume fraction is greater than in the case for which phoretic motions are enabled. This behavior is expected, as, in the absence of phoretic motion, the accumulation of oxide within the cloud is more important due to the absence of mechanisms driving the oxide outside of the system. However, similar to the temperature profiles, the discrepancies with the experimental results are still important. Given the importance of the condensation reaction of alumina during an Al droplet combustion, it is hypothesized that a more accurate model for this reaction would provide a more accurate prediction of the experimental results. It is assumed that a simplified Arrhenius law, with an activation energy of 0, and an empirically determined and constant pre-exponential factor is not sufficiently accurate. The next section will discuss the attempt of using the CNT to describe the condensation reaction of alumina.

7.4 Implementation of the CNT

Given the inability of the previous model to accurately predict the simulation results, improvements in this model have been proposed. While most aspects of combustion simulations are widely known and rely on well-known physical relations, such as kinetics, conservation equations, boundary conditions, etc., the changes are made in the condensation reaction of alumina. Indeed, although widespread, the use of an empirical Arrhenius law to describe the condensation reaction of alumina is oversimplified. Similar to the work of Beckstead [47] and Finke [51], an attempt is made to implement the Classical Nucleation Theory (CNT) as the driving mechanism of the condensation reaction.

7.4.1 Nucleation rate

As a first approach, with equations based on the work of Finke et al. [51], the reaction rate for alumina condensation is modified and considered as proportional to the nucleation rate given by the CNT. It is considered that when the supersaturation S is greater than unity (i.e., when the partial pressure of alumina monomers is greater than the equilibrium vapor pressure of alumina), the alumina monomers $(Al_2O_3)_m$ regroup to form an alumina nucleus of n monomers with:

$$n = \left(\frac{2\sigma s_m}{3k_b T \ln S} \right)^3 \quad (7.3)$$

With σ the surface tension of liquid alumina, s_m the surface area of an alumina monomer, k_b the Boltzmann's constant, T the temperature.

Denoting P_v the partial pressure of alumina monomers, v_m the volume of an alumina monomer and m_m the mass of an alumina monomer, the reaction rate k_{nuc} of the reaction $(Al_2O_3)_m \rightarrow (Al_2O_3)_{clusters}$, in $kg \cdot m^{-3} \cdot s^{-1}$ is written as:

$$k_{nuc} = \left(\frac{P_v}{k_b T} \right)^2 \sqrt{\frac{2\sigma}{\pi m_m}} \cdot e^{\left(\frac{-16\pi\sigma^2 v_m^3}{3k_b^3 T^3 \ln S^2} - \frac{s_m \sigma \ln S}{k_b T} \right)} \cdot m_m \cdot n \quad (7.4)$$

The nucleation rate extracted from Ref. [51], in number of nuclei per unit time per unit volume, is multiplied by the mass of a nucleus to obtain the reaction rate k_{nuc} in mass units. At this point, the growth and coagulation of alumina droplets are neglected. The comparison between an Arrhenius-based law and a nucleation rate- based law is made in the case of air at 1 bar. For the comparison, phoretic motions are enabled, the diffusivity of the oxide smoke is set at $10^{-5} m^2 \cdot s^{-1}$ and the pre-exponential factor of the Arrhenius condensation reaction is set at 10^{15} . The condensation reaction is considered reversible and alumina particles are considered to have a radius of 1 micron to evaluate their phoretic properties. Note that the nucleus size is only used to determine the condensation reaction rate, not the transport properties of alumina.

Figure 7.12 shows that upon using a reaction rate based on the nucleation rate of alumina, the temperature profile is significantly lower than the measured temperatures. The reaction rate computed from the nucleation rate being much lower than the empirical Arrhenius-based reaction rate, less heat is produced by alumina condensation leading to low temperatures. Figure 7.13 indeed shows a much lower alumina volume fraction in the cloud using the nucleation rate compared to the Arrhenius-based volume fraction. Nonetheless, the order of magnitude of alumina volume fractions simulated using the nucleation rate is close to the experimental volume fraction. The shape of the profile is, however, not accurately simulated.

Given the temperature profiles from Fig. 7.12, accounting for nucleation as the sole condensation process, is not sufficient. As a result condensation via nucleus growth is additionally implemented.

7.4.2 Nucleation rate & growth rates

Let us denote G the growth rate, in $m^3 \cdot s^{-1}$, of a single alumina nucleus. This growth rate had already been introduced in the previous chapter and will not be described here. Denoting

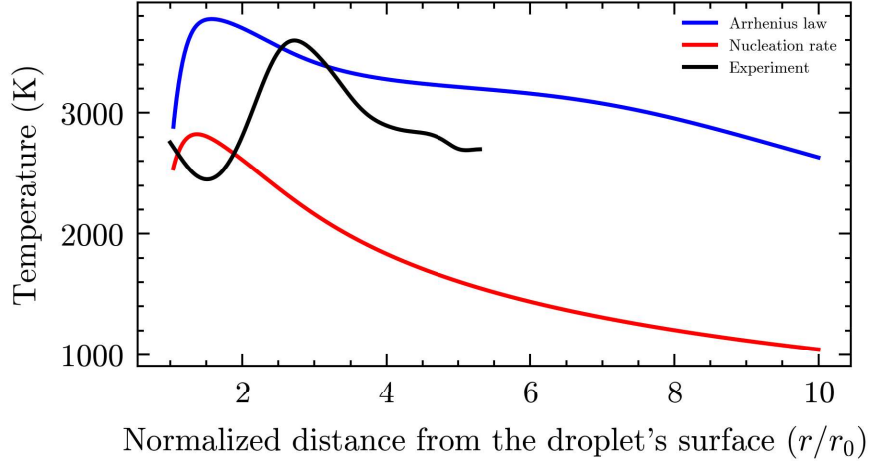


Figure 7.12: Temperature profiles obtained by simulations using an Arrhenius-based condensation law, a nucleation rate-based condensation law, and from experiments.

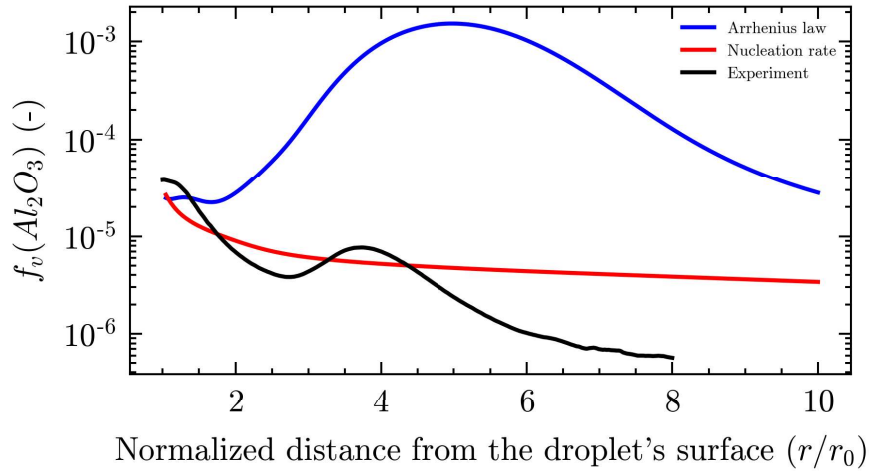


Figure 7.13: Alumina volume fraction profiles obtained by simulations using an Arrhenius-based condensation law, a nucleation rate-based condensation law, and from experiments.

the mass fraction of liquid alumina as $Y_{Al_2O_3}$ and the total density of the mixture as ρ_{tot} , then the number of nuclei per unit volume N_n is given by:

$$N_n = \frac{\rho_{tot} Y_{Al_2O_3}}{m_m n} \quad (7.5)$$

And the condensation rate due to the growth of alumina nuclei J^{Growth} is given, in $kg \cdot m^{-3} \cdot s^{-1}$ by:

$$J^{Growth} = G \cdot \rho_{Al_2O_3} \cdot N_n \quad (7.6)$$

With $\rho_{Al_2O_3}$ the density of liquid alumina. The term J^{Growth} is added to the nucleation-based condensation rate k as both mechanisms consist in condensing alumina monomers into

liquid alumina. It is important to note that Eq. 7.4 refers to a critical nucleus, of critical radius r_0 defined as:

$$r_0 = \frac{2\sigma v_m}{k_b T \ln S} \quad (7.7)$$

Finke et al. [51] mentioned that a nucleus with a radius smaller than this critical radius will undergo shrinking, while a nucleus with a radius larger than the critical radius will lead to the growth of this nucleus. At the critical nucleus size, of radius r_0 and composed of n monomers, the alumina nucleus remains at a constant size. In this first oversimplified model, the nucleus size is not locally tracked and a constant size approximation must be made. If all the alumina nuclei are considered at the critical size, the growth rate will not participate in the reaction rate. It is thus decided that the condensation rate will be computed with the assumption of nuclei of radii $r_{nuc} = A \cdot r_0$ with a factor A greater than unity to allow for growth. Various values of A have thus been tested in the case of atmospheric air.

Figure 7.14 displays the temperature profiles for various nucleus sizes. For $A = 1$, the growth rate is 0 and thus does not participate in the condensation reaction of alumina. The maximum temperature is reached for $A = 2$, and the temperature decreases for larger values of A . Similar trends are observed for the volume fraction in Fig. 7.15. Adding a growth-based condensation rate to the model doesn't appear to influence the shape of the temperature profile, and significant discrepancies are still observed, especially the location of the peak temperature. Note that any value of A provides acceptable orders of magnitudes for the temperature. Regarding the volume fraction, adding the growth-based condensation rate results in large volume fraction values at 6 to 10 droplet radii. Alumina production is then slower and is significantly produced further from the droplet surface.

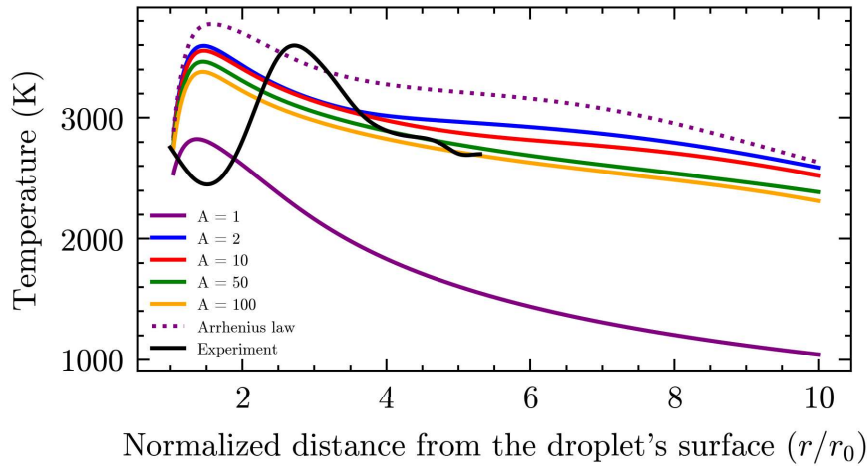


Figure 7.14: Temperature profiles obtained using a growth + nucleation based condensation rate in atmospheric air, for various nucleus sizes, and compared to experimental data.

However, ignoring the volume fractions at distances greater than 5 droplet radii from the droplet's surface as observed in Fig. 7.16, the shapes of the volume fraction profiles are encouraging, displaying a local maximum at the droplet's surface, with another local

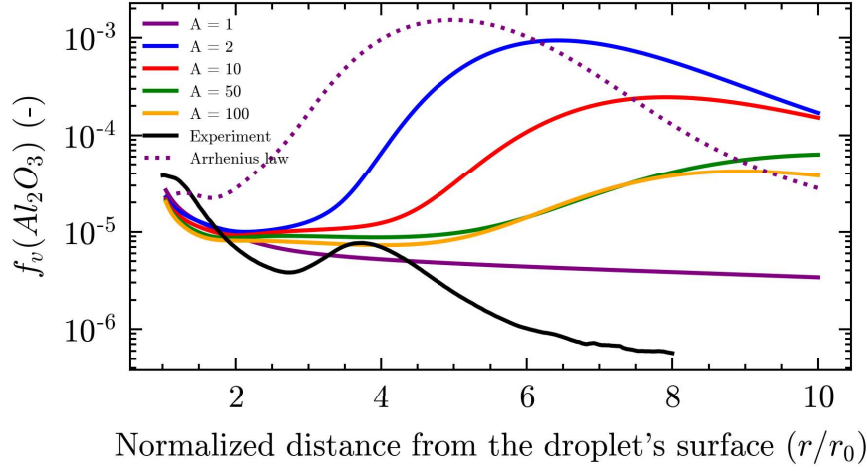


Figure 7.15: Alumina volume fraction profiles obtained using a growth + nucleation based condensation rate, for various nucleus sizes, and compared to experimental data.

maximum between 2 and 3 droplet radii, approaching the trend of the experimental volume fraction. The peak of alumina concentration at the droplet's surface is found to arise from both surface production of Al_2O_3 as well as thermophoresis.

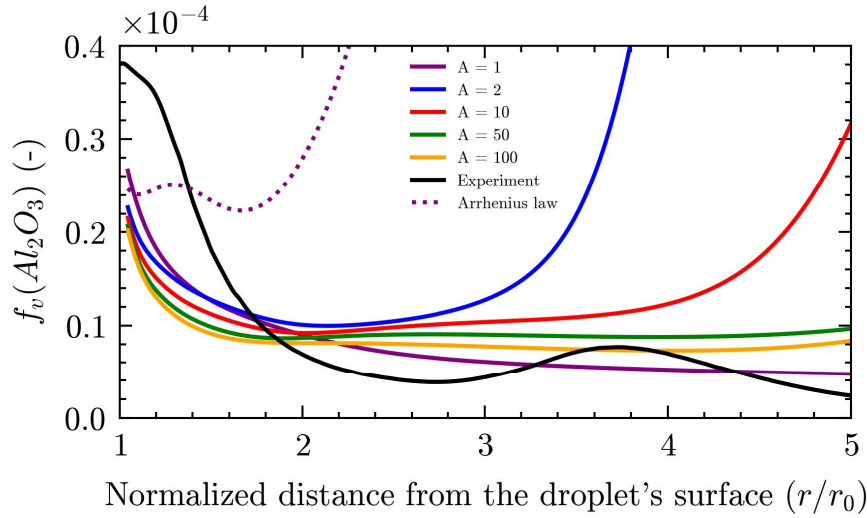


Figure 7.16: Close up version of Fig. 7.15.

Although strong discrepancies are observed between experiments and simulations, the results on the volume fraction profile while using the CNT are encouraging. Please note that simulations with the implementation of the CNT model have been performed for various other conditions, summarized in table 7.4. For all the simulated cases, the observations and conclusions are identical to the present ones.

It is believed that a more accurate model could be implemented, discarding the assumption of a unique nucleus size, and tracking the evolution of the nuclei in each cell. However, such

Table 7.4: Conditions for simulations performed using the CNT model.

Mixture	Pressure (bar)
Air	1
Air	3
Air	5
O_2/Ar (15/85) vol.	1
O_2/Ar (30/70) vol.	1
O_2/Ar (21/79) vol.	1
O_2/Ar (21/79) vol.	4
O_2/He (21/79) vol.	3
O_2/He (21/79) vol.	7
O_2/He (21/79) vol.	11

modifications of the model require a different structure of the model, with an additional species (alumina nuclei). This perspective has not been explored in the context of this work due to a lack of time.

7.5 Conclusions and perspectives

In this chapter, a brief state of art on the existing models of single aluminum particle combustion has been presented. Although each introduced model slightly differs from the other, none of these models present an accurate prediction of the experimental results of this work. Then, the numerical model from Refs. [49, 59] has been used to simulate the experimental results for identical burning conditions. At first, a brief parametric study was performed to identify the first order parameters of the model. While changing the reversibility of the condensation reaction or the size of the alumina particles (considered constant everywhere in the oxide smoke), no significant changes in the simulation results are observed. Upon enabling phoretic motions, changing the diffusivity of the oxide smoke, or changing the pre-exponential factor of the condensation reaction, more evident changes are observed in the simulation results, but none of the cases displayed a good correlation with the experimental results. In order to improve the accuracy of the model, it has been decided to replace the empirically determined Arrhenius law for the condensation reaction by a physical reaction based on the Classical Nucleation Theory. Upon using the nucleation rate of alumina, the production of condensed alumina is too slow and the results are not acceptable. The growth rate of alumina nuclei has subsequently been added as a condensation mechanism, resulting in larger condensation rates. Although no significant changes are observed in the temperature profiles, the alumina concentration profiles are significantly different from initial calculations, and are encouraging the use of the CNT to characterize the condensation reaction of alumina. Strong discrepancies remain, and a more accurate model is required to expect a good correlation to experimental results. At first, tracking the nucleus sizes throughout the oxide cloud could significantly enhance the accuracy of the

results. As an additional outcome of such a model, alumina particle size profiles could be obtained. Coagulation processes could also be implemented for an increased accuracy of the model.

Chapter 8

Conclusions & Perspectives

With 112 successful launches, the Ariane 5 and Ariane 6 launchers developed by ArianeGroup have provided Europe with wide access to space technologies. Both launchers are equipped with solid rocket motors, fueled with solid propellants that contain aluminum particles as energetic additives. In the presence of aluminum particles in the combustion chamber, the performances of the motor are drastically improved due to the high energy released by aluminum combustion. As a downside, the combustion products (alumina), as well as the aluminum droplets, are in the condensed phase, leading to potential two-phase flow losses, thermoacoustic instabilities, nozzle geometry modifications or weight accumulation in recirculation regions of the motor. To prevent performance losses from occurring, the combustion of aluminum, as well as the resulting combustion products, must be well characterized.

Note that although in the propulsion domain, condensed phase combustion products are a downside, benefits can be drawn from this characteristic in another field. The condensed nature of alumina significantly makes its capture significantly easier than capturing gaseous combustion products. Upon using an external energy supply that can be provided by renewable energies, the totality of the combustion products can theoretically be recycled back into aluminum. For these reasons, combined with a high energetic density and availability, aluminum has also gained a lot of interest as a clean energy carrier. The complete characterization of aluminum droplet combustion is necessary to assess the full efficiency of the clean energy cycle that can arise from the use of aluminum as an energy carrier.

Although numerous simulations of aluminum combustion are available in the literature, experimental data remains scarce. Various groups developed aluminum dust flame experiments to provide a macroscopic characterization of aluminum combustion, while single particle combustion experiments provide fundamental insights on the physics of combustion. The latter experiments are challenging, due to the micrometric scale of the particles, to the short timescales of the combustion, and to the difficulty to isolate a single particle in specific conditions. In this work, an electrodynamic levitator is used, allowing us to isolate a single micrometric aluminum particle in a controlled environment. A laser brings the particle to a thermal runaway, and the use of a high-speed camera enables the observation of the self-sustained combustion of an aluminum droplet.

To provide an accurate and complete characterization of the burning aluminum droplet and its condensed combustion products, new optical diagnostics have been developed. The already well-known light extinction method has been extended to a multispectral application.

The high-speed color camera, equipped with a triple band filter to enable a close to trichromatic observation, is coupled with a white LED that serves as a backlight. The backlit oxide cloud that surrounds the burning aluminum droplet is captured by the camera and compared to the non-backlit oxide cloud to assess its optical depth. The multispectral nature of the diagnostics enables the evaluation of the optical depth at three distinct wavelengths. Comparing the values of spectral optical depths results in the evaluation of the size of the nanometric alumina droplets, at any location in the oxide smoke. Subsequently, the volume fraction of these condensed combustion products is assessed at any location in the cloud. The size and concentration of the condensed combustion products providing crucial information on their optical properties, the light emissions of the oxide smoke can thus be modulated by its absorption and emission properties. The oxide smoke being composed of condensed alumina as well as gaseous species, a newly developed numerical method enables the splitting of the condensed phase light emissions from the gaseous phase light emissions. The condensed phase emissions following Planck's law, the temperature field surrounding the burning aluminum droplet is assessed. In addition, the gaseous emission profiles are measured and conclusions about the nature of these emissions can be drawn. Close to the aluminum droplet, blue emissions attributed to the presence of AlO are observed, while further from the droplet, hypothesized electronic emissions are measured.

With the use of these experimental results, the structure of the aluminum droplet burning in atmospheric air and its surrounding oxide cloud is clearly identified. To assess and quantify the impact of the environment on combustion, additional experiments have been performed in various oxygen concentrations, various diluents, various pressures, and using CO_2 as the oxidizer. Upon selecting key combustion parameters, such as maximum temperature or the flame stand-off ratio, the influence of the environment on the system is characterized. Empirical and physical correlations have been proposed for the identified key combustion parameters, highlighting the mechanisms at play during the combustion of an aluminum particle. These experimental results will also provide a crucial database of targets for simulation purposes.

The obtained experimental results are then compared to simulation results available in the literature, as well as to results obtained using the model developed in-house at ArianeGroup. A brief parametric study pinpoints the parameters that can significantly influence the simulation results, and displays the incapacity of the current model to predict experimental results. As the condensation reaction of alumina was considered to follow an Arrhenius law, an improvement in the model was proposed by using a condensation reaction rate based on the classical nucleation theory. A reaction rate based on the sum of the nucleation rate of alumina and of the growth rate of alumina nuclei has been developed. The resulting temperature profiles appear to be only slightly impacted by this implementation, while the concentration profiles of alumina are significantly impacted. While none of the configurations offered an acceptable correlation to the experimental results, the results are encouraging and the implementation of a more accurate model based on the classical nucleation theory is expected to provide a more accurate simulation of the combustion process.

Although the application of the developed optical diagnostics requires a stable and symmetric combustion, extending the set of experimental burning conditions, such as studying more oxygen concentrations, more pressures and more diluents would significantly enhance the quality of the correlations and could support the proposed observations and conclusions.

In addition to the parameters evaluated thanks to the optical diagnostics, various parameters remain unknown, such as the aluminum vapor concentration profile. The group of Professor Aldén and Professor Li in Lund recently developed a laser diagnostic to assess this concentration profile, and adding this to the current setup would further enhance the degree of characterization of the burning aluminum droplet. Quenching the aluminum droplet would also provide more insight into the nature and size of the condensed combustion products. In addition, the characterization proposed in this work could be extended to different metals burning in the vapor phase regime. The comparison of the combustion mechanisms between different metals could result in the identification of future fuels or clean energy carriers.

This work also sheds light on the discrepancies between the current models of aluminum droplet combustion and experimental results. Gaseous phase combustion is widespread and well known in the field of numerical simulations. However, solid combustion is more recent, and accounting for condensed combustion products appears as the real challenge. Generally, strong hypotheses must be made for alumina condensation, the released energy of alumina condensation or the transport mechanisms of this oxide smoke. In order to accurately predict the experimental result, the development of a complex numerical model is required. The implementation of the total classical nucleation theory to correctly account for alumina condensation appears mandatory. This is challenging because it requires a complex tracking of alumina nuclei, considering intermediate species and condensation rates that are a function of many parameters, in opposition to the previously considered Arrhenius law. It is, however, believed that this long-term perspective would provide a more accurate simulation of the system, paving the way for the simulation of different fuels or more complex systems.

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Appendix A

Electric field studies

A.1 Derivation of the electric field

In this section, it is proposed to derive the electric field generated by the set of 3 electrodes. Note that this section is greatly inspired by the work of Frickel, as mentioned in the manuscript. It is recalled that the upper and lower electrodes are supplied with a constant current, while the ring electrode is supplied with an alternating current. To express the total electric field, the alternating electric field E_{AC} associated to the potential Φ_{AC} will be determined first, ignoring the constant electric field. Then, the alternating electric field is ignored to derive the constant electric field E_{DC} and its associated electric potential Φ_{DC} . The total electric field will be considered to be the sum of the two determined electric fields. In order to simplify this study, and considering that this field only depends on the difference in potential between the electrodes, the 0 in terms of electric potential is chosen as the mean between the upper and lower electrodes. A vertical axis $\vec{z}$ is defined as the axis passing through the center of the ring electrode, and through the center of the upper and lower electrodes. A radial axis $\vec{r}$ is determined, perpendicular to the $\vec{z}$ axis, and with its center located at the center of the levitator.

In an axisymmetric system empty of charges, the electric potential has the following shape in spherical coordinates:

$$\Phi(r_s, \theta_s, t) = \sum_{n=0}^{\infty} A_n r_s^n P_n(\cos(\theta_s)) \quad (\text{A.1})$$

With A_n geometry dependent coefficients, t the time, and P_n the Legendre polynomials. Rewriting this equation in cylindrical coordinates and expressing the first terms of the sum gives:

$$\Phi(r_s, z, t) = A_0 - A_1 z + A_2 (r^2 - 2z^2) + A_3 (3zr^2 - 2z^3) + A_4 (3r^4 - 24z^2 r^2 - 8z^4) + \dots \quad (\text{A.2})$$

Given the geometry of the system, the alternating potential will be an even function of the z coordinate: $\Phi_{AC}(r, z, t) = \Phi_{AC}(r, -z, t)$. Similarly, the constant potential generated by the upper and lower electrodes will be an odd function of the z coordinate: $\Phi_{DC}(r, z, t) =$

$-\Phi_{DC}(r, -z, t)$. The AC potential will thus only contain the even terms of equation A.2, and the DC potential will only contain the odd terms of equation A.2. It thus comes:

$$\Phi_{AC}(r, z, t) = A_0 + A_2 (r^2 - 2z^2) + A_4 (3r^4 - 24z^2r^2 - 8z^4) + \dots \quad (\text{A.3})$$

The potential can be adjusted so that the term A_4 is 0, and upon neglecting the terms of higher orders, it comes for the alternating potential in the levitator:

$$\Phi_{AC}(r, z, t) = A_0 + A_2 (r^2 - 2z^2) \quad (\text{A.4})$$

The spatial point located on the upper electrode that is the closest to the center of the levitator is located at $r = 0$ and $z = z_0$. At the moment, the potential at this location is considered to be 0 as the DC potential is neglected in this first derivation:

$$\Phi_{AC}(0, z_0, t) = 0 = A_0 - 2A_2z_0^2 \quad (\text{A.5})$$

Similarly, the points of the circular electrode that are the closest to the center of the levitator are located at $r = r_0$ and $z = 0$ with r_0 the internal radius of the ring electrode. The circular electrode has an alternating non-zero potential:

$$\Phi_{AC}(r_0, 0, t) = -V \cos(2\pi ft) = A_0 + A_2r_0^2 \quad (\text{A.6})$$

With f the frequency of the alternative current and V the amplitude of the alternating potential. From equations A.5 and A.6, it comes that $A_2 = \frac{A_0}{2z_0^2}$ and $A_0 = \frac{-2V \cos 2\pi ft z_0^2}{2z_0^2 + r_0^2}$. The electric potential generated by the ring electrode alone, at any location of the levitator is thus expressed as:

$$\Phi_{AC}(r, z, t) = \frac{V \cos(2\pi ft) z_0^2}{2z_0^2 + r_0^2} (2z^2 - r^2 - 2z_0^2) \quad (\text{A.7})$$

The electric field that results from this potential is obtained by evaluating the opposite of the potential's gradient:

$$\overrightarrow{E_{AC}} = -\nabla \Phi_{AC} = \frac{4V \cos(2\pi ft)}{2z_0^2 + r_0^2} \left(\frac{r}{2} \overrightarrow{r} - z \overrightarrow{z} \right) \quad (\text{A.8})$$

If an offset V_1 is applied to the ring electrode, then the alternating electric field becomes:

$$\overrightarrow{E_{AC}} = -\nabla \Phi_{AC} = \frac{4(V_1 + V \cos(2\pi ft))}{2z_0^2 + r_0^2} \left(\frac{r}{2} \overrightarrow{r} - z \overrightarrow{z} \right) \quad (\text{A.9})$$

Let us now consider that the potential generated by the ring electrode is 0. As proposed in the literature, only the first degree is considered for the equation A.2 for the constant electric potential:

$$\Phi_{DC}(r, z, t) = -A_1 z \quad (\text{A.10})$$

And the resulting electric field is easily obtained:

$$\overrightarrow{E_{DC}} = A_1 \overrightarrow{z} \quad (\text{A.11})$$

According to Frickel, A_1 can be expressed as a function of the difference in potential between the upper and lower electrodes V_{DC} , multiplied by a geometrical constant C_{DC} :

$$\overrightarrow{E_{DC}} = C_{DC} V_{DC} \overrightarrow{z} \quad (\text{A.12})$$

The overall electric field $\overrightarrow{E}$ in the levitator is expressed as:

$$\overrightarrow{E} = \overrightarrow{E_{DC}} + \overrightarrow{E_{AC}} = \frac{4(V_1 + V \cos(2\pi ft))}{2z_0^2 + r_0^2} \frac{r}{2} \overrightarrow{r} - \left(\frac{4(V_1 + V \cos(2\pi ft))}{2z_0^2 + r_0^2} z - C_{DC} V_{DC} \right) \overrightarrow{z} \quad (\text{A.13})$$

A.2 Influence of the electric field on the combustion process

A.2.1 Induced magnetic field

Although the aluminum particle loses its charges when its temperature increases, it has to be noted that in the presence of an alternating electric field, an induced magnetic field is produced as predicted by Maxwell's laws. This section provides a brief calculation to demonstrate that the generated magnetic field does not impact the combustion process. The aluminum being paramagnetic, its magnetism is lost at high temperatures, and the magnetic field will have no effect on the droplet. However, alumina is diamagnetic and its magnetism is preserved at high temperature. This calculation thus evaluates the influence of the magnetic field on an alumina particle. As a first step, the induced magnetic field must be determined. Let us recall that for any vector $\overrightarrow{V}$:

$$\nabla \times (\nabla \times \overrightarrow{V}) = \nabla (\nabla \cdot \overrightarrow{V}) - \Delta \overrightarrow{V} \quad (\text{A.14})$$

Recalling that Maxwell's equations give, then for a magnetic field $\overrightarrow{B}$: $\nabla \cdot \overrightarrow{B} = 0$. Denoting the electric field as $\overrightarrow{E}$ and in absence of electric current density, then: $\nabla \times \overrightarrow{B} = \mu_0 \epsilon_0 \frac{\partial \overrightarrow{E}}{\partial t}$ with μ_0 the vacuum permeability and ϵ_0 the vacuum permittivity. Finally, $\nabla \times \overrightarrow{E} = -\frac{\partial \overrightarrow{B}}{\partial t}$. Recombining these equations, and recalling that the rotational and temporal derivation are interchangeable if the spatial parameters are constant with time, the following equation is obtained for the magnetic field:

$$\frac{1}{c^2} \frac{\partial^2 \overrightarrow{B}}{\partial t^2} - \Delta \overrightarrow{B} = 0 \quad (\text{A.15})$$

With c the celerity of light. The induced magnetic field being necessarily perpendicular to the electric field, and the electric field being along the $\overrightarrow{r}$ and $\overrightarrow{z}$ axes, the magnetic field is along the $\overrightarrow{\theta}$ axis. The cylindrical symmetry results in $\overrightarrow{B}$ being independent on θ , but depends on r , z and t . As a result, $\overrightarrow{B}$ can be written as $\overrightarrow{B} = B \overrightarrow{\theta}$. As the spatial and temporal parameters are independent, we write:

$$B = B_\theta(r, z) T(t) \quad (\text{A.16})$$

With B_θ and $T(t)$ two independent functions. Equation A.15 becomes:

$$\frac{1}{Tc^2} \frac{\partial^2 T}{\partial t^2} \vec{\theta} - \frac{1}{B_\theta} \Delta(B_\theta \vec{\theta}) = 0 \quad (\text{A.17})$$

As this equation represents the difference of two independent functions, and as this difference is equal to 0, both functions are equal to the same constant and we can write, with K a positive constant:

$$\frac{1}{Tc^2} \frac{\partial^2 T}{\partial t^2} \vec{\theta} = \frac{1}{B_\theta} \Delta(B_\theta \vec{\theta}) = -K \vec{\theta} \quad (\text{A.18})$$

The resolution for the function T is straightforward and we can write with T_1 and T_2 two constants:

$$T(t) = T_1 \sin(\sqrt{K}ct) + T_2 \cos(\sqrt{K}ct) \quad (\text{A.19})$$

Assuming the absence of magnetic field at the initial time $t = 0$, then:

$$T(t) = T_1 \sin(\sqrt{K}ct) \quad (\text{A.20})$$

The laplacian of the magnetic field is now written:

$$\Delta(B_\theta \vec{\theta}) = -\frac{2}{r^2} \frac{\partial B_\theta}{\partial \theta} \vec{r} + \left(\nabla^2 B_\theta - \frac{B_\theta}{r^2} \right) \vec{\theta} \quad (\text{A.21})$$

From the cylindrical symmetry:

$$\frac{1}{B_\theta} \left(\nabla^2 B_\theta - \frac{B_\theta}{r^2} \right) = -K \quad (\text{A.22})$$

That further develops to:

$$\frac{1}{B_\theta} \left(\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial B_\theta}{\partial r} \right) + \frac{\partial^2 B_\theta}{\partial z^2} - \frac{B_\theta}{r^2} \right) = -K \quad (\text{A.23})$$

Assuming that the r and z variables are independent, it can be written $B_\theta(r, z,) = R(r)Z(z)$ and equation A.23 simplifies to:

$$\frac{1}{Rr} \frac{\partial}{\partial r} \left(r \frac{\partial R}{\partial r} \right) + \frac{1}{Z} \frac{\partial^2 Z}{\partial z^2} - \frac{1}{r^2} = -K \quad (\text{A.24})$$

That further develops to:

$$\frac{1}{Rr} \frac{\partial R}{\partial r} + \frac{1}{R} \frac{\partial^2 R}{\partial r^2} + \frac{1}{Z} \frac{\partial^2 Z}{\partial z^2} - \frac{1}{r^2} = -K \quad (\text{A.25})$$

This equation can be written in a simplified form:

$$\frac{R'}{Rr} + \frac{R''}{R} - \frac{1}{r^2} + K = \frac{Z''}{Z} \quad (\text{A.26})$$

Once again, two independent functions are equal, and it is assumed that they are equation to a constant $-K_2$ with $K_2 > 0$ due to the impossibility for Z to vary exponentially with z . Then it can be written:

$$\frac{R'}{Rr} + \frac{R''}{R} - \frac{1}{r^2} + K = -K_2 \quad (\text{A.27})$$

That can be rewritten as:

$$r^2 R'' + r R' - R (1 - (K_2 + K) r^2) = 0 \quad (\text{A.28})$$

Introducing a variable $r_b = (K + K_2)^{1/2} r$ so that equation A.28 becomes:

$$r_b^2 R'' + r_b R' + R (r_b - 1) = 0 \quad (\text{A.29})$$

The solution to such an equation is the composition of a Bessel function of the first kind of order 1, J_1 with a Bessel function of the second kind of order, 1 Y_1 :

$$R = R_1 J_1 \left(\sqrt{K + K_2} r \right) + R_2 Y_1 \left(\sqrt{K + K_2} r \right) \quad (\text{A.30})$$

With R_1 and R_2 constants. As Y_1 diverges to $-\infty$ at $r = 0$ and as the magnetic field cannot be infinite at the center of the chamber, it comes $R_2 = 0$. Equation A.26 also led to:

$$\frac{Z''}{Z} = -K_2 \quad (\text{A.31})$$

For which the solution is:

$$Z = Z_1 \cos \left(\sqrt{K_2} z \right) + Z_2 \sin \left(\sqrt{K_2} z \right) \quad (\text{A.32})$$

With Z_1 and Z_2 constants. As the electric field is an odd function along z , the magnetic field must also be an odd function of z as the rotational does not invert the parity of a function. Then it comes $Z_1 = 0$ and the magnetic field can be written as:

$$B_\theta = R_1 J_1 \left(\sqrt{K + K_2} r \right) Z_1 \sin \left(\sqrt{K_2} z \right) T_1 \sin \left(\sqrt{K} ct \right) \quad (\text{A.33})$$

The temporal dependence of the magnetic field is the same as the temporal dependence of the electric field by a phase shift. Thus it is proposed to assume $\sqrt{K} = \frac{2\pi f}{c}$ and thus, $K \ll 1$. Similarly, if $\sqrt{K_2} \gg 1$, therefore the magnetic field along z will have several nodes while this is physically impossible to have magnetic nodes at a given height in the system. Thus, $K_2 \ll 1$. As a result, a series expansion of the magnetic field is performed:

$$J_1(x) \approx \frac{x}{2} (x \rightarrow 0) \quad (\text{A.34})$$

$$\sin(x) \approx x (x \rightarrow 0) \quad (\text{A.35})$$

Then a simplified formulation for the magnetic field is proposed, using a constant Cst :

$$B_\theta \approx Cst \cdot rz \sin(2\pi ft) \quad (\text{A.36})$$

Recalling that $\nabla \times \vec{B} = \mu_0 \epsilon_0 \frac{\partial \vec{E}}{\partial t}$, the constant can be identified, and thus:

$$B_\theta \approx -\frac{8V\mu_0\epsilon_0\pi f}{2z_0^2 + r_0^2} \cdot rz \sin(2\pi ft) \quad (\text{A.37})$$

It is important to note that the magnetic field was analytically determined by a derivation that did not require knowledge of the electric field in the chamber. The latter only allowed the identification of the constant giving the amplitude of the magnetic field. Since the electric field only presents the first terms of a sum, this may explain why the rotational of this electric field does not directly give the opposite of the partial derivative of the magnetic field with respect to time. This derivation is however considered sufficient to determine an order of magnitude of the amplitude of the force that applies on an alumina particle.

The magnetic force exerted on an alumina particle $\vec{F}_{mag}$ is expressed as:

$$\vec{F}_{mag} = \nabla (\vec{m} \cdot \vec{B}) \quad (\text{A.38})$$

With $\vec{m}$ the magnetic moment of the dipole, that can be expressed as:

$$\vec{m} = V\vec{M} = V\chi_v\vec{H} = \frac{V\chi_v}{\mu}\vec{B} \quad (\text{A.39})$$

With V the volume of the dipole, $\vec{M}$ its magnetization, χ_v its volume magnetic susceptibility, $\vec{H}$ the magnetic excitation and μ the magnetic permeability of the material. The magnetic force that applies on an alumina droplet of radius r_p is thus expressed as:

$$\overrightarrow{F_{Al_2O_3}} = \frac{4\pi r_p^3 \chi_v}{3\mu} \left[\left(\frac{8V\mu_0\epsilon_0\pi f}{2z_0^2 + r_0^2} \sin(2\pi ft) \right)^2 z^2 r \vec{r} + \left(\frac{8V\mu_0\epsilon_0\pi f}{2z_0^2 + r_0^2} \sin(2\pi ft) \right)^2 z r^2 \vec{z} \right] \quad (\text{A.40})$$

Let us consider, in the aim of overestimating the impact of the magnetic field, an alumina particle of 100 microns in radius. Let us use $\chi_v = -1.43 \cdot 10^{-6}$, $\mu \approx \mu_0 = 12.57 \cdot 10^{-7} \text{ kg.m.A}^{-2}.\text{s}^{-2}$, $V = 4000 \text{ V}$, $\epsilon_0 = 8.85 \cdot 10^{-12} \text{ A}^2.\text{s}^4.\text{kg}^{-1}.\text{m}^{-3}$, $f = 50 \text{ Hz}$, $z_0 = 1 \text{ cm}$ and $r_0 = 2.5 \text{ cm}$. In the aim to overestimate this force, it is considered that $r = r_0$, $z = z_0$ and that $\sin(2\pi ft) = 1$. With these assumptions, it comes:

$$\overrightarrow{F_{Al_2O_3}} = -5.47 \cdot 10^{-32} \vec{r} - 1.37 \cdot 10^{-31} \vec{z} \quad (\text{A.41})$$

In comparison, the weight of an alumina particle of identical size $P_{Al_2O_3}$ is evaluated at:

$$\overrightarrow{P_{Al_2O_3}} = -7.48 \cdot 10^{-8} \vec{z} \quad (\text{A.42})$$

As the weight of a particle is about 10^{23} orders of magnitudes larger than the magnetic force that results from the alternating electric field, the influence of the field on the condensed species during the combustion process is neglected. The influence of the electric field on the

lighter elements such as gases or electrons has not been theoretically evaluated. However, a brief set of experiments presented in the following section provides more insight regarding the influence of the electric field on the combustion process.

A.2.2 Influence of $\vec{E}$ on the observed combustion

In order to provide some insights on the influence of the electric field on the combustion process of a single aluminum droplet, the original experimental setup has been modified to enable the control of the electric field during combustion. A set of fast responding switches has been added to the power supply of the electrodes. During a first series of experiments, aluminum particles were burned in atmospheric air, and the electric field was turned off upon particle ignition (based on the laser shut-off signal). The images obtained in the absence of an electric field are compared to images in presence of the electric field in Fig. A.1. No diagnostics were applied to these data except for the visualization of the combustion. No discrepancies can be observed upon switching off the electric field, and the two combustion processes are similar.

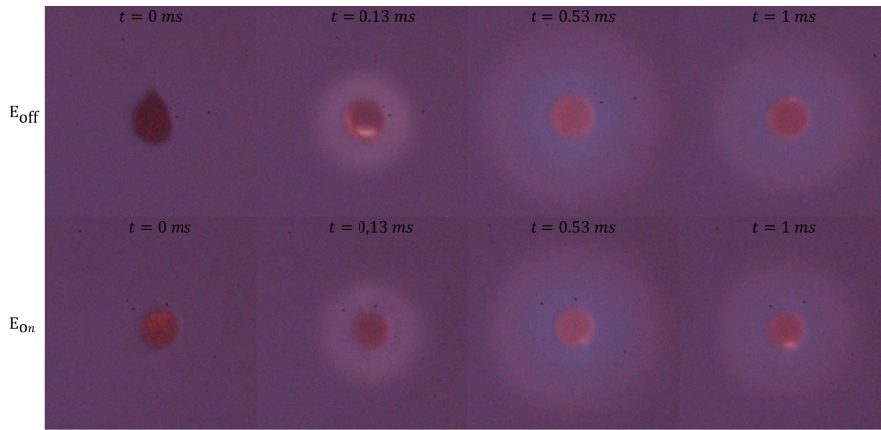


Figure A.1: Combustion images with the electric field (top) and without the electric field (bottom).

Another study has been performed in a pure oxygen mixture, to observe the influence of the electric field in the presence of greater concentration of condensed phase. In such conditions, the burning behaviour is highly chaotic and no conclusions could be drawn. Although no obvious observations were made regarding the influence of the electric field on the combustion process, a more in depth study is necessary. Performing multiple tests to obtain statistical result is a perspective that will be examined shortly.

Appendix B

Spectral response graphs

In this appendix, the spectral response graphs of various optical devices, introduced in the experimental setup chapter, are summarized.

B.1 Dichroic filters

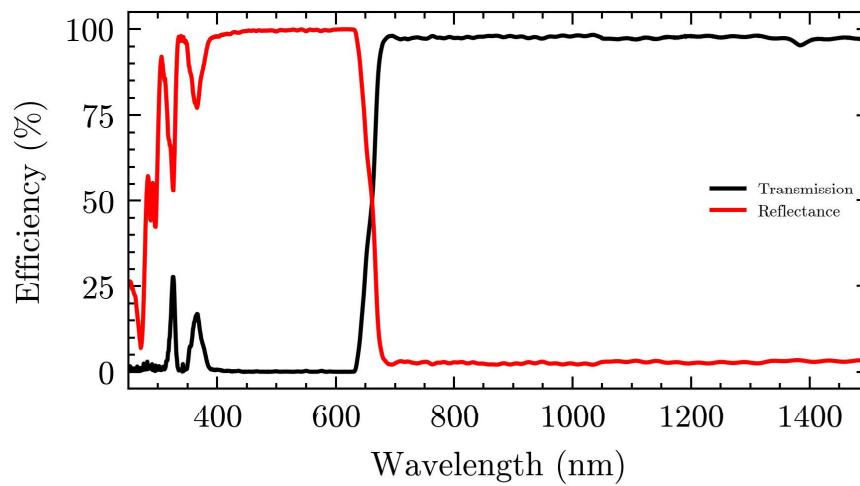


Figure B.1: Spectral transmission and reflectance efficiency of the DMLP650R dichroic filter .

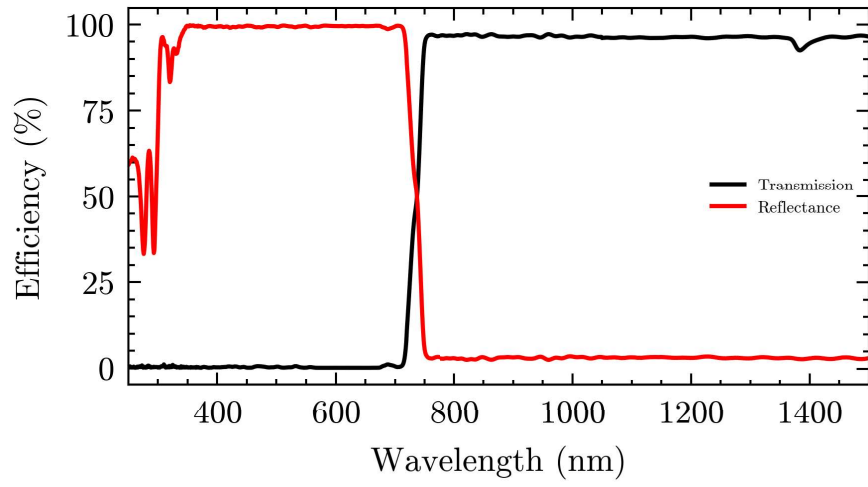


Figure B.2: Spectral transmission and reflectance efficiency of the DMLP735R dichroic filter .

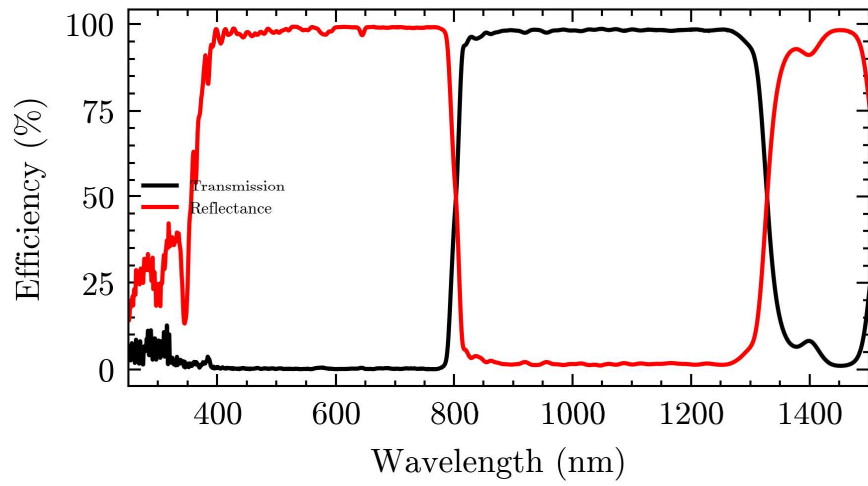


Figure B.3: Spectral transmission and reflectance efficiency of the DMLP805R dichroic filter .

B.2 Bandpass filters

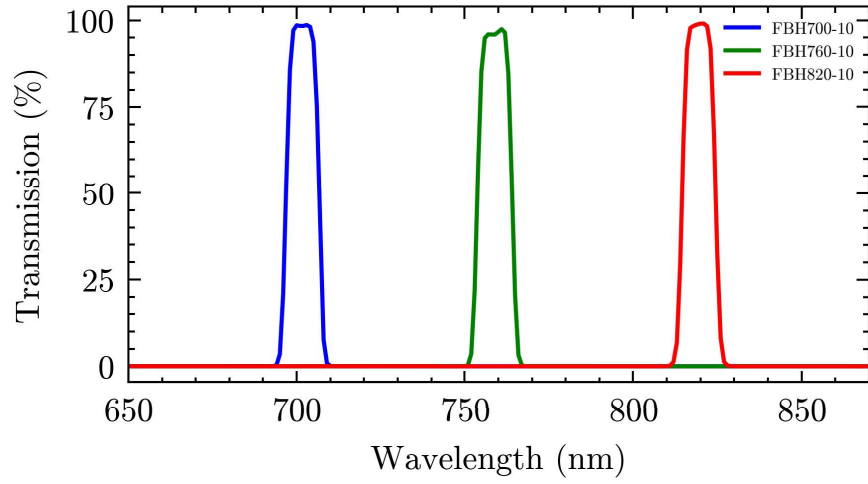


Figure B.4: Spectral transmission efficiency of the FBH700-10, FBH760-10 and FBH820-10 bandpass filters .

B.3 Photomultipliers

The three photomultipliers are from the identical model and the spectral response is the following:

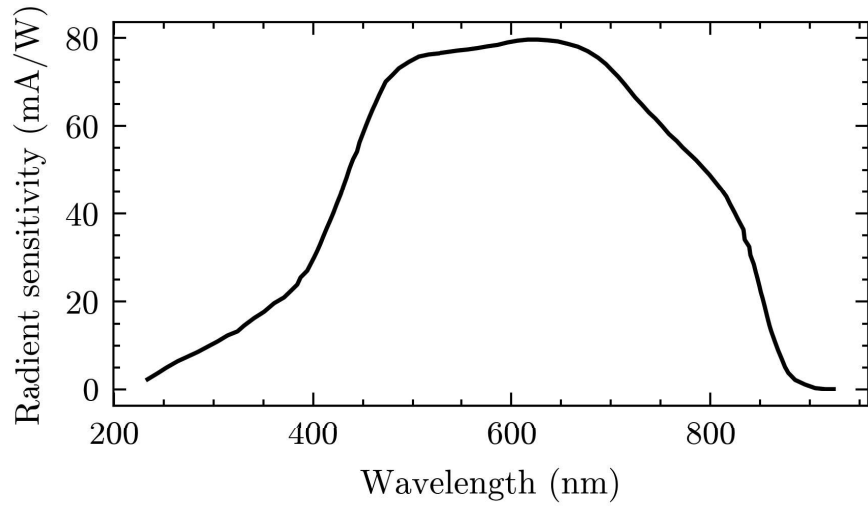


Figure B.5: Spectral radiant sensitivity of the PM1002.

B.4 Solis LED

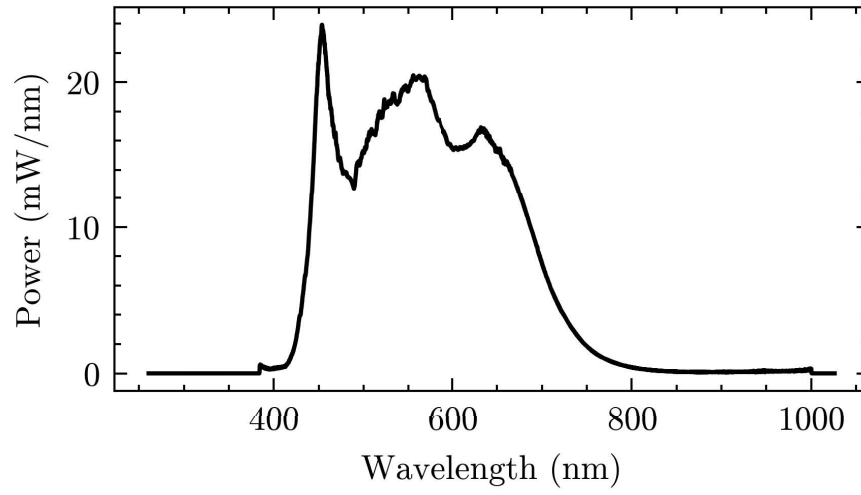


Figure B.6: Spectral emission profile of the SOLIS 3-C white LED .

Appendix C

Camera calibration

C.1 Spectral overlap calibration

The spectral responses of the R, G and B matrices of the camera are plotted as a function of the wavelength in Fig. C.1. From this figure, it is made obvious that spectral overlap will occur. As an example, assuming that a 1 W radiation at 530 nm impacts the camera's sensor. From Fig. C.1, it comes that a signal of about 0.27 A will be measured by the G matrix, a signal of about 0.07 will be measured by the B matrix, and a signal of about 0.02 will be measured by the R matrix. The signals on the B and G matrices are not expected nor desired as the radiation is expected to be entirely green and the signal is thus expected to only influence the G matrix.

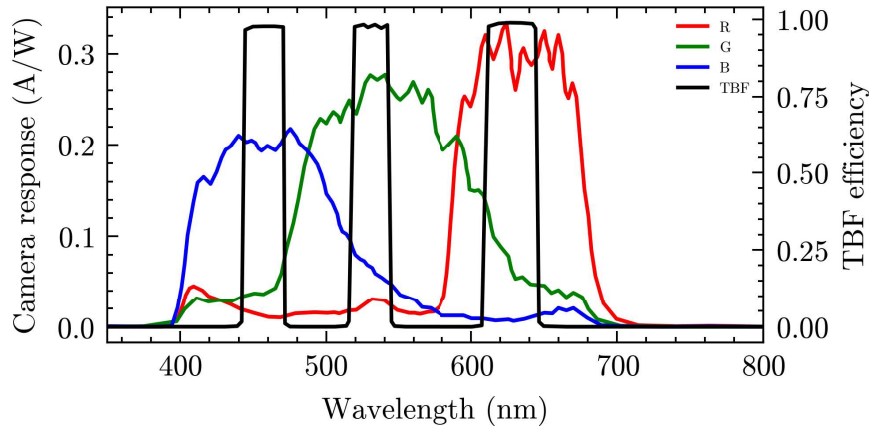


Figure C.1: Spectral response of the R, G and B matrices of the color camera. The efficiency of the Triple Band Filter (TBF) is also displayed.

To account for this spectral overlap, a calibration is performed. Let us define R_m , G_m and B_m the intensities measured by the camera on the R, G and B matrices. Let us define R_r , G_r and B_r the real emitted intensities that reach the camera's sensor. To account for spectral overlap, 6 constants $(K_n)_{n=1\dots 6}$ are determined so that the following system of equation is verified for any measurements with the triple band filter used with the camera:

$$\begin{cases} R_m = R_r + K_1 G_r + K_2 B_r \\ G_m = K_3 R_r + G_r + K_4 B_r \\ B_m = K_5 R_r + K_6 G_r + B_r \end{cases} \quad (C.1)$$

To determine the constants K , three calibration experiments are performed. The three experiments are represented in Fig. C.2.

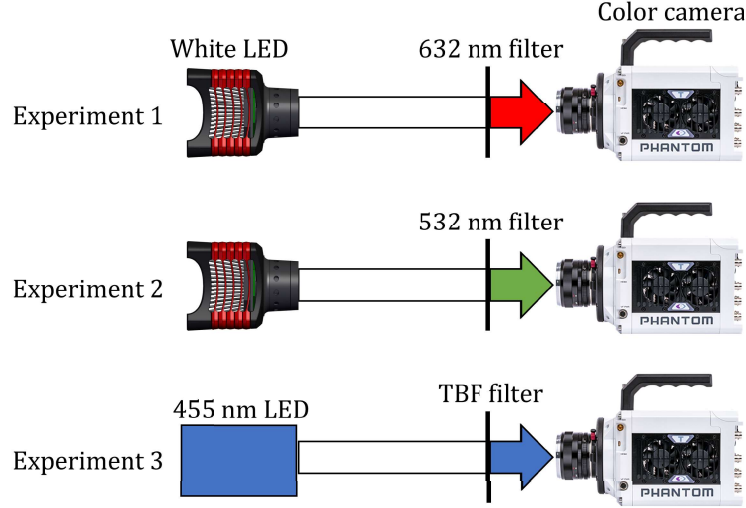


Figure C.2: Schematics of the experiments performed to determine the spectral overlap coefficients.

From experiment 1 in Fig. C.2, the system C.1 is simplified to:

$$\begin{cases} R_m = R_r \\ G_m = K_3 R_r \\ B_m = K_5 R_r \end{cases} \quad (C.2)$$

The parameters K_3 and K_5 are determined. From experiment 2 in Fig. C.2, the system C.1 is simplified to:

$$\begin{cases} R_m = K_1 G_r \\ G_m = G_r \\ B_m = K_6 G_r \end{cases} \quad (C.3)$$

The parameters K_1 and K_6 are determined. Finally, from experiment 3 in Fig. C.2, the system C.1 is simplified to:

$$\begin{cases} R_m = K_2 B_r \\ G_m = K_4 B_r \\ B_m = B_r \end{cases} \quad (C.4)$$

The last parameters K_2 and K_4 are assessed, and the spectral overlap can be correctly accounted for during experimental measurements, and during the blackbody calibration described next.

C.2 Blackbody calibration

Let us consider that the color camera, equipped with the triple band filter, is placed in front of a blackbody of known temperature T . Assuming that all of the pixel of the cameras have identical intensities R , G and B at the three wavelengths of the triple band filter, a calibration function C must be determined so that:

$$C(R, G, B) = T \quad (\text{C.5})$$

To obtain this calibration function, two blackbody calibrations have been carried out.

A first calibration process was performed on a 'low temperature' blackbody, which could reach a maximum temperature of 2000 K. A schematic of this calibration setup is presented in Fig. C.3. This setup ensures the use of the same optical setup as the one used for capturing aluminum combustion images. Below 2000 K, Planck's law predicts extremely low green and blue emissions while red emissions will prevail. In other words, below 2000 K, $R \gg G > B$. As a result, extrapolating the calibration results obtained with this calibration, to the temperatures expected during aluminum combustion, near 3600 K, is sensitive and challenging.

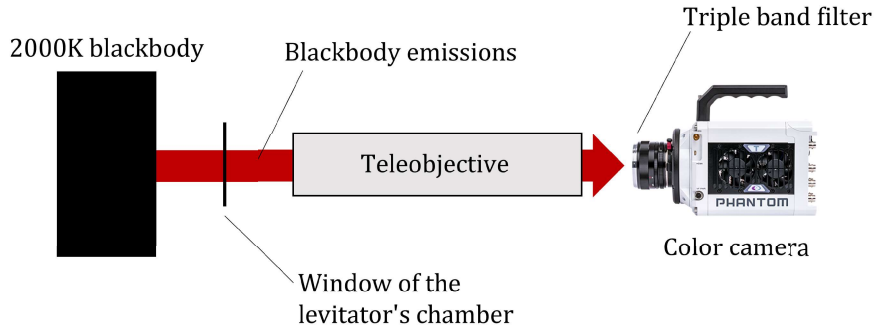


Figure C.3: Schematic of the 'low temperature' blackbody calibration setup.

To refine the calibration procedure and improve the accuracy of the extrapolation of the calibration law to high temperatures, a second calibration procedure is carried out with a 'hot temperature' blackbody, that reaches a maximum temperature of 3200 K. A schematic of this second calibration setup is presented in Fig. C.4. Practical difficulties prevented us from using the same optical setup as in Fig. C.3. Instead, a Nikon 105mm UV objective replaced the teleobjective, and density filters were used to limit the light intensity reaching the camera's sensor.

During both calibration procedures, images of the blackbody radiations were captured with the color camera at various blackbody temperatures. For each blackbody temperature, the exposure time, aperture of the objective, and neutral density filters were varied, to provide

various R , G , and B values at the same temperature. In the case of the 2000 K blackbody, the R , G and B values are obtained by averaging the values of all the pixels. In the case of the 3200 K blackbody, the magnification of the optical system being less important than with the teleobjective, only a portion of all the pixels was illuminated. The R , G and B values were obtained by averaging the values of the intensities of the illuminated pixels.

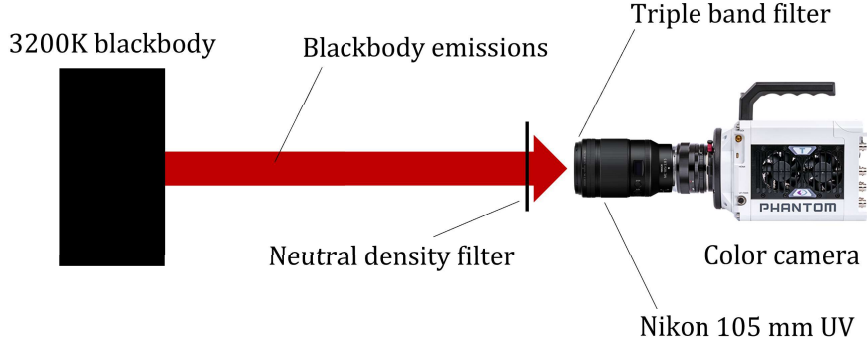


Figure C.4: Schematic of the 'high temperature' blackbody calibration setup.

With the hypothesis of an ideal sensor, at a fixed blackbody temperature, the R , G and B values are expected to be influenced by changes in apertures or exposure time. However, as these variations only influence the amount of light impinging the sensor, but do not influence the spectral nature of the emissions, the ratios R/G , R/B and G/B are expected to remain constant, as long as the blackbody temperature is fixed. However, the sensor of the camera appears to be sensitive to the amount of light impinging the sensor. As a result, a gray pixel value P_g defined as $P_g = R + G + B$ is used as a marker of the total emission intensity measured by the camera's sensor. Figure C.5 displays the values of the R/G ratio obtained in both calibration procedures, as a function of the pixel gray value P_g and of the blackbody's temperature. Similar graphs are displayed for the R/B ratio in Fig. C.6 and for the G/B ratio in Fig. C.7.

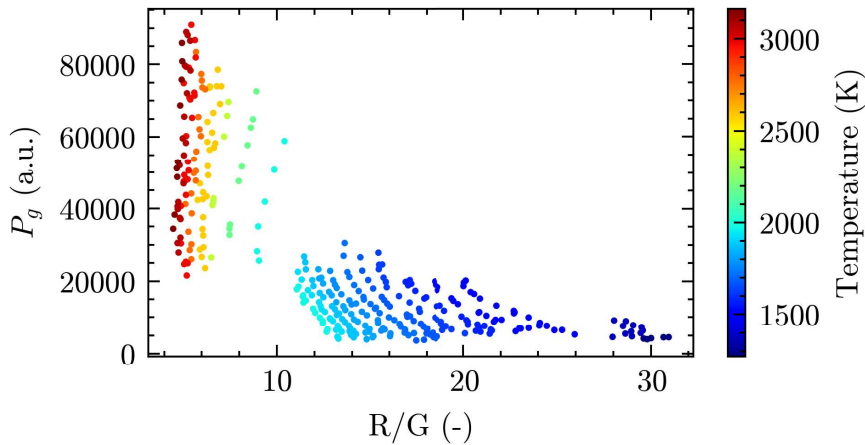


Figure C.5: R/G ratio for various pixel gray value P_g and blackbody temperature.

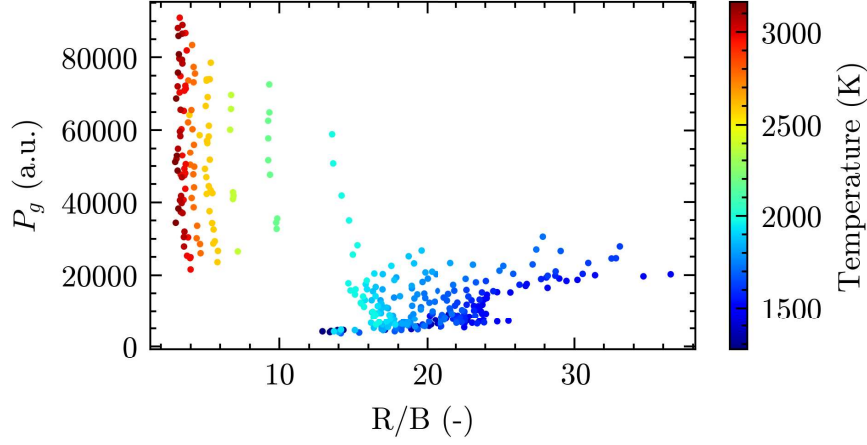


Figure C.6: R/B ratio for various pixel gray value P_g and blackbody temperature.

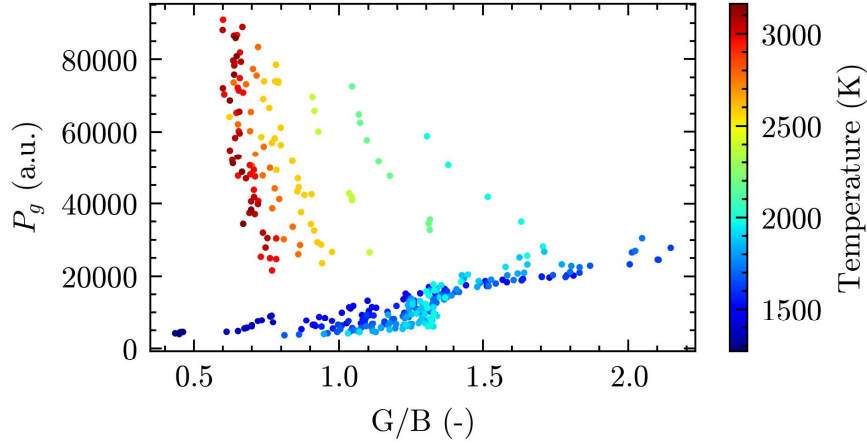


Figure C.7: G/B ratio for various pixel gray value P_g and blackbody temperature.

It is observed that calibration data for low temperatures (<2000 K) show unpredictable trends when the value of B is involved in the ratio. At high temperatures, the ratios show acceptable trends. Indeed, at low temperatures, low to no blue emissions are expected using Planck's law. Combining this effect to the low pixel gray values, it is assumed that the blue emissions are highly perturbed by noise. As a result, it is proposed at first to determine a calibration function based on the R/G ratio given its validity for all the calibration points. An arbitrary function is thus determined, relating R/G , P_g , and the temperature of the blackbody T :

$$R/G = \frac{(a \cdot T)^b}{(c \cdot P_g + d)^e} + \frac{(f \cdot T)^g}{(h \cdot P_g + i)^j} + k \quad (\text{C.6})$$

Where $a, b, c, d, e, f, g, h, i, j$ and k are optimized parameters for the calibration law. The calibration function for the R/G ratio is displayed in Fig. C.8. The calibration function appears to correctly predict the trend of the R/G ratio as a function of P_g and the blackbody

temperature. A quantitative evaluation of the efficiency of this calibration law is presented in Fig. C.9, where the relative error e between the experimental values of the R/G ratios are compared to the R/G ratios computed using the calibration law. A maximum relative error on the ratio of 18 % is measured, while the mean relative error is 3.28 %, supporting the validity of the calibration law.

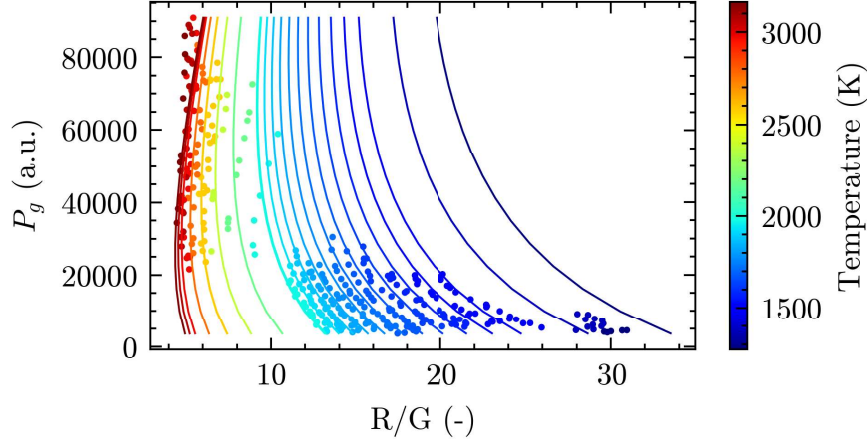


Figure C.8: Experimental R/G values (dots) and R/G values obtained using equation C.6 (solid lines).

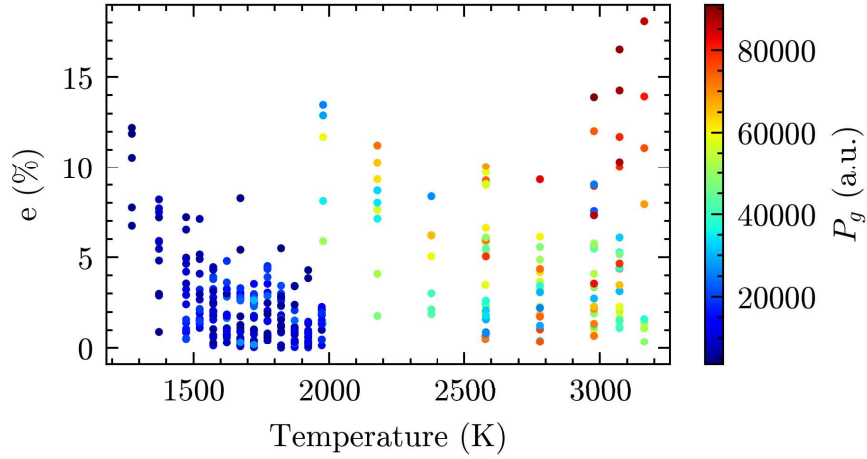


Figure C.9: Relative error between calibration data and the calibration law from equation C.6.

From Figures C.6 and C.7, it is rather obvious that a similar procedure cannot be performed, as no calibration law could be fitted to these data, due to the perturbations of the ratios at low temperatures. However, as the high temperature ratios show acceptable behaviors, it is proposed to correct the low temperature R/B and G/B ratios. To do so, a transfer law is established to correlate high temperatures R/B ratios to high temperatures

R/G ratios. This transfer law expressed R/B as a function of R/G , P_g , T , and optimized parameters $a, b, c, d, e, f, g, h, i, j$ and k :

$$R/B = \left(\frac{(a \cdot T)^b}{(c \cdot P_g + d)^e} + \frac{(f \cdot T)^g}{(h \cdot P_g + i)^j} + k \right) \cdot \frac{R}{G} \quad (\text{C.7})$$

Figure C.10 shows that the transfer law correctly predicts the R/B values at high temperatures as the triangles are superimposed with the dots. The low temperature ratios appear corrected, as a similar trend to the R/G ratios is observed.

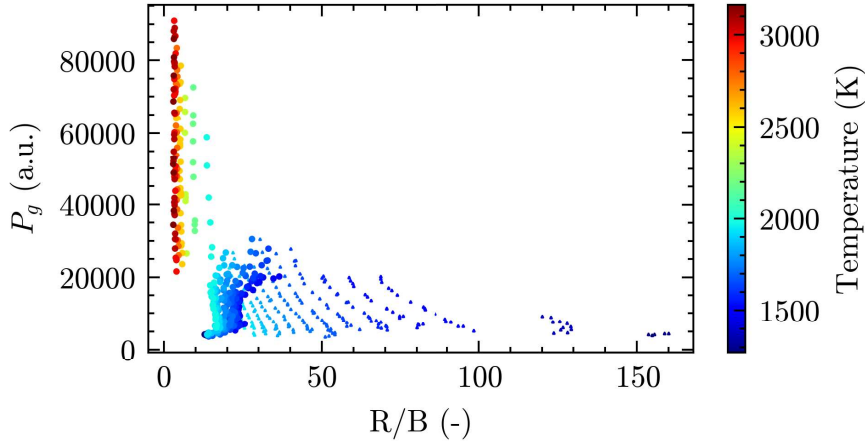


Figure C.10: Experimental R/B (dots) and corrected R/B using equation C.7 (triangles).

The mean relative error between the high temperature experimental R/B values and the reconstructed R/B values using equation C.6 is 2.84 % supporting the validity of the transfer law. Finally, to reconstruct the low temperature values of the G/B ratio, it is proposed to consider the most simple relation:

$$G/B = \frac{R}{B} \cdot \frac{1}{\frac{R}{G}} \quad (\text{C.8})$$

Similarly to the R/B reconstruction, the high temperature G/B ratios are correctly reconstructed with a mean error of 2.84 % between experimental values and the reconstructed values using equation C.8. The comparison between the experimental data and the reconstructed data is presented in Fig. C.11. Similarly to the reconstruction of the R/B ratio, the reconstructed high temperature ratios are superimposed to the experimental data, while the low temperature ratios appear to be correctly reconstructed.

With the use of equation C.6, a lookup table is generated, so that each pair $(R/G, P_g)$ corresponds to a blackbody temperature T_{RG} . Using equation C.7, the calibration ratios R/G provide the corrected calibration ratios R/B and a lookup table is generated, so that each pair $(R/B, P_g)$ corresponds to a blackbody temperature T_{RB} . Finally, using equation C.8, the calibration ratios R/G and R/B provide the corrected calibration ratios G/B , and a lookup table is generated so that each pair $(G/B, P_g)$ corresponds to a blackbody temperature T_{GB} .

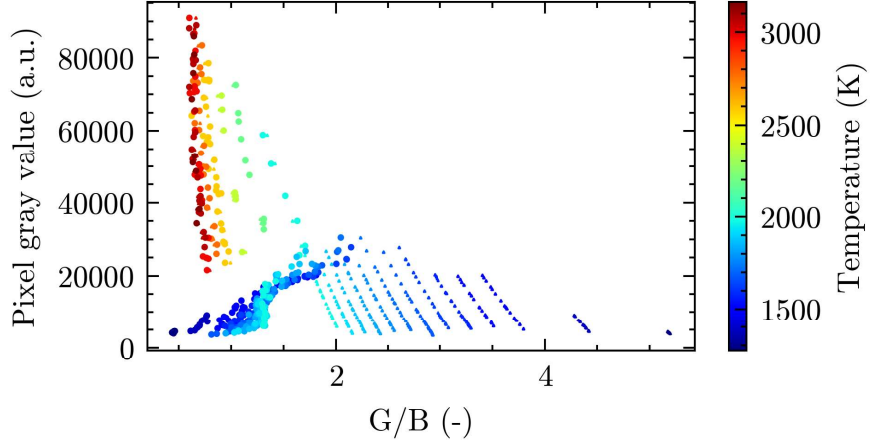


Figure C.11: Experimental G/B (dots) and corrected G/B using equation C.8 (triangles).

This calibration procedures thus provides three lookup tables, so that upon observing an emitting body and measuring three intensities R , G and B , three temperatures are obtained T_{RG} , T_{RB} and T_{GB} . If the observed body is a blackbody, or a greybody, then: $T_{RG} = T_{RB} = T_{GB}$.

Appendix D

Experimental profiles

All the obtained experimental results are presented in this appendix. Temperature, alumina concentration, alumina particle size and gaseous emission profiles are presented for all the experimental conditions studied. It is recalled that the Al droplet has for all cases an initial diameter of 70 microns.

D.1 Temperature profiles

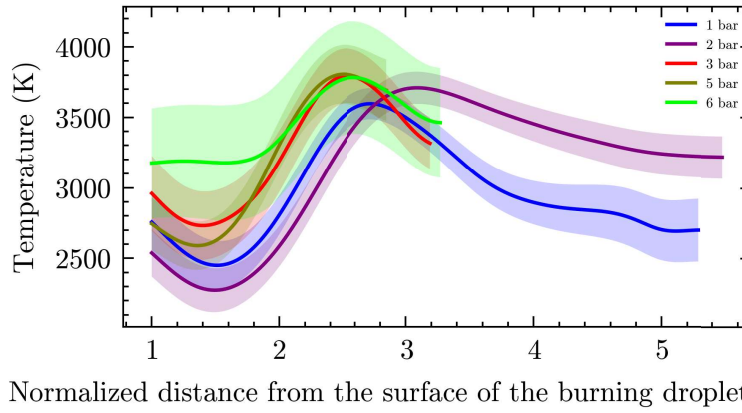


Figure D.1: Temperature profiles surrounding an Al droplet burning in air at various pressures

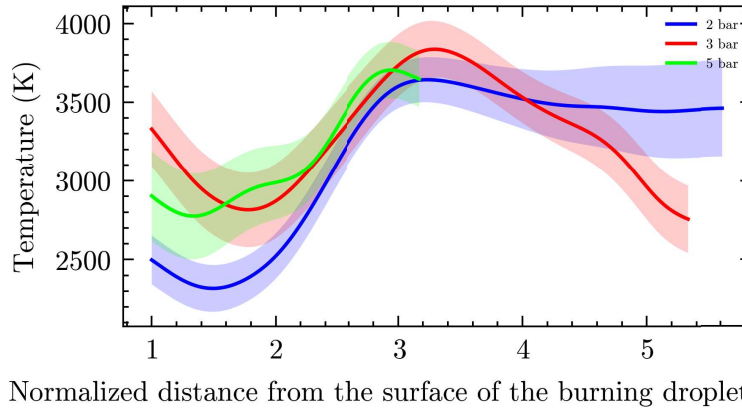


Figure D.2: Temperature profiles surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .

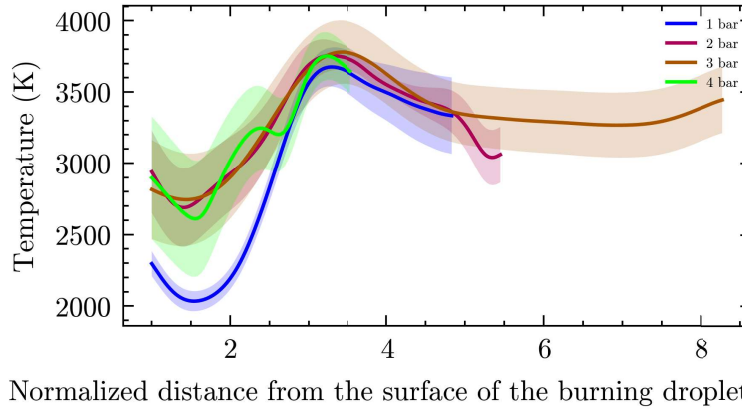


Figure D.3: Temperature profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures .

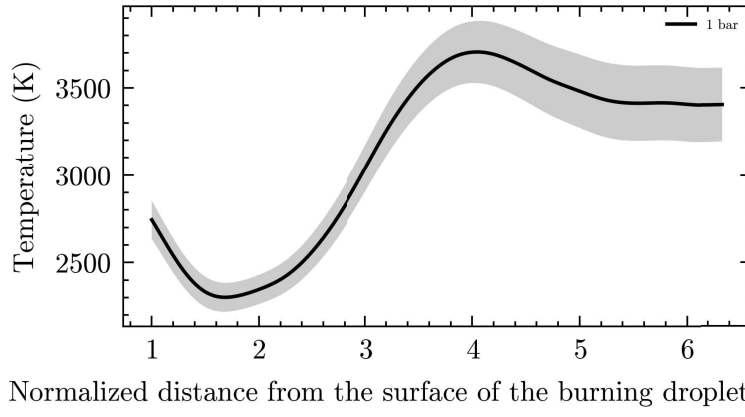


Figure D.4: Temperature profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .

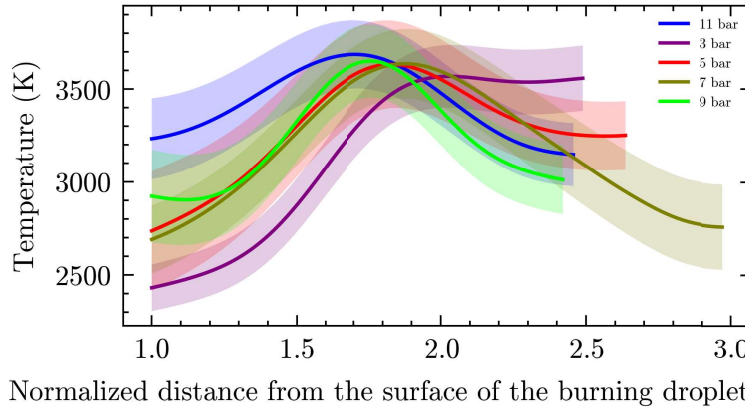


Figure D.5: Temperature profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures .

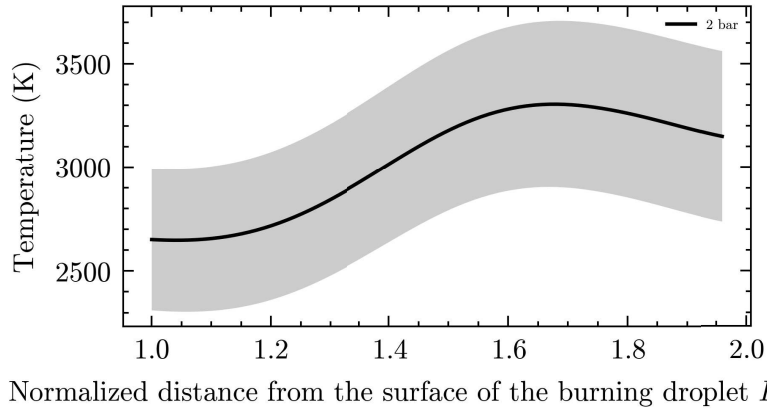


Figure D.6: Temperature profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars .

D.2 Alumina volume fraction profiles

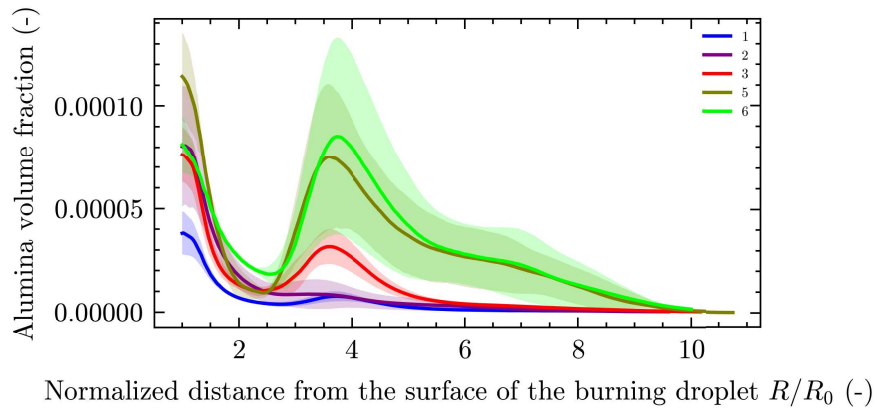


Figure D.7: Alumina volume fraction profiles surrounding an Al droplet burning in air at various pressures .

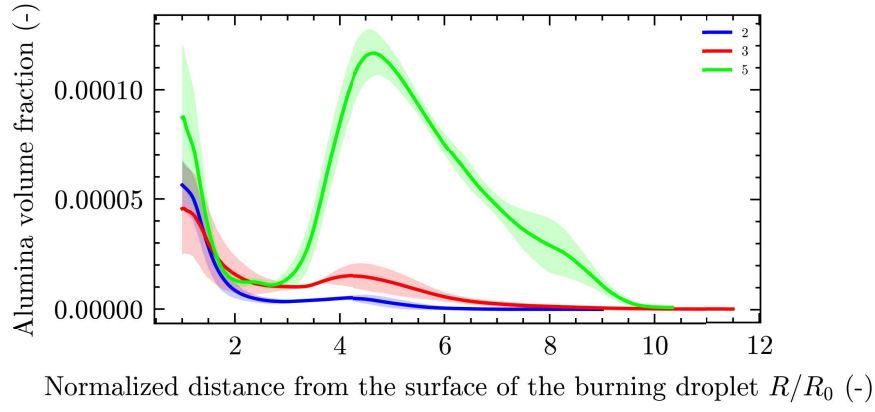


Figure D.8: Alumina volume fraction profiles surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .

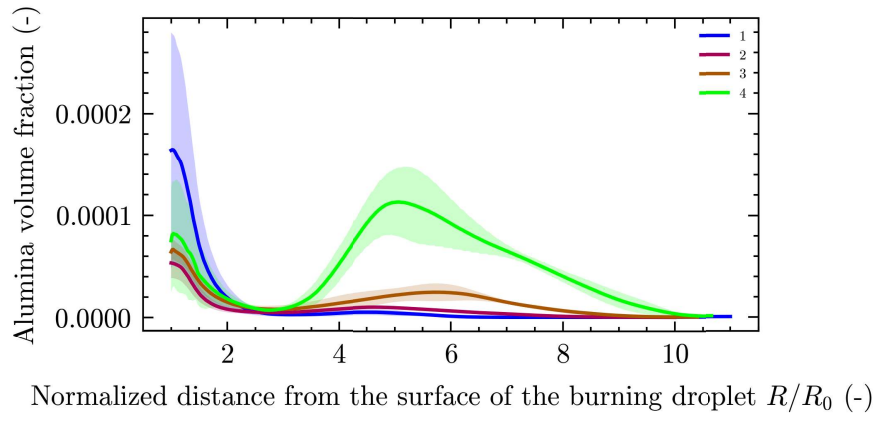


Figure D.9: Alumina volume fraction profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures .

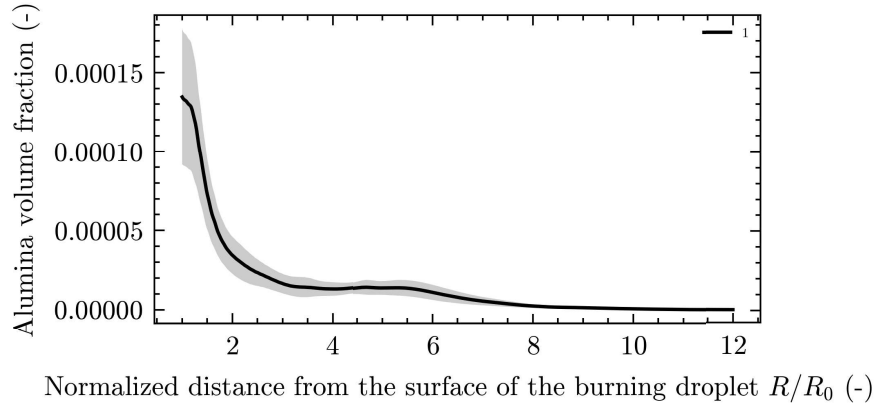


Figure D.10: Alumina volume fraction profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .

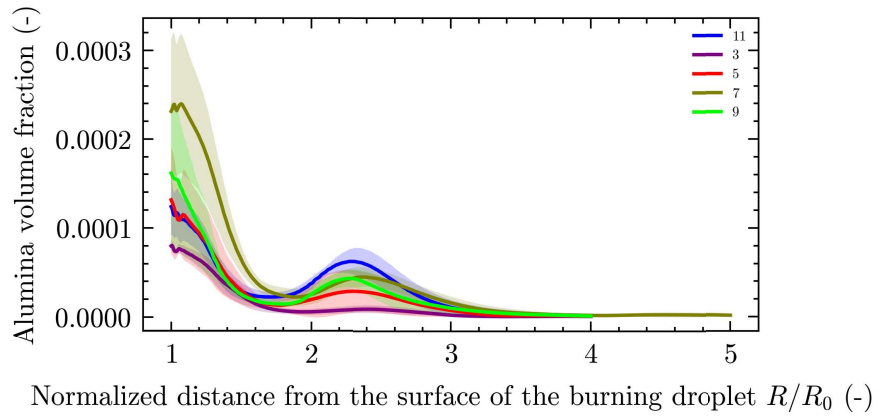


Figure D.11: Alumina volume fraction profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures .

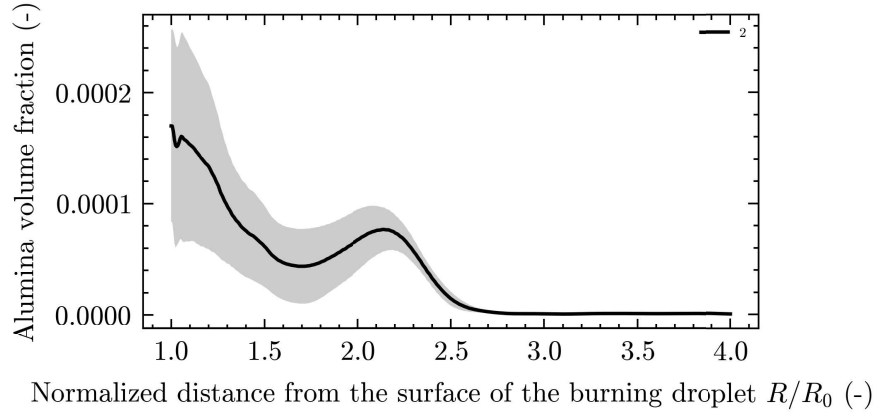


Figure D.12: Alumina volume fraction profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars .

D.3 Alumina particle size

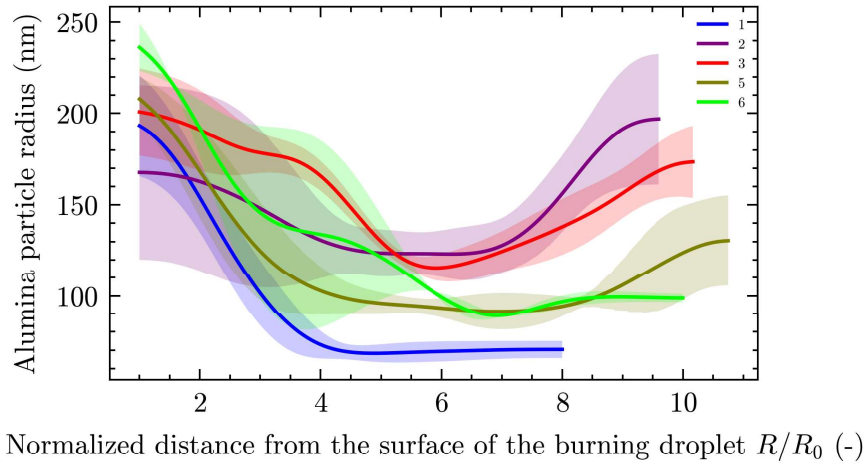


Figure D.13: Alumina particle size profiles surrounding an Al droplet burning in air at various pressures .

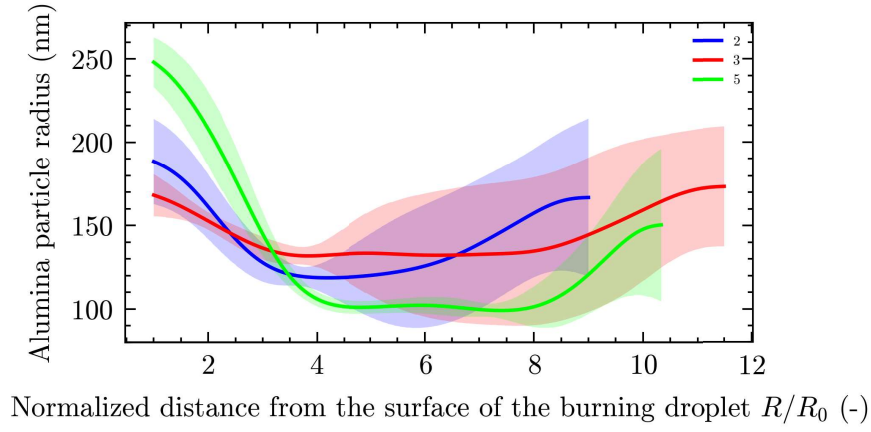


Figure D.14: Alumina particle size profiles surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .

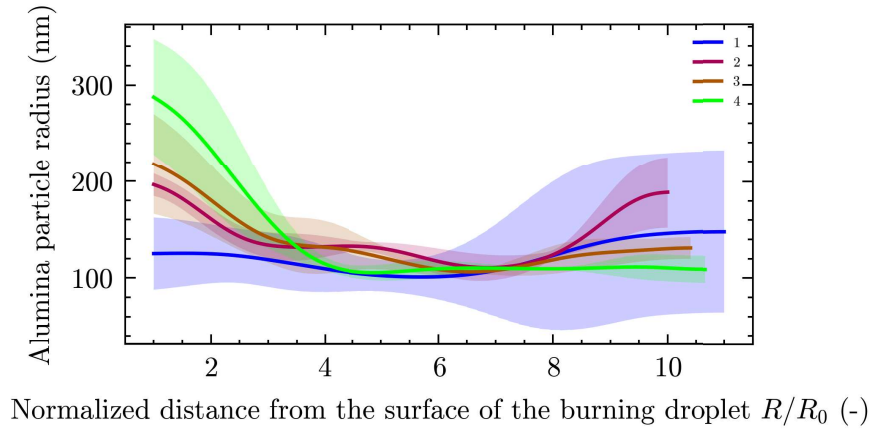


Figure D.15: Alumina particle size profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures .

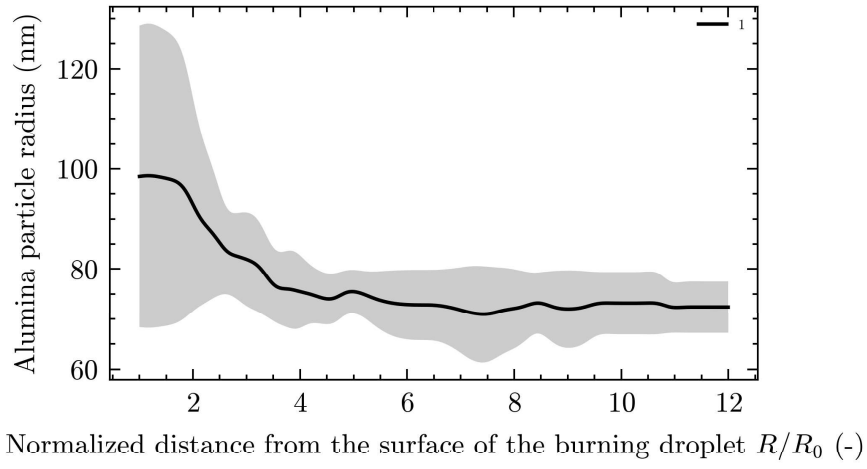


Figure D.16: Alumina particle size profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .

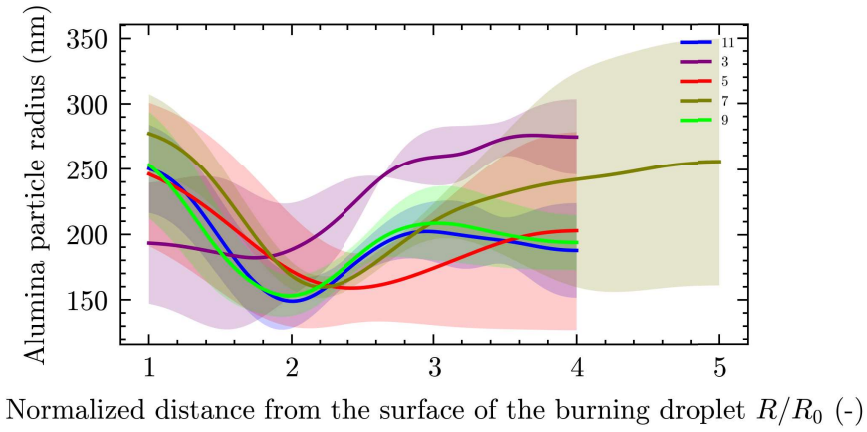


Figure D.17: Alumina particle size profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures .

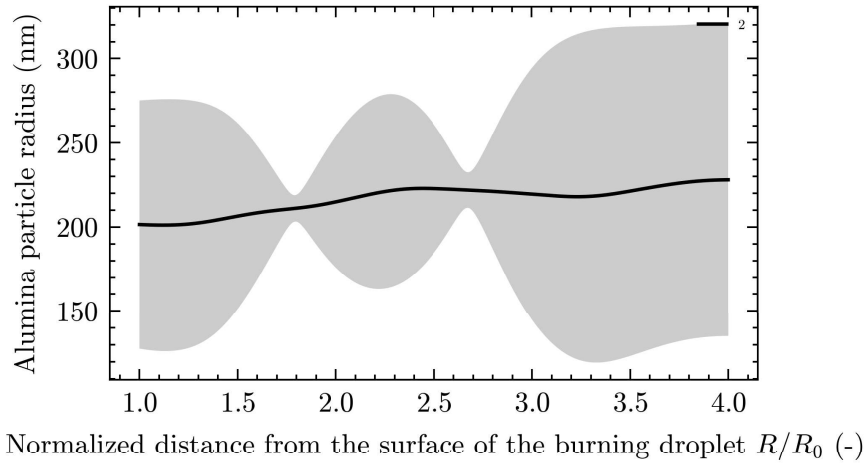


Figure D.18: Alumina particle size profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars .

D.4 Gaseous emissions profiles - R

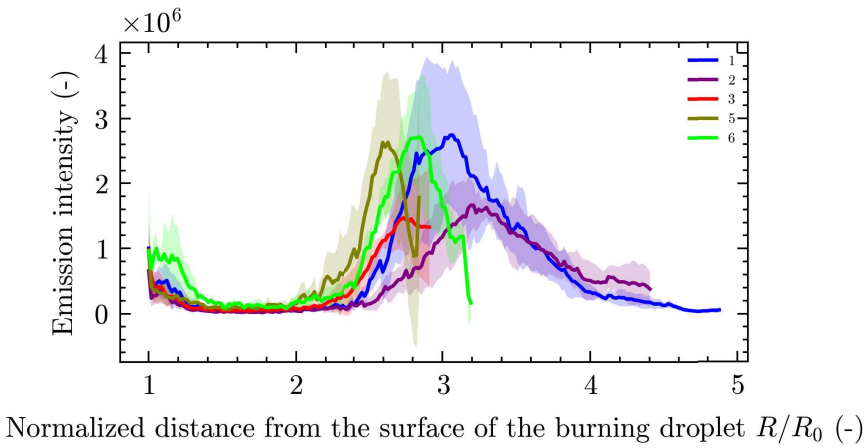
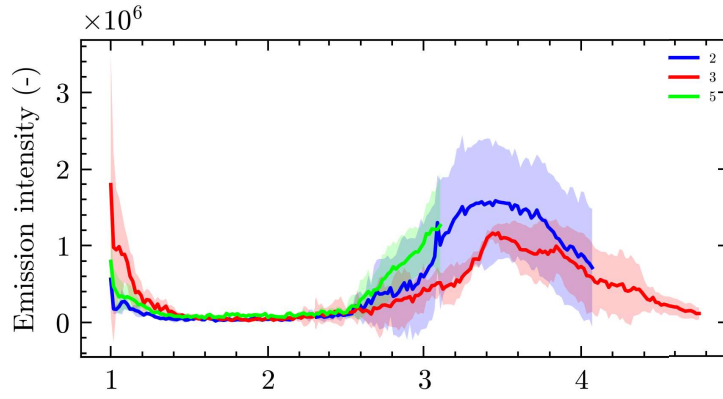
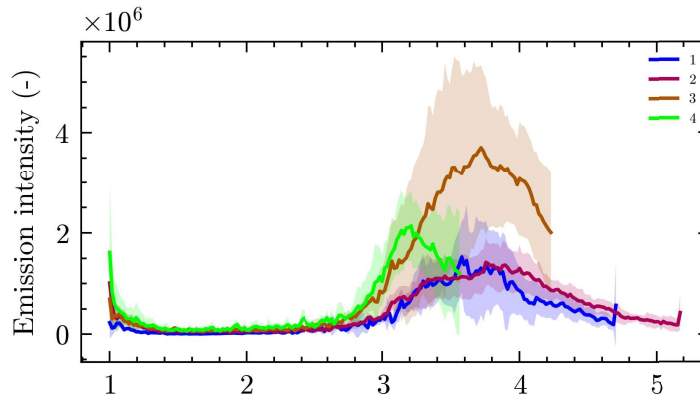


Figure D.19: Gaseous emissions profiles measured on the R matrix of the camera surrounding an Al droplet burning in air at various pressures .



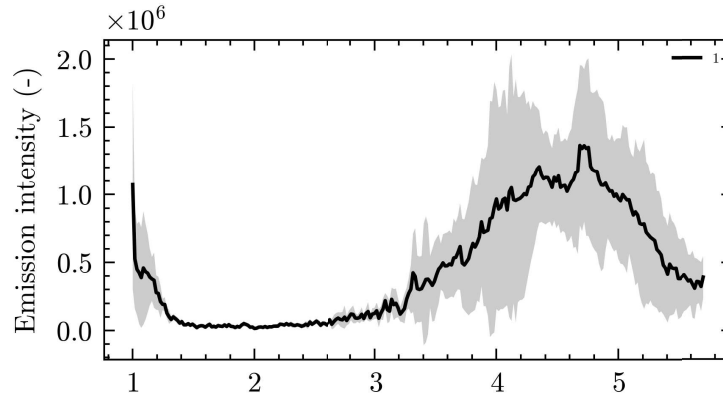
Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.20: Gaseous emissions profiles measured on the R matrix of the camera surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .



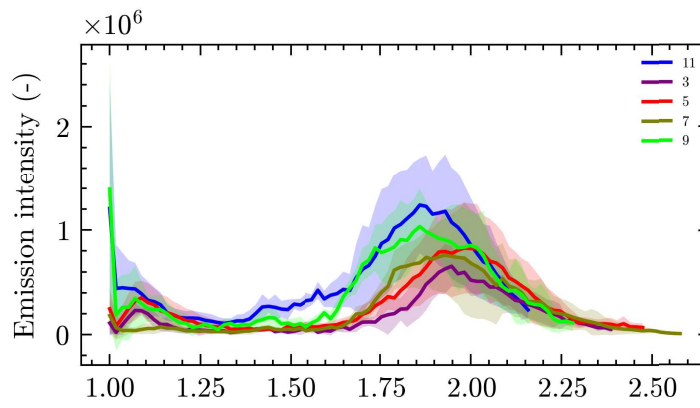
Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.21: Gaseous emissions profiles measured on the R matrix of the camera profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures .



Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.22: Gaseous emissions profiles measured on the R matrix of the camera profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .



Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.23: Gaseous emissions profiles measured on the R matrix of the camera profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures .

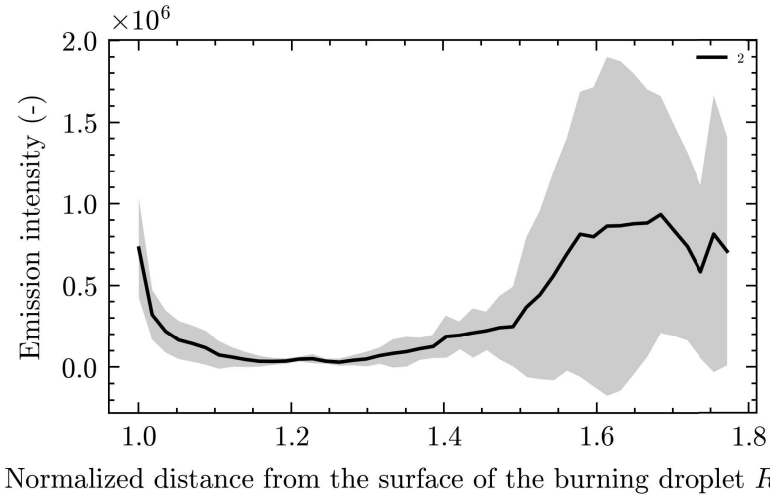


Figure D.24: Gaseous emissions profiles measured on the R matrix of the camera profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars .

D.5 Gaseous emissions profiles - G

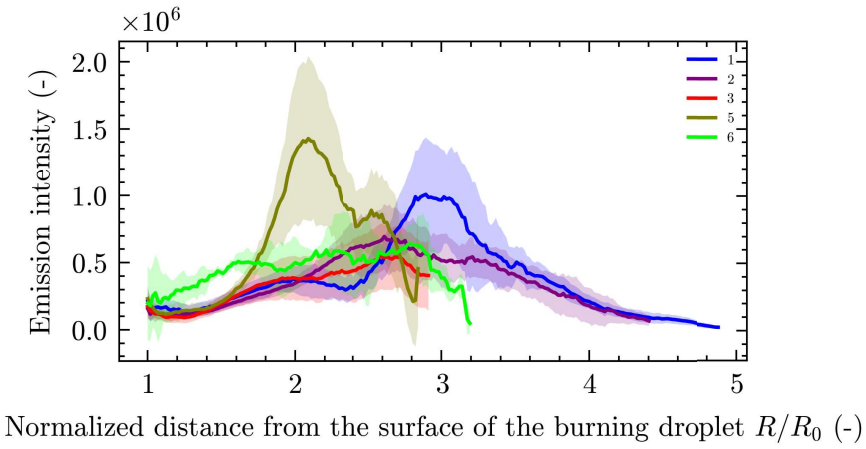


Figure D.25: Gaseous emissions profiles measured on the G matrix of the camera surrounding an Al droplet burning in air at various pressures .

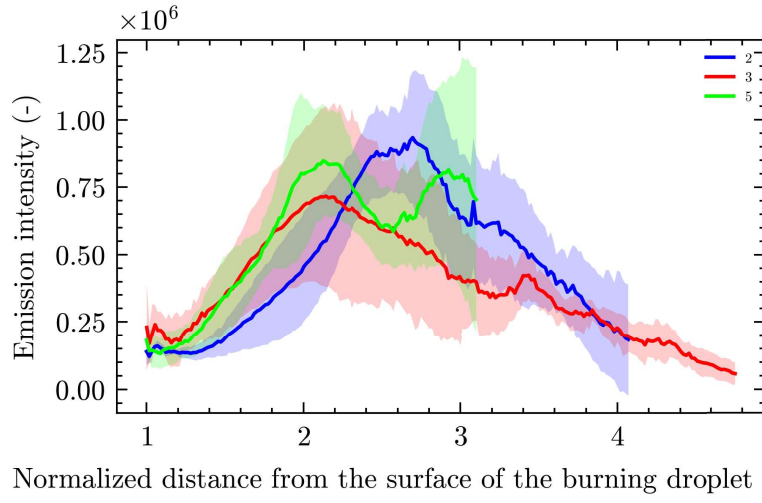


Figure D.26: Gaseous emissions profiles measured on the G matrix of the camera surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .

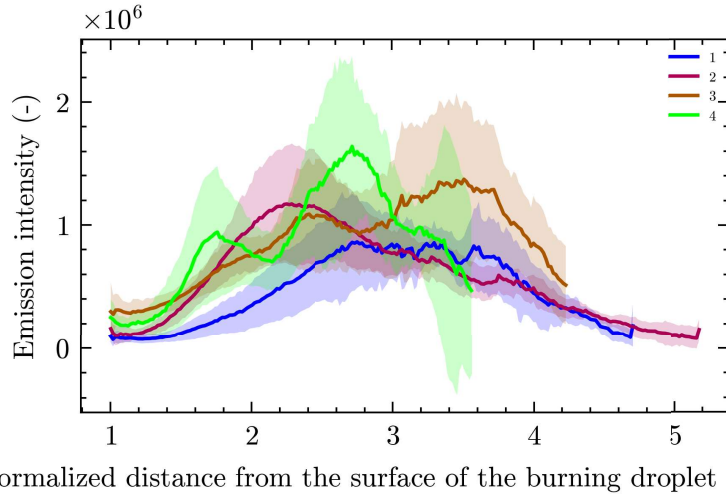
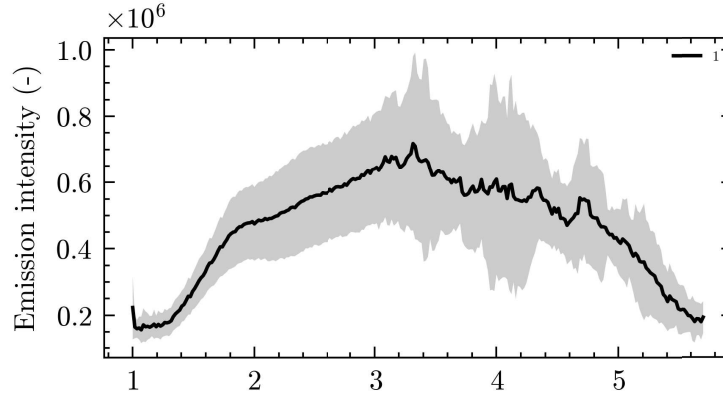
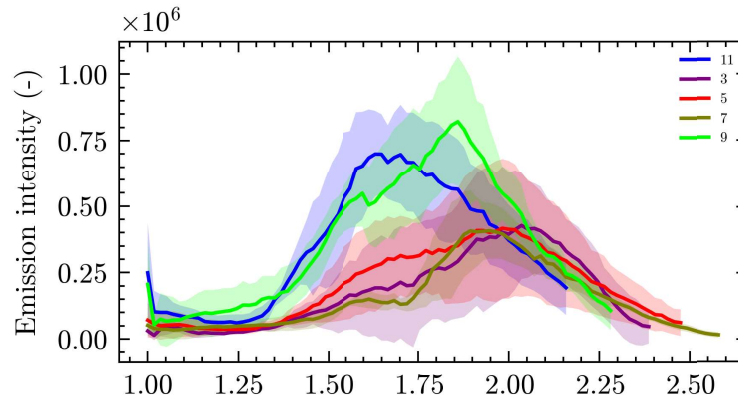


Figure D.27: Gaseous emissions profiles measured on the G matrix of the camera profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures .



Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.28: Gaseous emissions profiles measured on the G matrix of the camera profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .



Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.29: Gaseous emissions profiles measured on the G matrix of the camera profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures .

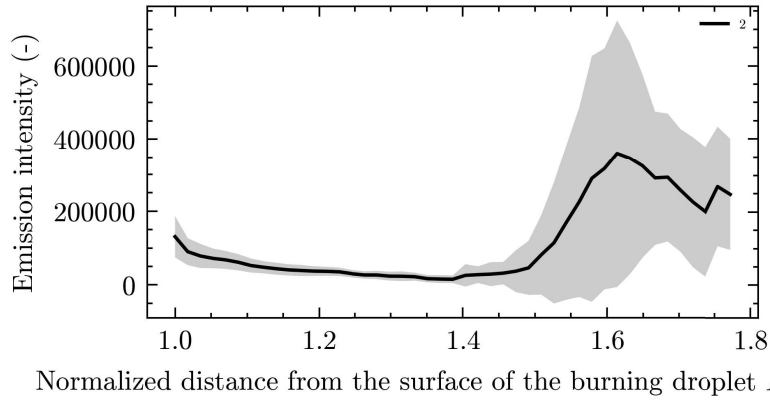


Figure D.30: Gaseous emissions profiles measured on the G matrix of the camera profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars .

D.6 Gaseous emissions profiles - B

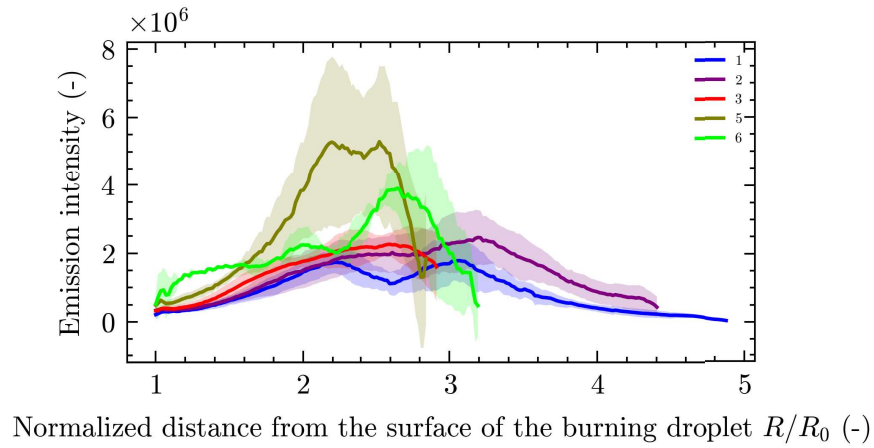


Figure D.31: Gaseous emissions profiles measured on the B matrix of the camera surrounding an Al droplet burning in air at various pressures .

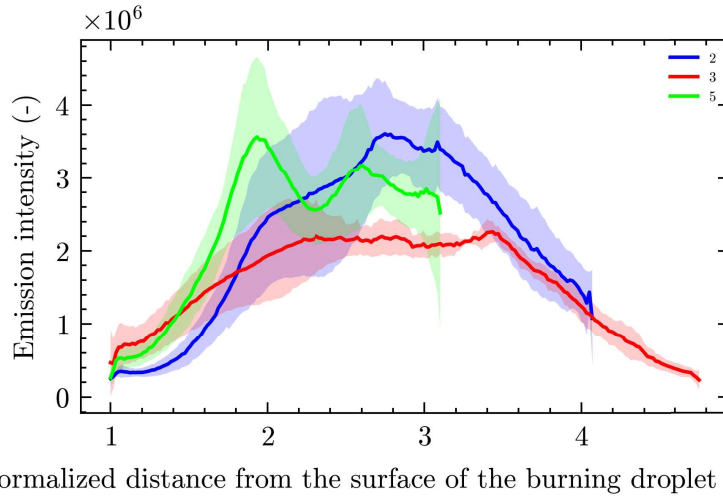


Figure D.32: Gaseous emissions profiles measured on the B matrix of the camera surrounding an Al droplet burning in an O_2/Ar (15/85) vol. mixture at various pressures .

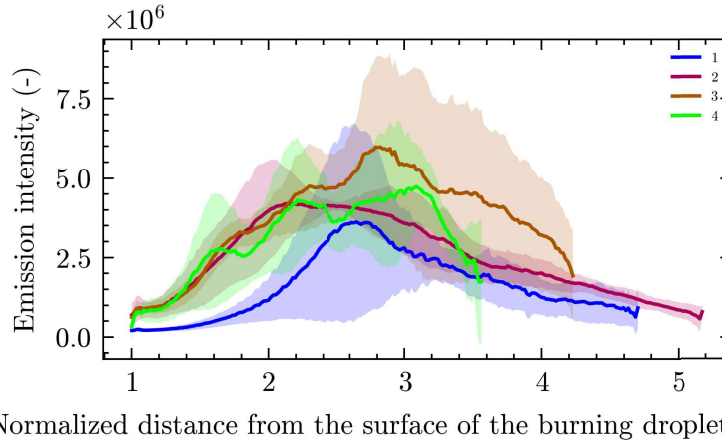
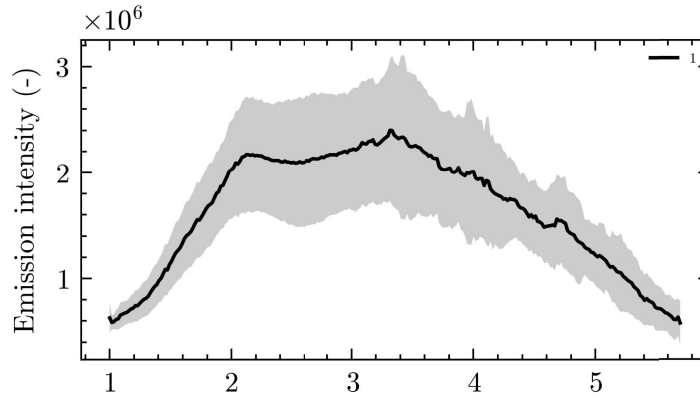
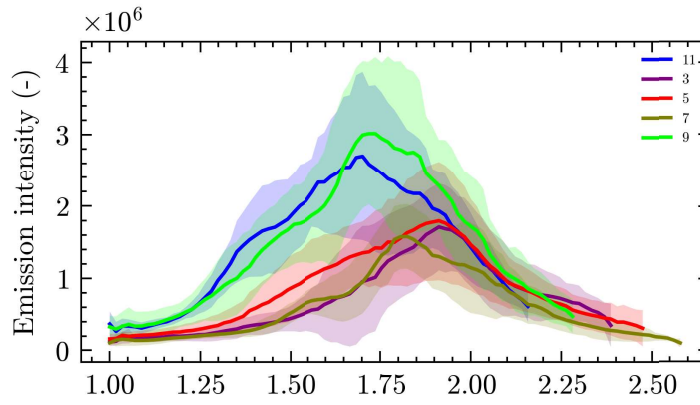


Figure D.33: Gaseous emissions profiles measured on the B matrix of the camera profiles surrounding an Al droplet burning in an O_2/Ar (21/79) vol. mixture at various pressures .



Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.34: Gaseous emissions profiles measured on the B matrix of the camera profile surrounding an Al droplet burning in an O_2/Ar (30/70) vol. mixture at 1 bar .



Normalized distance from the surface of the burning droplet R/R_0 (-)

Figure D.35: Gaseous emissions profiles measured on the B matrix of the camera profiles surrounding an Al droplet burning in an O_2/He (21/79) vol. mixture at various pressures .

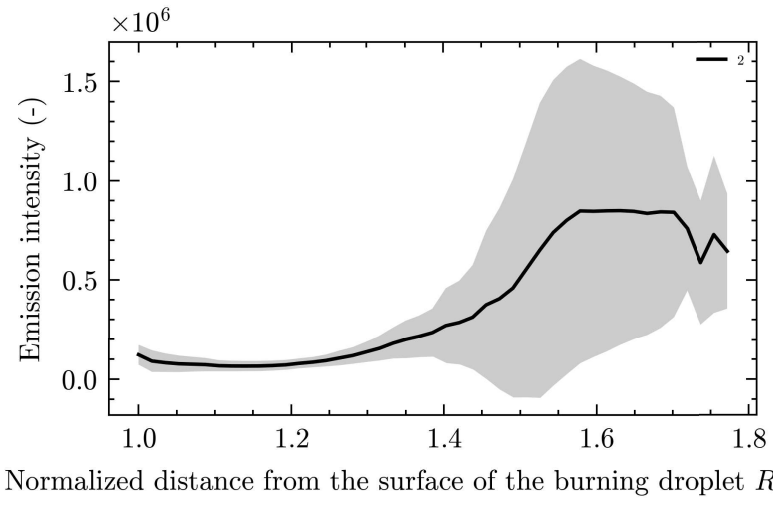


Figure D.36: Gaseous emissions profiles measured on the B matrix of the camera profile surrounding an Al droplet burning in an CO_2/N_2 (60/40) vol. mixture at 2 bars .

Résumé en français

Introduction

En 1971, le premier projet européen, « Europa », a été lancé afin de rendre l'espace plus accessible. Cependant, il n'a pas atteint ses objectifs, ce qui a conduit à la naissance du programme Ariane en 1973, initié par le CNES (Centre National d'Études Spatiales). Ce programme vise à permettre à la France de lancer ses propres satellites en orbite, indépendamment des États-Unis d'Amérique et de l'URSS, qui étaient à l'époque les pays leaders en matière d'accès à l'espace. Le premier lancement réussi d'Ariane 1, en 1979, a conduit au développement rapide de ses successeurs, Ariane 2 lancé en 1986, Ariane 3 lancé en 1984 et Ariane 4 lancé en 1988. Plus tard, en 1996, Ariane 5 est lancé, équipé de nouvelles technologies améliorant les performances et la charge utile. En 2015, Airbus et Safran ont formé conjointement ArianeGroup (anciennement Airbus Safran Launchers) dans le but de développer le lanceur Ariane 6, qui a récemment effectué son premier lancement réussi le 9 juillet 2024.

L'augmentation des performances et la réduction des coûts apportées par Ariane 5 et Ariane 6 sont liées à l'utilisation de moteurs à propergol solide, contrairement aux versions précédentes de ce lanceur qui utilisaient des propergols liquides. Cette technologie fournit une poussée importante au début du vol et permet de réduire les coûts de développement grâce à sa facilité d'utilisation, de production et de stockage. Avec le nombre croissant de satellites en orbite autour de la Terre, la réduction du coût des lancements et l'amélioration des performances des fusées sont au cœur de nombreuses études menées par ArianeGroup. Ce travail de thèse de doctorat est motivé par la nécessité d'améliorer les performances des moteurs d'ArianeGroup, en approfondissant notre compréhension générale de la combustion des propergols solides, et plus précisément de la combustion de l'aluminium. Les processus précis de combustion des particules d'aluminium, utilisées comme additifs énergétiques dans les propergols solides, ne sont pas encore entièrement compris et caractérisés. Un état des lieux des connaissances actuelles sur la combustion de l'aluminium et ses applications est présenté, soulignant l'intérêt des travaux de cette thèse de doctorat.

Ce travail vise à approfondir nos connaissances sur la combustion de particules d'aluminium individuelles, grâce à un dispositif expérimental unique associé à des diagnostics optiques récemment développés. L'application de ces diagnostics à la combustion d'une particule d'aluminium individuelle dans diverses conditions permet de mettre en évidence des phénomènes et des mécanismes cruciaux, et fournit ainsi une base de données inégalée pour les modèles numériques. De plus, la comparaison des données expérimentales obtenues avec le modèle numérique de combustion de l'aluminium d'ArianeGroup permet d'identifier les points à améliorer dans le modèle.

Les applications de la combustion d'Aluminium

Dans ce chapitre, les deux principaux domaines d'application de la combustion d'aluminium sont présentés. La description de l'aluminium en tant qu'additif énergétique dans les moteurs de fusées est faite avant d'introduire son application en tant que vecteur énergétique propre.

Dans les systèmes de propulsion, et principalement dans le domaine de l'exploration spatiale, les moteurs à propergol solide sont utilisés en raison de leur simplicité de conception et de leurs performances élevées, c'est-à-dire une poussée importante pendant de longues durées. La plupart des propergols solides contiennent des particules d'aluminium qui agissent comme un additif énergétique. Lors de la consommation du propergol, les particules d'aluminium s'enflamment et brûlent à des températures plus élevées que celles de la flamme HTPB-AP, ce qui améliore considérablement les performances du moteur.

La combustion de l'aluminium forme de l'oxyde d'aluminium qui se condense lors de sa génération, produisant de l'oxyde d'aluminium en phase condensée, l'alumine. La présence d'une phase condensée dans les moteurs de fusée entraîne des pertes de performance dues à un écoulement diphasique, à des instabilités de combustion et à une combustion incomplète, qui sont généralement compensées par une amélioration des performances. L'aluminium fait également l'objet d'études actives en tant que vecteur d'énergie propre, car l'alumine peut être capturée à la suite du processus de combustion hautement exothermique et facilement transportée vers un lieu où les énergies renouvelables permettent de la réduire en aluminium. L'aluminium réduit peut être transporté à nouveau vers des lieux où l'on a besoin d'énergie, où il peut être brûlé à nouveau pour produire l'énergie requise. Les difficultés liées à l'efficacité des cycles ou aux processus entièrement sans carbone sont au centre des études actuelles.

Les deux applications de la combustion de l'aluminium dans l'industrie ont montré l'importance de l'aluminium et le rôle crucial que joueront les développements futurs dans ce domaine. Pour ces deux applications, la forte exothermicité de la combustion de l'aluminium présente un intérêt majeur, tandis que la présence de produits de combustion condensés semble être soit bénéfique, soit néfaste. Après cette introduction aux applications de la combustion de l'aluminium dans le domaine industriel, il est essentiel de bien comprendre les mécanismes de la combustion de l'aluminium afin d'optimiser son utilisation.

La combustion d'aluminium

Ce chapitre fournit une description générale du processus de combustion d'une seule particule d'aluminium, de l'allumage à l'extinction. Les principales conclusions de ce chapitre peuvent être résumées selon la liste suivante:

- Les particules d'aluminium dont le diamètre est généralement supérieur à 10 microns, s'enflamment lors de la fusion de la couche d'oxyde, à 2326 K. Les particules plus petites atteignent des températures d'inflammation plus basses en raison des fissures dans la couche d'oxyde qui permettent la diffusion de l'oxydant vers l'aluminium.
- Une approche simpliste de la combustion de l'aluminium consiste à supposer que les petites particules (<10 microns) brûlent dans un régime limité par la cinétique chimique, les réactions se produisant à la surface de la gouttelette. Les particules plus grosses brûlent dans un régime limité par la diffusion, la flamme se formant à distance de la

surface de la gouttelette. L'apparition d'un régime de transition est observée et une caractérisation plus détaillée est nécessaire.

- Les sous-oxydes d'aluminium tels que AlO , Al_2O ou AlO_2 sont identifiés comme des intermédiaires de réaction cruciaux qui se forment avant l'alumine.
- Une fois formée, l'alumine se condense immédiatement pour former des produits de combustion condensés (CCP) dont la taille varie généralement entre 10 nm et 400 nm selon les conditions de combustion. Ces CCP peuvent soit diffuser hors du système, principalement en raison du flux de Stefan, soit diffuser vers la gouttelette, principalement en raison de la thermophorèse. Cette dernière peut entraîner l'accumulation de l'oxyde sur le lobe d'alumine localisé sur la goutte, conduisant à sa croissance.
- Il est prouvé que les réactions hétérogènes en surface jouent un rôle crucial lors de la combustion des particules d'aluminium, même pour les particules dont le diamètre peut atteindre 100 microns.
- Dans l'air atmosphérique, la température à la surface de la gouttelette devrait être d'environ 2500 K, tandis que la température de la flamme devrait être proche de 3600 K.
- Les particules d'aluminium ne suivent pas strictement la loi du d^2 . De nombreuses études ont évalué le diamètre des particules, la composition de l'oxydant, la pression et la température comme paramètres de premier ordre pour prédire le temps de combustion d'une particule d'aluminium.
- La croissance de la couche d'oxyde entraîne généralement la projection et la rotation de la gouttelette d'aluminium, ce qui conduit généralement à son extinction. En présence d'azote dans le gaz diluant, la formation de AlN sous forme de phase condensée à la surface de la gouttelette a été observée, entraînant un refroidissement rapide de la goutte.
- Une augmentation de la pression entraîne une diminution du stand off ratio (SOR), une augmentation de la concentration en alumine et une augmentation de la température de la flamme. La pression a une faible influence sur les temps de combustion.
- Les températures adiabatiques de la flamme et les temps de combustion devraient subir de faibles variations lorsque le diluant varie entre N_2 , Ar ou He , bien que la croissance de la couche d'oxyde soit favorisée par la présence de N_2 et que le SOR diminue lorsque He est utilisé.
- La combustion est fortement influencée par la nature de l'oxydant. Les rendements d'oxydation sont mesurés comme étant plus faibles pour la vapeur et le dioxyde de carbone. De plus, la présence d'azote ou de carbone est supposée entraîner la formation de AlN , $AlNO$ ou AlC en tant que CCP.
- La présence d'un champ électrique lors de la combustion d'une particule d'aluminium devrait influencer légèrement le processus de formation des CCP.

Bien que la compréhension globale de la combustion des particules d'aluminium se soit considérablement améliorée ces dernières années, ce qui ressort de cette introduction, espérons-le, c'est que très peu de données expérimentales spatialement résolues sont disponibles dans la littérature. Il existe peu, voire aucun, profils de température ou de concentration d'espèces entourant la gouttelette d'aluminium en combustion, sans parler des différentes conditions de combustion. Le manque de données spatialement résolues empêche la validation des nombreuses simulations disponibles dans la littérature. Ce travail de doctorat vise donc à fournir ces données. Pour répondre à ce besoin, un dispositif expérimental précédemment utilisé par Braconnier et Glasziou a été amélioré et est décrit en détail dans le chapitre suivant.

Dispositif expérimental

Dans ce chapitre, le dispositif expérimental utilisé tout au long de ce travail est présenté. Il contient une discussion sur les dispositifs expérimentaux que l'on trouve dans la littérature, ainsi qu'une introduction générale au dispositif utilisé dans ce travail. Une description détaillée du lévitateur électrodynamique est fournie. Ce dispositif qui permet d'isoler une unique particule d'aluminium micronique, sans contact avec une surface et dans un environnement contrôlé, est composé d'un ensemble de trois électrodes. Ces électrodes ainsi que leur alimentation sont décrites dans ce chapitre. Le dispositif d'allumage de la particule, composé d'un laser CO_2 est aussi détaillé. L'ensemble du dispositif est équipé de diagnostics optiques, tels que des tubes photomultiplicateurs (PMs) et une caméra rapide couleur couplée à un téléobjectif. Les PMs permettent des mesures d'intensité lumineuse avec une excellente résolution temporelle, tandis que la caméra permet d'observer la goutte d'aluminium en combustion avec une bonne résolution spatio-temporelle. L'équipement détaillé dans ce chapitre permet l'application d'une méthode nouvellement développée pour caractériser la combustion de l'aluminium, décrite dans le chapitre suivant.

Développement d'une nouvelle méthode d'analyse

Dans ce chapitre, le développement d'une nouvelle méthode d'analyse est entièrement décrit. Dans un premier temps, les méthodes d'analyse disponibles dans la littérature sont rappelées, en présentant notamment la méthode de fluorescence induite (PLIF), utilisée par Bucher sur la combustion d'aluminium, qui peut fournir un profil de température résolu spatialement, mais seulement basée sur les émissions de l'AlO. Cette méthode ne permet pas une caractérisation complète du système, et ne permet pas de caractériser la température de l'alumine.

La méthode proposée évite l'utilisation de l'hypothèse du corps gris pour caractériser l'alumine grâce à la connaissance des propriétés optiques de la fumée d'oxyde, qui peuvent être obtenues à l'aide d'une méthode multispectrale d'extinction de la lumière (M-LEM). Grâce à ces informations, une méthode de pyrométrie à résolution spatiale peut être améliorée pour tenir compte de l'émissivité et de l'absorption, ce qui donne lieu à une méthode appelée «Modulation d'émission par absorption» (MAE).

La méthode multispectrale d'extinction de la lumière repose sur le fait d'évaluer les propriétés optiques d'un milieu en évaluant son efficacité à atténuer la lumière. L'application de cette méthode à la combustion d'aluminium est entièrement décrite dans ce chapitre. Une

LED pulsée est couplée à une caméra couleur afin d'évaluer la profondeur optique intégrée du nuage d'oxyde, à trois longueurs d'onde distinctes. Le profil de profondeur optique locale est évalué à l'aide d'une procédure de déconvolution par pelage d'oignon (onion-peeling). L'approximation de Rayleigh, souvent utilisée pour les très petites particules, est écartée en raison de son invalidité, et la section efficace d'extinction d'une particule d'alumine est calculée à l'aide de la théorie de Mie. Ensuite, une procédure d'optimisation est appliquée aux profils de profondeur optique locale aux trois longueurs d'onde d'intérêt afin d'évaluer le rayon des particules d'alumine. Ce processus permet de valider que les grandeurs indépendantes de la longueur d'onde soient égales lorsque calculées sur les trois matrices de la caméra R,G et B. À partir de là, le profil de fraction volumique d'alumine est évalué. Un profil spatialement résolu de taille de particules d'alumine ainsi qu'un profil de concentration d'alumine sont alors obtenus.

Par la suite, l'émission de l'alumine est modulée par ses propriétés optiques via la méthode de modulation d'émission par absorption (MAE). Les images des intensités de combustion, sans rétroéclairage, sont mesurées par la caméra couleur. L'intensité de chaque pixel est obtenue à partir d'une somme de contributions locales. Les intensités locales sont constituées du produit de la radiance du corps noir, fonction de la température, et d'un coefficient d'émission mesuré via la M-LEM. Une méthode de déconvolution tenant compte de l'auto-extinction est utilisée pour récupérer les intensités locales à n'importe quel endroit du nuage. Grâce à la connaissance du coefficient d'émission, le produit d'une constante optique par la radiance du corps noir est connu. L'utilisation d'un étalonnage de la caméra via un corps noir permet d'évaluer le profil de température à l'aide du rapport des émissions corps noir locales. Cependant, la présence de sous-oxydes d'aluminium tels que l' AlO , émettant dans la partie bleue-verte du spectre visible, perturbe la mesure de température. Une procédure d'optimisation est mise au point pour séparer les émissions gazeuses des émissions du corps noir. À la suite de cette procédure, le profil de température spatialement résolu de l'alumine est évalué et les profils normalisés des émissions en phase gazeuse sont obtenus pour les trois longueurs d'onde du filtre triple bande.

Résultats expérimentaux

Dans ce chapitre, les résultats expérimentaux obtenus par l'application de la méthode d'analyse développée au cours de ces travaux sont présentés. Dans un premier temps, les résultats obtenus pour une combustion d'aluminium dans l'air atmosphérique sont présentés, permettant de mettre en avant la structure du système ainsi que certains résultats de référence. La zone de condensation de l'alumine est clairement identifiée, les régions d'accumulation de l'alumine sont mises en avant, et les régions d'émissions gazeuses et électroniques sont aussi mises en évidence. Par la suite, des paramètres de combustion clés (KCPs) sont introduits afin de permettre une étude rapide et simplifiée de l'influence des conditions de combustion sur les paramètres physiques de la combustion. Des KCPs tels que la température de flamme maximale ou la taille moyenne des particules d'alumine sont choisis. Par la suite, chaque KCP est évalué pour chaque condition expérimentale. 19 conditions expérimentales différentes ont été étudiées, en faisant varier la pression du milieu, la concentration en oxygène de l'oxydant, la nature du diluant, ou la nature de l'oxydant. L'évolution de chaque KCP en fonction des conditions est caractérisée et, si possible, des lois de corrélations sont proposées. Les

principales conclusions obtenues sont listées par la suite.

Température de flamme

- Lorsque la pression augmente, la température maximale augmente, jusqu'à une certaine pression. Si la pression continue d'augmenter, la température diminue.
- La température maximale est similaire lorsque l'argon et l'azote sont utilisées en tant que diluant, mais elle est plus faible dans le cas de l'hélium.
- La température maximale est plus faible lorsque le CO_2 est utilisé comme diluant, et la température maximale croît si la concentration en O_2 du milieu croît.
- Les pertes thermiques par conduction de la zone de condensation de l'alumine semblent contrôler l'évolution de la température maximale.

Température de goutte

- La température de la goutte augmente avec la pression du milieu.
- La température de la goutte est identique dans le cas de l'argon et dans le cas de l'azote, mais est plus faible dans le cas de l'hélium.
- La température de la goutte est reliée à une loi de Clausius-Clapeyron en considérant une pression partielle d'aluminium de 0.29 à la surface de la goutte.

Stand-off ratio (SOR)

- Le stand-off ratio est le plus faible dans le cas de l'hélium, le plus élevé dans le cas de l'argon, et entre ces deux valeurs dans le cas de l'azote.
- Le SOR est le plus faible dans le cas du CO_2 et augmente avec la concentration en O_2 du milieu.
- Une loi de corrélation, issue de la combustion de gouttes d'hydrocarbures et modifiée pour la combustion métallique, est extraite de la littérature.

Concentration de l'alumine

- Le pic de concentration d'alumine augmente avec la pression du milieu.
- Le pic de concentration d'alumine est le plus faible dans le cas de l'hélium, augmente dans le cas de l'azote, et augmente plus fortement dans le cas de l'argon.
- Le pic de concentration d'alumine augmente avec la concentration en O_2 et est le plus faible dans le cas du CO_2 . Une corrélation empirique est proposée.
- La localisation de ce pic d'alumine suit les mêmes observations que le SOR, et une corrélation fonction de ce SOR est proposée.

Taille des particules d'alumine

- La taille des particules d'alumine augmente avec la pression jusqu'à une certaine pression, au delà de laquelle, la taille des particules décroît si la pression augmente.
- Les particules d'alumine sont les plus grandes dans le cas de l'hélium, avec des tailles comparables pour l'azote et l'argon.
- La taille des particules d'alumine décroît lorsque la concentration en O_2 croît, avec une taille maximale dans le cas du CO_2 .
- Ces tendances peuvent être prédites avec la théorie de la nucléation en ayant la connaissance du temps de résidence des particules dans le nuage d'oxyde.

Émissions des phases non-condensées

- La localisation des émissions gazeuses suit les mêmes observations que le SOR, et une corrélation fonction de ce SOR est proposée.
- La localisation des émissions électroniques suit les mêmes observations que le SOR, et une corrélation fonction de ce SOR est proposée, avec une température électronique constante de 21 000K.

L'obtention de ces résultats expérimentaux constitue une base de données cible importante pour la simulation numérique. Par la suite, différents modèles numériques sont introduits, et un modèle existant est amélioré pour modéliser au mieux les résultats obtenus expérimentalement.

Simulation numérique

Dans ce chapitre, un bref état de l'art des modèles existants de combustion de particules d'aluminium individuelles est présenté. Bien que chaque modèle présenté diffère légèrement des autres, aucun d'entre eux ne permet de prédire avec précision les résultats expérimentaux de ce travail. Nous avons donc utilisé le modèle numérique d'ArianeGroup pour simuler les résultats expérimentaux dans des conditions de combustion identiques. Dans un premier temps, une brève étude paramétrique a été réalisée afin d'identifier les paramètres de premier ordre du modèle. En modifiant la réversibilité de la réaction de condensation ou la taille des particules d'alumine (considérée comme constante partout dans la fumée d'oxyde), aucun changement significatif n'a été observé dans les résultats de la simulation. En activant les mouvements phorétiques, en modifiant la diffusivité de la fumée d'oxyde ou en modifiant le facteur pré-exponentiel de la réaction de condensation, des changements plus évidents ont été observés dans les résultats de la simulation, mais aucun des cas n'a montré une bonne corrélation avec les résultats expérimentaux. Afin d'améliorer la précision du modèle, il a été décidé de remplacer la loi d'Arrhenius déterminée empiriquement pour la réaction de condensation par une réaction physique basée sur la théorie classique de la nucléation. En utilisant le taux de nucléation de l'alumine, la production d'alumine condensée est trop lente et les résultats ne sont pas acceptables. Le taux de croissance des noyaux d'alumine a ensuite été ajouté comme mécanisme de condensation, ce qui a permis d'obtenir des taux

de condensation plus élevés. Bien qu'aucun changement significatif ne soit observé dans les profils de température, les profils de concentration d'alumine sont très différents des calculs initiaux et encouragent l'utilisation du CNT pour caractériser la réaction de condensation de l'alumine. De fortes divergences subsistent et un modèle plus précis est nécessaire pour obtenir une bonne corrélation avec les résultats expérimentaux. Dans un premier temps, le suivi de la taille des noyaux dans le nuage d'oxyde pourrait améliorer considérablement la précision des résultats. Un tel modèle permettrait également d'obtenir les profils de taille des particules d'alumine. Des processus de coagulation pourraient également être mis en œuvre pour améliorer la précision du modèle.

Conclusion et perspectives

Avec 112 lancements réussis, les lanceurs Ariane 5 et Ariane 6 développés par ArianeGroup ont permis à l'Europe d'accéder largement aux technologies spatiales. Les deux lanceurs sont équipés de moteurs à propergol solide, alimentés par des combustibles solides contenant des particules d'aluminium comme additifs énergétiques. En présence de particules d'aluminium dans la chambre de combustion, les performances du moteur sont considérablement améliorées grâce à la forte énergie libérée par la combustion de l'aluminium. L'inconvénient est que les produits de combustion (alumine) ainsi que les gouttelettes d'aluminium se trouvent en phase condensée, ce qui peut entraîner des pertes d'écoulement diphasique, des instabilités thermoacoustiques, des modifications de la géométrie de la tuyère ou une accumulation de poids dans les zones de recirculation du moteur. Pour éviter toute perte de performance, la combustion de l'aluminium et les produits de combustion qui en résultent doivent être bien caractérisés.

Il convient de noter que, bien que dans le domaine de la propulsion, les produits de combustion en phase condensée constituent un inconvénient, cette caractéristique peut présenter des avantages dans un autre domaine. La nature condensée de l'alumine facilite considérablement sa capture par rapport à celle des produits de combustion gazeux. En utilisant une source d'énergie externe pouvant être fournie par des énergies renouvelables, la totalité des produits de combustion peut théoriquement être recyclée en aluminium. Pour ces raisons, combinées à une densité énergétique élevée et à une grande disponibilité, l'aluminium suscite également beaucoup d'intérêt en tant que vecteur d'énergie propre. La caractérisation complète de la combustion des gouttelettes d'aluminium est nécessaire pour évaluer l'efficacité totale du cycle d'énergie propre qui peut résulter de l'utilisation de l'aluminium comme vecteur d'énergie.

Bien que de nombreuses simulations de la combustion de l'aluminium soient disponibles dans la littérature, les données expérimentales restent rares. Divers groupes ont mis au point des expériences sur la flamme de poussière d'aluminium afin de fournir une caractérisation macroscopique de la combustion de l'aluminium, tandis que des expériences sur la combustion de particules individuelles fournissent des informations fondamentales sur la physique de la combustion. Ces dernières expériences sont difficiles à réaliser en raison de la taille micrométrique des particules, de la courte durée de la combustion et de la difficulté à isoler une seule particule dans des conditions spécifiques. Dans ce travail, un lévitateur électrodynamique est utilisé, ce qui nous permet d'isoler une seule particule micrométrique d'aluminium dans un environnement contrôlé. Un laser amène la particule à un emballement

thermique, et l'utilisation d'une caméra haute vitesse permet d'observer la combustion auto-entretenue d'une gouttelette d'aluminium.

Afin de fournir une caractérisation précise et complète de la gouttelette d'aluminium en combustion et de ses produits de combustion condensés, de nouveaux diagnostics optiques ont été développés. La méthode déjà bien connue d'extinction de la lumière a été étendue à une application multispectrale. La caméra couleur haute vitesse, équipée d'un filtre à triple bande permettant une observation quasi trichromatique, est couplée à une LED blanche qui sert de rétroéclairage. Le nuage d'oxyde rétroéclairé qui entoure la gouttelette d'aluminium en combustion est capturé par la caméra et comparé au nuage d'oxyde non rétroéclairé afin d'évaluer sa profondeur optique. La nature multispectrale des diagnostics permet d'évaluer la profondeur optique à trois longueurs d'onde distinctes. La comparaison des valeurs des profondeurs optiques spectrales permet d'évaluer la taille des gouttelettes d'alumine nanométriques, à n'importe quel endroit de la fumée d'oxyde. Ensuite, la fraction volumique de ces produits de combustion condensés est évaluée à n'importe quel endroit du nuage. La taille et la concentration des produits de combustion condensés fournissant des informations cruciales sur leurs propriétés optiques, les émissions lumineuses de la fumée d'oxyde peuvent ainsi être modulées par ses propriétés d'absorption et d'émission. La fumée d'oxyde étant composée d'alumine condensée ainsi que d'espèces gazeuses, une méthode numérique récemment mise au point permet de séparer les émissions lumineuses de la phase condensée de celles de la phase gazeuse. Les émissions de la phase condensée suivant la loi de Planck, le champ de température entourant la gouttelette d'aluminium en combustion est évalué. De plus, les profils d'émission gazeuse sont mesurés et des conclusions sur la nature de ces émissions peuvent être tirées. À proximité de la gouttelette d'aluminium, des émissions bleues attribuées à la présence de AlO sont observées, tandis que plus loin de la gouttelette, des émissions électroniques hypothétiques sont mesurées.

Grâce à ces résultats expérimentaux, la structure de la gouttelette d'aluminium brûlant dans l'air atmosphérique et le nuage d'oxyde qui l'entoure ont été clairement identifiés. Afin d'évaluer et de quantifier l'impact de l'environnement sur la combustion, des expériences supplémentaires ont été réalisées avec différentes concentrations d'oxygène, différents diluants, différentes pressions et en utilisant du CO_2 comme oxydant. Après avoir sélectionné les paramètres clés de la combustion, tels que la température maximale ou le rapport de distance de la flamme, l'influence de l'environnement sur le système a été caractérisée. Des corrélations empiriques et physiques ont été proposées pour les paramètres clés de combustion identifiés, mettant en évidence les mécanismes en jeu lors de la combustion d'une particule d'aluminium. Ces résultats expérimentaux fourniront également une base de données cruciale pour la simulation.

Les résultats expérimentaux obtenus sont ensuite comparés aux résultats de simulation disponibles dans la littérature, ainsi qu'aux résultats obtenus à l'aide du modèle développé en interne chez ArianeGroup. Une brève étude paramétrique met en évidence les paramètres qui peuvent influencer de manière significative les résultats de la simulation et montre l'incapacité du modèle actuel à prédire les résultats expérimentaux. La réaction de condensation de l'alumine étant considérée comme suivant la loi d'Arrhenius, une amélioration du modèle a été proposée en utilisant une vitesse de réaction de condensation basée sur la théorie classique de la nucléation. Une vitesse de réaction basée sur la somme de la vitesse de nucléation de l'alumine et de la vitesse de croissance des noyaux d'alumine a été développée. Les profils de

température résultants semblent n'être que légèrement affectés par cette mise en œuvre, tandis que les profils de concentration d'alumine sont considérablement affectés. Bien qu'aucune des configurations n'ait offert une corrélation acceptable avec les résultats expérimentaux, les résultats sont encourageants et la mise en œuvre d'un modèle plus précis basé sur la théorie classique de la nucléation devrait permettre une simulation plus précise du processus de combustion.

Bien que l'application des diagnostics optiques développés nécessite une combustion stable et symétrique, l'élargissement de l'ensemble des conditions expérimentales de combustion, par exemple en étudiant davantage de concentrations d'oxygène, de pressions et de diluants, améliorerait considérablement la qualité des corrélations et pourrait étayer les observations et conclusions proposées. Outre les paramètres évalués grâce aux diagnostics optiques, divers paramètres restent inconnus, tels que le profil de concentration de vapeur d'aluminium. Le groupe du professeur Aldén et du professeur Li à Lund a récemment mis au point un diagnostic laser pour évaluer ce profil de concentration, et l'ajout de celui-ci à la configuration actuelle améliorerait encore le degré de caractérisation de la gouttelette d'aluminium en combustion. La capture de la gouttelette d'aluminium par 'quenching' permettrait également de mieux comprendre la nature et la taille des produits de combustion condensés. En outre, la caractérisation proposée dans ce travail pourrait être étendue à différents métaux brûlant en phase vapeur. La comparaison des mécanismes de combustion entre différents métaux pourrait permettre d'identifier de futurs combustibles ou vecteurs d'énergie propre.

Ce travail met également en lumière les divergences entre les modèles actuels de combustion des gouttelettes d'aluminium et les résultats expérimentaux. La combustion en phase gazeuse est très répandue et bien connue dans le domaine des simulations numériques. Cependant, la combustion solide est plus récente, et la prise en compte des produits de combustion condensés apparaît comme un véritable défi. En général, des hypothèses fortes doivent être formulées concernant la condensation de l'alumine, l'énergie libérée par la condensation de l'alumine ou les mécanismes de transport de cette fumée d'oxyde. Afin de prédire avec précision le résultat expérimental, le développement d'un modèle numérique complexe est nécessaire. La mise en œuvre de la théorie classique de la nucléation pour tenir compte correctement de la condensation de l'alumine semble obligatoire. Cela représente un défi, car cela nécessite un suivi complexe des noyaux d'alumine, en tenant compte des espèces intermédiaires et des taux de condensation qui sont fonction de nombreux paramètres, contrairement à la loi d'Arrhenius précédemment considérée. On estime toutefois que cette perspective à long terme permettrait une simulation plus précise du système, ouvrant la voie à la simulation de différents combustibles ou de systèmes plus complexes.