Flame Coupling, De-Coupling, and Quenching in Multi-Front Flames

Jan Palecka

Master of Engineering
Mechanical Engineering

McGill University
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ABSTRACT

The propagation of hybrid flames formed by the combustion of aluminum suspensions in methane-air mixtures was studied through the flame quenching behavior in narrow channels in a flame tube apparatus. An experimental map of quenching distances was obtained for different metal powder concentrations. It reveals that at low concentration, the flame exists and quenches as a simple methane-air front. Above a critical concentration, a second aluminum front forms and couples with the first one into a two-front structure. The quenching of such flame can happen in two modes: at intermediate concentrations, the flame de-couples and the fronts quench individually, while at higher powder loadings, both fronts merge and quench simultaneously. A simplified physical model, with powder concentration and heat losses to channel walls as main parameters, has been developed to explain the experimental results. Each reaction, involving methane and aluminum respectively, starts at a pre-defined ignition temperature and is modelled as a single step constant heat release for a given combustion time. The experimentally observed propagation and quenching behaviors have been qualitatively reproduced by the model and the simplicity of the formulation allows for a clear explanation of this front interaction in terms of heat transfer.
ABRÉGÉ

La propagation des flammes hybrides formées lors de la combustion d’aluminium en suspension dans des mélanges méthane-air a été étudiée à travers l’étanchement de flamme dans un appareil expérimental à tube. Les distances d’étanchement ont été obtenues pour un éventail de concentrations de la poudre métallique. Ces résultats démontrent que pour une faible concentration, la flamme existe et s’éteint sous forme d’un seul front. Au-delà d’une concentration critique, un second front d’aluminium apparaît et se couple au premier dans une structure à double-fronts. L’étanchement d’une telle flamme peut s’effectuer sous deux formes: en cas de concentrations intermédiaires, la flamme se découpe et les fronts s’éteignent séparément, alors qu’à des teneurs en aluminium plus importantes, le chevauchement des fronts cause leur étanchement simultané. Un modèle physique simplifié, avec la concentration de poudre ainsi que les pertes de chaleur comme paramètres majeurs, a été développé pour expliquer les résultats expérimentaux. Chaque réaction, celle du méthane d’un côté et celle de l’aluminium de l’autre, débute à une température d’ignition prédéfinie et est représentée par une seule étape de production de chaleur à valeur constante pendant un temps de combustion donné. Le modèle reproduit de manière qualitative la propagation et l’étanchement, observés lors des expériences, et la simplicité de sa formulation mène à une explication claire de l’interaction des deux fronts en termes de transfert thermique.
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LIST OF SYMBOLS

\( D_i \) : Mass diffusion coefficient

\( E_a \) : Activation energy

\( F \) : Combustion time (non-dimensional)

\( Q_i \) : Heat of reaction \( i, i = 1,2 \)

\( R \) : Universal gas constant

\( R_i \) : Reaction \( i, i = 1,2 \)

\( S_i \) : Heat production term of reaction \( i, i = 1,2 \) (non-dimensional)

\( S_L \) : Laminar flame speed

\( T \) : Gas temperature

\( T_{ad} = T_b \) : Adiabatic (burned) temperature, used interchangeably depending on the reference

\( T_u \) : Temperature of unburned mixture

\( T_{ij} \) : Ignition temperature of fuel fraction \( j, j = 1,2 \) (dimensional)

\( V \) : Flame speed (dimensional)

\( Z \) : Distance between the onsets of each reaction (non-dimensional)

\( a = 1 + \nu \phi \) : Concentration dependent heat capacity of mixture (non-dimensional)

\( c \) : Heat capacity (dimensional)

\( d_q \) : Quenching distance

\( c_i \) : Integration constants

\( h \) : Heat exchange coefficient
$k_i$: Pre-exponential Arrhenius reaction factor of reaction $i$, $i = 1,2$

$q_j$: Rate of heat production (Arrhenius-related)

$r_j$: Heat loss related eigenvalue, $j = 1,2$

$x$: Spatial coordinate (density weighted and dimensional)

$x_F$: Location of 2$^{\text{nd}}$ front with respect to 1$^{\text{st}}$ one

$\Psi_i$: Rate of reaction $i$, $i = 1,2$ (dimensional)

$\Omega_i$: Non-dimensional heat transfer numbers (Arrhenius related)

$\alpha$: Heat diffusivity

$\beta_i$: Non-adiabatic flame speed coefficient (non-dimensional)

$\delta$: Flame thickness (dimensional)

$\zeta$: Heat production coefficient (non-dimensional)

$\eta$: Heat loss term (non-dimensional)

$\theta$: Temperature (non-dimensional)

$\theta_{ad_i}$: Adiabatic temperature (non-dimensional)

$\theta_{ij}$: Ignition temperature of fuel fraction $j$, $j = 1,2$ (non-dimensional)

$\kappa$: Flame speed (non-dimensional)

$\mu_i$: Non-dimensional heat transfer numbers (explained in section 4)

$\nu$: Heat capacity (dimensional)

$\lambda$: Heat conductivity

$\lambda_u$: Heat conductivity of unburned mixture
\( \xi: \) Spatial coordinate (non-dimensional)

\( \rho: \) Density

\( \rho_u: \) Density of unburned mixture

\( \tau_{\psi_i}: \) Combustion time of reaction \( i, i = 1,2 \)

\( \phi: \) concentration (non-dimensional)
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1 INTRODUCTION

The importance of the combustion process in modern times can be hardly overstated. Despite its association to pollution both of toxic and of greenhouse gases and particulates, it plays an essential role in energetics. Combustion of coal, oil products, natural gas, biomass, low calorific gases or hydrogen provides almost 92% of world primary energy supply [1]. Combustion systems offer a combination of high energy densities and high power densities, so far unrivaled by any other chemical system[2], [3]. They achieve this through the high activation energy of involved reactions [4] which allows, at the same time, for a stable energy storage at low temperatures and fast reaction rates at high temperatures. The central role combustion has played for more than a century has led to a solid fundamental understanding of flames through both experiments and conceptual models. Complex mechanisms are used to optimize performances of turbine combustors [5]–[7], piston engines [8], [9] or rocket boosters [10], [11].

Heterogeneous combustion, involving solid fuels, appears in a variety of applications. Coal and biomass already play a crucial role in energetics. Moreover, prevention and mitigation of explosions in industries handling fine powders, such as in pharmaceutics [12], [13], in food industry (grain elevators [14]–[16], etc.), or in paint production [17], as well as in mines [18], [19], are a strong motivation to study such fuels.

Metals form a distinct group of solid fuels. They are extensively used in many combustion applications. Metal particles enhance the performance of propellants and of slurry fuels [20]–[22], and have the potential of becoming an important fuel on their own [3], [23] in flames involving much faster oxidation than for example metal-air batteries [24]. The study of metals is also advantageous from a fundamental perspective, as they present much more uniform composition and properties than other solid materials such as coal and can thus be used to examine how a particular mixture burns, and to identify and to measure fundamental properties of the resulting flame. The combustion may be studied both on the level of single particles [25]–[27] as well as a frontal phenomenon [3], [28]–[32].

Despite this, the understanding of dust combustion in general is far less developed than in the case of gas. The main cause is the complexity of such flames, resulting from the non-uniform nature of
fuel mixtures. Unlike gases which can most often be considered as a continuum, heterogeneous mixtures, such as suspensions, offer interfaces between the gas and solid on the surface of particles, leading to different physical scales, both on the level of the individual particles and of the overall flame. Apart from complex chemical reactions, such systems are defined both by micro- and macro-scale processes such as thermal and mass diffusion, phase changes (melting and evaporation), transport phenomena, and many other coupled thermal and mechanical effects [33], [34]. This is moreover complicated by widely varying particle size distributions in these suspensions and the fact, that in real settings, the fuels may be in different phases, such as coal and methane in mine explosions.

Fuels with very different properties in mixtures of two distinct powder suspensions or of a suspension with a combustible gas, may often be involved in very distinct reactions. They may compete for the oxidizer or may even react sequentially, one reacting with the products of the other’s reaction with the primary oxidizer. This can in turn lead to a complex multi-frontal structure of the overall flame where the fronts may couple and de-couple depending on various parameters.

The objective of this thesis is to examine, both through experiments and through a qualitative model, the configurations such flames may adopt for various powder concentrations. The understanding of how such a flame propagates is important both in enabling its propagation in cases where it is needed (propulsion or energy production) as well as its preventions in accidents (mine and industrial setting explosions). From a fundamental point of view, this may provide a valuable tool in sorting out the main effects in such heterogeneous flames. The approach is to weaken the flame up to its propagation limit and see how it responds. This probing technique is based on the concept of the quenching distance and heat losses, introduced in the next section.

First, a concept and relevant literature review is presented, introducing the main topics of study, how they fit together and how the present thesis contributes to existing knowledge. The next section examines the experimental tests on the quenching behavior of hybrid aluminum methane-air flames. Then, the qualitative model, developed to explain the behavior of the flame, is introduced and derived. And the last section presents the comparison between the experiments, the model and other existing formulations. The goal of this last part is to identify which of the behaviors and explanations are really fundamental and not only model-dependent.
2 LITERATURE AND CONCEPT REVIEW

The topic of this work concerns a very particular area of combustion involving hybrid mixtures and leading to the formation of dual fronts. The interaction between these two fronts is studied by means of experimentally induced heat losses. Both the assumptions of the model and the results of the experiments rest on a series of concepts pertaining to:

- Heterogeneous, and in particular metal, combustion which studies the fast oxidation of individual solid particles and explains how and when this reaction can lead to the formation of a flame front, analogous to gas combustion
- The different representations of the flame either through ignition temperature and step kinetics or Arrhenius reaction rate
- Multi-frontal combustion in which the flame is treated as a combination of parallel and/or consecutive reactions forming individual fronts whose mutual interaction is the key to understanding the dynamics of the flame
- The presence of heat losses leading to multiple solutions to flame propagation and the phenomenon of quenching

The findings in each of these areas, relevant to this work, are presented below.

2.1 Metal combustion

2.1.1 Single particle combustion

The combustion of single particles can happen in different modes. The division is usually made using the so-called Glassman criterion [35] which classifies the modes of combustion of metal particles by means of comparing the adiabatic temperature of the stoichiometric metal-air reaction to its boiling temperature. If the flame temperature is lower, the metal burns in its solid phase heterogeneously with air on the surface of the particle, and if the flame temperature is higher than
boiling, the metal evaporates and then burns in vapor phase in a micro-flame on or further from the particle.

2.1.2 Concept of ignition

Depending on its size, the properties of the metal and its oxide, the temperature and the type of oxidizer, the particle combustion may either be limited by various physical diffusion processes or by chemical reaction kinetics, as discussed in [36].

Of a particular interest is the competition between surface kinetic reactions and heat diffusion away from the particle. A particle may start to burn in the kinetic mode which is relatively slow at low temperature, due to its exponential dependence on it. But as the heat produced by the reaction increases the temperature on the surface, this reaction rate increases. At a critical temperature, the rate is too high for the heat to be diffused away from the particle and heats it up sharply. This leads to an exponential growth of the rate analogous to a Semenov thermal runaway [37] and the particle quickly switches from a kinetic to a diffusion-limited mode. This critical transition is described as the ignition of the particle occurring at the ignition temperature.

2.1.3 Effect of concentration

Different from the single particle ignition is the concept of onset of reaction, described in [38]. One can imagine a gas flow which is seeded with increasing concentrations of metal particles. A single particle undergoes a slow oxidation reaction which takes a long time to completion and, assuming no heat losses, this raises the temperature of the gas infinitesimally over a very long time. As the concentration increases, the combined heating starts to have a higher total effect on the surrounding gas which in turn raises the temperature, i.e. the reaction rate, of the particles in a positive feedback loop. The particles react faster and the reaction time thus diminishes quickly to the point where it reaches the heat diffusion timescale. This fast reaction of particles, which, unlike ignition, does not necessitate any criticality, results in the formation of a front analogous to a gas flame.
2.2 Flame representation: Ignition versus Arrhenius kinetics

This discussion of how combustion starts and proceeds can be extended to a general gas flame. In 1881, in a first attempt to formulate a physical model for a flame, Mallard and LeChatelier [39] imagined it as a thin reaction front propagating through an unburnt mixture. The front preheats the gas ahead by conduction. As soon as this gas reaches a prescribed ignition temperature, the reaction starts. The propagation speed of the front is simply obtained by equating the rate of heat transfer through the front boundary to the rate of heat needed to bring the mixture from initial to ignition temperature in the preheat zone (Figure 2-1). This shall be discussed in more detail in the model section.

The Mallard and LeChatelier formulation was eventually dismissed, mainly due to the imposition of an external ignition temperature. This approach is inadequate [40] as it presumes the existence of a precise temperature, similar to boiling or evaporation, which a given mixture either reaches and reaction happens or does not and no flame is produced. Yet, a fuel-oxidizer gas mixture reacts at any temperature and the reaction rate follows the Arrhenius law, in an exponential dependence on temperature. Provided there are no heat losses to a system, it is only a matter of time when a mixture sets off a flame, first slowly raising the temperature and thereby the rate of reaction and then exponentially reaching a flame. For a low initial temperature $T_0$, this time is extremely long. But raising $T_0$ leads to an exponential decrease in this time, as calculated by Todes [37], [41]. This explains why a mixture of methane and oxygen at room temperature will never set off unless it is provided with an external heat source. The precise conditions of reactions then depend on a combination of temperature and species concentration evolution and are not unique for an initial mixture.

Thus models that followed used both a combination of energy and species conservation equations as well as an Arrhenius reaction rate approach. These were able to provide flame speed expressions, through matched asymptotic expansion, by making the generally realistic assumption of high activation energies for flames [37], [42]. This formulation is in some sense similar to a high ignition temperature assumption, where the gas has to reach fairly high temperatures before it quickly forms a flame, but without the constraining imposition of an actual critical temperature.
The flame then happens in an extremely thin zone around the flame adiabatic temperature (Figure 2-1).

![Figure 2-1: Schematics of representation of a flame through both ignition temperature and Arrhenius-type models.](image)

The absence of criticality in Arrhenius formulations and its presence in the case of ignition has important fundamental mathematical consequences. In Arrhenius this leads to a general smoothness in the system which favors numerical approximations and, provided the grid mesh is adapted to the quickly varying scales, one can obtain even accurate unsteady solutions for complex systems [43]. On the other hand, the term $e^{-E_a/RT}$ precludes any precise analytical formulation and approximations are obtained by expanding the exponent under strict and limiting conditions of large (or low) activation energies $E_a$. In the case of ignition, the situation is reversed. The contradiction between numerical approximation and the discontinuity induced by the sharp diving boundary of the step requires a lot of attention in numerical resolution [44] and may lead to the loss of important qualitative behavior. The advantage of this formulation, however, becomes clear in analytical terms. The sharp boundaries provide the system with clearly-defined domains over which the governing equations may be solved separately and then the solutions matched at individual boundaries[40], [45]–[47]. This can often lead to exact analytical closed-form solutions to the governing equations which allow the system to be completely defined in the space of all of
its parameters. Moreover, provided a reasonable choice of the ignition temperature, this formulation may achieve good quantitative agreement with Arrhenius and experiments while allowing to obtain a more complete picture of all propagation regimes [40].

This mathematical aspect of criticality shall further be discussed in the analytical model section as well as in the comparison of its findings with the Arrhenius approximate analysis.

### 2.3 Two-front flames

The present thesis examines the behavior of so-called hybrid flames involving metallic suspensions in methane-air mixtures. Such flames have been observed to exhibit two-front structures, notably in experiments with aluminum and iron performed at McGill University [29]–[32], [45], [48]. There, methane-air Bunsen-type flames were seeded with increasing concentrations of metal powder. At a critical concentration the suspension would undergo an onset of reaction and form a bright front. This would be accompanied by a significant increase in measured temperature to values close to aluminum/iron combustion in methane-air products. Also, a two-flame structure would be directly observed in the case of iron, aluminum combustion being too bright.

Indications of two-front flames have been indirectly observed in other heterogeneous systems, usually performed in closed bombs [49]–[51], by the presence of two consecutive pressure rises, probably corresponding to the impact of two consecutive fronts on the wall. Yet, two-front flames do not necessitate a heterogeneous mixture. It has been observed in a large variety of other flames involving mixtures of gases such as methyl nitrate, methyl nitrite, methane/nitrogen dioxide [52]–[54].

Theoretical studies pertaining to simplified systems of consecutive reactions were first comprehensively performed by Khaikin [55] and interpreted by Zeldovich [37]. In their analysis, the flame speeds were obtained by the method of matched asymptotic expansion, resting on the realistic assumption of very high activation energies of combustion reactions [56]. \( R_1 \) and \( R_2 \) form fronts whose respective speeds are \( S_{L_1}(T) \) and \( S_{L_2}(T) \). The temperature profile reaches a value \( T_{b_1} \) after \( R_1 \) and \( T_{b_2} \) after \( R_2 \) (Fig. 2-2). Depending on the relative values of all of the parameters
of $R_1$ and $R_2$, it was shown that the overall flame may adopt one of three configurations under the separation, control and merging regimes. These results were confirmed by other studies [37], [55], [57]–[61].

![Figure 2-2: Schematics of a two-front flame problem.](image)

As Merzhanov [62] explained, in a propagating flame this can be easily understood by comparing the frontal speeds of $R_1$ and $R_2$, namely the speeds of $R_1$ at $T_{b1}$ and $T_{b2}$, $S_{L1}(T_{b1})$ and $S_{L1}(T_{b2})$, as well as the speed of $R_2$ at $T_{b2}$, $S_{L2}(T_{b2})$.

Since $T_{b2} > T_{b1}$, the front of $R_1$ propagates faster at $T_{b2}$, i.e. $S_{L1}(T_{b2}) > S_{L1}(T_{b1})$.

The three configurations stem from the relative values of these three speeds:

- When $S_{L1}(T_{b1}) > S_{L2}(T_{b2})$, $R_2$ is not fast enough and separates from the $R_1$ which propagates independently. This is called the separation mode and the overall flame depends solely on the parameters of $R_1$, propagating at $S_{L1}(T_{b1})$.

- When $S_{L1}(T_{b2}) > S_{L2}(T_{b2}) > S_{L1}(T_{b1})$, $R_2$ at $T_{b2}$ moves faster than $R_1$ at $T_{b1}$ and approaches it. The heat flowing from $R_2$ to $R_1$ raises the temperature and the speed of $R_1$. But $R_2$ cannot reach $R_1$, because this would raise the temperature of $R_1$ to $T_{b2}$, and as $S_{L2}(T_{b2}) > S_{L1}(T_{b1})$, $R_1$ would accelerate and distance itself from $R_2$. So $R_1$ and $R_2$ settle
at a constant distance, controlled only by the heat flowing from $R_2$ which also dictates the speed of the overall flame at $S_{L_2}(T_{b_2})$. This is therefore called the control mode.

- When $S_{L_2}(T_{b_2}) > S_{L_1}(T_{b_2})$, $R_2$ is faster than $R_1$ even at $T_{b_2}$ and becomes attached to $R_1$. The two reactions become merged and both essentially happen at $T_{b_2}$ but the overall flame in this merged mode is limited by the speed of the first reaction $S_{L_1}(T_{b_2})$.

The aforementioned modes of flame propagation will be discussed later in the thesis and will be used to explain the experimental results.

The concept of two interacting fronts has also been extended to explain behaviors in heterogeneous flames [46]. This model, which is further developed in the present study, has been formulated primarily for the combustion of two very different solid suspensions. In this work, the results are supported by experiments performed on mixtures of aluminum and manganese micron-sized particles. Each front, i.e. the reaction between the oxidizer and each metal, starts when the mixture reaches the respective ignition temperature of each metal. This simplification can correspond to larger particles undergoing ignition. It was found that, depending on the respective concentrations of each solid fuel, the flame could exist as only one front, with one of the metals burning, two separated fronts or two overlapped fronts, similar to the separated and merged fronts in the Arrhenius analysis. The simplified formulation of the problem and the use of ignition temperature allowed the governing equations to be solved analytically and the flame speed was expressed in a set of algebraic equations for all concentrations.

### 2.4 Quenching distances

The previous discussion was conducted in the context of adiabatic flames. Yet, real flame systems are usually subjected to some level of heat losses. Often, the adiabatic approximation is close enough as these losses are minimized in engineering applications through insulation layers, concentric geometries [63] or heat recirculation designs [64]–[67]. Nevertheless, in many applications, heat losses are an important factor to be taken into account. For a given flame, these can happen through conductive and convective transport as well as through heat radiation to the surroundings.
Mathematically, heat losses provide the system with a criticality and the existence of multiple solutions [37]. It has been proven that an adiabatic diffusion-reaction system representing a flame has a unique solution for a given set of parameters. But the presence of heat losses allows the flame to adopt two equilibria, i.e. steady state configurations [37], [68]. Under heat losses, the flame temperature diminishes from the adiabatic value, independent of the rate of reaction, to a maximum burning temperature, a function of the flame speed. This new dependence creates one more degree of freedom which leads to the possibility of more than one solution to the flame profile and velocity combination [37].

The present discussion shall be limited to linear heat losses, induced in a flame by conduction to the walls surrounding the system. As was shown first by Spalding [68] and others [69]–[72], the application of linear heat losses leads to the existence of two solutions to the flame propagation, one higher and physical and one lower and unphysical. The physicality of the solution comes from the response of the flame to parameters such as, for example, the increase of heat losses. This will cause the physical speed to decrease while the unphysical will be enhanced. Increasing the heat losses further to a critical value leads to the two speeds to merge into a single one, called the quenching speed. For higher heat losses, no propagation solution exists. This bifurcation is a general phenomenon caused by losses to diffusion-reaction systems.

In practice, heat losses are raised by forcing the flame through tubes or channels. As the channel becomes narrower, the shrinking cross-section, i.e. the reactive region, of the flame becomes smaller relative to the perimeter, i.e. the surface where heat losses occur. This results in a slower physical speed decrease down to a minimum at quenching when the flame disappears. The channel width at that point is the quenching distance, denoted $d_q$ in figure 2-3. As the magnitude of heat losses sufficient to quench the flame are directly linked to its strength, its rate of reaction and therefore to its velocity, the quenching distance is inversely proportional to the burning velocity and is a fundamental property of the flame [68]. Quenching distances play a crucial role in engineering applications, allowing flame propagation in small engines [73] and its extinction in flame arrestors [74].

There is a significant difference in the quenching behavior observed between flames in gas and in heterogeneous mixtures. In the first case, quenching distances have been widely studied for various mixtures [75]–[77] over wide ranges of equivalence ratios. In gases, the flame is the strongest
around stoichiometry, at equivalence ratios of 1. For lower or higher values, one of the reactants is in excess and instead of participating in the heat production through combustion, higher fractions of it stay unreacted and only contribute to the heat capacity. Therefore, the flame becomes effectively weaker and at too low or too high equivalence ratios, it extinguishes, marking the lower and higher flammability limit of the flame. Therefore, as the flame approaches either of them, the flame needs less and less heat losses to disappear and the quenching distance sufficient to achieve this increases. This results in the quenching curve shown in fig. 2-3.

![Quenching Curve](image)

**Figure 2-3:** Qualitative shapes of quenching distance curves for gas and heterogeneous (powder) flames.

On the other hand, in the case of suspensions, one may change the equivalence ratio by varying the concentration of powder in air. At low concentrations, the particles cannot form a front, but after a critical value, the front forms and becomes stronger, reducing its quenching distances quickly. Around stoichiometry, the mixture achieves its minimum quenching distance. But, as was observed in previous tests with metal suspensions [45], [78], the metal particles, orders of magnitude higher in density than the air molecules, do not displace the air, so the increase in heat capacity of the mixture is well balanced by the increase of particle surface for reaction with air, resulting both in a steady burning velocities [79], [80] and quenching distances [45], [78], which
stay around the minimum value for richer mixtures. For very rich mixtures though, the dispersion of powder is limited by the capabilities of the experimental setup and the quenching behavior is unclear. These results were reproduced by a simplified model by Goroshin et al. [45], [46], on which the one in this thesis is closely based, and which shall be developed in the model section.
3  EXPERIMENTS

The following section presents the experimental tests and results. The experimental apparatus is a modification of previous designs [45], [78], [80]. It has been constructed in the framework of an undergraduate thesis where all the details can be found [81]. This work contains only the description necessary to understand the experiments and subsequent results. These experimental results have been presented and published in the Proceedings of the 35th Combustion Symposium [32].

3.1  Experimental apparatus and methods

3.1.1  Mixtures used

The experiments performed in this work involved well-defined suspensions of aluminum powder in methane-air mixtures. The powder was composed of atomized aluminum particles (Ampal 637, Ampal NJ) used in previous experiments with stabilized dust flames and aluminum flames in tubes [29]–[31], [82]. Ampal aluminum particles have a spheroidal or nodular shape. The Sauter mean diameter, d32, derived from the particle distribution (Fig. 3-1) obtained from SEM images of the powder, was about 5.6 μm, while measurements obtained using the light scattering technique with a Malvern Mastersizer 2000 yielded a value of 6.9 μm. Detailed information on the particle size distribution of the powder is given in [32]. Control tests also involved inert silicon carbide powder with an irregular particle shape and with a Sauter mean diameter instrument of about 4.2 μm.
As preliminary measurements with mixtures of air and methane in stoichiometric proportions yielded very low quenching distances, very close to minimal measurable values, inert nitrogen gas was used to dilute this mixture until the more easily measurable values (>2 mm) were obtained. The final gas oxygen concentration was about 16.3%.

3.1.2 Apparatus

The main purpose of the present experimental apparatus is to measure quenching distances in hybrid solid-gaseous fuel mixtures. As said before, it combines the basic features of earlier designs for flame quenching experiments in dust clouds [45], together with a number of novel features for the dust flow and flame diagnostics developed for the stabilized flame burner apparatus [31], [38], [82].

The apparatus is shown in Fig. 3-2. The metal powder is stored in a piston at the bottom of a generic dust dispersion system, used in previous designs [45], [78]. This system permits the creation of a uniform dust flow with micron-size powders of particles with a diameter less than 20 μm. The syringe-type piston feeds the powder through an inlet tube where the powder is impinged by a concentric supersonic “air knife” jet of gas, issued from a 50-μm annular slot. The initially turbulent flow is expanded and laminarized in a long 8° angle conical diffuser before flowing into the flame tube (see Fig. 3-2 C). The mixture flows up the 48-mm-inner-diameter, 1.5-m-long
vertical Pyrex flame tube (see Fig. 3-2 A) which, for safety reasons, is encased inside a larger diameter acrylic tube. The mixture flows through the upper open end of the flame tube, equipped with a short 25-mm-wide aluminum conical nozzle. Right above this exit, the powder concentration is monitored. The purpose of the nozzle is to smoothen the jet flow while the powder concentration is measured, minimizing the noise in the measurement signal, as well as to reduce disturbances to the flow inside the tube that otherwise are induced by the ventilation flow. An electrically-heated tungsten filament is stretched across the flow below the nozzle. During the experiment, it is used to ignite the mixture. Immediately after ignition, the flow in the tube is stopped by closing the solenoid valve in the gas supply line and the flame propagates down to the closed end of the tube in the quiescent two-phase fuel-air mixture.
The quenching distances of flames were measured by observing flames propagating through assemblies of equally-spaced steel or brass plates (Fig. 3-2 B). These are positioned in the upper part of the tube at about 50 cm from the upper open end. The plate assembly is supported inside the glass tube by resting on a loop of spring wire. The positioning of the quenching channels at a relatively short distance from the point of the dust concentration monitoring (laser beam) minimizes both the error in estimating the dust concentration inside the quenching channels and
the influence of the flame acoustic as well as other oscillations [83] on flame propagation and quenching. The 0.5-mm-thick and 52-mm-long brass or steel plates can be positioned from each other at an equal distance varying from 4 to 20 mm for steel assemblies and 2 to 5 mm for brass assemblies. The steel quenching plates are soldered together by four thin rods passing through the blades, whereas the brass plates are held together between lower and upper brass rings having precision-milled 0.5 mm slots (Fig. 3-2 B).

3.1.3 Concentration measurements

An important feature of the apparatus is the concentration monitoring system. Most experiments involving dust combustion lack the ability of directly measuring the powder concentration, which has to be estimated a priori [49]–[51], [84], [85]. On the other hand, the present system is equipped with a concentration measuring system which allows it to follow the concentration evolution in time. The system comprises a laser-light-attenuation probe consisting of a diode laser, cylindrical lenses forming a rectangular 10 × 2 mm beam, a narrow band-pass filter and spatial filters, and lenses to focus the laser beam onto a photodiode sensor. The laser beam traverses the powder-seeded flow along the flow jet diameter right above the nozzle exit at the upper end of the tube. The output of the photodiode is constantly recorded by a data acquisition system and is also displayed on the monitor for real-time observation. The system is calibrated by aspiration of the dusty flow through a set of fine multilayered filters with a vacuum pump for about 5 seconds. The average dust concentration during the measurement time is then determined by dividing the total mass of the collected dust by the flow volume, i.e., the volume of gas fed through the dispersion system during the same time.

3.1.4 Experimental procedure

Once the piston starts pushing powder into the apparatus, there is a time interval during which the concentration of aluminum in the laminar stream of the hybrid fuel mixture ascending the tube is transient. This concentration reaches an approximate plateau after about 45–50 s as indicated by the concentration monitoring system at the exit of the tube (see Fig. 3-3). The duration of the
plateau is usually about 3–5 s. With a powder mixture flow of about 25 cm/s, the concentration is uniform over about 75–100 cm of the upper tube length. Once the monitoring system indicates the plateau has been reached, the operator cuts the flow and, with a short time delay, ignites the mixture. Consequently, the flame propagates down through the quiescent mixture with a sufficiently uniform powder concentration stretching from the point of ignition down past the quenching channels. The accuracy of the concentration measurements is estimated to be in the range of ±10% of the measured value. Upon reaching the plate assembly, the flame either extinguishes or separates into flamelets, propagating through individual channels of the quenching plate assembly. The flame propagation in the tube and through the quenching plate assembly is recorded by a high-speed camera at a rate from 300 to 600 frames/s.

![Concentration monitoring system](image)

**Figure 3-3:** Concentration monitoring system, its location in the apparatus and powder concentration plateau reached before the mixture is ignited.

### 3.2 Results

#### 3.2.1 Inert Silicon Carbide tests

The goal of the experimental tests was to understand the behavior of hybrid flames resulting from the combustion of solid (aluminum) suspensions in methane-air mixtures. As will be discussed later, seeding a mixture with reactive powders increases the reactivity of the overall blend but also its heat capacity. In order to study and assess the relative contribution of both of these effects, the methane-air mixture was first seeded with increasing concentrations of inert silicon carbide (SiC)
powder of a comparable size and heat capacity to the aluminum one. The relative inertness of SiC in methane-air flames was demonstrated in previous tests [29], [31], [82]. These tests allowed to isolate the effect of heat capacity increase. The results on quenching experiments with methane flames seeded with SiC powders are presented in Fig. 3-4. These flame display an orange appearance similar to the light emitted by soot in the flame during gas combustion.

As Fig. 3-4 shows, the quenching distances obtained for pure gaseous methane-air-nitrogen flames are in the range of 3–3.5 mm. As the reduction of oxygen leads to higher quenching distances [77], this result is consistent with the reported value of 2–2.5 mm for a stoichiometric methane-air flame quenching within rectangular channels at the normal oxygen content of 21% [86]. During the tests, when the flame propagated through the channels, the speed of the flamelet in each channel was usually different, due to local fluctuations in dust concentration. The quenching distance increases with the increase of SiC content from about 3.5 mm for an unseeded flame to about 9.5 mm for concentrations around 270 g/m$^3$ for which flame propagation in the tube is still possible. At higher concentrations of SiC powder above 270 g/m$^3$, the seeded flame fails to
propagate down the tube. This is in agreement with previous observations of the influence of SiC seeding on quenching of the stabilized stoichiometric methane-air flame [29]. There, the tip of the Bunsen methane flame seeded with SiC started to be open at dust concentrations of about 200 g/m$^3$ and the flame completely disappeared at SiC concentrations above 300 g/m$^3$. Therefore, the increase in inert powder loading leads to higher quenching distances until the flame cannot propagate at all.

3.2.2 Aluminum tests

In a second series of tests, SiC powder was replaced by reactive aluminum particles. Again, the mixture was seeded with increasing concentrations of powder and the quenching distances of resulting flames were measured.

Similar to stabilized Bunsen methane-aluminum flames, the freely-propagating flames in mixtures of aluminum suspensions demonstrated two significantly different flame propagation modes, one characterized by a dimmer orange flame and one by a very bright white flame. The overall results are displayed on an experimental map (Fig. 3-5). The upper region of the map corresponds to the flame propagation within the tube, i.e. outside of the channel assemblies, whereas the lower part shows the flame behavior within the channel assemblies for varying channel widths and aluminum concentration.

First, within the main tube, at aluminum concentrations below the critical value, which was about 300 g/m$^3$ in the present experiments, most of the aluminum oxidizes relatively slowly in the products of the methane flame over a long distance without substantially raising the flame temperature [29], [31]. A fraction of particles may sometimes undergo a more vigorous combustion, forming isolated streaks within the flame. Otherwise, the flame appearance and color are very similar to the appearance of the flame seeded with inert SiC powder, and the flame quenching distance, as shown in Fig. 3-5, similarly increases with an increase of powder loading. The somewhat higher quenching distance of the flame seeded with SiC, in comparison to aluminum, can be explained by the smaller size of the SiC particles that are in closer thermal equilibrium with the gas than aluminum and, therefore, provide a more effective heat sink.
Moreover, the previously described slow oxidation of aluminum may, in a very limited way, contribute to the overall heat production.

At some critical concentration of aluminum, a rapid transformation of the flame appearance is observed. This occurs around 150–200 g/m³ for the stationary Bunsen flame in methane-air mixtures studied in [31] and at about 300 g/m³ in the present experiments performed with freely-propagating flames at reduced oxygen concentration. This qualitative transformation, as was shown in [29], [30], is associated with the formation of an aluminum dust-flame front that becomes coupled to the methane flame. This behavior is in accordance with the previous test on Bunsen flames [29], [31], [82]. There, this change in brightness of the flame was accompanied by a sharp decrease in the visible combustion zone of aluminum from the order of cm to 1–2 mm, by an increase in the completeness of aluminum combustion from about 50–70% to 100%, and by a sudden jump in temperature from values close to the methane flame with inert additives to thermodynamically-predicted values of aluminum reacting with water and CO₂ in the methane combustion products [29].

The heat from the combustion of aluminum enhances the overall flame, enabling its propagation for much larger powder loading than in the inert SiC case. The upper limit in concentration in the present tests, 600 g/ m³, is only due to the limitation of the current dispersion system and the flame exhibited a propensity to propagate at even higher concentrations.
The coupled very bright methane-aluminum flame exhibits two different types of quenching behavior depending on the aluminum concentration and the width of the channel. As Fig. 3-5 shows, at aluminum concentrations below 450 g/m$^3$, upon entering the channels, the bright flame disappeared and only the dim orange one, similar to the ones observed for SiC and at low concentration of Al, propagates through the assembly. The bright aluminum flame, however, re-
appears in less than a tube diameter after the flame emerges from the quenching channels (see Fig. 3-6).

![Flame propagating through the quenching plates, displaying a de-coupling behavior.](image)

Figure 3-6: Flame propagating through the quenching plates, displaying a de-coupling behavior.

At aluminum powder concentrations above 450 g/m$^3$, no change in brightness is observed in the flame as it enters the narrow channels (Fig. 3-7), the flame does not transfer to a dim orange color. It is also interesting to note that the bright white flame inside the channel seems to demonstrate a weaker dependence of the quenching distance on concentration than the decoupled methane-aluminum flames (Fig. 3-5).

The apparent dimmer color of Fig. 3-7 comes from the different camera settings which attenuate the brightness of the flame. In reality no significant difference in brightness between flames of the type of Fig. 3-6 and of Fig. 3-7 have been observed.
One interesting behavior that one can observe on Fig. 3-7 is the difference in propagation velocities of individual flamelets in the channels. This is due to the fluctuations of powder concentration in the tube along with some residual turbulence in the flow. These are inherent in this kind of tests and have to be accounted for. These variations in concentration lead to the difference in local reaction rates of the flamelets, causing the variation in speed.

The consequence of this is that some of the flamelets (the leftmost one in the figure) reach the end of the channels first. Once this occurs, the flamelets quickly expands in the tube into a full-fledged flame. By doing so, it pushes the mixture in front of this expansion back into the main tube as well.
as into the other channels. Thus, the slower flamelets are actually pushed back into the tube, resulting in their blow-off back into the upper part of the tube. Sometimes, this push is so vigorous that it forces a considerable amount of fresh mixture into the upper part of the tube. This often causes a vigorous short-lived flame in a supposedly burnt region. A good example can be seen in Fig. 3-8 where the expansion of the flame forces a significant amount of powder up one channel into the product side, causing a momentary flash, which lasts for less than a second. One can also notice how the color of this powder pocket switches from orange to bright white. This is an example of the uncertainty inherent to powder combustion in general.

The aforementioned results provide a range of varying flame behaviors that require more explanation and interpretation through simple combustion concepts. This lead to the development of the simplified model, described in the following section. The model results are then used to interpret the experiments in the subsequent section.
4 ANALYTICAL MODEL

4.1 Model formulation and assumptions

The aim of the present model is to study the behavior of dual-front flames from a qualitative perspective. This will allow to clearly explain why and when the flame adopts its different configurations observed in the last chapter. For the sake of simplicity, the structure of the flame has been greatly simplified by means of assumptions listed below. While such formulation inevitably leads to relatively large quantitative disagreement with experimental values, it is very useful in isolating and underlining main qualitative causes of flame coupling.

In this work, the flame is represented as a one-dimensional steady-state structure propagating into an unburned mixture of an oxidizer and two very different fuels. The frame of reference is placed on top of the flame so that the flame appears to be stationary. This formulation corresponds to the schematics presented on the left side of Fig. 2-1. The flame is set to start at x=0.

As discussed in the Literature Review section, merging in flames with double fronts has been most often associated to thermal communication between both fronts. Most of the previously-discussed models [55], [59], [87] represent flame fronts as reaction-diffusion waves. As has been shown, the chemistry of the reaction is simplified, leading to simple parallel or sequential schemes of two independent reactions $R_1$ and $R_2$ of the form:

\[
\begin{align*}
A & \xrightarrow{R_1} C \quad \text{(parallel)} \quad \text{or} \quad A \xrightarrow{R_1} E \xrightarrow{R_2} F \quad \text{(sequential)} \quad (4-1)
\end{align*}
\]

Where A, B, C, D, E, and F represent different species. These schemes usually study only the conservation of the limiting reactants A and B, and of the intermediary species E. The steady-state flame profile is usually obtained by solving a system of differential equations comprising an energy equation across the flame coupled to a set of species conservation equations for each of the reactants. As there is no pressure increase across the flame, the momentum conservation equation can be neglected [56]. The set of equations then become:
Table 4-1: Systems of reaction-diffusion equations for two-reaction fronts

<table>
<thead>
<tr>
<th>Parallel</th>
<th>Sequential</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \lambda_u \frac{d^2 T}{dx^2} - V \rho c \frac{dT}{dx} + Q_1 \psi_1(a) + Q_2 \psi_2(b) = 0 )</td>
<td>( \lambda_u \frac{d^2 T}{dx^2} - V \rho c \frac{dT}{dx} + Q_1 \psi_1(a) + Q_2 \psi_2(e) = 0 )</td>
</tr>
<tr>
<td>( D_A \frac{d^2 a}{dx^2} - V \frac{da}{dx} - \psi_1(a) = 0 )</td>
<td>( D_A \frac{d^2 a}{dx^2} - V \frac{da}{dx} - \psi_1(a) = 0 )</td>
</tr>
<tr>
<td>( D_B \frac{d^2 b}{dx^2} - V \frac{db}{dx} - \psi_2(b) = 0 )</td>
<td>( D_E \frac{d^2 e}{dx^2} - V \frac{de}{dx} + \psi_1(a) - \psi_2(e) = 0 )</td>
</tr>
</tbody>
</table>

Here, \( V \) is the propagation velocity of the flame, and \( a, b, \) and \( e \) are the respective concentrations of species A, B and E. \( \psi_1 \) and \( \psi_2 \) are the rates of reaction of \( R_1 \) and \( R_2 \) and are depending on the limiting reactants of each equation. The limiting reactant can be either the fuel or the oxidizer. The mixture is assumed homogeneous at every point across the flame leading to a uniform temperature profile \( T \).

The present model further simplifies this formulation. Similar to [45], [46], each reaction is modeled to happen in a purely diffusion regime. Moreover, the mixture is assumed fuel-lean, meaning there is enough oxidizer for both reactions and its depletion has thus no influence on either reaction rate. Thus, each reaction starts as soon as the temperature reaches a given ignition value and proceeds at a constant rate for a given combustion time. Both reaction rates can therefore be represented as Heaviside functions. The ignition temperature and the combustion time are external parameters analogous to the Mallard and LeChatelier formulation. Even though, as has been mentioned in the review section, this approach has been dismissed in the gas flame literature [40] as very inaccurate and replaced by Arrhenius kinetics, it is much more applicable to combustion of suspensions where the onset of reaction can be a critical phenomenon.

The independence of \( \psi_1 \) and \( \psi_2 \) on the concentrations of the limiting reactants leads to the decoupling of the differential equations. Therefore, both reaction schemes collapse into a single energy equation:

\[
\lambda_u \frac{d^2 T}{dx^2} - V \rho c \frac{dT}{dx} + Q_1 \psi_1 + Q_2 \psi_2 = 0
\]  
(4-2)
This equation has been obtained by means of some further assumptions, discussed in [45], [46]:

- The spatial variable $x$ is in fact scaled with respect to density in a so-called ‘Spalding transformation’ [88], [89].

$$x = \int_{0}^{X'} \frac{\rho}{\rho_u} dx$$  \hspace{1cm} (4-3)

This eliminates the effect of gas expansion caused by the sharp increase of the flame temperature. As the equations under consideration are steady-state, one can easily retrieve the actual spatial flame profile (with respect to $x'$) through an inverse transformation.

- The conductivity in the range of 300 – 3000 K is assumed linear with respect to temperature.

$$\lambda = \lambda_u \left( \frac{T}{T_u} \right) = \lambda_u \left( \frac{\rho_u}{\rho} \right)$$  \hspace{1cm} (4-4)

This allows the temperature diffusion term to become $\lambda_u \frac{d^2T}{dx^2}$ in the present coordinate system.

This work also examines how heat losses, due to quenching channels, affect the flame speed and structure. These are represented in a same way as in [45]. Similar to other fundamental work involving heat losses [43], [47], [68], conduction to the channel walls is considered to be the main vector and the losses are represented as a simple linear function between the gas and the cold channel temperature $T_u$ (also the temperature of the unburned mixture) scaled by a heat transfer coefficient $h$. The equation then becomes:

$$\lambda \frac{d^2T}{dx^2} - V \rho c \frac{dT}{dx} + Q_1 \psi_1 + Q_2 \psi_2 - h(T - T_u) = 0$$  \hspace{1cm} (4-5)

In this first approach, radiation is neglected for sake of simplicity. In reality, for such hot flames (~3000 K) radiation heat losses may play some role. It has been shown [30], that for such small
experimental settings as in the tube apparatus, radiation has a negligible effect on the preheat zone and can be considered solely as a heat loss.

In equation (4-5), the reaction rates $\psi_1$ and $\psi_2$ are Heaviside steps. As the gas is heated up in the preheat zone (from $x \to \infty$ to $x = 0$), it first reaches the lower ignition temperature $T_{i1}$ of reaction 1. This is set to happen at $x=0$. The mixture is further heated until it reaches the second ignition temperature at $x = -x_f$.

The rates can be thus expressed in the following manner:

$$
\psi_1 = \frac{1}{\tau_{c1}} [H(x) - H(x + \tau_{c1}V)]
$$
$$
\psi_2 = \frac{1}{\tau_{c2}} [H(x - x_f) - H(x - (x_f - \tau_{c2}V))] \quad (4-6)
$$

Far ahead of the flame, the mixture does not feel the effect of the flame and far behind it, the mixture reaches equilibrium conditions. The appropriate boundary conditions are:

$$
x \to -\infty \quad T(x) \text{ is finite} \quad \frac{dT}{dx}(x) = 0
$$
$$
x \to +\infty \quad T(x) = T_u \quad \frac{dT}{dx}(x) = 0 \quad (4-7)
$$

4.2 Non-dimensionalization

Following the same logic as in [45], [46], equations (4-5) and (4-6) are cast into a non-dimensional form with all parameters listed in table 4-2.
Table 4-2: Non-dimensional numbers

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \theta = \frac{T}{T_u} )</td>
<td>Non-dimensional temperature</td>
</tr>
<tr>
<td>( \xi = \frac{x}{V \tau_c} )</td>
<td>Non-dimensional spatial coordinate</td>
</tr>
<tr>
<td>( \kappa = \sqrt{\frac{\alpha}{\tau_c}} )</td>
<td>Non-dimensional flame speed ( \where \alpha = \frac{\lambda}{c_1 \rho_1} )</td>
</tr>
<tr>
<td>( \eta = \frac{h \tau_c}{c_1 \rho_1} )</td>
<td>Non-dimensional heat loss term</td>
</tr>
<tr>
<td>( \phi = \frac{\rho_2}{\rho_1} )</td>
<td>Non-dimensional concentration ratio of the second fuel fraction</td>
</tr>
<tr>
<td>( \nu = \frac{c_2}{c_1} )</td>
<td>Non-dimensional heat capacity ratio</td>
</tr>
<tr>
<td>( F = \frac{\tau_{c_2}}{\tau_{c_1}} )</td>
<td>Non-dimensional combustion time ratio</td>
</tr>
<tr>
<td>( S_1 = \frac{Q_1}{c_1 T_u} )</td>
<td>Non-dimensional heat of 1\textsuperscript{st} reaction</td>
</tr>
<tr>
<td>( S_2 = \frac{Q_2 F \phi}{c_1 T_u} = \zeta \phi )</td>
<td>Non-dimensional heat of 2\textsuperscript{nd} reaction</td>
</tr>
<tr>
<td>( Z = \frac{x_f}{V \tau_c} )</td>
<td>Position of second front</td>
</tr>
</tbody>
</table>

The non-dimensional form of equation (4-5) and the corresponding boundary conditions are then:

\[
\frac{1}{\kappa^2} \frac{d^2 \theta}{d \xi^2} + (1 + \nu \phi) \frac{d \theta}{d \xi} - \eta (\theta - 1) = \Psi_{1nd} + \Psi_{2nd}
\]

\[
\Psi_{1nd} = S_1 [H(\xi) - H(\xi + 1)] \quad \Psi_{2nd} = S_2 [H(\xi - Z) - H(\xi - Z - F)]
\]

\[
\xi \to -\infty \quad \theta(\xi) \text{ is finite} \quad \frac{d \theta}{d \xi}(\xi) = 0
\]

\[
\xi \to +\infty \quad \theta(\xi) = 1 \quad \frac{d \theta}{d \xi}(\xi) = 0
\]

In the equation above, there are two unknowns: the position of ignition of the second reaction, \( Z \), and the flame propagation velocity, \( \kappa \). Both of these are eigenvalues of the differential equation.
and will be determined once the latter is solved over the spatial domain, i.e. \( x \in (-\infty, \infty) \), with appropriate constraints. These will be given by the possible flame configurations, as discussed in the next section.

The second order linear differential equation (4-8) has a simple closed-form general solution over \((-\infty, \infty)\):

\[
\eta = 0 \text{ (adiabatic):} \quad \theta(\xi) = c_i + c_j e^{-\kappa^2(1+\nu\phi)\xi} - \frac{S}{1+\nu\phi} \xi + 1 \\
\eta \neq 0 \text{ (heat losses):} \quad \theta(\xi) = c_i e^{-r_1 \xi} + c_j e^{-r_2 \xi} - \frac{S}{1+\nu\phi} + 1
\tag{4-10}
\]

Where

\[
r_{1,2} = \frac{1}{2} \left[ \kappa^2 (1 + \nu\phi) \mp \sqrt{\kappa^4 (1 + \nu\phi)^2 + 4\kappa^2 \eta} \right]
\tag{4-11}
\]

The heat source term \( S \) being:

\[
S = S_1[H(\xi) - H(\xi + 1)] + S_2[H(\xi - Z) - H(\xi - (Z + F))]
\tag{4-12}
\]

### 4.3 Cases studied and solution procedure

As mentioned earlier, the goal of this work is to explain flame coupling and decoupling observed during the experiments. In the tests, two parameters are varied: the concentration of aluminum and the heat losses (through the width of quenching channels).

From previous experimental and theoretical work [29], [31], the onset of aluminum combustion at the given size of particles (<10 μm) generally appears slower than the methane one. Moreover, it has been shown that aluminum reacts with products of methane-air reaction, namely water and carbon dioxide. Therefore, reaction 1 (with a lower ignition temperature) was attributed to methane and reaction 2 to aluminum combustion. Reducing the channel width increases the effective heat losses to the flame. The experiments were thus reproduced by varying \( \phi \) (aluminum concentration) and \( \eta \) (heat losses).
In the experiments, these variations lead to changes in flame configurations. The solution approach was to find all such conceivable configurations and to determine under what ranges of \( \phi \) and \( \eta \) these were possible. Six different cases, displayed on Fig. 4-1, were examined. These depend on whether the flame exists only as one or two fronts, whether these fronts are separated or overlapped and finally whether the flame is adiabatic or subjected to heat losses.

As one can see at the bottom of fig. 4-1, the actual location of the second front lead to different profiles of the reaction term \( S \). Each of these profiles yielded a different domain of integration over which equation (4-8) was solved. Figure 4-1 displays the temperature profiles corresponding to each configuration. These are direct results of calculations explained in the following section. Each configuration and corresponding temperature profile will be discussed in corresponding subsections and will be interpreted in experimental terms in section 5.

![Figure 4-1: Flame configurations studied in the present model corresponding to different spatial representations of the heat production term, the existence of heat losses, along with resulting temperature profiles.](image)

In each case, the domain was divided according to the values of \( S \) into different zones, leading to a pair of \( c_i \) and \( c_j \) for every zone. Their values were determined using the boundary conditions and the continuity of both the temperature profile and its derivative between contiguous zones. Applying these conditions for 5 zones (3 in the one-front case), one obtains 12 equations for 10...
unknowns (7 for 6 unknowns in the adiabatic case). This yielded 2 extra equations (1 in the adiabatic case) in each case. These algebraic equations, shown in table 4-3 for each case, were then used to find an exact expression for both eigenvalues of equation (4-8), the velocity $\kappa$ and the value of $Z$, the separation between the onsets of each reaction.

Once $Z$ and $\kappa$ were determined using the extra 1 or 2 equations, the expressions for $c_i$ and $c_j$ (listed in the appendix) were used to obtain the temperature profiles over the domain.

### 4.4 Finding $\kappa$ and $Z$ with extra equations

The following section analyzes the results of the extra algebraic equation(s) for each case. For the sake of clarity, the results have been divided between the one-front case and the two-front case.

#### 4.4.1 One-front flame

##### 4.4.1.1 Adiabatic case ($\eta = 0$)

#### 4.4.1.1.1 Solution

The one-front case has been previously analyzed and solved in [45]. In the present work, this solution to the equation has been analyzed further and recast from an implicit to an explicit form for both the adiabatic and non-adiabatic case. This has allowed a more physical interpretation in actual heat transfer terms. It has also allowed to reinterpret the Mallard and LeChatelier result [39] for flame speed.

In this case, the extra equation was found to be:

$$\kappa^2 a^2 (\theta_{l_1} - 1) = S_1 (1 - e^{-\kappa^2 a})$$

Using the 0-branch of the so-called Lambert function $W_0(x)$ [90], an exact explicit formula for the flame speed has been obtained:

$$\kappa_1^2 = \frac{1}{a} [W_0(-\mu_1 e^{-\mu_1}) + \mu_1] \quad \text{where} \quad \mu_1 = \frac{S_1}{(1+\nu\phi)(\theta_{l_1} - 1)} = \frac{S_1}{a(\theta_{l_1} - 1)}$$

(4-14)
The corresponding curve of the flame speed $\kappa$ vs. the concentration $\phi$ is plotted on Fig. 4-2. One can observe that as the concentration of $\phi$ increases, the flame speed of one front decreases. This is to be expected as in this case, the 2$^{nd}$ fuel, represented by $\phi$ does not undergo combustion and does not participate in the heat production. Its only contribution is to increase the heat capacity of the mixture, which requires more and more heat to achieve an increase in temperature. Therefore, the flame is weakened and slows down. The adiabatic temperature $\theta_{ad_1}$ of this one-front flame is:

$$\theta_{ad_1} = 1 + \frac{\dot{s}_1}{a} = \mu_1 (\theta_1 - 1)$$  \hspace{1cm} (4-15)

At a critical value of $\phi$, the flame is too weakened and all the heat produced in the reaction cannot bring the flame up to the ignition temperature. The flame stops. This happens when $\theta_{t_1} = \theta_{ad_1}$ at:

$$\phi_{0c} = \frac{1}{\nu} \left( \frac{S_1}{\theta_{t_1} - 1} - 1 \right)$$  \hspace{1cm} (4-16)

Figure 4-2: Flame speed curves for one-front (blue) and two separated- (red) and merged- (green) front adiabatic flames.
4.4.1.1.2 Comparison with Mallard and LeChatelier analysis

At the end of the 19th century, Mallard and LeChatelier [39] proposed an expression for the flame speed using an external ignition temperature formulation. In this approach, it was assumed that the steady state flame ignites when the gas reaches a certain temperature. While this formulation has been abandoned in most flame models, considered to be inadequate for reasons explained in the concept review section, it is still in use in some particular cases involving diffusion combustion [40], [45], [46], metal-water propellant combustion [91], and has been even reintroduced for some instances of high activation energy gas combustion [92]. In their analysis, the authors postulated that the velocity can be obtained by considering the heat flux across the flame front. By this reasoning, the rate of heat needed to bring the mixture from ambient temperature $T_u$ to the ignition temperature $T_i$ is equal to the rate of heat transfer conducted from the flame to the preheat zone. One can then obtain a simple expression for the flame speed:

$$ \dot{m}c_p(T_{ig} - T_u) = \lambda A \frac{dT}{dx}(x_{ignition}) $$  \hspace{1cm} (4-17)

$$ \rho V A c_p(T_{ig} - T_u) = \lambda A \frac{dT}{dx}(x_{ignition}) $$  \hspace{1cm} (4-18)

As Fig. 4-3 (a) shows, by using the fact that the flame happens in a very narrow zone $\delta$, $\frac{dT}{dx}(x_{ignition})$ can be approximated as $\frac{T_b - T_i}{\delta}$, yielding:

$$ \rho V c_p(T_i - T_u) = \lambda \frac{T_b - T_i}{\delta} = \lambda \frac{T_b - T_i}{V \tau_c} $$  \hspace{1cm} (4-19)

$$ V_{MLC} = \frac{\lambda}{\rho c_p} \frac{T_b - T_i}{T_i - T_u} \frac{1}{\tau_c} = \frac{\lambda}{\rho c_p} \frac{T_b - T_i}{T_i - T_u} \dot{\omega} $$  \hspace{1cm} (4-20)
One can express this result in terms of non-dimensional numbers used in the present model. This yields:

\[ \kappa_{MLC}^2 = \frac{\theta_{ad1}^{-1} - 1}{\theta_{i1}^{-1}} = \mu_1 - 1 \] (4-21)

Figure 4-3: Comparison between the approximations of the slope in the Mallard and LeChatelier formulation, the illustrative case, and the present model.

Now, one can consider the other ‘illustrative’ case where the actual \( \frac{dT}{dx} (x_{ignition}) \) is approximated now by the slope of the preheat zone \( \frac{T_b-T_u}{\delta} \). This purely theoretical construction, in Fig. 4-3 (b), will be used to illustrate the link between the Mallard and LeChatelier approach and the present model. Taking this approximation, one arrives at:

\[ V_{EC} = \sqrt{\frac{\lambda}{\rho c_p}} \frac{T_b-T_u}{Ti-Tu} = \sqrt{\frac{\lambda}{\rho c_p}} \frac{T_b-T_u}{Ti-Tu} \dot{\omega} \] (4-22)

Which in term of non-dimensional numbers becomes:

\[ \kappa_{EC}^2 = \mu_1 \] (4-23)
One of the main simplifications in the Mallard and LeChatelier approach consists in expressing the slope $\frac{dT}{dx}(x_{\text{ignition}})$ in a linear form as $\frac{T_{\text{ad}} - T_i}{\delta}$. This approximation may seem accurate enough for flames happening in a thin sheet region (small $\delta$) happening close to the adiabatic temperature, i.e. for flames with high activation energies.

Since the present model gives the flame speed as an exact explicit solution to the conservation of energy across the whole flame, it is clear that the role of the Lambert inverse function is to exactly track the slope $\frac{dT}{dx}(x_{\text{ignition}})$ as is shown on Fig. 4-3 (c).

One can in fact examine how the Mallard and LeChatelier approximation, based on no actual conservation equation but only on heat transfer across the flame, can be reconciled with an energy conservation formulation. Fig. 4-4 shows the deviations for different locations of the ignition temperature. In the limit, where $T_i = T_{\text{ad}}$, none of the formulations leads to propagation ($V = 0$). But as soon as $T_i < T_{\text{ad}}$, the Mallard and LeChatelier underestimates the flame speed. As Fig. 4-4. shows, the relative error instantaneously jumps to over 29% and only very slowly decreases as the ignition temperature slides towards $T_u$. This variation is in many ways analogous to lowering the effective activation energy of the reaction. The slope at the point of ignition slowly approaches $\frac{T_i - T_u}{\delta}$ as $T_i$ approaches $T_u$.

The interesting observation is that beside the errors due to its simplified formulation, the Mallard and LeChatelier approximation contains an error of almost 30% with respect to the step-ignition kinetics in which it is defined. This error is at its highest the closer the ignition temperature is to the adiabatic one, i.e. at high activation energies. Thus, even in cases where the ignition temperature can be invoked, Mallard and LeChatelier is not an accurate solution and should be avoided.
Figure 4-4: Comparison between the Mallard and LeChatelier flame speed expression and the exact solution and corresponding percentage error.

One can also use a Mallard and LeChatelier-like analysis to interpret the results of the present model. From the previous discussion, it is possible to think of the non-dimensional number $\mu_1 = \frac{\theta_{ad}^{-1}}{\theta_{t_1}^{-1}}$ as the magnitude of that heat transfer. The higher the value of $\mu_1$, the higher the difference between the ignition and the final, adiabatic, temperature, meaning that a lower proportion of heat is used to preheat the flame. This allows the flame to ‘start’ earlier and, consequently, to propagate faster. The flame speed is therefore a measure of the rate of energy the reaction has to transfer to its preheat zone. The lower this energy proportion, the higher the propagation speed.
4.4.1.2 Non-adiabatic case ($\eta \neq 0$)

The extra equation in the non-adiabatic case is slightly more complicated than its adiabatic counterpart:

$$(ar_{2} + 2\eta)(\theta_{i_{1}} - 1) = S_{1}(1 - e_{2}^{-r_{2}})$$  \hspace{1cm} (4-24)

As has been shown [68], the introduction of heat losses introduces the existence of multiple solutions to the flame speed. In the case of a simple one-front flame, there are two solutions, one physical and one unphysical. This is nicely captured by the inverse Lambert function $W(x)$ which is double-valued in the real space, with branches $W_{-1}(x)$ and $W_{0}(x)$. Its use leads again to an exact explicit formulation of the flame speeds $\kappa_{-1}$ and $\kappa_{0}$:

$$\kappa_{-1,0} = \frac{\beta_{-1,0}^{2}}{a\beta_{-1,0} + \eta} \quad \text{where} \quad \beta_{-1,0} = W_{-1,0}[-\mu_{1}e^{2\eta a}/a] + \mu_{1} - \frac{2\eta}{a}$$

with $\mu_{1} = \frac{\theta_{ad_{1}} - 1}{\theta_{i_{1}} - 1}$  \hspace{1cm} (4-25)

The curves corresponding to this case are shown on Fig. 4-5. The physical and unphysical nature of $\kappa_{0}$ and $\kappa_{-1}$ stems from how they change with increasing concentration $\phi$ of the non-reacting component. Similar to the adiabatic case, increasing $\phi$ leads to an increase of the heat capacity of the mixture. The mixture consumes more energy before it reaches ignition temperature. This weakens the flame and reduces the flame speed. This is captured by $\kappa_{0}$. On the other hand, $\kappa_{-1}$ shows that the flame is enhanced by the addition of inert material – a clearly unphysical behavior. Both curves meet and merge at a bifurcation point. For higher values of $\phi$, no solution exists – the flame does not propagate at all. This transition from 2 flame speeds to one and then to none is commonly known as flame quenching. This happens at a finite speed $\kappa_{q1}$ and concentration $\phi_{q1}$:

$$\kappa_{q1} = \frac{\beta_{q}^{2}}{a\beta_{q} + \eta} \quad \text{where} \quad \beta_{q} = \frac{S_{1}}{a(\theta_{i_{1}} - 1)} - \frac{2\eta}{a} - 1$$  \hspace{1cm} (4-26)
\[
\phi_{q1} = \frac{1}{\nu} \left[ \frac{2\eta(\theta_{i_1} - 1) - S_1}{(\theta_{i_1} - 1)w_1} \left[ \frac{2\eta(\theta_{i_1} - 1) - S_1}{S_1\epsilon} \right] - 1 \right]
\]

(4.27)

As Fig. 4-5 shows, in the adiabatic case, as \( \phi \) is raised, the flame slows down until it stops. In the non-adiabatic case \( (\eta \neq 0) \), this weakening of the flame is intensified by the heat losses. Not only is the heat produced by the flame used to raise the temperature of the mixture, but it is also dissipated to the environment. This means that for a given value of \( \eta \), the heat loss term, the flame starts already at a lower velocity at \( \phi = 0 \) than its adiabatic counterpart. As \( \phi \) increases, the flame slows down faster. A key consequence of the presence of heat losses is that the flame is so much weakened that it even cannot slow down past a certain value, the speed at quenching. For any lower propagation speed, the flame simply disintegrates. As the heat losses intensify, i.e. \( \eta \) increases, a lower concentration \( \phi_{q1} \) is sufficient to quench the flame while the corresponding minimum propagation speed \( \kappa_{q1} \) increases. At a sufficiently high \( \eta \), this one-front flame is simply not able to exist at all, even for \( \phi = 0 \).

The line connecting the quenching points for different values of \( \eta \) is called the quenching curve and is shown on Fig. 4-5.
Figure 4-5: Flame Speed Curve for Non-Adiabatic One-Front Flames for Different Values of $\eta$.

The temperature profile corresponding to this non-adiabatic one-front case is shown on Fig. 4-1. The heat losses prevent the flame from reaching the adiabatic temperature. It only achieves a maximum value within the front and then decreases as the post-flame zone is subjected to heat losses. The expression for the maximum temperature is:

$$\theta_{max_1} = 1 + \frac{s_1}{\eta} - \frac{s_1}{\eta} \left[ 1 - \frac{(\theta_{1_1} - 1)(2\eta + ar_2)}{s_1} \right]^{\frac{\eta}{2\eta + ar_2}}$$

$$\kappa_{max_1} = \frac{1}{r_2 - r_1} \ln \left[ 1 - \frac{(2\eta + ar_2)(\theta_{1_1} - 1)}{s_1} \right]$$  \hspace{1cm} (4-28)

One can clearly see that, as mentioned in the literature review, the final temperature becomes dependent on the flame speed $\kappa$, which is an indication of the multiplicity of solutions in the presence of heat losses.

Finally, in the context of the Mallard and LeChatelier analysis, the expression for the flame speed (equation 4-22) shows that the propagation velocity is again a consequence of the heat transfer
between the front and the preheat zone, expressed by the ratio \( \mu_i = \frac{\theta_{ad_i}}{\theta_{ig_i}} \) of the adiabatic temperature with respect to the ignition temperature, but in the non-adiabatic case, this key number becomes \( \mu_{\eta_i} = \mu_i - \frac{2\eta}{a} \) directly showing the weakening effect of the heat losses \( \eta \).

4.4.2 Two-front flame

4.4.2.1 Analytical expressions

Let us now turn to the more complex case of two-front flames. Here, the main assumption is that the second fuel fraction, represented by the concentration term \( \phi \), is now reactive and will participate in the heat production. Its reaction is brought about whenever the temperature reaches the second ignition temperature \( \theta_{ig_2} \). The key difference, caused by the presence of the second front, is that each flame system has one more degree of freedom and is now characterized by two equations and two corresponding eigenvalues: the flame speed \( \kappa \) and the distance between the onset of each reaction \( Z \).

In total, one obtains four different systems which are adiabatic \((\eta = 0)\) or non-adiabatic \((\eta \neq 0)\) and where the flame fronts are clearly separated \((Z > 1)\) or overlapped \((Z < 1)\).

The systems of equations defining each system are tabulated below:

<table>
<thead>
<tr>
<th>( \eta = 0 )</th>
<th>( Z &gt; 1 )</th>
<th>( Z &lt; 1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \kappa^2 a^2 (\theta_{i_1} - 1) + S_1 (e^{-\kappa^2 a Z} + \kappa^2 a S_1) = \kappa^2 a^2 (\theta_{i_2} - 1) )</td>
<td>( \kappa^2 a^2 (\theta_{i_1} - 1) + S_1 (e^{-\kappa^2 a - 1} + S_2 e^{-\kappa^2 a Z} - 1) = 0 )</td>
<td>( \kappa^2 a^2 (\theta_{i_1} - 1) + S_1 (e^{-\kappa^2 a - 1} + S_2 e^{-\kappa^2 a Z} - 1) = 0 )</td>
</tr>
<tr>
<td>( \kappa^2 a^2 (\theta_{i_1} - 1) + S_1 (e^{-\kappa^2 a - 1} + S_2 e^{-\kappa^2 a Z} - 1) = 0 )</td>
<td>( \kappa^2 a^2 (\theta_{i_1} - 1) + S_1 (e^{-\kappa^2 a - 1} + S_2 e^{-\kappa^2 a Z} - 1) = 0 )</td>
<td>( \kappa^2 a^2 (\theta_{i_1} - 1) + S_1 (e^{-\kappa^2 a - 1} + S_2 e^{-\kappa^2 a Z} - 1) = 0 )</td>
</tr>
</tbody>
</table>

Table 4-3: Systems of algebraic equations for two-front flame configurations
With some more algebra, one can easily verify that the expression for separated fronts ($Z > 1$) and for the overlapped fronts ($Z < 1$) become equal when $Z = 1$. Moreover, one can verify that for the limit of $\eta \to 0$, the non-adiabatic expressions become the same as the expressions for $\eta = 0$.

For each of the four cases, one can use the second equation to obtain an explicit expression for the flame separation $Z$ in terms of the burning velocity $\kappa$. From table 4-3., it is clear that equations $E_2$ and $E_4$, as well as $E_6$ and $E_8$ are the same. This yields one expression for $Z$ in the adiabatic case and one in the non-adiabatic case:

$$\eta = 0 \quad Z = \frac{1}{\kappa^2 a} \ln \left[ \frac{S_2(1-e^{-\kappa^2 a F})}{\kappa^2 a^2 (\theta_{11}-1) - S_1(1-e^{-\kappa^2 a})} \right]$$  \hspace{1cm} (4-29)

$$\eta \neq 0 \quad Z = \frac{1}{r_2} \ln \left[ \frac{S_2(1-e^{-r_2 F})}{(2\eta + ar_2)(\theta_{11}-1) - S_1(1-e^{-r_2})} \right]$$  \hspace{1cm} (4-30)

For all the systems just described, the problem has been reduced from a differential equation to a set of highly non-linear transcendental algebraic equations. Even though these are impossible to recast in an explicit form for the velocity, with the exception of system 1 (as we shall see in the next section), this change of the nature of the problem is of an important consequence. The transition from differential to purely algebraic relations means that it is possible to obtain all the roots of the equations with certainty, albeit through numerical root finding algorithms. This is one of the main strengths of the step-ignition formulation mentioned previously [40].

With some extra analysis, one may even show that the range of permissible values of $\kappa$ is bounded, for all systems, by a maximum value given by the domain of definition of the expression for $Z$. This rather lengthy analysis is avoided here for sake of clarity and is summarily shown in the appendix.

The next section considers the only case of two-front systems for which the flame speed can be expressed explicitly. This, in turn, gives an interesting physical insight into the communication between two separated fronts.

4.4.2.2 Adiabatic case with separated fronts

The simplest case of the two-front configurations is the adiabatic system $E_1$ and $E_2$ of two separated fronts (Fig. 4-1. b). One can use the expression for $Z$ (equation (4-29)) and plug it back
into equation $E_1$. Then, using again the Lambert inverse function, one obtains the following explicit formula for the flame speed:

$$
\kappa_2 = \sqrt{\frac{1}{a F} W_0 \left(-\frac{1}{a} \mu_2 e^{-\frac{1}{a \mu_2}}\right) + \frac{1}{a^2 F} \mu_2} \quad \text{where} \quad \mu_2 = \frac{\theta_{ad_2} - \theta_{ad_1}}{\theta_{i_2} - \theta_{ad_1}} \quad (4-31)
$$

Here, the adiabatic flame temperature of the two-front case, $\theta_{ad_2}$, is:

$$
\theta_{ad_2} = 1 + \frac{S_2}{a} + \frac{S_2 F}{a} \quad (4-32)
$$

The curve corresponding to this expression is shown on fig. 4-2. The most striking observation is that it seems to stem directly from the adiabatic one-front case curve at $(\phi_{2c}, \kappa_{2c})$. This is no coincidence, as this results directly from the analysis of the domain of definition of the expression for $Z$ (equation 4-29). There, the lower limit for the value of $\kappa$ is given by the limit of the expression in the logarithm. One may show that, for $Z$ to be real, the value of $\kappa$ has to be higher than $\kappa_1$, precisely the value of propagation velocity of the adiabatic one-front case. If one were to follow solely the curve of equation (4-31), without considering the value of $Z$, one would get the red dotted curve shown on Fig. 4-2. The dotted part corresponds to the case for which $Z$ is a complex number. As soon as the dotted line crosses the one of $\kappa_1$, $Z$ becomes real and very quickly, almost exponentially, decreases from $+\infty$ to 1 as $\phi$ and consequently $\kappa_2$ increases. The crossing happens at $\phi_2$ for which no explicit solution has been found.

Physically, one may give the following interpretation. Here, the second fuel fraction (fraction 2), represented by the concentration $\phi$, is assumed reactive. At very low concentrations ($0 < \phi < \phi_2$), there is not enough fraction 2 to form a front, i.e. to release enough heat to reach $\theta_{i_2}$. But, at the critical concentration $\phi_{2c}$, there is enough of fraction 2 to reach that potential and a steady state 2nd front may form in an infinitely distant wake of the first one. As the concentration is further raised, the speed of the 2nd front rapidly increases and this front quickly approaches the first one up to the point where they become contiguous ($Z = 1$).

One may again look at the flame speed expression for $\kappa_2$ (equation (4-31)) in terms of heat transfer, similar to the Mallard and LeChatelier formulation. The expression for $\kappa_2$ is analogous to the adiabatic one-front case, $\kappa_1$, but here, the non-dimensional number $\mu_1 = \frac{\theta_{ad_1} - 1}{\theta_{i_1} - 1}$ has been replaced
by $\mu_2 = \frac{\theta_{ad_2} - \theta_{ad_1}}{\theta_{i_2} - \theta_{ad_1}}$ and the expression for $\kappa_2$ is also scaled by the relative thickness of the second front $F$.

In the one-front case, the term $\mu_1$ originated from the balance between the rate of heat transfer from the front to the preheat zone and the rate of heat needed to bring the mixture from the initial temperature $\theta_u = 1$ to the ignition temperature $\theta_{i_1}$. As Fig. 4-6. shows, this approach can be analogously applied on $\mu_2$. As the form of the expression is almost exactly the same for $\kappa_1$ as for $\kappa_2$, one can simply replace the roles of $\theta_u = 1$ by $\theta_{ad_1}$, $\theta_{i_1}$ by $\theta_{i_2}$, and $\theta_{ad_1}$ by $\theta_{ad_2}$ respectively. In this light, one may say that the expression for $\kappa_2$ is a balance between the rate of heat transfer from the second front to its preheat zone and the rate of heat required to bring the mixture from the adiabatic temperature of the one-front flame to the ignition temperature of the second front, i.e. the extra amount of heat added by the reaction of the second front. Raising the concentration $\phi$ leads to an increase in the reaction term $S_2$ and consequently to a higher $\theta_{ad_2}$ and $\mu_2$. The second front then provides more heat to its own preheat zone, making it easier for the temperature to reach $\theta_{i_2}$, and thus advances relatively closer towards the first front. This accrued rate of heat production also has an overall enhancing effect on the whole flame, which then accelerates.
4.4.2.3 Adiabatic case with overlapped fronts

The two adiabatic fronts can also be considered in an overlapped configuration. This corresponds to the system of equations $E_3$ and $E_4$ in table 4-3. As mentioned before, even though $E_4$ yields an explicit expression of $Z$ in terms of $\kappa$, no way has been found to isolate $\kappa$ and to express it in terms of the other variables. Therefore, the roots of the present system have been found by means of root-finding algorithms on MATLAB (using the function fzero). Analyzing the expression for $Z$, given by equation (4-29), it is possible to show that the admissible values of $\kappa$ are bounded both below and above. This overlapped system requires that $1 < Z < 0$, which yields both the upper and lower values of the range over which the roots are to be found.

The root finding algorithm yielded the curve shown on Fig. 4-2. The curve is directly continuous with the one obtained in the previous section for the overlapped case, which is not a surprise, as it was shown before that at $Z = 1$, both the overlapped and separated systems became the same set of equations.

Physically, one may continue the explanation from the previous section. After the formation of the second front at a critical concentration $\phi_2$, the distance between the fronts promptly diminishes up to the moment when the fronts become contiguous ($Z = 1$) at $(\phi_{Z=1}, \kappa_{Z=1})$. Increasing the concentration further leads to the second front overlapping with the first one. As the concentration is increased even further, the second front advances more into the first one ($Z$ decreases even more) and the flame speed increases even though its acceleration decreases.

A side-note can be also made by recasting equation $E_3$ in terms of non-dimensional numbers from Fig. 4-6.

Here, it is interesting to note that the solution to the flame speed of the adiabatic merged fronts system depends on the relative values of $\mu_1$ and $\mu_4$, the ratios of $T_{ad_1} - T_u$ and $T_{i2} - T_u$ to $T_{i2} - T_u$. This shall be further discussed in the next section.
4.4.2.4 Interpretation of Z

Another side-note that will be of importance later on is the expression of $Z$ in terms of non-dimensional numbers from Fig. 4-6:

$$Z = \frac{1}{\alpha^2} \ln \left[ \frac{1}{F} \frac{(\mu_3 - \mu_1)(1-e^{-\kappa^2 a F})}{\kappa^2 a^2 - \mu_1 (1-e^{-\kappa^2 a})} \right]$$

(4-34)

Here, one can notice that the separation between the fronts, $Z$, depends on the non-dimensional numbers $\mu_1$ and $\mu_3$, relating, respectively, the adiabatic temperature of the first front $T_{ad_1}$ and the adiabatic temperature of the combined fronts, $T_{ad_2}$, to the ignition temperature of the first front $T_{ig_1}$.

The logarithmic expression for $Z$ has been observed in previous studies [46], [58], [59] pertaining to both Arrhenius-type and ignition kinetics. In order for $Z$ to be real and positive (as the formulation requires), the expression within the logarithm has to be greater than 1. This means that:

$$\frac{1}{F} \frac{(\mu_3 - \mu_1)(1-e^{-\kappa^2 a F})}{\kappa^2 a^2 - \mu_1 (1-e^{-\kappa^2 a})} > 1$$

(4-35)

It can be noted that the denominator of this equation is nothing else than the expression contained in equation (4-13), used to find the speed of the adiabatic one-front flame. Equating it to 0 leads to the one-front flame solution obtained in equation (4-14). This is another proof of the self-consistency of the model, which predicts that for a value of $Z \to \infty$, the flame speed is that of a one-front flame (a fully separated regime).

Equation (4-35) states that for a separation between the fronts $Z$ to exist, one must have:

$$\frac{(\mu_3 - \mu_1)(1-e^{-\kappa^2 a F})}{F} > \kappa^2 a^2 - \mu_1 (1-e^{-\kappa^2 a})$$

(4-36)

i.e., that, for a given width of the 2nd front, $F$, the difference $T_{ad_2} - T_{ad_1}$, represented by $\mu_3 - \mu_1$ must be high enough with respect to the kinetics of the first reaction alone for the second front to exist.
4.4.2.5 Non-adiabatic cases

Finally, one can add heat losses to the two-front systems. Similar to the one-front case, this change leads to the existence of multiple solutions for the combination of $\kappa$ and $Z$. Here, the systems of equations $E_5$-$E_6$ and $E_7$-$E_8$ are even more complex than their adiabatic counterparts. As discussed previously, both $E_6$ and $E_8$ lead to the same explicit expression for $Z$. One then employs root finding methods to obtain all the velocities for every concentration $\phi$. The following section is applicable to heat loss values $\eta > \eta_{critical}$, where the critical value is fairly low. The case of $\eta < \eta_{critical}$ will be covered in the next section.

The corresponding curves are shown on Fig. 4-7. One can again observe the existence of two flame speeds for a given concentration. Here though, the behavior is precisely opposite to the one-front case. There, fraction 2 was assumed not to react, but in the two-front case it actively contributes to the heat produced by the second front. It is easier to picture the behavior of the flame as one starts at high concentrations $\phi$ and observes what happens as this concentration is lowered. Of the two possible flame speeds of curve $C_1$, one increases and the other one decreases until both curves meet at a critical concentration $\phi_{q2}$. At lower concentrations, there is not enough of fraction 2 to form a second front and the flame cannot exist as a two-front structure for the given value of heat losses $\eta$. Out of these speeds, the upper curve can again be thought of as physical and the lower one as unphysical. This stems from the fact that, in this case, decreasing $\phi$ leads to lowering the reaction rate, the maximum temperature and consequently the flame speed of the mixture. The increase in flame speed with reducing $\phi$, displayed by the lower curve, is an unphysical behavior. The point of encounter of both curves represents again quenching, induced by the combination of heat losses and too low a concentration $\phi$. As the value of $\eta$ is increased, higher concentrations $\phi_{q2}$ are necessary for the flame to even exist and the point of quenching also occurs at increasing finite flame speeds $\kappa_{q2}$. The flame speed curves are therefore observed to ‘retreat’ towards the right in the same way the one-front ones retreated towards the left with increasing heat losses. One can again trace a curve, starting at $\phi_2$, the point where the adiabatic two-front flame first appeared, and link all the quenching points. This is the so-called quenching curve for the two-front case.
Coupled to this is another important feature, the transition between separated to overlapped fronts. One can see that for a given heat loss term \( \eta \), the folded curve \( C_1 \) one obtains over \( \phi \) is separated into two regions. For lower flame speeds, the system is defined by separated fronts. Then, above a flame speed \( \kappa_{Z=1} \), happening when \( Z = 1 \) at \( \phi_{Z=1} \), the flame transitions into two overlapped fronts.

Plotting a series of curves for increasing heat loss terms \( \eta \) (Fig. 4-7) reveals that the points \( (\phi_{Z=1}, \kappa_{Z=1}) \) form another curve. This one originates at the point where \( Z = 1 \) in the adiabatic two-front flame case. As the value of \( \eta \) is increased, \( \phi_{Z=1} \) increases while \( \kappa_{Z=1} \) decreases. This leads to a very important point where this curve of \( Z = 1 \) crosses the quenching curve, denoted by \( (\phi_{qZ}, \kappa_{qZ}) \).

For \( \phi_{qZ} < \phi_C \), the fold, i.e. quenching, happens while the flame exists as two separated fronts, while for \( \phi_{qZ} > \phi_C \), the flame quenches as two overlapped fronts.

Figure 4-7: Flame speed curves of non-adiabatic two-front flames (above \( \eta_{critical} \)).
4.4.2.6 Existence of multiple propagation speeds

The general solution of a non-adiabatic two-front system, discussed above, in which the flame speed was simply described by a two-branched curve with a fold bifurcation at the quenching point, is applicable heat loss terms $\eta > \eta_{\text{critical}}$. However, for smaller heat losses, i.e. $\eta < \eta_{\text{critical}}$, a more complex behavior has been observed. This complexity emanates from the study of the expression for $Z$ (equation (4-30)). By performing the same analysis on $Z$ as in the adiabatic two-front cases, one can come up with the domain of definition of $Z$ in terms of $\kappa$. In previous sections, it was found that this expression was bounded above and below, restricting the range in which the roots of the system of algebraic equations are to be found. But, below $\eta_{\text{critical}}$, a region where $Z$ ceases to be real develops within this range. The domain of definition of $Z$ becomes disjoint, leaving out a region $[\kappa_-; \kappa_+]$. Within this region, $Z$ is complex.

In fact, the values of $\kappa_-$ and $\kappa_+$ correspond exactly to the values of $\kappa_{-1}$ and $\kappa_0$ (equation (4-25)), i.e. the flame speeds of the non-adiabatic one front configuration. Therefore, it appears the one-front case is a natural limit of the two-front separated configuration in both the adiabatic and the non-adiabatic case. Fig. 4-8 illustrates dynamically how the flame speed curves of the one-front and two-front configurations are related for increasing values of heat losses. One starts with the adiabatic system (Fig 4-8 a.), described earlier in Fig. 4-5. For a very low value of $\eta$ (Fig. 4-8 b.), the one-front double-branched curve is perfectly wedged into the two-front flame speed. The latter is actually composed of a total of 3 folds wrapped around the one-front curve. As the heat losses are increased (Fig. 4-8 c.), the one-front curve retreats towards the left, in a behavior described earlier in section 4.a.ii. The two-front curve maintains its grip over it and the 3 folds begin to approach each other. This continues for increasing $\eta$ up until the quenching point (the fold) of one-front case reaches a line connecting the uppermost and the lowest fold of the two-front curve (Fig. 4.8 d.). For even higher $\eta$, the curves separate, each retreating into an opposite direction. Now far from the disturbance caused by the one-front case, the folds of the two-front curve coalesce together into a single fold, described in the previous section (Fig. 4-8 e.). A further increase in heat losses leads to the behavior described in the previous section, where the flame quenches as two separated fronts and (Fig. 4-8 f. to h.) and then as two overlapped fronts (Fig. 4-8 g.). The previously described behavior points out to the existence of more than two
solutions for some regions of parameters for this non-adiabatic two-front step-ignition configuration. In fact, for example in Fig. 4-8 b., when the concentration is at values between the lowest and the middle fold, there are three different steady-state flame speeds which may describe the separated front system and four, if one adds the solution represented by the overlapped configuration. One may actually directly study the different flame configurations by means of the different values of Z corresponding to each of these four branches of the two-front flame speed curve.

Figure 4-8: Flame speed curves for two separated front flames for $\eta < \eta_{critical}$ showing the possible existence of up to 4 solutions for some concentration ranges.

Fig. 4-9 shows how the flame configuration changes with the different values of flame speed. The three-dimensional curve illustrates the relation between the flame speed $\kappa$, the concentration $\phi$, and the distance between the onsets of reaction $Z$, and is plotted against its two-dimensional shadows on the $\kappa - \phi$, $\kappa - Z$, and $Z - \phi$ planes. Plane $\kappa - \phi$ corresponds to figure 4-8 c. Points 1 to 9 describe different positions on the curve. The figure also displays the temperature profiles corresponding to each point.
Starting at point 1, the fronts are contiguous \((Z = 1)\). As one follows the first branch, the concentration is decreased and, in accordance with the previous discussion, the fronts get further apart \((Z \text{ increases})\). Then, the first fold is reached (between 2 and 3). Continuing along the second branch, the concentration is raised with the fronts moving very far apart (point 4). The ‘flame’ stretches and allows for a large dip in temperature between both fronts. One then reaches the second fold and continues along the third branch, decreasing the concentration again. The second front approaches the first, ‘filling up’ the temperature dip between them (points 5 to 7). Finally, after the 3 fold is crossed, the concentration is raised again and the fronts move apart again, but now with no noticeable temperature dip.
The different configurations observed above witness how the system operates with the different degrees of freedom induced by the presence of heat losses. Also, it is noteworthy that the temperature profiles on branches 2 and 3, where the two-front solution follows the one-front one, are very similar to the one-front case: the temperature drops behind the first front into the temperature dip described above, as it would in the one-front case, but right before the second front, it grows again very fast. These temperature profiles illustrate the different equilibria that the flame may reach depending on the external conditions. A logical question concerns the stability of each equilibrium. A simple numerical assessment, based on unsteady formulations, presented in the appendix, has been attempted but, mainly due to the difficulties linked to the numerical representation of with step functions [44], it did not yield any conclusive results.

It appears that these different profiles may be nothing else than a curiosity induced by the specific formulation of the present model, a mathematical artefact with no counterpart in real life, and thus worth mentioning only in the appendix, were it not for the crucial role of the three-fold curve in explaining the experiments. As will be shown in the next section, the existence of this four-solution region is imperative to explain the transition from a bright flame in the tube to a dimmer one in the narrow channels. This can happen only in regions where the $\phi_{q1} > \phi_{q2}$, i.e. the region of the triple-fold discussed above.
5 COMPARISON BETWEEN EXPERIMENTS AND MODEL

The goal of this section is to reconcile the experimental tests with the results of the model. As explained in the model section, the nature of this comparison is purely qualitative. The numerous assumptions and simplifications of the model make it stray relatively far from actual flame speed values. Nevertheless, the same formulation frees the model from many constraints and dependencies that prohibit analytical solutions while keeping the general behavior. This will be attested by the qualitative agreement between the analytical and experimental findings as well as by a comparison with an Arrhenius approach to two-front flames. The agreement between the two mathematical formulations will permit a more general interpretation of how and when two fronts have a tendency to form and under what conditions they merge or stabilize within a finite distance of each other.

5.1 From flame speed to quenching distances

5.1.1 Overall flame speed graph

In the previous section, flame speed results for every configuration were each accompanied by a small paragraph with an appropriate physical interpretation. Now, this information will be assembled into a coherent explanation of how the flame behaves as:

- the concentration is raised for an adiabatic flame
- the concentration is set to a certain value and heat losses are increased

These changes are both indicated by paths on the flame speed versus concentration map on Fig. 5-1 a. The variation of concentration corresponds to path 1-2-3-4. The subsequent changes in heat losses are shown for two different values of concentration, path a-b-c-d for a lower and path e-f-g for a higher concentration.

Path 1-2-3-4 corresponds to the behavior of an adiabatic flame, i.e. a flame where the effect of heat losses is relatively small on the propagation and, for all practical purposes, may be neglected. In terms of experiments, this corresponds to the propagation of the flame in the main tube where the inner diameter of the tube (48 mm) is an order of magnitude higher than the typical front thickness of a flame in a metal suspension [45]. Therefore, going from 1 to 2 to 3 to 4 corresponds to
performing consecutive flame propagation measurements for increasing powder loadings of the flame in a tube with no plate assemblies. At a critical concentration of the 2nd fraction, the 2nd flame front forms and couples to the primary front.

![Quenching curves and corresponding quenching map](image)

**Figure 5-1**: a. Quenching curves obtained in section 4 with paths corresponding to experiments and b. corresponding quenching map.

Fig. 5-1 a. shows the quenching curves derived in the previous section for all flame configurations. There, one can divide the graph into regions where a certain type of flame is defined, i.e. each flame configuration can exist only within a specific combinations of heat losses and concentrations. So, for a given mixture composition and a channel width, one may predict whether the flame will propagate or not and describe its structure.

So, as indicated by the adiabatic path 1-2-3-4 on fig. 5-1 a., at $\phi = 0$, a one-front flame (from the reaction of the 1st fraction) exists. As the concentration is raised (1-2), the 2nd fraction increases the heat capacity of the mixture and forces the flame speed to decrease. For these low concentrations, the flame can have only one front. Above the critical value of $\phi = \phi_2$, the flame can exist in two forms:

- Either the 2nd fraction is inert (as in the case of SiC tests) and the flame still exists as a one front that needs to heat a mixture with an increasing heat capacity, so both its adiabatic temperature and flame speed decrease with concentration, until the flame stops propagating.
- Or, the 2nd fraction acts as a fuel and can form its own front separated from the first one.

In this case, raising the concentration leads to a decrease in the value of \( Z \), i.e. the fronts approach, and the speed increases (2-3). At a particular concentration, the 2nd front reaches the 1st one (\( Z = 1 \)). A further increase leads to the merging of the fronts (3-4) and an increase of the flame speed.

On the other hand, paths a-b-c-d and e-f-g correspond to fixing the powder flow rate and performing tests with decreasing channel widths (increasing number of plates) in quenching assemblies. For each path, the concentration is set to a constant value \( \phi_A \) and \( \phi_B \). In the first case, the adiabatic flame is composed of two separated fronts (point a). As the heat losses are increased (a-b), the thermal communication between the fronts is weakened, the flame speed decreases and the fronts move further apart, i.e. \( Z \) increases, until the second front completely vanishes (b). At this point, the overall energy of the flame suddenly drops, which is manifested by a sharp drop in flame speed and maximum temperature (b-c). Further increases in heat losses (c-d) lead to the quenching of this primary flame (d). The second example, e-f-g, shows a flame at higher concentrations of the second fuel, where two merged fronts exist in the adiabatic case (e). Increasing the heat losses (e-f) leads to the decrease in flame speed and to a gradual increase in \( Z \) up to the point where \( Z = 1 \) (f). At this point, a further increase in heat losses (f-g) leads to the unmerging and separation of the fronts, and later, to the quenching of the merged flames (g). At this concentration, the heat of the second front is vital to the propagation of the whole flame. A single one front seeded with so much unignited powder, cannot propagate and the entire flame structure quenches in unison.

5.1.2 Quenching distance map

The flame speed graph from Fig. 5-1 a. can be modified and recast into a form analogous to the experimental results. These show (Fig. 5-1 b.) the flame behavior for different channel widths \( d \) versus aluminum concentration. As previously discussed in section 4, the latter can be represented by the concentration term \( \phi \). According to the model, by far the main effect of the channel width on the flame is through the heat loss term \( \eta \). All other possible ways, including the boundary layer
growth or chemical species quenching effects have been neglected. From [45], the heat loss term is:

\[ \eta = \frac{h c_e}{c_1 \rho_1} \]  

(5-1)

From [45] and [93], the heat transfer coefficient \( h \), corresponding to conduction losses to the wall is proportional to \( 1/d^2 \). So:

\[ \eta = A_\eta \frac{1}{d^2} \rightarrow d = \frac{A_\eta}{\sqrt{\eta}} \]  

(5-2)

Where \( A_\eta \) is a constant.

This relation allows one to replot the main curves, delimiting the regions of Fig. 5-1 a., on a graph of \( d \) vs. \( \phi \). This is shown on Fig. 5-1 b and Fig. 5-2 a.

![Figure 5-2: Comparison between experiments (a.) and model (b.) on a same type of quenching map.](image)

Fig. 5-1 b. reproduces all regions of definition of the different flame configurations. The strip region on the top of the graph shows the case where \( d \to \infty \), i.e. where \( \eta \to 0 \). It is the adiabatic case, in other words the case where the effect of losses to the walls is negligible. This corresponds to the flame configuration before it enters the narrow channels. The said region reproduces the
sudden appearance of the second front at a critical concentration \( \phi_2 \) as well as the front merging at \( \phi_{Z=1} \).

One can also recover the quenching behavior. The zone of existence of each configuration is bounded below by its quenching curve. For lower channel widths, the heat loss term \( \eta \) becomes too substantial and the flame cannot propagate.

Fig. 5-2 shows the similarity in the quenching graphs predicted by the model and obtained in the experiments, as discussed in the next section.

5.2 Interpretation of experimental results through model

5.2.1 Agreement between experiments and model

As previous work [29], [31], [82] as well as the current one [32] has indicated, the difference in brightness in the flames has been explained through the relative participation of aluminum particles on the overall combustion. On the one side, the dim orange flame, so similar to the SiC cases, indicates, that the overwhelming majority of particles undergo a slow oxidation and do not participate in the heat production. The flame can therefore be effectively thought of as a single methane-air front, seeded with either virtually inert particles or particles whose combustion time is much longer than the combustion time of the methane-air flame.

The sudden jump in the brightness at a critical temperature of the flame has been associated with the onset of reaction [38] of a metal flame front through the collective effect of individual particles, as explained in the concept review. The disappearance and reappearance of the bright flame as it enters and leaves the quenching channels can therefore be associated directly with the ability of the particles to burn in a front.

A clear qualitative correspondence between the two plots on Fig. 5-2 leads to a physical interpretation of the quenching behavior of the aluminum-methane-air flames in terms of the binary mixture of two reactive fractions. First, at low concentrations, only a methane-air flame, seeded with aluminum particles forms in the main tube and propagates through the narrow channels before it quenches. As more slowly reacting aluminum is added, its main effect is to raise the heat capacity of the mixture, thus lowering its flame speed and heat release, and making it
susceptible to quench at higher channel widths, similar to the inert SiC. In terms of combustion models, this can be thought of as the separation mode. The sporadic combustion of aluminum has virtually no effect on the methane-air front as it happens on a much larger timescale and is thus unable to stabilize any front even in the tube, where heat losses lead to a decrease in temperature after the methane-air front has passed.

At a critical concentration, about 320 g/m$^3$, the collective aluminum reaction is sufficient to form a front in the main tube. As the model shows, the two fronts are at first separated by some physical distance (this was directly observed in [82]) and it is relatively easy to cut away the secondary front through heat loss, corresponding to path a-d in Fig. 5-1 a. This is most analogous to the control mode. The aluminum front is not physically linked to the methane-air one, but its existence helps to counter-act the effects of increasing heat capacity and slow the quenching tendencies of the flame, leading to what can be approximated as an inflection point on the experimental quenching curve.

As was observed in Fig. 5-1 a., as the aluminum concentration further increases, the fronts in the main tube merge (3). The response of such a flame to heat losses is first a front separation, and then again a quenching of the second front, with the primary front still propagating up to an even higher heat loss value (path e-g in Fig. 5-1 a. This logic matches with the experimental observations of the sudden aluminum flame extinction at the entrance of narrow channels, the sole propagation of the methane flame between the parallel plates. As the primary methane flame emerges from the plates into the main tube, the decrease in heat loss allows the secondary front to reappear and to re-couple to the primary flame [32]. As the heat release from the aluminum reaction increases with powder concentration, the coupled flame improves its resistance to extinction and propagates through narrower channels. This trend is opposite to the behavior of a single methane-air flame loaded with higher levels of non-reacting particles, whose quenching distances keep increasing with concentration. At some concentration $\phi_Z=1$, the two quenching curves cross. For higher concentrations, only the combined effect of the two fronts can counter-act the heat losses and the high heat capacity of the mixture. This can obviously be associated with the merging mode where the aluminum front now fully dictates whether and how the flame will propagate.

At such high concentrations, however, the model must be considered with caution, as some of its initial assumptions, such as the fuel-lean nature of the mixture or the external ignition temperature
for each reaction, may no longer be reasonable approximations. Therefore, a more complex analysis should be developed to build upon the understanding developed in this work.

5.2.2 Role of the 4-solution region

In the previous section, it was noted that for heat losses lower than a critical value, the constant heat two-separated-front curves underwent three instead of one fold bifurcations. This, sometimes lead to an overall of three, and with the merged fronts four, flame speeds for a given combination of heat losses and concentration.

![Diagram](image)

Figure 5-3: Crossing of the quenching curves of the one- and two-front configurations and the existence of 4 solutions at low heat loss term $\eta$.

One of the interesting comments is that the crossing of the quenching curves, explaining the change of behavior within the quenching plates as the concentration is raised, happens at the heat loss value for which the concentrations $\phi_{q_1}$ (Fig. 4-5) and $\phi_{q_2}$ (Fig. 4-7) are equal. This crossing is
illustrated in Fig 5-3, where the heat has been raised from a low heat loss value ($\eta_I$) to a high one ($\eta_{II}$). In the low heat case, i.e., in a large channel, one can see that there is a range of concentrations $[\phi_{q2,I}, \phi_{q1,I}]$ for which the flame may propagate at up to four different speeds (Fig. 5-3 b.). As the channel width is narrowed down and the heat losses reach the value of $\eta_{II}$ (Fig. 5-3 c), both the flame speed curves for one- and two-front flames leave each other and stop touching (Fig. 5-3 c), i.e. there is no more possibility of 3 or 4 speed and, for a given concentration, the flame can propagate only at a stable (full lines in Fig. 5-3 c) or unstable velocity (dashed lines). An interesting observation is that in these regions, there is a range of concentrations $[\phi_{q1,II}, \phi_{q2,II}]$ for which the flame is not able to propagate at all. It would seem that in this range, the combined effect of too high a heat capacity and too low a heat production potential cannot withstand the effect of heat losses. For lower concentrations ($< \phi_{q1,II}$), the lower heat capacity allows the one-front flame to propagate, and for higher concentrations ($> \phi_{q2,II}$), the heat production is high enough to sustain a two-front flame despite the heat capacity and heat losses.

It would therefore appear that the appearance of 3 or 4 solutions to the flame propagation arises from the possibility of co-existence of one- and two-front flames for a given concentration. Such a co-existence is imaginable for example in unstable systems, where the flame may oscillate between a one-front and two-front structure, in a series of ‘ignitions’ and ‘extinctions’ of the second front. Paradoxically, the heat losses could play a stabilizing role, creating a concentration gap between the possibility of existence of one and two fronts.

5.3 Comparison with an Arrhenius Formulation

5.3.1 Arrhenius approximate analysis of front approaching

The experiments were interpreted by means of a simplified model with ignition temperatures and step kinetics. The behavior of the flame was explained in terms of parameters such as the ignition temperature, the heat release, etc. In this part, the goal is to assess how much generality this explanation holds. This can be achieved by seeing how a more Arrhenius approach can be used to explain the behaviors and how much these comparisons resemble the previous ones with a different model. If these are of the same nature, it is clear that the interpretation is of a more fundamental nature and it is not just a parameter fitting exercise.
As was stated in the literature review, one cannot obtain a fully analytic solution with an Arrhenius formulation. On the other hand, it may be helpful to perform an approximate analysis. In the case below, one can use the main parameters of an Arrhenius formulation and do a simple parametric study to assess what changes will bring two separated fronts together and lead to merging, a dependence not as clearly explained in previous literature.

One can start by considering a parallel non-competing system of reactions $R_1$ and $R_2$ similar to the ones developed in the analytical section:

\[
\begin{align*}
\frac{d^2 T}{dx^2} - U \frac{dT}{dx} + Q_1 \phi_1 + Q_2 \phi_2 &= 0 \\
\frac{d^2 a_1}{dx^2} - U \frac{da_1}{dx} - \phi_1 &= 0 \\
\frac{d^2 a_2}{dx^2} - U \frac{da_2}{dx} - \phi_2 &= 0 \\
\end{align*}
\]  

(5-3)

Where $U$ is the steady-state propagation of the overall flame, $a_1$ and $a_2$ the concentrations of each fuel (here, the oxidizer is assumed again in excess), $Q_1$ and $Q_2$ the heats of reaction of each front, $\phi_1$ and $\phi_2$ the two reaction rates (here with a simplest Arrhenius formulation), $k_1$ and $k_2$ the pre-exponential factors, and $E_{a_1}$ and $E_{a_2}$ the respective activation energies of $R_1$ and $R_2$. Such a system is shown on Fig. 5-4.
Figure 5-4: Schematic of the two-flame system considered in the approximate Arrhenius approach.

One can start at a moment when both of the fronts exist and are in a separation mode, \( R_2 \) lagging very far behind \( R_1 \). There is a large spatial separation around point \( x_a \) where, as [62] confirms, the second reaction has virtually not taken place yet, while the first one is practically finished. This means that the first reactant has basically fully reacted and the second reactant has almost not reacted at all:

\[
a_1(x_a) \approx 0 \quad \& \quad a_2(x_a) \approx 1
\]  

(5-4)

Also, the temperature is virtually equal to the adiabatic reaction of the first front:

\[
T \approx T_{ad_1} = T_u + Q_1/c
\]  

(5-5)

This happens on the temperature plateau between the first increase in \( T \) and the second one. Here the temperature slope is increasing very slightly and only from the second reaction as the first one is almost done.

The approach is to observe how the heat production changes with changes in temperature. As \( T \) rises, the reaction rate rises as well up to the point where suddenly the exponential dependence on \( T \) leads to the full onset of the second reaction. The overall rate of heat production from \( R_2 \), is:
\[ q_2 = Q_2 \Phi_2 = a_2 Q_2 k_2 e^{\frac{E_{a_2}}{RT}} \]  
(5-6)

And the change of \( q_2 \) with respect to temperature is:

\[ \frac{dq_2}{dT} = \frac{a_2 Q_2 k_2 E_{a_2}}{RT^2} e^{-\frac{E_{a_2}}{RT}} \approx \frac{Q_2 k_2 E_{a_2}}{RT^2} e^{-\frac{E_{a_2}}{RT}} \quad (\text{as } a_2 \approx 1) \]  
(5-7)

Moreover, knowing that \( T_{ad_1} = T_u + Q_1/c \), one can say that:

\[ \frac{dq_2}{dT}(T_{ad_1}) = \frac{Q_2 k_2 E_{a_2}}{R T_{ad_1}^2} e^{-\frac{E_{a_2}}{RT_{ad_1}}} = \frac{Q_2 k_2 E_{a_2}}{R (T_u+Q_1/c)^2} e^{-\frac{E_{a_2}}{R(T_u+Q_1/c)}} \]  
(5-8)

This gives a dependence of the readiness of the 2\textsuperscript{nd} reaction to happen in terms of external parameters. One can lump these into two independent parameters:

\[ \frac{dq_2}{dT}(T_{ad_1}) = \frac{Q_2 k_2}{T_u+Q_1/c} e^{-\frac{E_{a_2}}{R(T_u+Q_1/c)}} = \Omega_1 \Omega_2 e^{-\Omega_2} \]  
(5-9)

where \( \Omega_1 = \frac{Q_2 k_2}{T_u+Q_1} = \frac{Q_2 k_2}{T_{ad_1}} \) is the ratio of the heat transfer from the second front (where \( Q_2 \) is the amount of heat and \( k_2 \) is the speed at which it is produced) to the temperature of the first front. It can be thought of as the rate at which energy is transferred from the 2\textsuperscript{nd} front to the 1\textsuperscript{st} one relative to the heat produced by the first reaction.

And \( \Omega_2 = \frac{E_{a_2}}{R (T_u+Q_1)} = \frac{T_{a_2}}{T_{ad_1}} \) is the ratio now of the activation temperature of the second front \( T_{a_2} \) to the adiabatic temperature of the first front. It is the expression of the easiness with which the 2\textsuperscript{nd} reaction happens relative to the final conditions of the 1\textsuperscript{st} one, measuring how much more energy has to be added between the end of the first and the beginning of the 2\textsuperscript{nd} reaction.

Fig.5-5 shows an overall map of how \( \frac{dq_2}{dT}(T_{ad_1}) \) evolves with different values of \( \Omega_1 \) and \( \Omega_2 \). In order to achieve merging of separated fronts, one is looking for very fast increase in \( q_2 \). This clearly happens for a combination of high \( \Omega_1 \) and low \( \Omega_2 \). This is a combination of high heat transfer between the fronts and a low relative threshold to cross between the two fronts, a fairly obvious result. More interestingly, the expressions give values of \( Q_2, k_2, \text{ and } E_{a_2} \) which lead to merging, which will happen for higher \( \frac{dq_2}{dT}(T_{ad_1}) \). For low values, on the contrary, the second reaction at \( T_{ad_1} \) will basically produce only very small changes in \( q_2 \) and will drag for a long time.
before it can actually pick up. An interesting observation is that the relative separation between the fronts does not depend on the activation energy of the first front $E_{a_1}$. It essentially does not matter how promptly the first reaction happens as long as the thermodynamically determined $T_{ad_1}$ is the same. The importance lies on how much energy the second reaction has to deliver and how fast. If the reaction is hard to attain, with too high activation energies $E_{a_2}$ or if it does not high rates of energy, such as for example aluminum not burning in a front, the flames are well separated. But, as the reaction happens faster or delivers more energy at higher rates, the fronts approach almost exponentially fast as the sharp increase in $\frac{dq_2}{dT}(T_{ad_1})$ on Fig.5.5 indicates. At this point this analysis, based mainly on a large separation between the fronts cannot be used anymore. As intuition suggests, the large amount of heat now flowing towards $R_1$ start to influence its kinetics and one enters into a control and later into a merging regime.

![Diagram](image)

**Figure 5-5:** Change in $q_2$ for different values of $\Omega_1$ and $\Omega_2$ indicating for which reaction parameters both separated fronts have the tendency to merge.
5.3.2 Comparison with step model

When compared to the step function model, some analogy can be found between $\Omega_1$ and $\Omega_2$ and the non-dimensional numbers in the ignition-step formulation. One has to bear in mind that the formulations of these two cases are different and so are the forms of the exact or approximate flame speed solutions, but in both of these pairs, the non-dimensional numbers of the ignition-step case have a very similar effect on the expressions as the ones in the Arrhenius case.

- $\Omega_2 = \frac{E_{a2}}{R T_{ad1}}$ and $\frac{\mu_4}{\mu_1} = \frac{\theta_{i2}^{-1}}{\theta_{ad1}^{-1}}$: Both are essentially ratios of the propensity of the second reaction to happen over the final conditions of reaction 1. One can conclude that to large extents, the ignition temperature and the activation of the second reaction are chemical switches which dictate how far behind the first reaction the second one occur. Both of these external parameters play qualitatively the same role.

- $\Omega_1 = \frac{Q_2 k_2}{T_{ad1}}$ and $\frac{\mu_2}{\mu_1} = \frac{\theta_{ad2} - \theta_{ad1}}{\theta_{ad1}^{-1}} \frac{\theta_{i1}^{-1}}{\theta_{i2} - \theta_{ad1}}$: Here the comparison is not as straightforward. In the Arrhenius term, $\Omega_1$ is the ratio of the rate of energy produced by the second reaction to how to the exit conditions of the first one. One may recover these terms, just in a slightly different formula in $\mu_2$. There, the difference between the adiabatic temperature of the overall flame and the adiabatic temperature of the 1st front alone, i.e. the amount of heat given off by the 2nd reaction, is similar to $Q_2$. The rate part is also present. In fact the higher $\frac{\theta_{i1}^{-1}}{\theta_{i2} - \theta_{ad1}}$, is the faster the second reaction happens relative to the speed of the first one, being in fact similar to $k_2$.

Thus one recovers both of the main factors influencing merging behaviors and one can also see that from previous discussions, both pairs, $\Omega_2$ and $\frac{\mu_4}{\mu_1}$ as well as $\Omega_1$ and $\frac{\mu_2}{\mu_1}$ are relatively comparable on their effect on the merging of the flame. This common ground attests that the separation to merging behavior of adiabatic two-front flames explanation is not dependent on the formulation of the model but stems from more fundamental heat transfer considerations. One can demonstrate that the two-front configurations are indeed governed by mutual heat transfer and heat losses. One important point to notice is that chemical and many physical effects (nature of reactions, diffusion...
etc.), neglected by the present models, is in fact contained in a very simplified way in the external switching parameters, the ignition temperatures for the step model and the activation energies for the Arrhenius approach.
6 CONCLUSIONS

Mixtures of two very distinct fuels, such as different suspensions or a suspension in a combustible gas, appear in various practical applications and their combustion often leads to flames with complex behaviors. The first goal of the present thesis is to obtain the different physical configurations of such flames by studying their quenching behavior. The tests are performed for well-defined mixtures of aluminum powder in methane-air gas.

The main results obtained in this thesis are two-fold.

- Experimental:
  o A flame tube apparatus, previously constructed in the scope of an undergraduate thesis, was used to obtain quenching distances for aluminum-methane-air mixtures for different concentrations of aluminum powder.

  o At low concentrations, only a methane air front propagates and quenches in the tube. It was observed that an increase in powder loading leads to an increase in quenching distances, with a slower rise than for inert particles.

  o In accordance with previous tests, at a critical concentration, a second aluminum front appears and couples with the methane-air one. At intermediate concentrations, this coupling is first weak and upon reaching the quenching channels, the fronts de-couple, quenching individually, while at higher concentration, the coupling is stronger and the fronts act as a single flame, either propagating or quenching together,

- Analytical:
  o A simplified model, based on previous formulations, has been used to represent the aluminum-methane air flame as two individual reaction fronts, set off by ignition and proceeding a constant rate for a given combustion time.
The model qualitatively reproduces the behavior of the flame both at low aluminum concentrations, predicting only one weakening front, as well as the appearance of the second, aluminum, front at a critical concentration and its coupling to the methane-air one.

The addition of heat losses to the walls allows the model to capture all major stages of front quenching observed in the experiments. The coupling and decoupling behaviors are explained simply through the configuration of the flame. The existence of one- and two-front flames in the channels is explained through the strength of coupling between the fronts and of the overall flame.

The model explains the nature of this coupling/de-coupling behavior. The second front can exist only if it is strong enough to enhance the first one. The separation between the fronts is controlled by the strength (rate of heat produced versus rate of heat lost) of the second reaction versus the chemical threshold that needs to be overcome, all relative to the strength of the first front. And finally, a weak coupling becomes strong when the second front reaches the first one and the only limit to propagation is the activation/ignition of the first reaction.

Apart from these major findings, the work developed in this thesis presents two other conclusions:

- The derivation of the model leads to an explicit formulation of exact solutions pertaining to the problem first introduced by Mallard and LeChatelier. In the light of this new exact formulation, the still commonly used Mallard and LeChatelier expression of flame speed appears to have an error of 30% resulting from the linear approximation of the temperature profile in the flame region.

- The findings of the step model are further confirmed by an approximate Arrhenius formulation of two well separated fronts. This analysis examines what changes in parameters of either reaction will lead to the fronts approach each other. A comparison of non-dimensional numbers in both formulations shows that flame
coupling and de-coupling is governed by the same parameters irrespective of the formulation. This emphasizes that flame coupling is a fundamental phenomenon in flames of mixtures with very different fuels.
7 APPENDIX

7.1 Unsteady Calculations

Some work has been performed to assess the stability of the different solutions described by the steady-state model. The goal was to develop an unsteady formulation of the problem and, using a simple Eulerian finite difference code, to see whether an initially perturbed flame profile would recover its steady state appearance and, in the case of multiple possible solutions, which one of the configurations the solution would converge to.

The code was used on all configurations of the flame and always seemed to converge to the uppermost branch of the flame speed graph (Figs. 4-2, 4-5, and 4-7), suggesting the stable nature of this branch for adiabatic/non-adiabatic and one-front/two-front configurations.

The results are, however, not presented in the main section of the thesis because the fact that the discontinuity of the step function cannot be precisely represented by any numerical approximation. There is a possibility that the approximation of the discontinuous step, no matter how fine the mesh is, does not reproduce the dynamics of a step front and the time dependent changes of the temperature profile may not correspond to how a perturbed step would propagate into a mixture. Further investigation into this approach is required.

Below is a derivation of the unsteady formulation. One starts with a time-dependent energy governing equation:

\[ c \rho \frac{\partial T}{\partial t} = \lambda \frac{\partial^2 T}{\partial x^2} + q - \alpha (T - T_u) \]

\[ (\rho_g c_g + B c_s) \frac{\partial T}{\partial t} = \lambda \frac{\partial^2 T}{\partial x^2} + \frac{B Q}{\tau_c} - \alpha (T - T_u) \]

Which, using the dimensionless expression of temperature (\( \theta = \frac{T}{T_u} \)) and time (\( \tau = \frac{t}{\tau_c} \)) becomes:

\[ (\rho_g c_g + B c_s) \frac{\partial \theta}{\partial \tau} = \lambda \tau_c \frac{\partial^2 \theta}{\partial x^2} + \frac{B Q}{T_u} - \alpha \tau_c (\theta - 1) \]
And, using the spatial non-dimensionalization: \( \xi = \frac{x}{V_{ss} \tau_c} \) where \( V_{ss} \): steady state velocity

\[
(\rho_g c_g + B c_s) \frac{\partial \theta}{\partial \tau} = \frac{\lambda}{V_{ss}^2 \tau_c} \frac{\partial^2 \theta}{\partial \xi^2} + \frac{B Q}{T_u} - \alpha \tau_c (\theta - 1)
\]

The final expression then is:

\[
\frac{\partial \theta}{\partial \tau} = \frac{1}{\kappa_{ss}^2 (1 + \nu \phi)} \frac{\partial^2 \theta}{\partial \xi^2} + \frac{S}{1 + \nu \phi} - \frac{\eta (\theta - 1)}{1 + \nu \phi}
\]

Below is an example of a conversion of a perturbed two-front non-adiabatic flame to its steady-state configuration:
7.2 Four solutions – additional graphs

The results of section 4 show the flame speed and $Z$ variations for different concentrations for a given heat loss term $\eta$. At low heat losses, the flame speed exhibits a possibility of up to 4 solutions for the same initial parameters. Here are some graphs showing what happens as the heat loss term is varied.

Below is a graph of flame speeds showing the adiabatic curves (Fig. 4-2) and curves for one value of $\eta$ of both one-front configuration ($\beta_1$ and $\beta_2$) from Fig. 4-5 and a fold curve from Fig. 4-7.
It was interesting to see how the value of $Z$ varies for different concentrations and different heat loss terms. Such a case is shown in Fig. 4-9, but this is only for a heat loss term very close to $\eta_{\text{critical}}$.

When the heat loss term decreases much more, the one-front and two-front cases become very entangled and the flame speeds of the lower three branches of the two-separated front curve become almost exactly equal to the one-front flame ones, which are in fact a mathematical limit of definition of the two-front expression for $Z$.

When these curves are so close to each other, it is very hard to apply root searching techniques to obtain precise values of the flame speed $\kappa$. The existence of these points was proven only by performing tedious checks for individual concentration values whose details have been left out from the thesis.

Since it is impossible obtain actual two-front flame speed values, the values of $Z$ were estimated by getting the analytical expressions for the one-front case and since these are extremely close to the actual values of two-front flame speeds, they were then used in the $Z$ expression instead. But, as these one–front case velocities are limits of definition of the expression for $Z$, i.e. their values yield precisely a 0 in the denominator of the $Z$ expression (equation 4-30), one had to find the closest expression possible for which $Z$ is defined. Thus, the smallest possible increment that MATLAB can actually handle ($\delta = 10^{-16}$ in this particular code) was used and was added/subtracted to $\beta_1$ and $\beta_2$ and these were used to obtain the values of $Z$, shown in the figure below. As the values of $Z(\beta_1 - \delta)$ and $Z(\beta_2 + \delta)$ are only best possible approximations to the actual $Z$, these form a scatter (green and brown on the next figure respectively).
The previous figure shows how $Z$ varies with concentration, in a curve analogous to the one on the $\phi - Z$ plane of Fig. 4-9. Another curve, now corresponding to the one on plane $\kappa - Z$, now seeing how $Z$ varies with changes in flame speed, was also obtained below:

In both previous figures, the red and curves correspond to the values of $Z$ of the other branches of the two-separated front curve, where it was possible to easily find the two-front flame speed and plug it into the $Z$ expression. The blue curve, corresponds to the merged-front case, yielding a $Z$ below 1 (as the fronts are merged).
It is very interesting to notice how the values of $Z$ dramatically change as soon as the two-front curve hits the one-front one (corresponding to the red curve hitting the regions of green and brown scatter in the two figures above). This really indicates that the one-front speed curve has a disruptive influence on the two-separated triple-fold flame speed curve.

After that, the two previous figures were obtained for different heat loss terms and the comparative curves are plotted on the next two figures. Here, the scatters observed in the previous two figures have been replaced by dotted trend curves using Excel.

Here, as the heat loss term decreases towards 0, i.e. asymptotes towards the adiabatic case ($\eta = 0$), the values of $Z$, corresponding to the distance between the two fronts become very much larger. This means that the flame, subjected to lower and lower heat losses can spread much more over the space without being weakened by the heat losses.

The next figure shows how the heat loss reduction affects the values of $Z$ with respect to the flame speed. Again, the same observation as on the previous figure can be made, i.e., that lower heat
losses allow the flame to spread further. Moreover, on this figure, a very interesting effect can be observed: All of the $Z$ values from the scatters (i.e. the colored dotted lines in the previous figure) collapse onto a single curve (black dotted curve), which is serves as a limit no matter how small the heat losses are.

*Points obtained by using $\kappa$ from non-adiabatic one-front configurations in $Z$ expressions*
REFERENCES


[72] L. A. Lovachev, V. S. Babkin, V. A. Bunev, A. V. V’Yun, V. N. Krivulin, and A. N.


