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Spray-Flame Oscillations in Two-Fuel Lean Mixture

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Abstract

We analytically and numerically analyse the stability of a spray-flame propagating through a homogeneous three-component lean premixture composed of fuel droplets (typically a heavy alkane with low vapour pressure), a second gaseous fuel (typically a light alkane "stimulating" combustion) and air in excess. The fuel vapour evaporating from the droplets has a Lewis number larger than one (i.e. fuel-1 with $Le_1 > 1$), while the light gaseous fuel-2 has a Lewis number smaller than one ($Le_2 < 1$). The initial mass fraction of fuel-1 (under liquid phase), $(Y_{l_1})_u$, expresses the spray liquid loading, while $(Y_{g_2})_u$ is the initial mass fraction of gaseous fuel-2, the overall fuel amount in fresh mixture therefore being $(Y_{l_1})_u + (Y_{g_2})_u$. For relative liquid loading, $\delta = (Y_{l_1})_u / \{(Y_{l_1})_u + (Y_{g_2})_u\}$ higher than 50%, an excellent agreement between both numerical and analytical predictions on the intrinsic spray-flame instability is found. Conversely, poor agreement between numerics and asymptotics is found for the predicted threshold of diffusional-thermal pulsating instabilities in fully gaseous premixed flames at large Lewis number. This difference comes from the fact that the mechanism leading to spray-flame oscillations is intrinsic, and not based on differential diffusional effects. When increasing $(Y_{l_1})_u$, spray-flame instability threshold decreases and for vanishing $(Y_{g_2})_u$ pulsations are found to occur for conditions that can easily be met in experiments on large carbon number fuel sprays.

1. Introduction

The paper deals with plane two-phase flame. It has been observed [1-2] that two-phase flame can exhibit an oscillatory regime of propagation. Recent analyses [3-5] have shown that the phenomenon results from an intrinsic mechanism which we briefly describe below. The present paper treats of the particular case where spray-flame propagates through a two-fuel aerosol.

Spray-flame is known for long [6-14] to exhibit large departures from one-phase premixed flame (i.e. the flames propagating in a gaseous mixture). Recent experiments conducted under microgravity brought some clarification with respect to the part played by droplets on spray-flame speed promotion [15] and supplied new phenomena on pulsating flame propagation in particle-laden gas [1]. Oscillatory spray-flames have equally been observed on the ground [2] for lean and stoichiometric one-fuel spray, whereas oscillatory spreading does not occur for the equivalent gaseous premixed flame.

The existence of a robust intrinsic instability leading to oscillatory spray-flames has been pointed out by a numerical investigation [3], first. Then, an analytical work [4], using the simple framework of the constant density model, complemented with a vaporization rate independent of gas temperature, has highlighted its domain of existence and clarified the mechanism responsible. The latter does not invoke any differential diffusive phenomena effects (it occurs for $Le = 1$) or heat loss (it occurs for vanishing latent heat). It however requires that the Zeldovich number (*Ze*) be sufficiently high and that the Damkoehler number (*Da*), the ratio of chemical reaction rate to vaporization rate, be sufficiently small: vaporisation time not larger than reaction time. Basically, the mechanism invokes the coupling between the thermal profile in the pre-heating zone and the fresh gaseous species profile, through the vaporisation process. We have also studied [5] the differential diffusive effects on the spray-flame intrinsic instability.

In the framework of microgravity experiments, Nunome *et al.* [16] studied spray-flame propagation in an air/*n*-decane mixture: flame propagation only happened for sufficiently large *n*-decane droplets. These authors also considered a feeble addition of methane by keeping the overall equivalence ratio constant; they observed a flame speed enhancement, in such a way that combustion seemed to have been "stimulated" by a light fuel. Such a "doped" combustion could have been carried out with hydrogen, too.

The paper purpose is to consider such a situation of two fuels composing a lean two-phase mixture; our goal is not to investigate the reasons of the flame speed promotion, but to predict how an additional volatile fuel -with a Lewis number smaller than one- can affect the onset of the intrinsic instability of spray-flames. Although experimental results on two-phase flame pulsations [1-2] also

report wrinkles, we shall restrict the present contribution to stability results relatively to plane perturbations ($k = 0$). This allows us to use 1-D numerical computations, reaching thus a quasi-unlimited numerical accuracy. The results presented in the sequel nevertheless have to be considered as an upper bound of threshold, because some unstable modes with $k \neq 0$ might develop at a lower threshold.

2. Modelling

This joint numerical/analytical work has to consider a minimal model. Its choice results from a compromise between considerations of easy analytical handling and the fact that it must contain the above described mechanism. In Ref. [3], we have reported parametric numerical studies with respect to more complex models. It has been observed that the spray-flame oscillatory properties are maintained if successive simplifications of the initial model are performed. Even if the pulsation frequency changes or the onset threshold moves, the spray-flame oscillations persist with : a rough vaporization law, a unity Lewis number, zero latent heat and even with a constant gas density. In other words, oscillatory spray-flame does exist within the framework of diffusive-thermal model. Moreover, as soon as droplets are small enough (i.e. $Da < 1$, see below) and their boiling temperature is sufficiently low (i.e. $\theta_v \leq 0.2$) spray-flame pulsating properties appear quite independent of these parameters. This is the reason why both parameters are set to $Da = 0.1$ and $\theta_v = 0.1$ in the whole paper. Furthermore, any heat loss is neglected (as a matter of fact, alkane latent heat amounts to a few percents of reaction heat).

As a result, the model we investigate for studying the stability analysis of the plane spray-flame is the simplest one of the literature on spray combustion [17]. Let us remind briefly its main characteristics (for more detail, see Ref. [3 ,4 ,14]): we suppose that combustion and spray length scales are well separated in the sense that there exists an intermediate scale between droplet inter-distance and spray-flame thickness. Under these conditions, averaging on the intermediate scale leads to conservation laws for each fluid. Further simplifications -i.e. neglecting droplet inertia and their part in thermal budget- lead to consider the liquid fuel as an additional species, which cannot burn without prior vaporization. Droplets are small, so that they have the same velocity as the host gas. Chemical kinetics is described by two one-step exothermic irreversible reactions, described by Arrhenius law. Furthermore, the mixture being supposed globally fuel-lean, reaction is controlled by both limiting fuel species (i.e both gaseous and vaporized fuels). Vaporization phenomenon is simply described [17] with a vaporization rate independent of temperature. The vaporization starts as soon as the mixture temperature reaches T_v, a threshold value which may be compared to the boiling temperature of the liquid fuel.

Under these conditions, T_b , the adiabatic spray-flame temperature is given by

$$T_b = T_u + \frac{Q}{C_p}\left(Y_{g_2}\right)_u + \frac{Q - L_v}{C_p}\left(Y_{l_1}\right)_u \qquad (1)$$

where T_u is the fresh mixture temperature, $\left(Y_{l_1}\right)_u$ and $\left(Y_{g_2}\right)_u$ being respectively the fuel mass fraction of liquid and gas in the fresh mixture. Additional assumptions concern the vanishing latent heat ($L_v = 0$) and that both fuels have the same reaction heat per mass unit Q , as well as the same activation energy E .

Using the dimensional scales relative to the Zeldovich Frank-Kamenetski theory of one-fuel lean premixed flame (at temperature T_b , with unity Lewis number and referred as ZFK), we shall present the conservation laws under the classic non-dimensional form; we first introduce $\theta(x, y, t)$, the reduced temperature such that $T = T_u + (T_b - T_u)\theta$; furthermore, all fuel mass fractions Y_{l1} , Y_{g1} and Y_{g2} are reduced with the initial overall fuel mass fraction $\left(Y_{l_1}\right)_u + \left(Y_{g_2}\right)_u$; time and length units are respectively selected as $\tau_{chem} = (D_{th})_b / U_{ZFK}^2$ and $L_{ZFK} = (D_{th})_b / U_{ZFK}$ where U_{ZFK} is the ZFK-flame speed defined [18] as the following reaction-diffusion speed for a fuel with $Le = 1$

$$U_{ZFK} = \sqrt{\frac{2\lambda \rho_b^2 Y_{O_2} B}{C_p \rho_u^2 Ze^2}} \exp\left(-\frac{E}{2RT_b}\right) \qquad (2)$$

where the non-dimensioning process has led to define the Zeldovich number as the reduced activation energy derived from E as

$$Ze = \frac{E}{RT_b^2}(T_b - T_u) \qquad (3)$$

Thermal diffusivity is defined as $D_{th} = \lambda/(\rho C_p)$, while D_i stands for the binary diffusion coefficient of the fuel-i gaseous limiting component and Le_i for $Le_i = D_{th}/D_i$. This process for non-dimensioning leads to define Da , the Damkoehler number, as the ratio of vaporisation characteristic time to chemical reaction time:

$$Da = \frac{\tau_{vap}}{\tau_{chem}} = \frac{\tau_{vap} U_{ZFK}^2}{(D_{th})_b} \qquad (4)$$

where τ_{chem} is the characteristic reaction time at temperature T_b and τ_{vap} stands for the characteristic time for vaporization. In [4] we have

shown that the spray-flame speed is identical to (2) for unity Lewis number. This result can be kept identical if an additional assumption is formulated: both fuel Lewis numbers satisfy [19] the condition

$$Ze\left(Le_i - 1\right) = O(1) \qquad (5)$$

When deriving asymptotic solution, we shall indeed suppose that condition (5) is fulfilled.

Within the above diffusive-thermal context, it is interesting to write the system of conservation laws in a "perturbed frame" moving with the flame speed affected by a generic perturbation of the type

$$x_{SF}(y,t) = -U_{ZFK}\,t + \varepsilon \frac{D_{th}}{U_{ZFK}} \Phi(\eta,\tau) \qquad (6.a)$$

where ε is a (infinitely small) dimensionless amplitude of the space-time front corrugation $\Phi(\eta,\tau)$, which is supposed to possess the form

$$\Phi(\eta,\tau) = \exp(\omega\tau + ik\eta) \qquad (6.b)$$

We then introduce the following non-dimensional space-time coordinates (ξ,η,τ) defined as

$$\{\xi,\eta,\tau\} = \left\{ \left(x - x_{SF}(y,t)\right)\frac{U_{ZFK}}{D_{th}}, y\frac{U_{ZFK}}{D_{th}}, t\frac{U_{ZFK}^2}{D_{th}} \right\} \qquad (6.c)$$

Under the new set of variables (5), conservation laws read

$$\frac{\partial\theta}{\partial\tau} + \left[1 - \varepsilon\frac{\partial\Phi}{\partial\tau}\right]\frac{\partial\theta}{\partial\xi} = \Delta_\varepsilon\theta + \dot{\omega}_{1chem} + \dot{\omega}_{2chem} \qquad (7.a)$$

$$\frac{\partial Y_{l1}}{\partial\tau} + \left[1 - \varepsilon\frac{\partial\Phi}{\partial\tau}\right]\frac{\partial Y_{l1}}{\partial\xi} = -\dot{\omega}_{vap} \qquad (7.b)$$

$$\frac{\partial Y_{g1}}{\partial\tau} + \left[1 - \varepsilon\frac{\partial\Phi}{\partial\tau}\right]\frac{\partial Y_{g1}}{\partial\xi} = \frac{1}{Le_1}\Delta_\varepsilon Y_{g1} - \dot{\omega}_{1chem} + \dot{\omega}_{vap} \qquad (7.c)$$

$$\frac{\partial Y_{g2}}{\partial\tau} + \left[1 - \varepsilon\frac{\partial\Phi}{\partial\tau}\right]\frac{\partial Y_{g2}}{\partial\xi} = \frac{1}{Le_2}\Delta_\varepsilon Y_{g2} - \dot{\omega}_{2chem} \qquad (7.d)$$

with

$$\dot{\omega}_{1chem} = \frac{Ze^2}{2} Y_{g1} \exp\left[-Ze\left(1-\theta\right)\right] \qquad (8.a)$$

$$\dot{\omega}_{2chem} = \frac{Ze^2}{2} Y_{g2} \exp\left[-Ze\left(1-\theta\right)\right] \qquad (8.b)$$

$$\dot{\omega}_{vap} = \frac{Y_{l1}}{Da} H(\theta - \theta_v) \qquad (8.c)$$

where H stands for the Heaviside function. For the standard heavy alkane spray-flames, θ_v, the threshold value for vaporization is of the order of 10^{-1}. As for Δ_ε, the perturbed Laplace operator, admits the form

$$\Delta_\varepsilon = \frac{\partial^2}{\partial\xi^2} + \frac{\partial}{\partial\eta}\left(\frac{\partial}{\partial\eta} - \varepsilon\frac{\partial\Phi}{\partial\eta}\frac{\partial}{\partial\xi}\right) - \varepsilon\frac{\partial\Phi}{\partial\eta}\frac{\partial}{\partial\xi}\left(\frac{\partial}{\partial\eta} - \varepsilon\frac{\partial\Phi}{\partial\eta}\frac{\partial}{\partial\xi}\right) \qquad (9)$$

Problem (7) is closed with the following boundary conditions :

$$\xi \to -\infty, \theta = 0, Y_{l1} = \left(Y_{l1}\right)_u, Y_{g1} = 0, Y_{g2} = 1 - \left(Y_{l1}\right)_u \qquad (10.a)$$

$$\xi \to +\infty,\ \theta = 1\ , Y_{l1} = 0\ , Y_{g1} = 0\ \ , Y_{g2} = 0 \qquad (10.b)$$

Time-dependent solutions are investigated using the method of small perturbations. All variables are written as the sum of the steady state solution and a small harmonic perturbation

$$\theta = \overline{\theta}(\xi) + \varepsilon\,\Phi(\eta,\tau)\,\hat{\theta}(\xi) \qquad (11.a)$$

$$Y_{l1} = \overline{Y}_{l1}(\xi) + \varepsilon\,\Phi(\eta,\tau)\,\hat{Y}_{l1}(\xi) \qquad (11.b)$$

$$Y_{g1} = \overline{Y}_{g1}(\xi) + \varepsilon\,\Phi(\eta,\tau)\,\hat{Y}_{g1}(\xi) \qquad (11.c)$$

$$Y_{g2} = \overline{Y}_{g2}(\xi) + \varepsilon\,\Phi(\eta,\tau)\,\hat{Y}_{g2}(\xi) \qquad (11.d)$$

the first terms in the RHS of (11.a-c) corresponding to the flame structure at steady state.

3. Plane Spray-Flame Stability

To analyse the linear stability of spray-flame propagation we use the method of matched asymptotic expansions (in the limit $Ze \to \infty$) to solve system (7) as in ref [4-5], where the inner zone treatment is inspired from [19]. As soon as $\theta_v^{1/Da}$ is a negligible quantity: a vaporization domain exits inside the preheating region, producing gaseous fuel, and well-separated from the reaction front where gaseous fuels are consumed. With the assumption that reaction and vaporisation zones do not overlap, system (7) can be solved. In this calculation, four separated zones are used and the different local solutions are matched in the limit $Ze \to \infty$. We first compute the steady solution which confirms that spray-flame speed is identical to *ZFK* gaseous flame speed, for any value of $\left(Y_{l1}\right)_u$. Then, the resolution of the perturbed system leads to the following relation of dispersion

$$\left(Y_{l1}\right)_u\left(1 - \frac{2r^-}{Ze}\right)\frac{\left[\omega + s_1^- + Da\left(\omega^2 - k^2\right)\right]e^{-(s_1^- + r^-)\xi_v}}{(kDa)^2 - Le_1 Da - (\omega Da + 1)^2} + \frac{2r^-}{Ze}\left(r^- - r^+\right) + \left(s_1^-\left(Y_{l1}\right)_u + s_2^-\left(1 - \left(Y_{l1}\right)_u\right) - r^-\right) = 0 \qquad (12)$$

with $r^\pm = \dfrac{1 \pm \sqrt{1 + 4(\omega + k^2)}}{2}$, $\Delta = (Da)^{-1}$,

$s_i^{\pm} = Le_i \dfrac{1 \pm \sqrt{1 + 4\ell_i(\omega + \ell_i k^2)}}{2}$ and $\ell_i = (Le_i)^{-1}$.

We recall that θ_v is the fixed value of the reduced boiling temperature; through $\theta_v = e^{\xi_v}$ it determines ξ_v, the position where the vaporisation front starts. This point is subjected to fluctuate with the perturbation of the temperature field. The fluctuation of the locus where vaporisation starts is given by $\hat{\xi}_v = (\theta_v^{-r^-} - 1) - \frac{2r^-}{Ze}\theta_v^{-r^-}$.

Dispersion relation (12) describes the stability properties of spray-flames for various Le_i and for different liquid fuel loadings $(Y_{l_1})_u$. A necessary condition that a given perturbation with $\{\omega \in C, k \in R\}$ leads to a new propagation mode is that $\omega_r \equiv \mathrm{Re}(\omega)$ is a positive quantity. Thus, $\omega_r = 0$ corresponds to the curve of neutral stability that delimitates a domain in the space of physical parameters, where the steady solution is unstable. A pulsating propagation (or oscillatory instability) will occur if it is found $\omega_i \equiv \mathrm{Im}(\omega) \neq 0$, simultaneous- ly.

4. Quantitative Results

We are interested in the determination of the critical Zeldovich number that characterises the onset threshold of the pulsating instability. For that, we have to interpret the dispersion relation in terms of the set $\{Le_1, Le_2, Ze, (Y_{l_1})_u\}$, which corresponds the main parameters of the study. We shall however restrict our interest to certain pairs of Lewis numbers. The first one $\{Le_1 = 1.25, Le_2 = 0.8\}$ satisfies fairly well the conditions $|Le_i - 1| << 1$. The second one $\{Le_1 = 1.5, Le_2 = 0.66\}$ lies quite far from $Le = 1$, while the third one $\{Le_1 = 1.8, Le_2 = 0.8\}$ could be related to the couple *n*-decane/methane.

The last couple has its first Lewis number [i.e. Le_1=1.8] large enough to observe the same diffusive differential effects as those that are responsible of the one-phase premixed flame oscillatory instability: we perform the same analytical predictions for a pure gaseous fresh mixture -i.e. gaseous fuels of the same Lewis numbers-, the composition of which is defined as $\{(Y_{l_1}) = 0\ ;\ (Y_{g_1})\ ;\ (Y_{g_2}) = 1 - (Y_{g_1})\}$. For $k = 0$, we report the comparison in **Fig.1**: the critical Zeldovich numbers obtained for both one-phase and two-phase instabilities is plotted as a function of the fuel-1 mass fraction . We furthermore supply both theoretical and numerical predictions. In **Fig. 1**, the contrast between spray-flame instability and premixed diffusive-thermal pulsation instability is quite striking. For a maximum liquid loading (on the plain curves), spray-flame oscillations occur at a rather low Zeldovich number (i.e. $(Ze)_{crit.} = 13.25$). Furthermore, both numerical and analytical (plain) curves are in agreement.

Now, if the initial liquid loading is replaced by vapour of the same Lewis number ($Le_1 = 1.8$), we first note a large discrepancy between numerics and asymptotics. As for the actual threshold, the numerical approach shows it is strongly shifted to $(Ze)_{crit.} = 23$.

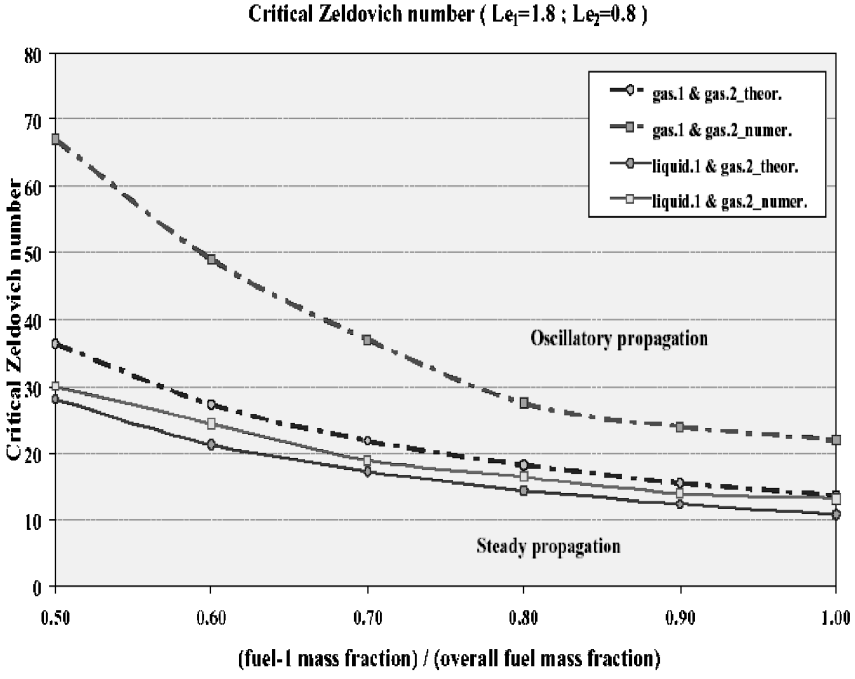

Fig.1. Neutral curve of oscillatory instabilities for the couple {Le_1=1.8,Le_2=0.8 } and $k = 0$, as a function of reduced fuel-1 mass fraction; critical Zeldovich numbers are determined from analytical analysis (circle symbols) or from numerical simulations (square symbols). Two different cases of fresh mixture are considered: all fuel-1 is initially under liquid phase in small droplets (plain curves, which are thus plotted as a function of the liquid loading) or under vapour (dotted curves). The other parameters are $Da = 0.1$ and $\theta_v = 0.1$.

On the one hand, we are led to conclude that

asymptotics supplies poor predictions for the one-phase instability if $|Le_1 - 1| << 1$ is not satisfied. This is due to the fact that the one-phase oscillatory instability results from a differential diffusive effect and the predictions are very sensitive to the treatment of diffusion in the inner zone, which is in fact a weak point of asymptotics. On the other hand, the discrepancy between both one-phase and two-phase thresholds shows that spray-flame instability is related to a particular (or intrinsic) mechanism, which is moreover weakly sensitive to diffusional effects.

We next consider for a short while the case of a pure spray-flame (as obtained with a unique low volatile fuel); we consider the results for $(Y_{l_1})_u = 1$ (i.e. for a maximum spray liquid loading, which also means $(Y_{g_2})_u = 0$). The numerical approach predicts the spray-flame oscillatory instability thresholds as follows for various Lewis number:

$\{Le_1 = 1.25 \rightarrow (Ze)_{crit} = 14.15\}$

$\{Le_1 = 1.5 \rightarrow (Ze)_{crit} = 13.75\}$

$\{Le_1 = 1.8 \rightarrow (Ze)_{crit} = 13.25\}$

These results are in agreement with our previous work [5] which predicted that spray-flame becomes more unstable with increasing Lewis number. Note that these critical Zeldovich numbers are here obtained for $k = 0$. If non plane perturbations were considered (i.e. $k \neq 0$), the above series might provide lower thresholds.

Now, if $(Y_{l_1})_u$ is progressively decreased by substituting fuel-2 gas with low Lewis number for fuel-1 liquid droplets, we observe an increase in critical Zeldovich numbers. These results are reported in **Fig.2**, where we notice a substantially enhancement of threshold. This confirms that spray liquid loading is a significant parameter of pulsating spray-flames. More precisely, Fig. 2 focuses on the results that concern the part played intrinsically by the presence of a large amount of liquid. In Fig.2, critical Zeld' ovich numbers are plotted with respect to liquid loading for various pairs $\{Le_1, Le_2\}$ of fuel Lewis numbers. Two general trends are noticeable: spray-flame oscillation becomes unlikely as liquid loading diminishes, whereas asymptotic theory slightly underestimates the actual threshold. When liquid loading diminishes, the threshold enhancement is more pronounced, all the more so since Le_2 is low (both curves with triangular symbols in Fig.2).

If low Lewis number gaseous fuel is further substituted for spray liquid loading, the pulsating spray-flame disappears at a non-zero $(Y_{l_1})_u$ [not reported in Fig. 2]. In between, the situation can become seriously intricate because a spray-flame with a low Lewis number fuel is unstable with respect to the (steady) cellular instability [5]. When both pulsating and cellular instabilities overlap –this indeed arises for $k \neq 0$–, the competition between unstable modes deserves to be studied, but it requires computational means that we have not used in the present work

This set of results allows us to formulate the spray-flame oscillatory instability as resulting from the following sequence:

a) let us suppose the flame accelerates [resp. decelerates] for some reason; it then results a strengthening [resp. smoothing] of pre-heating temperature gradient.

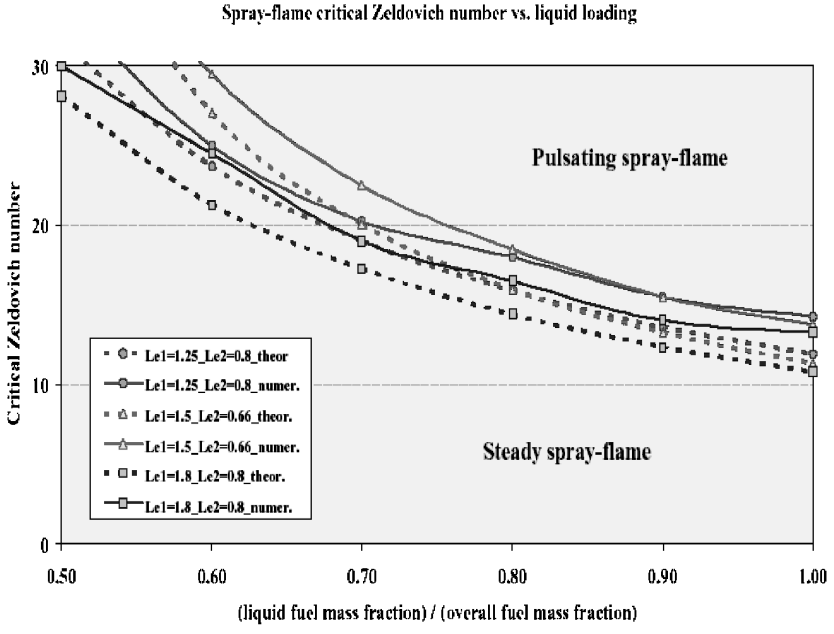

Fig. 2: Neutral curve of intrinsic oscillatory instability of spray-flame:. critical Zeldovich number vs. spray liquid loading for various pairs of Lewis numbers, where Le_1 [resp. Le_2] is the Lewis number of the non volatile fuel [resp. the gaseous fuel]. Theoretical critical Zeldovich numbers are dotted lines. The other parameters are Da = 0.1 and θ_v = 0.1.

b) vaporization occurs closer to [resp. farther from] reaction zone if droplets are small enough (Da<1).

c) gaseous fuel gradient enhances [diminishes] in reaction zone, if fuel mobility is low (i.e. *Le* not too small)
d) heat release increases [resp. decreases]
e) flame propagation speeds up [resp. slows down], if Ze is enough large.
Step e) insures the instability feed-back, and step c) is at the root of the non-linear saturation, as vaporized fuel becomes exhausted [resp. maximum], which is unstable in its turn with respect to deceleration [resp. acceleration]. Note additionally that step b) leads qualitatively to the same consequences as those induced by high Lewis number effects on single-phase flame: gaseous fuel enhancement [resp. diminishing] in reaction zone. However, the difference between both single-phase and two-phase oscillatory instabilities is situated in the fact that differential diffusive effects are not invoked in the two-phase flame oscillatory instability. In the former case of high Lewis number, gaseous fuel gradient is increased only because the far field fuel profile is unaffected by temperature profile steepening.

5 . Conclusion

The present analytical / numerical joint approach treats of pulsating spray-flames, which are doped with a low Lewis number gaseous fuel, as can be met in recent experiments. When Zeldovich number is large enough (but substantially lower than the critical Zeldovich number of diffusive-thermal oscillatory instability in one-phase premixture) oscillations of spray-flames propagating in an aerosol with a sufficient liquid loading are predicted. This threshold in Zeldovich number substancially increases as the liquid loading is diminished (i.e. $(Y_{g2})_u$ is enhanced) in spray, as indicated in Fig. 2. Furthermore, the threshold in Zeldovich number is found to slightly decrease as Le_1 , the Lewis number of the liquid fuel, increases. In the same way, increasing the mobility of the gaseous fuel (i.e. decreasing Le_2) enhances the onset threshold, but does not produce dramatic effects.

As a result, the role played by differential diffusive effects is found weaker the one by liquid loading. This explains why asymptotic theory is more successful in predicting spray-flame intrinsic instability than in studying diffusive-thermal oscillatory instability when Lewis numbers differ from 1. Note that the departure between theory [20] and numerics in predicting the threshold of diffusive-thermal oscillation of premixed (gaseous) flames had been underlined in Ref. [21-23]. We interpret these large departures in terms of the difficulties to theoretically handle non-unity Lewis number species in the inner zone; this is why we found this weakness less decisive for studying the coupling between vaporisation and reaction, than between differential diffusional effects.

To summarise, we stress that the sequence mentioned above, that describes the coupling between vaporization zone and reaction zone corresponds to a robust mechanism (intrinsic to sprays) leading to pulsating spray-flame propagation. For operating, it nevertheless requires that spray liquid loading and Zeldovich number are sufficient.

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