## THE ONSET OF OSCILLATIONS IN NON-PREMIXED COMBUSTION

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In a microgravity environment, molecular diffusion is the primary mechanism by which fuel and oxidizer that are initially separated are brought together to the reaction zone. Combustion systems in microgravity are therefore primed to diffusive-thermal instabilities. One such instability appears in the form of spontaneous oscillations. Oscillations were observed in condensed-phase fuels [1] and gas-jet diffusion flames [2], and in microgravity jet-flames [3], candle flames [4]-[5] and spherical flames surrounding large fiber-supported fuel droplets [6]. The nature of oscillations is quite different in each of these cases: the droplet flame exhibits radial oscillations, the edge of the candle flame is seen to move back and forth along the hemispherical flame surface and the jet-flame oscillations are primarily up and down along the axis. Despite these differences, associated mainly with the mode of oscillation, one may identify some common factors: in all cases the flame exhibits low-frequency oscillations, oscillations are only observed in special mixtures and their onset occur only at near-extinction conditions.

As in premixed systems, the disparity between the thermal diffusivity of the mixture and the molecular diffusivities of the fuel and oxidizer is responsible for the development of flame instabilities. While thermal diffusion tends to nullify temperature differences, and hence has a stabilizing influence, molecular diffusion may enhance these differences. It is known that a premixed system exhibits a cellular instability when the effective Lewis number<sup>1</sup> is below some critical value less than one, and a pulsating instability when the Lewis number is above a critical value larger than one. The situation in non-premixed systems, however, is more complex because there are now two effective Lewis numbers, associated with the fuel and oxidizer, and because the structure of a diffusion flame varies significantly with the Damköhler number D, representing the ratio of the diffusion to the chemical reaction times. When the Damköhler number is large, the reactants are completely consumed at a reaction sheet that separates a region of oxidant but no fuel from a region where there is only fuel. Since complete combustion dictated by the short chemical reaction time can only occur along the stoichiometric surface, any slight disturbance of the reaction sheet is obliterated and the flame is stable irrespective of the value of the Lewis numbers. For moderate values of the Damköhler number  $D_{ext} < D < \infty$ , with  $D_{ext}$  representing the extinction value, there is incomplete combustion with significant reactants leakage through the reaction zone. Preferential and differential diffusion, associated with non-unity and unequal Lewis numbers, may now lead to various forms of instability.

In this presentation, we summarize results based on a simple one-dimensional configuration in which one reactant (the fuel, say) is supplied in a uniform stream and the other (the oxidizer) is diffusing against the stream [8]. The objective has been to identify the basic mechanisms

<sup>&</sup>lt;sup>1</sup> Based on the thermal diffusivity of the mixture, determined by the abundant component, and the molecular diffusivity of the deficient reactant.

responsible for the onset of oscillations. The simplicity of the model permits a comprehensive study that maps the entire range of parameters. In particular, we have examined the conditions for the onset of oscillations and their dependence on (i) the supply conditions, namely the mixture strength and the temperatures at which the fuel and oxidizer are introduced, (ii) the diffusion properties of the reactants represented by two distinct Lewis numbers,  $L_F$  and  $L_X$ , for the fuel and oxidizer, respectively and (iii) the dynamic properties lumped into a single parameter - the Damköhler number D.

An important auxiliary parameter that determines whether oscillations occur or not, is the heat transfer parameter  $\gamma$ , defined as the ratio of the excess heat conducted to one side of the reaction sheet to the total heat generated in the reaction zone. Generally,  $-1 < \gamma < 1$  and when  $\gamma = 0$  there are equal fluxes of heat directed away from the reaction sheet. More heat is transported to the fuel side when  $\gamma < 0$  and to the oxidizer side when  $\gamma > 0$ . The degree of reactants leakage through the reaction sheet is determined by  $\gamma$  and the Lewis numbers; generally speaking, the fuel is the more completely consumed reactant when  $\gamma < 0$  and the oxidizer is the more completely consumed reactant when  $\gamma < 0$  and the mixture strength  $\phi$ , defined as the ratio of the initial fuel to oxidizer mass fraction normalized by their stoichiometric proportions, and is given by  $\gamma = (\phi - 1)/(\phi + 1)$ . Thus, a fuel "rich" system corresponds to  $\gamma > 0$ ; a fuel "lean" system to  $\gamma < 0$ . In general,  $\gamma$  depends on the Lewis numbers and on the supply-temperature differential  $\Delta T$ .



Our results indicate that oscillations occur when the reactant diffusing against the stream (the oxidizer in the present configuration) is the more completely consumed reactant, corresponding to a situation for which  $\gamma > 0$ , or a relatively "rich" system<sup>2</sup>. Thus, for a given set of parameters corresponding to  $\gamma > 0$  there exists a value of the Damköhler number  $D^* > D_{ext}$  which identifies the onset of oscillations. The stable flames are =

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those extending from the Burke-Schumann solution with complete consumption  $(D \rightarrow \infty)$  down to the marginally stable state  $D^*$ , with the unstable states limited to  $D_{\text{ext}} < D < D^*$ . This is shown schematically in Fig. 1 where the dependence of the flame temperature  $T_f$  on the Damköhler number D is presented. Note that oscillations  $d_{\text{exc}}$  and  $T_f$  prior to but near extinction with the range of unstable states depending on the remaining parameters.

The primary mechanism responsible for oscillations appears to be the inability of the reactants to diffuse quickly enough to keep up with temperature fluctuations, namely when the Lewis numbers are sufficiently large (typically, but not necessarily larger then one). Neutral stability curves identifying regions of stable (above the curve) from unstable (below the curve) states, as a

162

<sup>&</sup>lt;sup>2</sup> In "lean" systems, corresponding to  $\gamma < 0$ , another type of instability occur; namely the development of cellular flames discussed in [7].

function of the Lewis numbers, are shown in Fig. 2 for  $\phi = 1$  and  $\Delta T = 0$ . (The corresponding values of  $\gamma$  are in the range  $0 < \gamma < 0.38$ , depending on  $L_X$ ). The figure shows, in particular, the range of unstable states expressed in terms of the Damköhler number relative to the extinction state. All states are stable when  $D \rightarrow \infty$ , as expected. For a given  $L_X$ , oscillations are possible only for a restricted range of  $L_F$ , and the range of unstable states grows as  $L_X$  increases.



For conditions corresponding to  $\gamma > 0$ , the reaction sheet is located at a distance from the stoichiometric surface on the oxidizer side. If the reaction sheet is slightly perturbed toward the stoichiometric surface, the burning intensity increases with more heat generated. Due to the relatively large thermal diffusivity of the mixture, the temperature at the sheet drops at a faster rate than mass is being supplied to it, and for  $\gamma > 0$  this imbalance is more pronounced on the oxidizer side. As a result, the reaction sheet tends to move back towards the stoichiometric surface, and possibly beyond its original location. Here the chemical reaction will be starving for fuel and the tendency, therefore, will be for the sheet to move back to the stoichiometric surface where again the burning intensity increases and the cycle continues. Heating the fuel relative to the temperature at which the oxidizer is supplied ( $\Delta T < 0$ ) results in more heat transported to the oxidizer side and, according to the mechanism described, promotes the onset of oscillations. The reverse is expected when heating the oxidizer ( $\Delta T > 0$ ). These tendencies are consistent with our predictions.

The predicted frequencies of oscillations at the onset of the instability are typically found in the range of 1-6 Hz, in accord with experiments. They grow larger when the onset occurs near the extinction limit ( $D^* \approx D_{ext}$ ) and when the Lewis numbers are both near one.

An important conclusion from the present study is the importance of the mixture strength  $\phi$  in promoting/suppressing the oscillations, as seen in Fig. 3. Stability curves, similar to the one presented in Fig. 2, show the effect of varying  $\phi$  for selected values of  $L_X$  (drawn for  $L_F = 1$  and  $\Delta T = 0$ ). Similar to the experimental observations [2], it is seen that oscillations are more likely to occur  $\phi$  is relatively large. It is evident from this figure that a small change in  $\phi$  causes a much larger effect on promoting the oscillation, than any realistic variation in Lewis number would cause.

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Experimental results [2] indicate that, for  $\phi$  sufficiently large, oscillations will occur even if one of the Lewis numbers is slightly less than one. For example, oscillations were observed when pure methane was supplied in the fuel stream and the oxidizer stream was diluted with nitrogen, conditions corresponding to  $\phi = 21.68$ ,  $L_X = 1.11$  and  $L_F = 0.81$ . These results are not inconsistent with our theory; in fact, Fig. 1 clearly suggests that as  $L_X$  increases, for example, the range of unstable states will extend into regions where  $L_F < 1$ . We note that a similar behavior was also detected in [7] where the boundary for the cellular instability appears often to extend into a region of Lewis number bigger than one. A closer examination shows that this is indeed the case and that the critical fuel Lewis number,  $L_F^{cr}$  say, decreases with increasing  $L_X$ . This illustrates the importance of both Lewis numbers in identifying the conditions for the occurrence of oscillations.

The importance of heat losses in promoting flame oscillations has been previously recognized; see for example [9]-[11]. Indeed, our theory shows that a small amount of heat losses may induce oscillations under conditions where, in their absence, oscillations are not likely to occur. The effects of heat loss appear more pronounced especially when the mixture strength is relatively small, namely when the likelihood for oscillations is small.

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