Diffusive-Thermal Instabilities of High Lewis Number Flames in Micro Flow Reactor

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1 Introduction

Investigation of flame structure and dynamics in micro channels is expected to contribute to practical applications of microscale system such as fuel diagnostics [1], verification of reaction mechanisms [2] and microcombustion device [3]. In experimental studies of combustion in micro channels with temperature gradient of the walls, it was demonstrated that three modes of combustion exist [4]. For high mixture flow rates, stable normal flame regime which corresponds to preheated freely propagating flame is observed. Stable weak flame characterized by low flame temperature can be found in the range of small inlet velocities. At the intermediate flow rates the dynamical flame behavior with repetitive ignition and extinction (FREI) occurs.

These flame regimes and their qualitative properties are well described in the frame of thermal-diffusion model with one step Arrhenius kinetics [5]. Flame oscillations in FREI mode are induced by heat exchange between combustible gas mixture and solid walls with nonuniform temperature distribution. At the same time, combustion of gas mixtures with large Lewis numbers are prone to pulsating diffusive-thermal instabilities [5], however, emergence of diffusive-thermal instabilities in micro channels with temperature gradient of the walls has not been investigated to date. At the same time, fundamental knowledge on manifestation of intrinsic flame instability in microchannels is important for further development of microcombustors and fuel diagnostic techniques.

In this work, we study diffusive-thermal flame instabilities in mixtures with Lewis number larger than unity in micro flow reactor with controlled temperature profile. We apply the thermal-diffusion model for combustion of rich hydrogen-air mixtures with reduced two-step kinetics [6].

2 Model

Fresh hydrogen-air gas mixture enters the reactor from the cold end of the tube and travels along the positive x-axis direction toward the hot part of the tube with specified velocity V. The temperature distribution in the reactor is described by given function θ. We consider a thermal-diffusion model [6] with reduced two-step chemistry for rich hydrogen-air flames which includes two steps: chain

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branching $3H_2 + O_2 \to 2H_2O + 2H$ and recombination $H + H + M \to H_2 + M$. The rates of the gross reactions are controlled by the rates of elementary reactions as follows: $\omega_I = w_1$, $\omega_{II} = w_4 + w_5$, where w_1 is the rate of $H + O_2 \to OH + O$ reaction, w_4 is the rate of $H + O_2 + M \to HO_2 + M$ reaction and w_5 is the rate of $H + H + M \to H_2 + M$ reaction. Following [5, 6] the governing equations for dimensionless temperature, T, concentration of oxygen, Y_{O_2}, and H-radicals, Y_R, can be written in spatially one dimension as

$$\frac{\partial T}{\partial t} = -V\frac{\partial T}{\partial x} + \frac{\partial^2 T}{\partial x^2} + K(\theta(x) - T) + R_4 Y_R Y_{O_2} e^{\beta - 1/T_b} + R_5 Y_R^2 e^{\beta - 1/T_b},$$

$$\frac{\partial Y_{O_2}}{\partial t} = -V\frac{\partial Y_{O_2}}{\partial x} + L_{O_2}^{-1}\frac{\partial^2 Y_{O_2}}{\partial x^2} - \beta Y_R Y_{O_2} e^{\beta - 1/T},$$

$$\frac{\partial Y_R}{\partial t} = -V\frac{\partial Y_R}{\partial x} + L_R^{-1}\frac{\partial^2 Y_R}{\partial x^2} + \beta Y_R Y_{O_2} e^{\beta - 1/T} - \beta R_5 Y_R^2 e^{\beta - 1/T_b} - \beta R_4 Y_R Y_{O_2} e^{\beta - 1/T_b},$$

where x and t are dimensionless spatial coordinate in units of thermal width of the flame and time respectively; K is the dimensionless coefficient of heat exchange between the gas mixture and the wall of the reactor; L_{O_2}, L_R are the Lewis numbers for O_2 and H-radicals respectively; $R_{4,5}$ are the reaction constants; V is the gas flow velocity in units of the velocity of a planar adiabatic flame; T_b is the downstream temperature of the burned mixture; β is the dimensionless activation energy; θ is the temperature of the reactor walls defined in the following way:

$$\theta = \begin{cases} T_0 + (\theta_{\max} - T_0)e^{\eta x} & x \le 0, \\ \theta_{\max} & x > 0 \end{cases}$$

where $1/\eta = const$ is the characteristic length of the temperature drop in the tube wall. Here we note that temperature is measured in the unit of the activation temperature of the branching reaction. Further details of the non-dimensionalisation can be found in [6] and are omitted here for brevity. The boundary conditions are defined as follows. The length of the reactor is considered to be infinite in both positive and negative directions of x-axis. On the left-hand side the temperature is equal to the ambient temperature T_0, oxygen has not been consumed yet, no radicals have been produced. On the right-hand side, no reaction occurs, thus the zero flux conditions for T, Y_{O_2} and zero radical concentration Y_R are imposed:

$$T = T_0,\ Y_{O_2} = 1,\ Y_R = 0 \text{ for } x \to -\infty,$$

$$\frac{\partial T}{\partial x} = 0,\ \frac{\partial Y_{O_2}}{\partial x} = 1,\ Y_R = 0 \text{ for } x \to +\infty.$$

The governing equations are solved numerically using the upwind explicit finite-difference scheme of the first order of accuracy in time and second order of accuracy in space. The solution is sought on a finite interval over x coordinate [L_1, L_2], where $L_{1,2}$ are chosen in such way as to minimize the influence of boundary conditions on the resulting flame characteristics. The grid size is taken to be sufficiently small and its further decrease affects the results in the third significant digit. For our calculations we fix the maximum temperature of the reactor walls to be equal to 1200 K, the initial temperature of gas mixture 300 K and parameter η = 1.5 cm^{-1} in dimensional quantities which typical for experiments on micro flow reactor [4].

3 Results

The diffusive-thermal instabilities are expected to occur for sufficiently large values of the equivalence ratio. In this paper we consider the mixtures with ϕ exceeding 7. We note here that current two step mechanism was already validated in the past study for such fuel rich conditions [6]. We have found that two types of instabilities can emerge: FREI and diffusive-thermal instability, which lead to formation of pulsating solutions with different structure. These solutions are illustrated in Figs. 1 and 2, where the temperature and radical concentration profiles are plotted as functions of coordinate, x, for $\phi = 7.4$. In Fig. 1, the diffusive-thermal pulsations are shown for $V = 1.82$. This velocity condition is close to the boundary between normal flame and FREI and corresponds to the midpoint of diffusive-thermal pulsations range. In this case, the distribution of $T(x, t)$ and $Y_R(x, t)$ are periodic functions of time. In Fig. 1, they are sampled at two moments of time along one period of oscillations, $T \sim 6.5$, corresponding to solutions with the largest and the lowest instant peak values of the radical concentration, which are presented with the dashed and solid lines, respectively. In the diffusive-thermal regime of flame oscillations, the amplitude of pulsations of T and Y_R remains limited so that quenching is not observed. This is in contrast to the regime of FREI pulsations which is illustrated in Fig. 2 for $V = 1.65$, where the distribution of $Y_R(x)$ is plotted for three moments of time along a single period of oscillations, $T \sim 16.4$. It is clearly seen in Fig. 2, that during FREI regime the peak instant concentration of radicals can drop to very small values and flame quenching and reignition is sequentially observed during a period of oscillations. The $Y_R(x)$ profile at time $t = 15.2$ corresponds to the developed upstream propagating combustion wave and $t = 20.2$ shows the upstream edge of reaction zone movement.

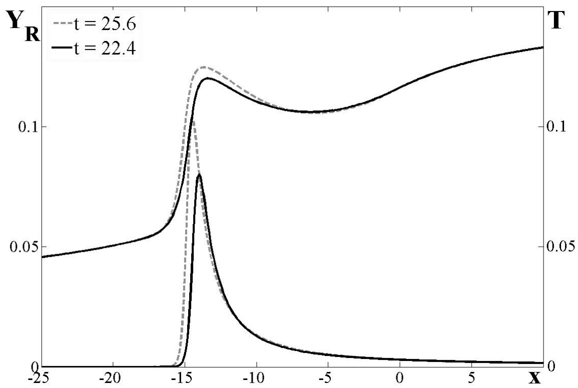

Fig. 1. Distribution of $T(x)$ and $Y_R(x)$ for diffusive-thermal pulsations for $\phi = 7.4$ and $V = 1.82$.

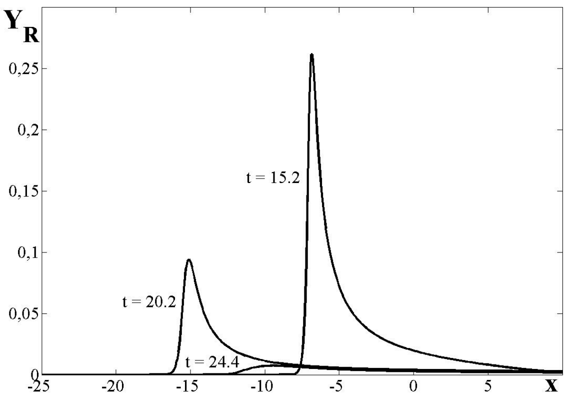

Fig.2. Distribution of $Y_R(x)$ for FREI for $\phi = 7.4$ and $V = 1.65$.

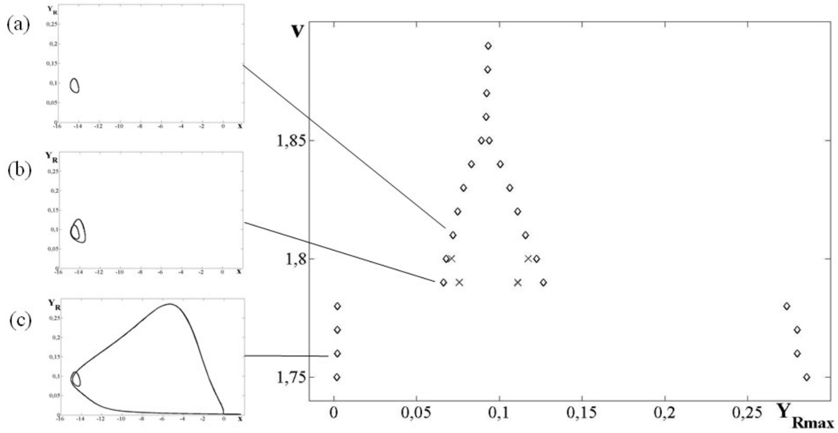

Fig. 3 Bifurcation diagram on V vs Y_{Rmax} plane showing normal flame, diffusive-thermal oscillations and FREI regimes. In the insets, dynamics of flame pulsations are illustrated in the phase plane (x_f, Y_R) for diffusive-thermal oscillations (a, b) and FREI (c).

Next we investigate the nature of bifurcations leading to the emergence of different pulsating regimes in greater detail. The equivalence ratio is kept constant and equal to 7.4, while the flow rate, V, is taken as a bifurcation parameter and is varied. It is convenient to characterize the oscillatory regimes by the location of the flame front, x_f, which we define as the position of local maximum of Y_R in space, and the value of the local maximum of Y_{Rmax}. These quantities compose the observable dynamic variables, which we further discuss on. In Fig. 3 the dependence of minimum and maximum values of the instant Y_{Rmax}, versus the flow rate, V, is plotted. For sufficiently high flow velocities, the normal flame branch which is a stationary solution is observed. And thus it is represented by a single point in the diagram for each value of V. As V is decreased at certain critical value of velocity, there emerges the diffusive-thermal regime of oscillations. It corresponds to the limit cycle in the phase plane x_f vs Y_{Rmax}, which is shown in the inset (a) in the Fig. 3. Further decrease of V results in the growth of the amplitude of pulsations according to the root-law, which is typical for supercritical Andronov-Hopf bifurcation. In other words, the oscillations are excited in soft regime and the amplitude grows in a continuous way as the bifurcation parameter is increased. A period doubling bifurcation occurs at certain value of V along the diffusional-thermal pulsating branch. This bifurcation leads to the formation of the solutions of period two, which demonstrate a pulsating behavior consisted of dual cycle with different amplitude. The solution of period two is demonstrated in the inset (b) in Fig. 3. For this case the additional local extrema of the peak radical concentration Y_{Rmax} are also shown with «x» symbols in bifurcation diagram. When V is around 1.79, a drastic change in the dynamics of flame oscillations is observed: FREI emerges in hard excitation regime from the diffusional-thermal pulsating branch as a result of the subcritical bifurcation. This transition manifests itself as a sudden jump of the amplitude and period of oscillations. Comparing the phase diagram in Fig. 3 (a-c), the difference between two regimes of flame pulsations becomes obvious. For diffusional-thermal pulsations the flame position and local maximum of Y_R oscillate with limited and relatively small amplitude near the average values corresponding to the stationary normal flame solutions. The variation of x_f is comparable to the flame thickness. In FREI regime, the extend of x_f variation is governed by the length scale of the temperature gradient on the wall of reactor i.e. $1/\eta$. During the extinction stage, flame temperature, normal velocity and Y_R dramatically decrease and flame is quenched by cold wall of the tube. Furthermore, it is blown-off to the hot part of the reactor with velocity close to the fresh mixture velocity V, i.e., x_f increases (bottom-right part of Fig. 3 (c)). As the flame moves downstream, the local temperature begin to rise due to heat exchange between gas and tube wall and due to the presence of residuals of radicals, the branching reaction is triggered on, Y_R starts to build-up and recombine, leading to the further increase of local temperature, flame ignition and upstream motion of the combustion front. It is interesting to note that a trace of diffusional-thermal pulsations is still present in Fig. 3 (c) as a small loop in the left wing of the diagram even in FREI regime.

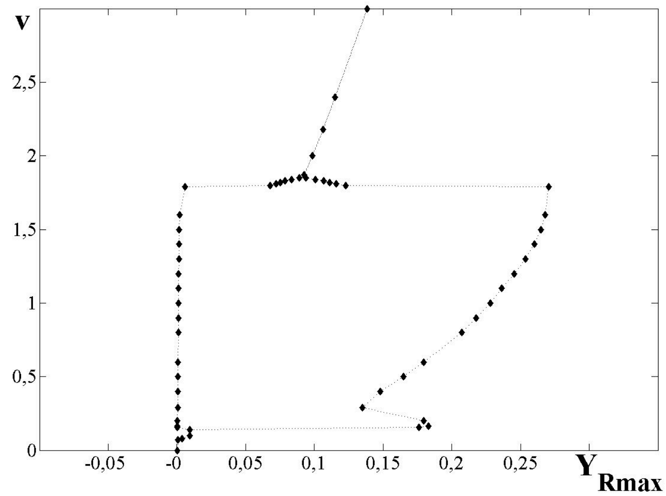

Fig. 4. Bifurcation diagram in the V vs Y_{Rmax} parameter plane for $\phi = 7.4$.

Figure 4 shows the full-scale bifurcation diagram in the plane of parameters V versus Y_{Rmax} for $\phi = 7.4$. For large and small flow velocities the normal and weak flame branches are clearly seen. The normal flame branch is replaced by the diffusional-thermal pulsations as a result of the supercritical Andronov-Hopf bifurcation. This flame regime occupies relatively narrow range of flow velocities from 1.8 to, 1.85, below which at $V \sim 1.8$, the FREI regime emerge due to subcritical bifurcation. The other subcritical bifurcation takes place at low values of V and causes the transition from FREI to weak flame branch.

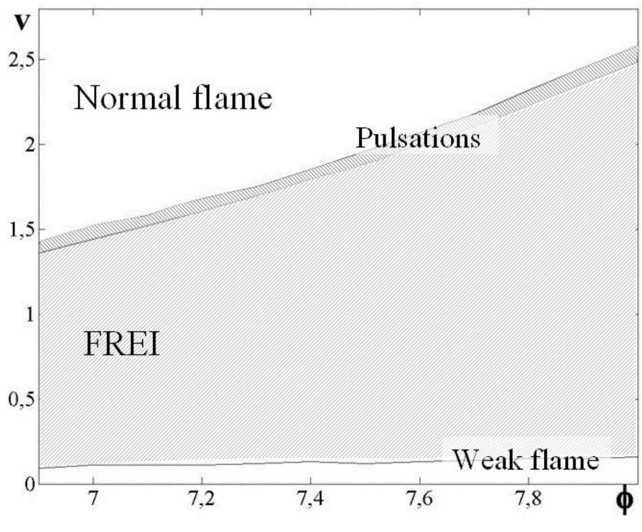

Fig. 5. Diagram of different flame regimes in the V vs. ϕ plane of parameters.

Figure 5 summarized the regions of existence of different combustion regimes in the plane of parameters V versus the equivalence ratio. The diffusional-thermal pulsations present in the narrow strip near the upper boundary between the normal flame and FRIE regimes. In terms of freely propagating flames, the diffusional-thermal instabilities are also predicted near the flammability boundaries of rich hydrogen-air flames. However instead of FREI, there is just flame extinction in this case. Thus we can expect that the regime of diffusional-thermal pulsations in micro flow reactor studied in this paper is the direct manifestation of diffusional-thermal instabilities of combustion waves with large Lewis numbers. The weak flame regime is caused by the flame-wall interaction has no analogies with freely propagating combustion waves. Interestingly, we have not found the diffusional-thermal pulsations near the boundary of weak flame and FREI regimes.

4 Conclusions

In this work we study the diffusive-thermal instabilities of rich hydrogen-air flames in micro flow reactor with controlled temperature profile. The main conclusions are as follows.

1) It is demonstrated that pulsating flame regime emerge as a result of diffusive-thermal instability via the supercritical Hopf bifurcation near the turning point for the normal flame regime.
2) The qualitative difference between FREI and diffusive-thermal pulsations is identified. It is shown that for the latter regime, the radical concentration remains finite and oscillates with certain amplitude near the average value corresponding to normal flame, whereas for the FREI regime, the radical concentration vanishes during the extinction stage and reappears during the ignition stage of the process.
3) The FREI regime emerge due to the subcritical bifurcations either from the diffusive-thermal pulsations or directly from the weak flame regime. The critical parameter values for the existence of FREI and diffusive-thermal oscillation regimes are found in terms of equivalence ratio and flow velocity.

Acknowledgments

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