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FLAME PROPAGATION OVER LIQUID ALCOHOLS
Part III. Pulsating regime

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The pulsating regime of flame spreading liquid fuels has been experimentally characterized. The mechanism that produces this oscillating behavior has been proposed, that correlates very well with our experimental data. The existence of a preheated region preceding the flame has been found; the characteristic horizontal length has also been experimentally measured. The transition temperatures have been found to possess common features for all fuels and geometrical configurations used in our experiments that can be used to improve fire safety in fuel containers.

Keywords: *combustion, flame spreading, fuels, heat transfer, temperature*

Introduction

Pulsating flame spreading over liquid fuels is a well known phenomenon [1]) whose mechanism is still now a controversial subject [2–4]. Recently, by interpreting to the plot of the flame spreading velocity *vs.* the initial temperature of the liquid fuel (T_0) in a long channel as a bifurcation diagram, we have identified the appearance of five different spreading regimes:

- for $T_0>T_1$, being T_1 a critical temperature of the system, flame spreading (v_f) is uniform, almost constant and of order 100 cm s^{-1}
- for $T_2<T_0<T_1$, where T_2 is a second critical temperature of the system, flame spreading is still uniform; flame velocities oscillate from 100 cm s^{-1} (for values of T_0 close to T_1) to 15 cm s^{-1} (for values close to T_2); the slope of the T_0–v_f diagram is of order 10 cm s^{-1} °C^{-1}
- for $T_3<T_0<T_2$, flame spreading is uniform, but in this case the slope is of order 1 cm s^{-1} °C^{-1}
- for $T_4<T_0<T_3$, a pulsating regime appears. By further decreasing the initial fuel temperature, $T_0<T_3$, the non-linear character of the spreading velocity oscillations becomes more pronounced, leading to relaxation oscillations with increasingly oscillation period, that diverges for $T_0=T_4$
- finally, for $T_0<T_4$, a new slow motion uniform regime is observed. Flame velocity is almost constant, of order 1 cm s^{-1} (pseudo uniform regime)

The critical transition temperature T_1 has been characterized as a steady state transition point, while T_2 is a transcritical bifurcation; T_3 corresponds to a Hopf bifurcation, and T_4 is a homoclinic connection. The analysis of recent experiments, carried out in our laboratory with aliphatic alcohols, leads us to propose a universal criterion for the beginning of the pulsations, as well as to advance a qualitative explanation for the mechanism of these oscillations.

Liquid phase preheating

When the liquid fuel temperature is below T_2, flame spreading proceeds thanks to the increase in the fuel vapour pressure, produced by the liquid phase preheating. The preheating of the fuel is mainly due to radiation and heat conduction from the flame through the gas phase. Below the flame anchoring position a large temperature difference is induced in the liquid phase, close to T_b–T_0, with T_b being the fuel boiling temperature. The surface temperature difference extends, in the liquid phase, over a length of the order *L*. This length that can be estimated in our experiments. Using a surface thermocouple measurement technique, an approximate value of *L* as a function of T_0 is given in Fig. 1. This case corresponds to a flame spreading over ethanol in a 100 cm long channel. No increase has been detected for $T_0>T_2$. For $T_3<T_0<T_2$ a small preheated region is observed, of order 1 cm; this region is even bigger in the pulsating region (up to 10 cm) and it is close to 15 cm in the pseudo uniform regime. Although the measurement technique used has a limited accuracy, it is a clear proof of the existence of a preheated region ahead of the flame.

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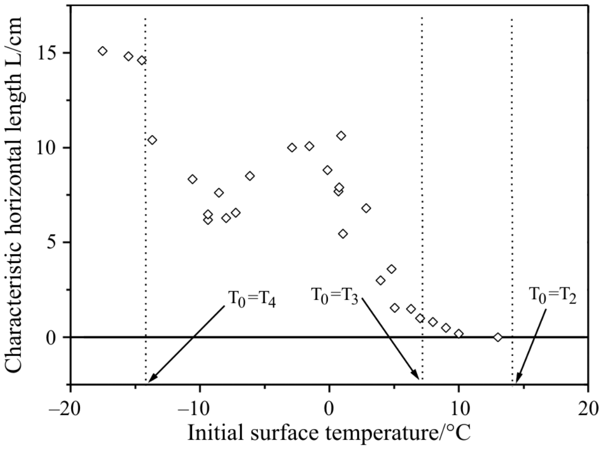

Fig. 1 Approximated horizontal length L of the vortex *vs.* initial temperature

For values of T_0 close to T_2 (and for bigger values too) the thickness of the preheating zone can not be detected with our experimental setup; it should be of the same size of the flame thermal thickness in the gas phase, $d_f=\chi_g/v_f$, where χ_g is the gas thermal diffusivity and v_f is the flame propagation velocity.

The temperature gradients generate stresses at the fuel surface due to the induced gradient in the surface tension (σ). The balance of these surface stresses by viscosity can lead to surface currents in the liquid fuel of characteristic velocity [5] given by:

$$u_s = \left(\left| \frac{d\sigma}{dT} \right| \frac{\Delta T}{2\sqrt{\rho \mu L}} \right)^{2/3} \qquad (1)$$

where ΔT is the liquid surface temperature difference established along the distance L, ρ is the fuel density and μ its dynamic viscosity.

Transition temperature T_2

For temperatures close to T_2, $\Delta T \approx T_b - T_0$ and $L \approx d_f$; then Eq. (1) leads to the law $u_s \propto v_f^{1/3}$. This relation provides a crossover value such that for larger values of the flame propagation velocity (corresponding to $T_0 > T_2$) the induced surface current velocity is unable to generate detectable capillary flows preceding the flame. On the contrary, when the fuel temperature is decreased so as to have flame propagation velocities smaller than the crossover value, the capillary flow can surpass the flame anchoring position and generates a thermocapillary recirculating region of warm liquid, preceding the flame that enhances flame spreading. We are then led to consider the speed ratio

$$S \equiv u_s / v_f = \left(\left| \frac{d\sigma}{dT} \right| \frac{\Delta T}{2 v_f \sqrt{\rho \mu \chi_g}} \right)^{2/3} \qquad (2)$$

as a control parameter for describing the transition from the purely diffusive spreading regime (controlled by the gas phase, like in the solid fuel case) to the assisted regime, where thermocapillary convection brings hot fuel ahead of the flame that enhances its spreading. We expect this transition to occur when some critical value S_2, of order unity, is reached for $T_0=T_2$.

Table 1 shows the critical fuel temperatures T_2 and flame spreading velocity for four aliphatic alcohols, measured in flame spreading experiments conducted in a 40 cm long channel, with two different lateral walls (duralumin and Pyrex, respectively). The second and the fourth lines of Table 1 correspond to the critical value obtained for the parameter S_2 in each case. It is worthwhile to notice that the four alcohols exhibit very close values of S_2, being the average value 1.05. The experiment uncertainties (mainly due to the influence of room air disturbances) are enough to explain the observed dispersion.

This result supports the existence of a universal critical value S_2 at which thermocapillary convection assistance starts. This critical value would depend on other parameters, such as the heat and chemical species looses, gravity effects, etc., but for a given experimental configuration S_2 must be a constant, the same for all the fuels.

Transition temperature T_3

Thermocapillary convection assists flame spreading for $T_3<T_0<T_2$. As we approach T_3, this enhancement is stronger and, finally, overstability may occur (and it does indeed!). For $T_0=T_3$, the assistance becomes unstable, producing an oscillatory behavior. Table 2 shows the experimental values of the parameter S for this critical temperature. All the results are of the same order (the mean value is in this case 3.4) but we cannot conclude that this critical value S_3 is the same for all the fuels and experimental configurations.

Table 1 Critical values of S_2 for aliphatic alcohols in a 40 cm long channel

Lateral walls		Methanol	Ethanol	Isopropanol	Butanol
Aluminium	T_2	9.3	9.5	17.5	36.1
	S_2	1.15	1.05	0.80	1.10
Pyrex	T_2	9.7	14.7	17.5	35.8
	S_2	1.25	1.10	0.9	1.05

Table 2 Critical values of S_3 for aliphatic alcohols in two different channels sizes

Lateral walls		Methanol	Ethanol	Isopropanol	Butanol
40 cm long channel	T_3	–1.9	3.5	5.9	24.1
	S_3	4.03	3.32	3.12	4.05
100 cm long channel	T_3	–1.0	5.8	7.8	32.2
	S_3	3.04	3.08	3.15	3.11

Strongly non-linear pulsations

Far away from the Hopf bifurcation point, non-linearity leads to relaxation oscillations in the spreading velocity. The period *P* of these oscillations can be calculated *A* typical plot in this region is shown in Fig. 2 (ethanol, 100 cm long channel). As we can see, there is a slow motion (induction period τ). Following this induction time, we can observe a quick motion interval, which is almost constant, of order 0.2 s (and never exceeds 1 s). Therefore, the induction period corresponds, mainly with the pulsation period.

The pulsating spreading has been accounted for by Di Blasi *et al*. [3], in a numerical integration of the corresponding governing equations; however, no explanation exists for the physical mechanism of such oscillations.

A mechanism can be argued based on the existence of a flame-to-liquid quenching distance, d_q. The phenomenological relation $d_q=a/v_f$ that links this distance to the flame velocity by means of some diffusivity coefficient, *a*, has strong experimental and (indirectly) theoretical support ([5] p. 269 and 284).

On the other hand, the flame velocity, being proportional to some power of the local fuel concentration, is expected to be a decreasing function of the height over the liquid surface. The intersection points of the graphs of these two relations determine the allowed pairs of values for the position and velocity of the flame leading edge. Usually, two intersection points appear, being unstable, corresponding to the upper position and stable the lower one. So, the position of closest approach to the fuel surface should be the one taken by the flame. When the regime is strongly supercritical, the generation of the preheated fuel region by capillary flows distorts the fuel concentration profile in the gas phase and a fuel enriched layer develops above the preheated region. The enriched layer will thicken in time as the fuel vapour diffuse upwards and eventually will intersect the quenching curve in two new points, allowing for two new flame positions, a stable one corresponding to a minimum flame-fuel distance but a larger velocity v_f^{max} and an unstable position, at the border of this enriched layer. This border will move upwards by diffusion, until it reaches the actual flame position. After the collapse, both intersection points disappear and the only remaining possibility for the flame is to reach the high velocity state corresponding to v_f^{max}. Then, the flame will advance quickly through the enriched region until, approaching the border, the fuel profile returns back to the initial shape that only allows the slow flame spreading regime v_f^{min}, and the cycle restarts.

Thus, in this scenario, the time needed by the enriched fuel layer to diffuse vertically upwards up to reach the quenching distance corresponding to the initial stable flame position (the induction period, which corresponds mainly to the pulsation period) that will be given by

$$\tau \approx C_3 \frac{d_q^2}{D} = \frac{C_3 a^2}{D(v_f^{min})^2} \tag{3}$$

where *D* is the diffusion coefficient of the fuel and C_3 some characteristic constant. The resulting formula provides a very good correlation of the experimental results as can be seen in Fig. 3. This plot corresponds to ethanol (100 cm long channel); the dotted line is the correlation given by Eq. (3) with C_3a^2/D=2.163.

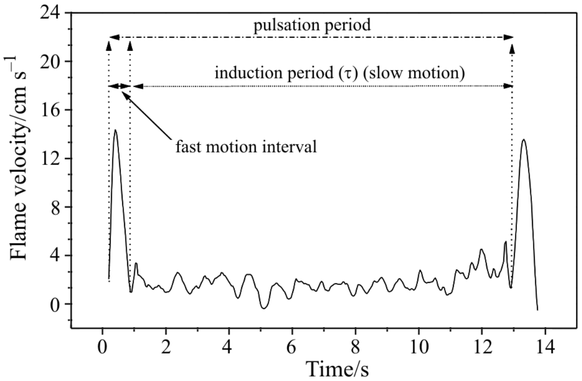

Fig. 2 Pulsation period (ethanol, 100 cm long channel)

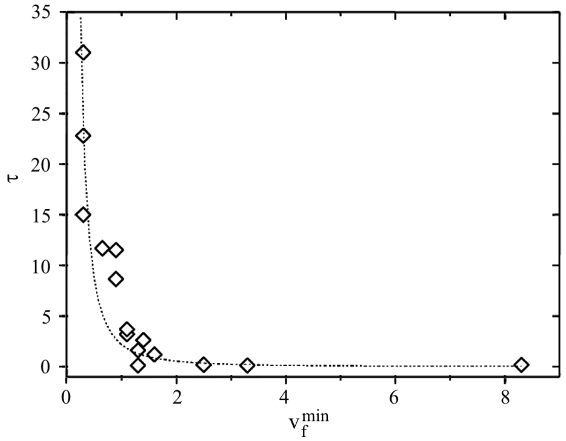

Fig. 3 The induction period along the oscillatory branch

Equation (3) also gives a first estimate of the critical value of the pulsation period at the onset of the oscillations, by taking $v_{\mathrm{f}}^{\min} = v_{\mathrm{f2}}$ (flame velocity for T_0=T_2).

Conclusions

The beginning of the assistance through convection to flame spreading over liquid fuels has been shown to possess universal features, corresponding to the transcritical bifurcation. The relevant control parameter has been defined and the critical value has been experimentally measured for four aliphatic alcohols. Also, a physical mechanism for the occurrence of the oscillations has been proposed and the predicted divergence of the pulsation period with the spreading velocity has been successfully tested.

Acknowledgments

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