



Soviet-era science, translated into English

Physical Chemistry

E. S. Semenov and A. S. Sokolik

1962

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Abstract

Full Text

Physical Chemistry

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Characteristics of Spherical Flames in the Formation Stage

(Presented by Academician V. N. Kondrat'ev, March 15, 1962)

There is an obvious contradiction in current ideas about the formation stage of spherical flames. On the one hand, during combustion in a closed vessel, within considerable limits of variation of the flame radius—up to 0.4 of the bomb radius—the rise in pressure and, correspondingly, the temperature of the fresh mixture is so small that at this stage one might suppose all flame characteristics to remain unchanged: the burning velocity and temperature, the width of the combustion zone, etc. Accordingly, in all existing thermal theories of spark ignition it is assumed that immediately after spark ignition a spherical combustion kernel is formed with all the characteristics of a stationary plane flame front (¹, ²) (see also (³), § 15). On the other hand, there are direct observations showing the gradual establishment of the stationary burning velocity, and indirect observations giving grounds to suppose the existence at this stage of a reduced flame temperature, in comparison with the adiabatic combustion temperature (⁴, ⁵) (see also (³), p. 239 ff.). This makes it necessary to establish, by direct observational methods, how the flame characteristics change in the stage of its formation, down to the minimum distance from the ignition point.

Fig. 1. Oscillograms of ionization current recorded at a probe potential of 2 V at distances from the ignition point (in mm): 1–30, 2–25, 3–20, 4–15, 5–10.

The experiments were carried out with a propane-air mixture of composition 4.16% C₃H₈ at $P = 250$ mm Hg, in a spherical bomb of radius $R = 90$ mm, with ignition at the center by a condenser discharge at a voltage of 9.5 kV and an energy of 35–50 millijoules. The flame velocity was determined by the Schlieren cinematography method through plane-parallel glass windows of the bomb, with a filming frequency of about 1500 frames/sec. With the aid of a single-electrode probe, the design of which was described earlier (⁶), and an OK-24 MKB cath-

Fig. 2. Concentration profiles.

Figure 2: Fig. 2. Concentration profiles.

ode oscillograph, oscillograms of the ionization current in the passing flame were recorded at various probe voltages. Intersections of these oscillograms at different instants of time (as shown in Fig. 1) gave probe characteristics, on the basis of which the profile of ion concentration with time $N(t)$ was determined; this profile could be recalculated, with the aid of the visible flame velocity U_v , into a profile with respect to distance $N(x)$. As the origin for measuring the width of the reaction zone, the point at which the current curve separated from the zero line was taken.

To eliminate the influence on the potential of the flame plasma of the residual voltage on the ignition electrode, the spark condenser was discharged to zero immediately after the closing spark was applied, by means of a spring contact device. At radii of less than 10 mm, removal of the residual voltage had to be carried out in a time of less than

2-3 msec. For this purpose a chopper was developed, actuated by a spring that was released by a trigger electromagnet. A comparison of the oscillograms themselves (Fig. 1), recorded at the same probe potential, equal to 2 V, at different distances r (mm) from the ignition point, already shows that, as the center of the bomb is approached, the oscillograms are stretched in time, with a noticeable decrease in the maximum ionization current. The corresponding concentration profiles $N(x)$ are shown in Fig. 2*. For each flame section, in analogy with the heat- and mass-balance equation, one may write an ion-balance equation for the processes of ion formation (q), diffusion $D \left(\frac{\partial^2 N}{\partial r^2} + \frac{2}{r} \frac{\partial N}{\partial r} \right)$ and convective transport $U_1 \frac{\partial N}{\partial r}$, and recombination (ρN^2), where D is the coefficient of ambipolar diffusion of the ions, U_1 is the local gas velocity corresponding to the temperature in the given section, and ρ is the recombination coefficient:

Fig. 2. Concentration profiles. $a-r = 30$, $b-r = 25$, $v-r = 15$, $g-r = 10$ mm

$$\frac{\partial N}{\partial t} = q + D \frac{\partial^2 N}{\partial r^2} - \left(U_1 - \frac{2D}{r} \right) \frac{\partial N}{\partial r} - \rho N^2 \text{ [cm}^{-3} \cdot \text{sec}^{-1}\text{]}. \quad (1)$$

Details of the procedure for determining the quantities D , U_1 , and ρ are given in (6). In the present case, Eq. (1) is applied to what is essentially a nonstationary combustion regime—the stage of flame formation. However, even in this case, over approximately 0.8 of the pulse length, $\left(\frac{\partial N}{\partial t} \right)$ is very small. Thus, for example, at $r = 10$ mm $\left(\frac{\partial N}{\partial t} \right)$ is not more than 13%, and at $r = 15$ mm already less than 5%, of the value of the largest term on the right-hand side of

Fig. 3. Profiles of the ion-formation rate.

Figure 3: Fig. 3. Profiles of the ion-formation rate.

Eq. (1). On this basis one may set

$$\frac{\partial N}{\partial t} = 0$$

and determine q from the equation

$$q = \rho N^2 + \left(U_1 - \frac{2D}{r} \right) \frac{\partial N}{\partial r} - D \frac{\partial^2 N}{\partial r^2}. \quad (2)$$

Fig. 3. Profiles of the ion-formation rate: $a-r = 30$, $b-r = 25$, $v-r = 15$, $g-r = 10$ mm

It is also necessary to take into account the possible error due to using the temperature profile of a stationary flame in calculations of the characteristics of the flame zone at the stage of its formation, since, on the basis of our own experiments, one may suppose that at small radii both the maximum combustion temperatures and the temperature gradients are considerably reduced. But, as was shown earlier ⁽⁶⁾, the inaccuracy in determining the temperature profile T itself has only a relatively weak effect on the results of calculating the profiles N and q . The profiles of the ion-formation rate $q(x)$, calculated—

* Here, as in Fig. 3, x is the distance from the beginning of the ion-formation zone.

data from equation (2) using the curves of Fig. 2 are presented in Fig. 3.

At the present time it has been firmly established that, in hydrocarbon flames, the overwhelming part of the observed ionization is of nonthermal origin, i.e., that the thermally equilibrium ionization of the combustion products is several orders of magnitude lower than the maximum ionization in the flame front. This gives grounds for assuming that the width of the chemical-transformation zone in the flame should in any case not be less than the width of the ion-formation zone. Similarly, the maximum q in the flame should reflect the maximum reaction rate. In calculating the width of the reaction zone, the directly measured visible flame velocity U was used. As follows from Fig. 4, it decreases continuously as the ignition point is approached, while the width of the reaction zone increases, already at $r = 10$ mm exceeding the stationary value ($\delta_p \approx 1.2$ mm at $r \geq 35$ mm) almost tenfold. At the same time, the maximum rate of ion formation at $r = 10$ mm is reduced by a factor of 4-5. Although exact measurements of q at $r < 10$ mm are already impossible because of the difficulties of completely eliminating the influence of disturbances from the spark,

Fig. 4

Figure 4: Fig. 4

nevertheless semi-quantitative observations indicate a further increase of δ_p as the ignition point is approached.

Fig. 4

It follows from these observations that, in the initial phase, the width of the reaction zone is comparable with the radius of the flame sphere, i.e., the spherical flame here represents a source of volumetric reaction. But when the width of the reaction zone is comparable with the radius of curvature of the flame, the volume of reacting gas from which heat is drawn is noticeably smaller than the volume of gas to which this heat is transferred; this must lead to a lowering of the combustion temperature, as compared with the adiabatic temperature of a plane flame, and, correspondingly, to a slowing of the burning velocity (see (7), p. 111). The smaller the radius of the sphere, the more significant this lowering of the combustion temperature. Thus, for example, at $r = 10$ mm the temperature of the combustion products is 250–300° lower than the temperature of a plane flame. Thus, only gradually, as the difference between the volumes of the heat-release and preheating zones is smoothed out, do the temperature and burning velocity approach their values in a plane flame. The stage of flame formation consists in a gradual transition from the initial source of volumetric reaction to a stationary plane flame (a sphere with negligible curvature). If earlier it was assumed that the effect of curvature appears for dimensions of a spherical flame comparable with the width of the stationary flame front, i.e., at $r < 1$ mm, then, as follows from the data obtained, this effect persists up to considerably larger flame dimensions, under the present conditions up to $r \approx 30$ mm.

An additional lowering of the temperature in the flame may also occur as a result of heat transfer to the ignition electrodes, but, as calculation shows, the magnitude of this heat removal does not exceed 3% of the heat release in the combustion source. The insignificance of this factor in lowering the temperature in a spherical flame is also confirmed by the fact that experiments carried out with electrodes of diameter 1.5 and 0.4 mm gave practically identical values of the width of the reaction zone and of the flame propagation velocity in the stage of its formation. The lowering of the temperature caused by the curvature of the spherical flame may be significantly intensified by secondary-

retarding effect of such stages as the oxidation $\text{CO} \rightarrow \text{CO}_2$. This should be the subject of a special investigation.

Institute of Chemical Physics
Academy of Sciences of the USSR

Received

16 I 1962

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