SOME CONTRIBUTIONS TO LAMINAR FLAME THEORY*

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1. INTRODUCTION

The fundamental problem of laminar flame theory consists of determining the structure and properties, specially the propagation velocity, of a combustion wave which advances through a homogeneous combustible mixture at rest, of given thermodynamic state and chemical composition.

In recent years important progress has been accomplished in the study of this problem, including its formulation, mathematical methods of solution, application to specific cases and development of experimental techniques.

Though a significant effort has also been devoted to the study of other problems of the theory, such as those of quenching, internal stability of the wave, ignition and flammability limits, the progress realized in them has been, in general, considerably smaller.

In particular, with respect to the existence of the inflammability limits, whose origin is yet unknown, in 1957 D. B. Spalding(1) proposed as their cause the heat losses which can occur in the flame either by convection or by radiation effects. The most important result of his work consisted in showing that such heat losses can produce two different propagation velocities for the flame which approach each other when the heat loss increases and finally coincide for a limit value of it, above which the combustion does not propagate through the mixture. According to Spalding, the point of coincidence of them would determine the burning velocity corresponding to the limit of inflammability of the mixture while the lower of both velocities cannot usually be observed because it is unstable. Similar results have been also obtained by von Kármán and Penner(2), for a simplified model in which the influence of diffusion is neglected and the rate of chemical reaction is constant; while Zeldovich and Barenblatt(3), starting from an unsteady state and by numerical integration of the flame equations, have also obtained a limit velocity determined by the heat loss, even when in their solutions the double velocity of Spalding does not appear. Finally,

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Hirschfelder[4] has shown lately that when the heat losses are localized in a porous stabilizer located at the cold limit of the flame, two propagation velocities are also obtained for each value of the heat transfered to the stabilizer and he has tried to relate this result with those of Spalding who, on his side, has published experimental measurements[5] which seem to confirm the real existence of both velocities.

The present work constitutes a theoretical analysis ot the problem, with the object of clearing, by means of a systematic formulation and discussion of the different cases considered, until what point the existence of both velocities depends on the choice of the boundary conditions or the use of the adequate parameters. The study is performed on a simplified flame model, easily integrable in exact form, with two unique chemical species, reactants and products. The effect of the diffusion as well as the influence of the concentration in the reaction rate is taken into account, but not, at least in a systematic way, that of the activation energy. However, qualitative conclusions can be deduced about the influence of the same, on the basis that its value does not alter the multiplicity nor the properties of the solutions.

The cases considered in this study and the results obtained are the following:

1. Flame with heat loss localized at the stabilizer. It is shown that the two velocities of Hirschfelder reduce to only one, by means of the choice of the adequate parameter, pointing out the apparent contradiction between Spalding's experimental results and the theoretical conclusions.

2. Flame with distributed heat losses. It is shown that the two velocities of Spalding reduce to only one by varying slightly the boundary conditions at the hot limit, both when an ignition temperature at the cold boundary is assumed as well as when the porous stabilizer of Hirschfelder is used. This result is particularly significant when the activation energy is different from zero, because then such modification is made imperceptible and the lower of both velocities is very small.

3. Finally, as a new cause of disturbance, for the same flame model, the effect of a dilution of the mixture produced by the lateral diffusion of the active species is considered, obtaining the result that the dilution diminishes the flame velocity, which vanishes for a limit value of the lateral diffusion coefficient.

2. ADIABATIC FLAME

With the conventional assumptions[6] and the notation which is specified in the Annex and assuming a first order chemical reaction rate n of the Arrhenius type:

$$n = We^{-\theta_\alpha/\theta}(1-Y) \tag{1}$$

the system of differential equations for an adiabatic flame, written in dimensionless form and referred to a coordinate system ξ which propagates with the flame is the following:

(a) Energy equation:

$$\frac{d\theta}{d\xi} = \theta - 1 + (1-\theta_0)(1-\varepsilon) \tag{2}$$

(b) Diffusion equation:

$$\frac{dY}{d\xi} = L(Y-\varepsilon) \tag{3}$$

(c) Reaction equation:

$$\frac{d\varepsilon}{d\xi} = \frac{1}{\varphi^2} e^{-\theta_\alpha \frac{1-\theta}{\theta}} (1-Y) \tag{4}$$

In this last equation,

$$\varphi \equiv m \sqrt{\frac{C_p}{\lambda W e^{-\theta_\alpha}}} \tag{5}$$

is an unknown parameter which measures the velocity of propagation of the flame in dimensionless form.

The problem consists in determining the eigenvalue of φ which makes compatible all the boundary conditions that must be satisfied by these equations at its cold and hot limits, as well as the solution of the system corresponding to this value.

As it is known[6], for solving this problem it is neccessary to assume the existence of an ignition temperature θ_i, greater than that of the cold gas θ_0, such that the rate of the chemical reaction be zero for values of θ lower than θ_i. If the origin of distances is chosen at the point where $\theta = \theta_i$ and the wave propagates in the negative direction then the above system of equations is only valid for the reaction zone of the flame $\xi > 0$, while for the heating zone $\xi < 0$, equation (4) must to substituted by the following:

$$\xi < 0, \quad \varepsilon \equiv 0 \tag{6}$$

so that in this region the variable ε disappears and the only unknown quantities are θ and Y.

Moreover, the solutions corresponding to both regions must join without discontinuity at the point $\xi = 0$ where the chemical reaction starts.

With these hypothesis, the boundary conditions of the system are the following:

For the reaction zone $\xi > 0$:

$$\left.\begin{array}{lll} \xi = 0, & \theta = \theta_i, & \varepsilon = 0 \\ \xi = \infty, & \theta = 1, & \varepsilon = Y = 1 \end{array}\right\} \tag{7}$$

For the heating zone $\xi < 0$:

$$\left.\begin{array}{lll} \xi = 0, & \theta = \theta_i & \\ \xi = -\infty, & \theta = \theta_\theta, & Y = 0 \end{array}\right\} \qquad (8)$$

The condition of continuity of Y at the origin is:

$$\xi = 0, \quad Y_i^- = Y_i^+ \qquad (9)$$

The continuity of θ and ε is automatically satisfied, due to the preceding boundary conditions.

The solution of such system determines a unique eigenvalue of φ for each value of θ_i.

Figure 1 shows, in the continuous line, the variation of φ with θ_i for a reaction with zero activation energy $\theta_a = 0$, in which case the complete solution of the system can be obtained in explicit from. In this figure, the

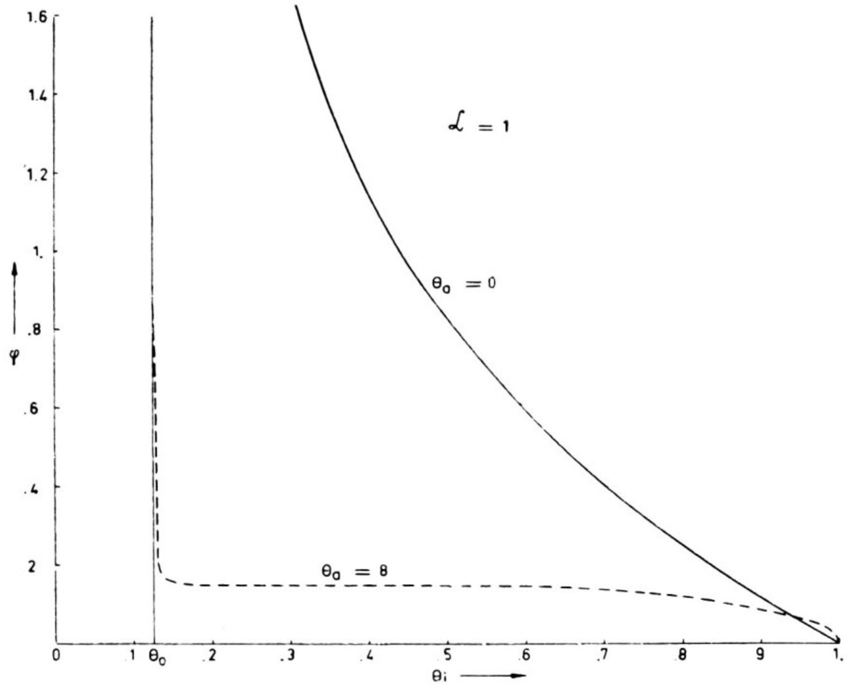

FIG. 1. Adiabatic flame. Variation of the burning velocity with θ_i for $\theta_a = 0$ and $\theta_a = 8$.

dotted line represents the solution corresponding to a typical value of the activation energy $\theta_\alpha = 8$, obtained by an approximate semi-analytical method. Here it can be seen that the activation energy does not alter the number of the solutions and it only does determine, among the infinite values of φ corresponding to the different values of θ_i, which is the adequate one. This value results in being independent of the unknown value of θ_i. Similarly, Fig. 2 shows the curves of variation of θ and Y as functions of ε, corresponding to these solutions.

The existence of ignition temperature can be substituted, like Hirschfelder has done[4], by that of a porous stabilizer located in the boundary of the flame $\xi = 0$, at the temperature θ_0 of the cold gas, which absorbs a given amount q of heat which substitutes in this model the unknown

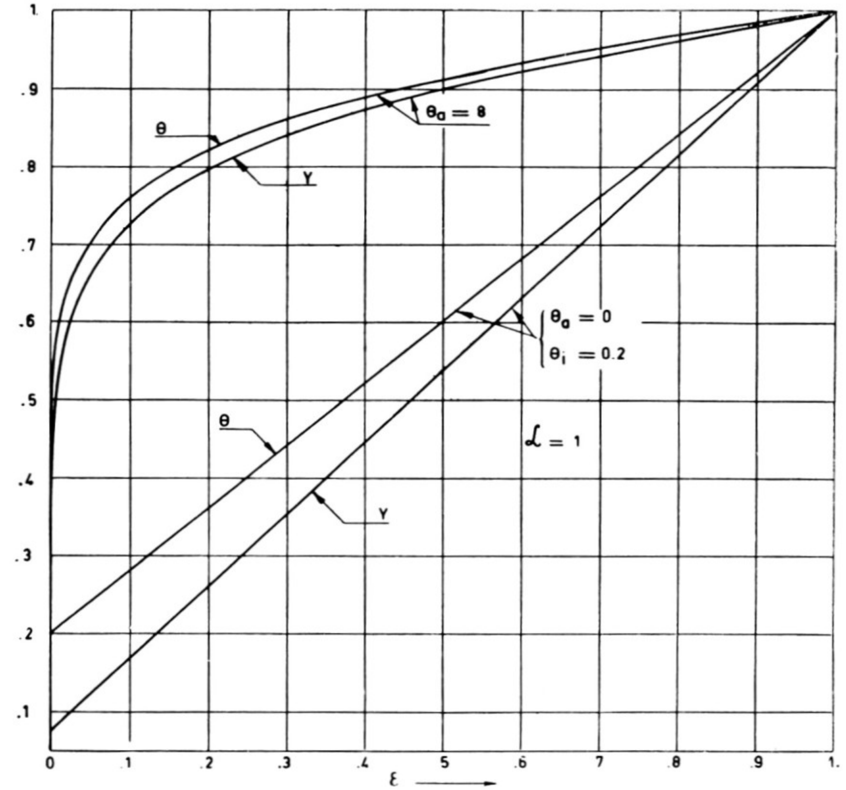

FIG. 2. Adiabatic flame. Variation of Y and θ with ε for $\theta_a = 0$ and $\theta_a = 8$.

ignition temperature. The conclusions obtained in this case are similar to the previous ones. In fact, when the heat transfered to the stabilizer is very small with respect to the heat released by the flame and the activation energy of the reaction has an appreciable value, the flame velocity takes a definite value, which coincides with that obtained in the model of the ignition temperatures and that, as in this, results in being independent of the amount of heat transferred to the stabilizer.

On the contrary, when the heat transfered to the stabilizer is appreciable, the velocity of propagation of the flame depends, naturally, on it. In this case, Hirschfelder[4] has shown recently that two different propagation velocities of the flame exist for each value of q, suggesting a close relation of this fact with the two velocities obtained by Spalding[1] and by von Kármán and Penner[2], when the heat losses are continuously distributed along the flame. In the following paragraph this case is studied and it is shown that the duplicity of solutions of Hirschfelder is due to an inadequate choice of the parameter of reference. Moreover, in the following paragraphs it is shown that this case and the one of heat losses distributed along the flame are essentially different, so that one of them cannot be justified by means of the other.

3. FLAME WITH HEAT LOSSES LOCALIZED AT THE COLD BOUNDARY

In this case, the equation (2) of the flame system must be replaced by the following:

$$\frac{d\theta}{d\xi} = \theta - 1 + (1-\theta_0)(1-\varepsilon) + \delta \tag{10}$$

the equations (3) and (4) remaining invariables.

In this equation the parameter δ is a dimensionless measure of the heat q transferred to the stabilizer per unit area and per unit time, defined by the following expression:

$$\delta \equiv \left(\frac{d\theta}{d\xi}\right)_{\xi=0} \equiv \frac{q}{m C_p T_{fa}} \tag{11}$$

It is evident that, instead of q, the significant parameter for the process is δ, since it measures the fraction of the heat released by the combustion which is transferred to the stabilizer. δ plays here the same role as θ_0 for the previous model.

The boundary conditions corresponding to the model of Hirschfelder are the following:

$$\left.\begin{aligned} \xi &= 0, \quad \theta = \theta_0, \quad \varepsilon = 0 \\ \xi &= \infty, \quad \theta = 1-\delta, \quad \varepsilon = Y = 1 \end{aligned}\right\} \tag{12}$$

The solution of the differential system also determines here, as in the case of θ_i, a unique value of φ for each value of δ. In particular, for zero activation energy, in which case the exact solution of the differential system can also be obtained in explicit form, the value of φ corresponding each value of δ is given by the following expression:

$$\sqrt{1+\frac{4}{\varphi^2 L}} = 1 + \frac{2}{L}\frac{\delta}{1-\delta-\theta_0} \tag{13}$$

Similarly to what was done in the case of the adiabatic flame, in Figs. 3 and 4 the value of φ as a function of δ and those of θ and Y as functions of ε respectively, are represented. Also here it can be shown that the presence of an activation energy different from zero does not alter the number of solutions and it only determines, as has been said before, the value of φ corresponding to the adiabatic model $\delta \leqslant 1$.

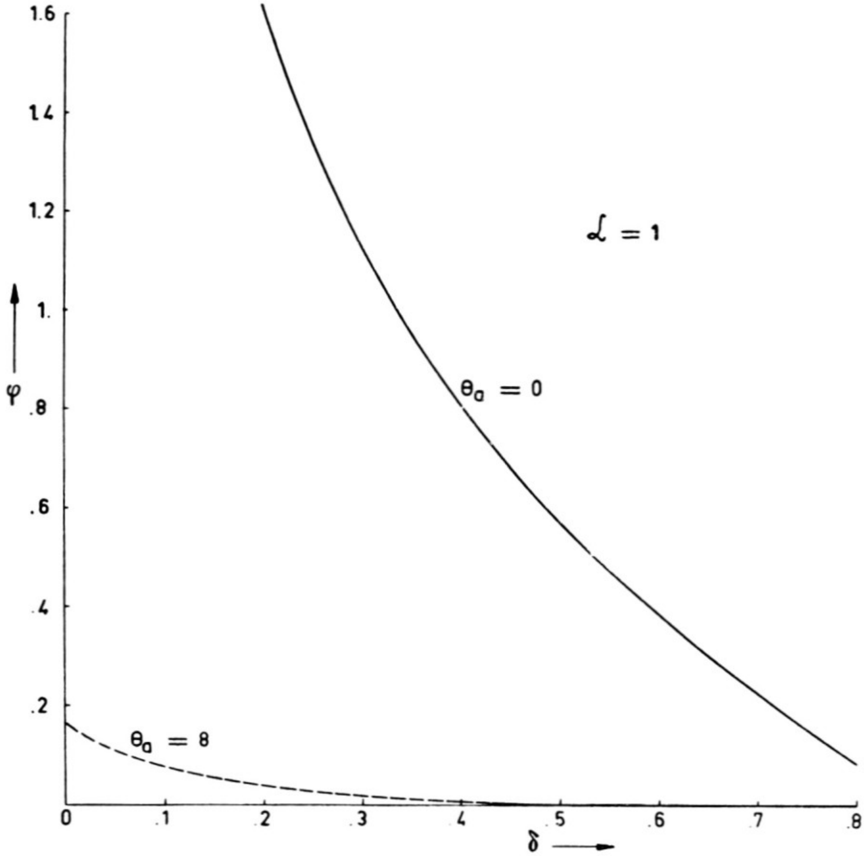

FIG. 3. Flame with localized heat loss. Variation of the burning velocity with δ for $\theta_a = 0$ and $\theta_a = 8$.

If we want now to obtain the two velocities of Hirschfelder from the preceding results it will suffice to represent φ not as function of δ, but as function of q, or of any dimensionless measure of it that does not depend on the propagation velocity of the flame. Such dimensionless measure independent of φ is the parameter γ defined by the expression

$$\gamma \equiv \frac{q}{T_{fa}\sqrt{\lambda C_p W}} \tag{14}$$

It is easily realized that between γ, δ and φ the following relation exists:

$$\gamma \equiv \delta\varphi \tag{15}$$

which allows us to express the results as functions of γ, from the previous solutions. Such results, for $\theta_\alpha = 0$, are given in Fig. 5, where it can be seen that the two values of the flame velocity corresponding to each value of the heat γ transferred to the stabilizer are a consequence of the two values of δ corresponding to each value of γ.

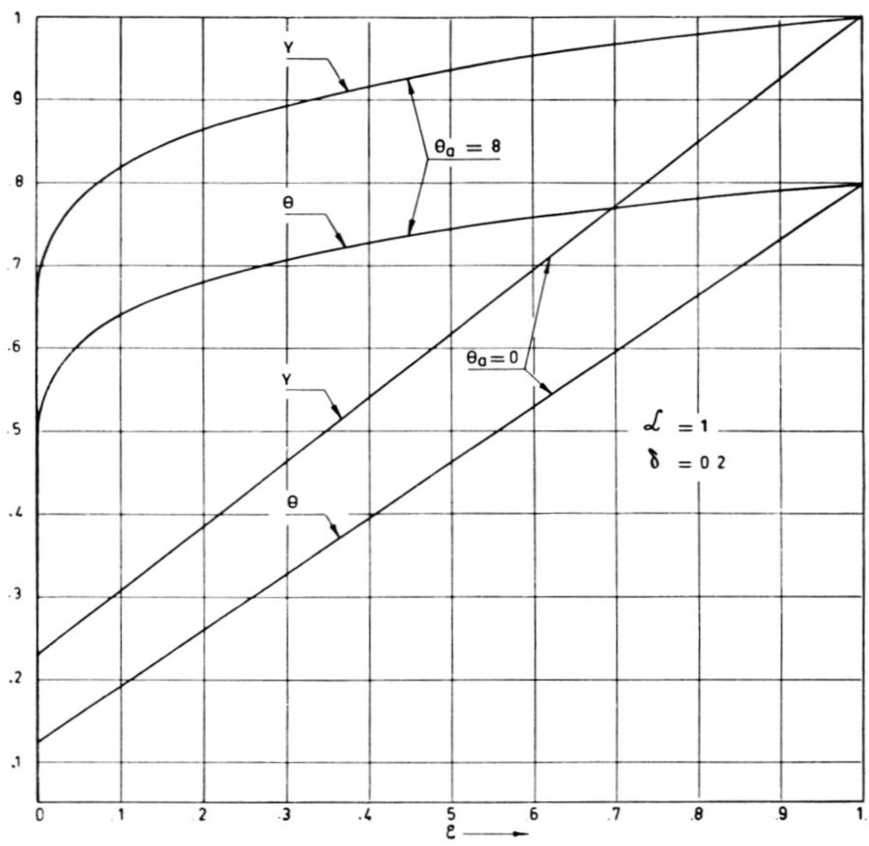

FIG. 4. Flame with localized heat loss. Variation of Y and θ with ε for $\theta_a = 0$ and $\theta_a = 8$.

Lately, Spalding[5] has published the experimental results of the measurements performed with a porous stabilizer which intended to realize physically the model of Hirschfelder. Apparently, he obtained two different propagation velocities of the flame for each value of the heat fraction transferred to the stabilizer. Essentially, his graphs, contrarily to those of Hirschfelder, are equivalent to a representation of φ vs. δ, so that the

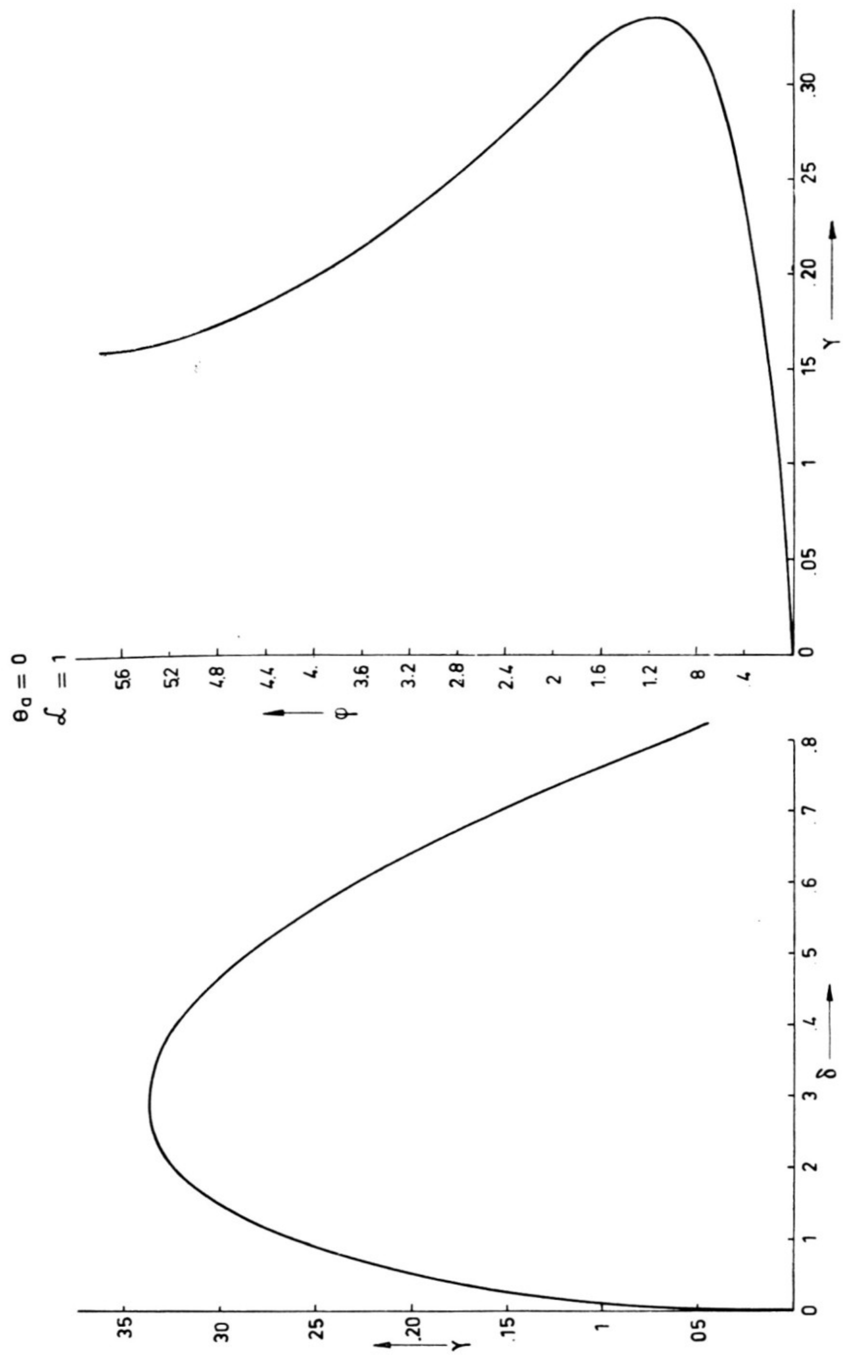

FIG. 5. Flame with localized heat loss. Explanation of the existence of two burning velocities.

two solutions obtained cannot be attributed to the action of the heat transferred to the stabilizer, since, in such case the solution should be unique, as it has been shown.

On the other hand, the two solutions which Hirschfelder obtains when the flame velocity is represented as function of γ have very little in common with the duplicity which results also in the case of the heat losses distributed along the flame, so that its experimental verification does not serve to justify the existence of two velocities in the case of distributed losses which, as it is shown in the following paragraph, is really very doubtful, since it suffices a small modification of the boundary conditions at the hot limit, imperceptible in the flames with appreciable activation energy, for the two solutions to be reduced to a single one.

4. HEAT LOSSES DISTRIBUTED ALONG THE FLAME

Let T_f be the final temperature of the flame and suppose that the local heat loss q_l, per unit length and per unit time, due, for example, to the lateral transfer of heat or to radiation, is given by the expression

$$q_l = k(T-T_f) \tag{16}$$

where k is a coefficient which is constant.

In this case, the energy equation of the flame system must be substituted by the following:

$$\frac{d\theta}{d\xi} = \theta - 1 + (1-\theta_0)(1-\varepsilon) + \frac{K}{\varphi^2}\int_{\xi_0}^{\xi}(\theta-\theta_f)\,d\xi \tag{17}$$

while the equations (2) and (3) remain invariables.

In equation (17), K is a dimensionless coefficient of heat loss, which is given by the expression

$$K \equiv \frac{k}{We^{-\theta_\alpha} C_p} \tag{18}$$

and ξ_0 is the point where the loss starts. For example, if the final temperature coincides with that of the cold gases, the only case considered by Spalding, and the loss takes place through all the flame, then is $\theta_f = \theta_0$, $\xi_0 = -\infty$.

As for the boundary conditions, those corresponding to the cold limit are the same for the adiabatic flame, while those of the hot limit must be substituted by the following:

$$\xi = \infty, \quad \theta = \theta_f, \quad Y = \varepsilon = 1 \tag{19}$$

As it results from (17), between θ_f and the total heat lost by the flame there exists the following relation:

$$\theta_f = 1 - \frac{K}{\varphi^2} \int_{\xi_0}^{\infty} (\theta - \theta_f)\, d\xi \tag{20}$$

Similarly to what occurs in the previous cases, also in this, when the activation energy is zero, exact solutions can be obtained in explicit form, which

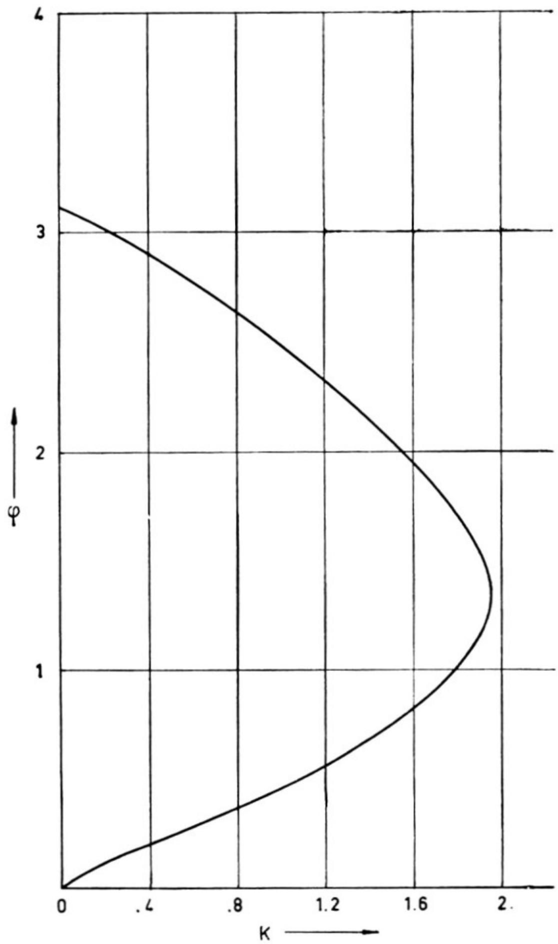

FIG. 6. Flame with distributed heat loss. Variation of the double burning velocity with K, for a final temperature equal to θ_0.

permits easy discussion of the results and extension of the conclusions to the case in which the activation energy is different from zero, since the value of θ_a does not alter, as it has been seen, the number of the solutions which result for $\theta_a = 0$. Such solutions are obtained below for the two cases of greater importance in which the final temperature coincides, respectively, with that of the cold gases θ_0, case of Spalding and von Kármán, and with the ignition temperature θ_i.

(a) $\theta_f = \theta_0$.—In this case, the solution of the system of the flame equations shows that for each value of the coefficient of heat loss K, two different values of the velocity φ exist, which are given by the equation

$$\frac{\frac{u}{2}\left[\sqrt{1+\frac{4K}{\varphi^2}}+1\right]+(L-1)u-L}{K+(L-1)u-L\sqrt{1+\frac{4K}{\varphi^2}}} = \frac{\theta_i-\theta_0}{1-\theta_0} \tag{21}$$

being

$$u = \frac{\varphi^2 L}{2}\left[\sqrt{1+\frac{4}{\varphi^2 L}}-1\right] \tag{22}$$

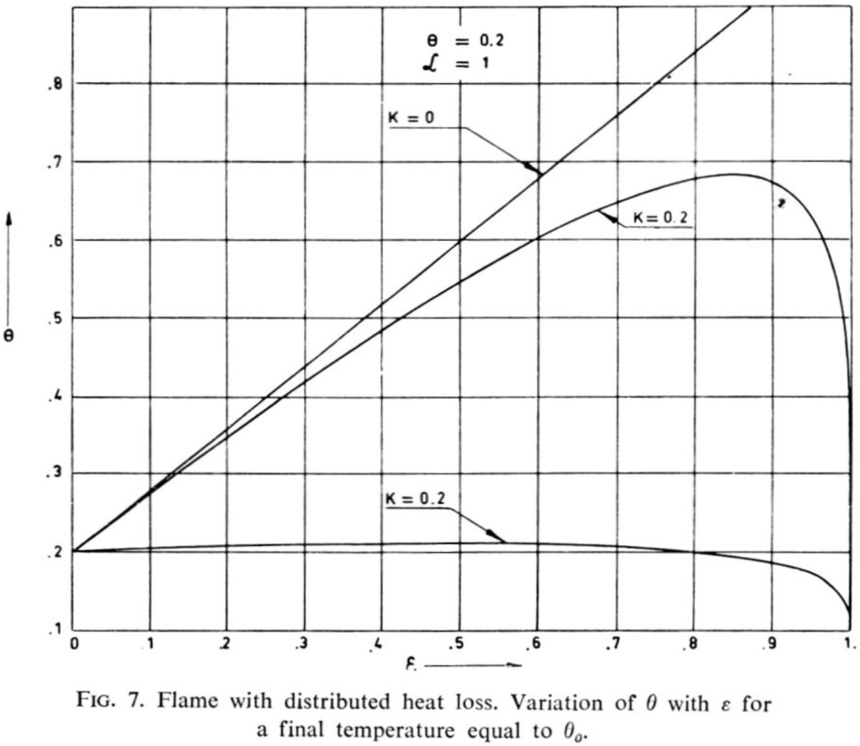

FIG. 7. Flame with distributed heat loss. Variation of θ with ε for a final temperature equal to θ_o.

Contrary to what happened when the heat losses were localized at the stabilizer, the two velocities corresponding to each value of the coefficient K cannot be reduced here to a single one by means of an adequate definition of this parameter. On the contrary, for obtaining a single velocity it would be necessary to assume that the losses take place according to a different law which would make the value of k dependent on that of m, that is, on the flame velocity.

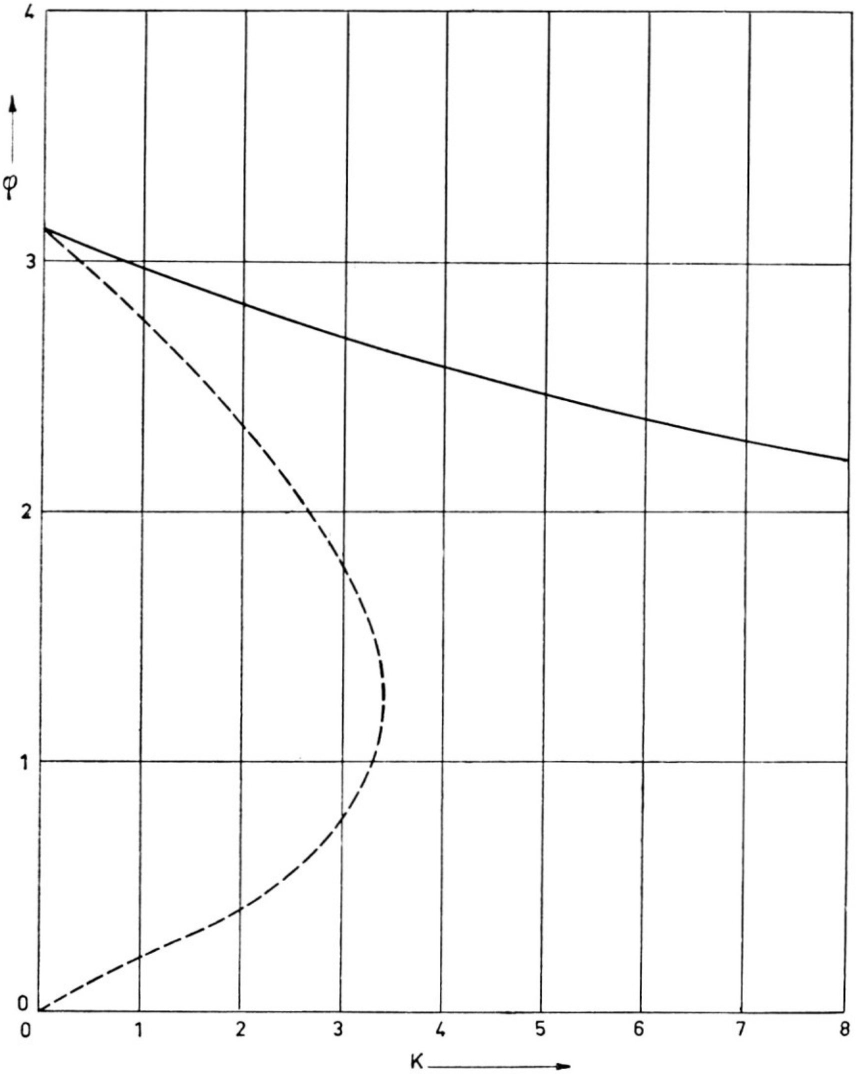

FIG. 8. Flame with distributed heat loss. Variation of the unique burning velocity with K for a final temperature equal to θ_i.

Figures 6 and 7 show the corresponding results for a typical case.

The previous conclusions are equally valid when the heat losses only take place in the reaction zone as well as when they occur through all the flame.

(b) $\theta_f = \theta_i$.—When the temperature of the burnt gases coincides with the ignition temperature, contrary to what occurs in the previous case, it results that a single propagation velocity of the flame exists for each value of the coefficient of heat loss. The value of this velocity is given by the system

$$\frac{\frac{K}{u}}{\frac{K}{u}-\left(1+\frac{u}{\varphi^2}\right)}\left[1-\frac{2\frac{u}{\varphi^2}}{\sqrt{1+\frac{4K}{\varphi^2}}-1}\right]=\frac{1-\theta_i}{1-\theta_0} \tag{23}$$

$$u=\frac{\varphi^2 L}{2}\left[\sqrt{1+\frac{4}{\varphi^2 L}}-1\right] \tag{24}$$

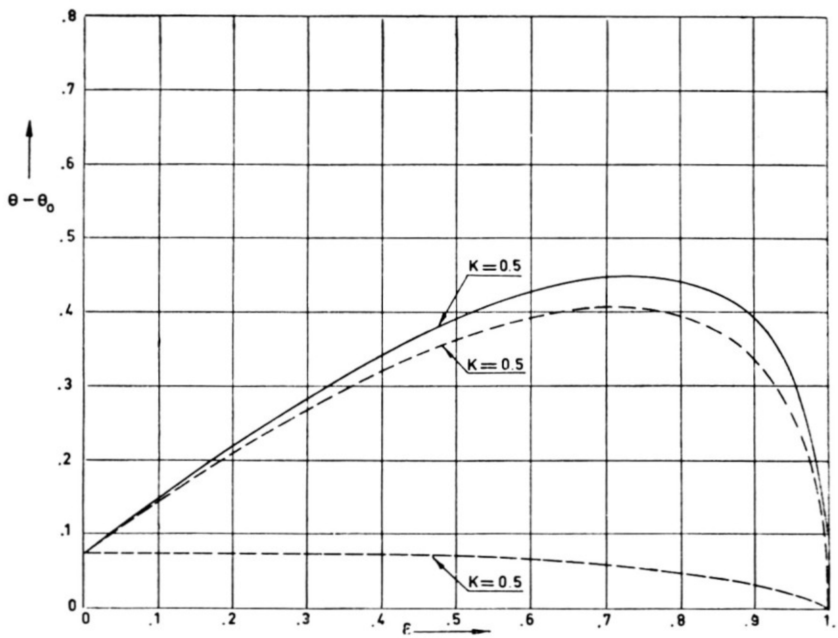

FIG. 9. Flame with distributed heat loss. Variation of θ with ε for a final temperature equal to θ_i.

Figures 8 and 9 show the corresponding results for this case. In them, the dotted lines show, for comparison, the two solutions corresponding to the case $\theta_f = \theta_0$.

The preceding conclusions exist wholly, as can be verified easily, when the diffusion effects are omitted as well as those of the influence of concentration in the reaction rate, by assuming it constant, as has been done in previous works already mentioned. Also, it can be proved that, in general, when $\theta_f < \theta_i$, two flame velocities exist, which reduce to only one for $\theta_f \geqslant \theta_i$.

Moreover when the ignition temperature condition at the cold boundary is substituted by the porous plug of Hirschfelder and distributed heat losses are assumed with final temperature θ_0 equal to the temperature of the plug, a single propagation velocity is obtained for each couple of values of δ and K.

The significance of these conclusions lies in the fact that a small variation in the boundary conditions at the hot limit, since θ_i can be chosen arbitrarily close to θ_0, reduce the two propagation velocities to a single one. Such conclusion reduces considerably the practical interest of the double solution, specially when the influence of the activation energy is taken into consideration. In fact, in such case, while the larger of the two velocities corresponding to each value of K for $\theta_f = \theta_0$ (actually the only one that has been experimentally observed with complete certitude) is independent of θ_i as occurs in the adiabatic case, this does not happen for the smaller velocity which must depend on the value assumed for the ignition temperature and vanishes when the final temperature is equa to θ_i.

5. FLAME WITH LATERAL DIFFUSION

A possible cause of disturbance in the behavior of the flame with respect to the ideal adiabatic solution considered in paragraph 2, which seems not to have been analysed yet, lies in the lateral diffusion of active chemical species which are substituted by the inert gases surrounding the flame, giving place to a dilution of the mixture.

This problem can be studied in similar form to the previous cases of heat losses, when the cooling term $k(\theta-\theta_f)$ of the energy equation is substituted by a term of lateral diffusion, whose action must be included as well in the reaction equation. If it is assumed, as in the case of distributed heat losses, that the local loss of active species by effect of the lateral diffusion, per unit length and per unit time, is proportional to its concentration $r(1-Y)$ where r is a constant coefficient, and if moreover, it is assumed that the activation energy is zero, in which case the exact solution of the problem can also be obtained, the equations of the flame for this case are the following:

(a) Energy equation:

$$\frac{d\theta}{d\xi} = \theta - 1 + (1-\theta_0)(1-\varepsilon) + \frac{1-\theta_0}{\varphi^2}\,\frac{1-\mu}{\mu}\int_0^{\xi}(1-Y)\,d\xi \tag{25}$$

(b) Diffusion equation:

$$\frac{dY}{d\xi} = L(Y-\varepsilon) \tag{26}$$

(c) Reaction equation:

$$\frac{d\varepsilon}{d\xi} = \frac{1}{\varphi^2\mu}(1-Y) \tag{27}$$

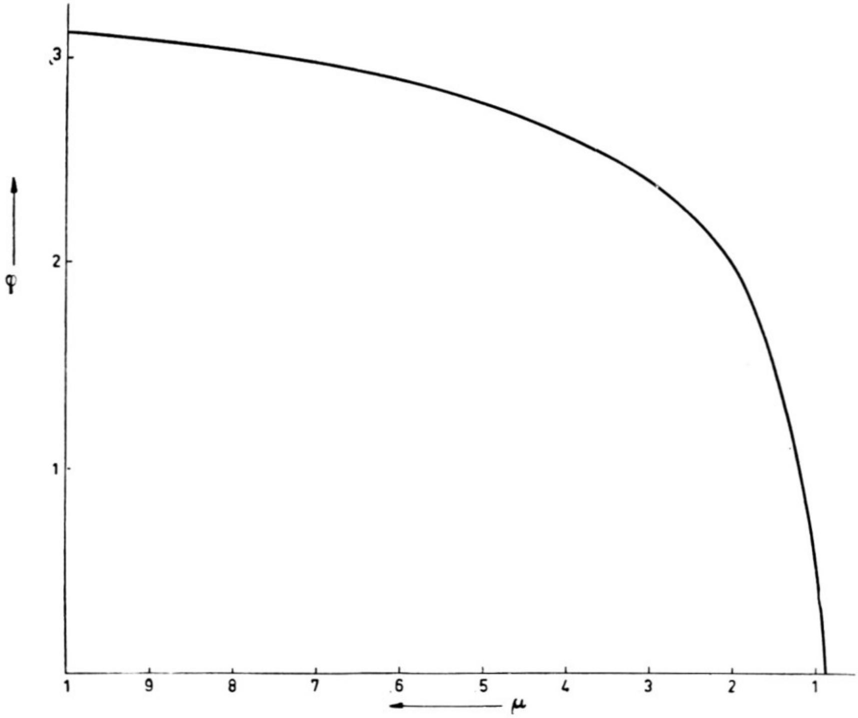

FIG. 10. Flame with lateral diffusion. Variation of the burning velocity with μ for $\theta_a = 0$.

In the equations (25) and (27), μ is a dimensionless measure of the effect of lateral diffusion, defined by the expression

$$\mu = \frac{W}{W+r} \tag{28}$$

The elimination of $(1-Y)$ between equations (25) and (27) allows the substitution of the energy equation by the following:

$$\frac{d\theta}{d\xi} = \theta-\theta_0-(1-\theta_0)\mu\varepsilon \tag{29}$$

The boundary conditions that must satisfy this system are the same as in the adiabatic case, except for the final temperature of the burnt gases, which, evidently, must be smaller than the adiabatic one, since part of the reactant is lost by lateral diffusion without being burnt. Such temperature θ_0 is related with μ by the expression

$$\theta_f = 1-(1-\theta_0)(1-\mu) \tag{30}$$

The solution on the differential system leads to the following expression for the flame velocity:

$$\varphi = \frac{2}{\sqrt{\mu L}}\left[\left(1+\frac{\frac{2}{L}}{\frac{1-\theta_0}{\theta_i-\theta_0}\mu-1}\right)^2-1\right]^{-1/2} \tag{31}$$

Consequently, it results that the lateral diffusion does not alter the unicity of the solution corresponding to the adiabatic case, but reduces the flame velocity when the diffusion increases. The velocity of the flame vanishes for a value of the coefficient of lateral diffusion which is given by the expression

$$\mu = \frac{\theta_i-\theta_0}{1-\theta_0} \tag{32}$$

Figure 10 shows the variation of φ as a function of μ for a typical case.

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APPENDIX

NOTATION

C_p = specific heat at constant pressure.
D = diffusion coefficient.
E = activation energy of the chemical reaction.

k = heat-loss coefficient.

$K \equiv \dfrac{k}{C_p W e^{-\theta_\alpha}}$ = dimensionless heat-loss coefficient.

$L \equiv \dfrac{\lambda}{\varrho D C_p}$ = Lewis-Semenov number.

$m \equiv \varrho v$ = Constant = mass flow rate.

q = heat transferred to the stabilizer.

q_l = local heat-loss.

r = coefficient of lateral diffussion.

R = molar gas constant.

T = temperature.

T_f = temperature of the burnt gases.

T_{fa} = temperature of the burnt gases for the adiabatic flame.

v = velocity.

n = reaction rate per unit volume.

W = frequency factor of the chemical reaction.

x = coordinate normal to the combustion wave.

Y = mass fraction of combustion products.

$\delta \equiv \dfrac{q}{m C_p T_{fa}}$ = dimensionless fraction of heat transferred to the stabilizer.

$\gamma \equiv \dfrac{q}{T_{fa}\sqrt{\lambda W Cp}}$ = dimensionless heat transferred to the stabilizer.

ε = mass flow rate fraction of combustion products

λ = thermal conductivity.

ϱ = density.

$\theta \equiv \dfrac{T}{T_{fa}}$ = dimensionless temperature.

$\theta_a \equiv \dfrac{E}{R T_{fa}}$ = dimensionless activation energy.

$\theta_f \equiv \dfrac{T_f}{T_{fa}}$ = dimensionless temperature of combustion products.

θ_i = dimensionless ignition temperature.

θ_0 = dimensionless temperature of cold gases.

$\xi \equiv \dfrac{m C_p}{\lambda} x$ = dimensionless coordinate.

$\varphi \equiv m \sqrt{\dfrac{C_p}{\lambda W e^{-\theta_\alpha}}}$ = dimensionless propagation velocity of the flame.

$\mu \equiv \dfrac{W}{W + r}$ = dimensionless coefficient of lateral diffusion.