

**THEORY OF
DROPLET VAPORIZATION
AND COMBUSTION**

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ABSTRACT

The classical theory of vaporization and combustion of fuel droplets is examined for low Reynolds numbers, when the effects of the motion of the droplet relative to the ambient atmosphere can be neglected, and the process can be considered spherically-symmetrical.

The large value of the ratio of the liquid density to the typical gas density justifies the quasi-steady approximation in the analysis of the gas phase process.

The large value of the ratio of the heat of vaporization per unit mass of fuel to the typical specific thermal energy of the liquid results in the existence of a first stage of droplet heating without vaporization, and a second stage when vaporization occurs at a nearly constant wet-bulb temperature, close to the boiling temperature.

The large values of typical activation energies of the gas phase reactions, leads to the possibility of multiple solutions of the quasi-steady conservation equations for the gas phase. These include solutions corresponding to a nearly frozen mode of vaporization, that can not exist above an "ignition" value of the ambient temperature, and a mode of vaporization with diffusion controlled combustion that can not exist for ambient temperatures below an "extinction" value.

I.- INTRODUCTION AND FORMULATION

Combustion of liquid fuels occurs only after atomization of the liquid fuel in the combustion chamber, that results in a spray of fuel droplets in a gaseous oxidizing atmosphere. The recirculating motion of the combustion gases will play a significant role in the vaporization process. See for example the reviews by Chigier (1976), and Williams (1984).

An important characteristic of the spray combustion process is associated with the fact that the ratio of the typical gas density ρ_o to the liquid density ρ_c is a small number $\rho_o/\rho_l = \epsilon \ll 1$. For combustion under normal pressure ϵ is of the order of 10^{-3} , but the ratio grows to values much more close to 1 for combustion at high or supercritical pressures. If we consider combustion of a typical hydrocarbon fuel with air, under conditions not far from stoichiometry on the rich side, the amount of air required per unit mass of fuel v_a is large, of the order of 15. Then we can conclude that the volume fraction of liquid in the spray is $(a/d)^3 \sim \rho_o/\rho_l v_A \ll 1$. Here a is the characteristic droplet radius, and $d \gg a$ is the mean distance of a droplet to its neighbour.

When the temperature and concentration field around a droplet is described, we find that the important changes occur at distances from the droplet center $r \sim a$. The changes far from the droplet, occurring at characteristic distances very large compared with a , are the result of the collective effects

of all the droplets; they should be calculated by some type of homogenization technique, as it is done in typical spray combustion analysis. In these analysis the response of a droplet in its local environment must be taken into account to calculate the effects of the droplets as sources of heat and mass for the gaseous media; this is the topic of these notes. For other reviews of this topic see Rosner (1972), Williams (1976) and Williams (1984).

The response of the droplet depends very much on the Reynolds, or on the Peclet number $P_e = Ua/\alpha_o$, based on the relative velocity of the droplet to the local gaseous environment, and on the thermal diffusivity $\alpha_o = k_o/\rho_o c_p$, equal to the ratio of the typical gas heat conductivity k_o , density ρ_o and specific heat c_p . If $P_e \gg 1$ the fuel vapor coming from the droplet surface to form a wake. The problem of vaporization and combustion in a boundary layer has received wide attention in the literature. If the gaseous velocity keeps a constant value the droplet loses rapidly its relative velocity (in the time required for the droplet to sweep a volume of gas with a mass of the order of that of the droplet); in that time only a small fraction $\sqrt{1/P_e}$ of the droplet is vaporized, so that, unless the ambient gas velocity is fluctuating due to turbulence and if the droplet has large radius, the vaporization will take place when the Peclet number has been reduced to values small compared with unity.

If we confine our attention to these low Peclet number cases, then the effect of the relative velocity of the ambient gas to the droplet can be neglected and then the droplet and the surrounding concentration and temperature field is spherically symmetrical. The conservation equations that describe the process in the gas phase, that we shall consider stagnant and with constant concentration and temperature to begin with, are as follows:

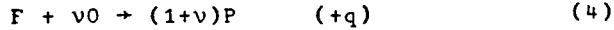
$$\frac{\partial \rho}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \rho v) = 0 \quad (1)$$

$$\rho \frac{\partial Y_\alpha}{\partial t} + \rho v \frac{\partial Y_\alpha}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 \rho D_\alpha \frac{\partial Y_\alpha}{\partial r}) = \dot{w}_\alpha \quad (2)$$

$$\rho c_p \frac{\partial T}{\partial t} + \rho v c_p \frac{\partial T}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 k \frac{\partial T}{\partial r}) = -q \dot{w}_1 \quad (3)$$

Here v is the radial velocity, Y_α is the mass fraction of the species α , T is the temperature, \dot{w}_α is the mass production rate per unit volume of specie α due to the chemical reaction. We have used Fick's law to describe the diffusion flux, with a diffusion coefficient D_α for species α . In the energy equation we use the so called combustion approximation neglecting the work due to pressure gradients and viscous forces. In addition we consider a constant specific heat c_p for the gas in order to simplify the presentation.

We shall also consider in these notes a simplified kinetic mechanism resulting in an overall irreversible one step reaction between a fuel F (subscript 1), oxidizer O (subscript 2) yielding a product P (subscript 3)



where ν represents a mass stoichiometric coefficient, or amount of oxidizer required to burn the unit mass of fuel to yield a mass $1+\nu$ of products with a heat release q per unit mass of fuel.

For the production rates \dot{w}_α we shall use an overall Arrhenius expression of the form involving a frequency factor B ,

$$w_2/\nu = -w_3/(1+\nu) = w_1 = -\rho B Y_1^n Y_2^m \exp(-E/RT) \quad (5)$$

involving a frequency factor B , that we shall consider for simplicity in the presentation to be constant, an activation energy E , or activation temperature E/R , and the reaction orders m and n .

From the momentum equations we can estimate the spatial pressure variations in the flow field around the droplet; they turn out to be so small that in the equation of state for the gas mixture we can replace p by its value p_∞ in the gas environment far from the droplet. If we consider the gas to be a mixture of perfect gases the equation of state becomes

$$p_\infty = \rho RT \{ \sum (Y_\alpha / M_\alpha) \} \quad (6)$$

where M_α is the molecular mass of the species α , R is the universal gas constant.

For the conservation equations we need initial and boundary conditions at the droplet gas interface and at infinity. For the moment, we shall consider the boundary conditions at infinity to be time independent, although we shall relax this condition afterwards.

The boundary conditions at infinity are:

$$r \rightarrow \infty: Y_{\alpha} = Y_{\alpha\infty}, T = T_{\infty} \quad (7)$$

When dealing with the reacting cases, we shall assume that $Y_{1\infty} = 0$, so as to be consistent with time independent conditions at infinity.

The boundary conditions at the droplet surface $r=a(t)$ are obtained from mass and energy conservation considerations at the liquid gas interface. It is required that at $r=a(t)$:

$$\rho\{v - (da/dt)\} = -\rho_1 (da/dt) \quad (8)$$

$$\rho(1-Y_1)\{v-(da/dt)\} + \rho D_1(\partial Y_1/\partial r) = 0 \quad (9)$$

$$\rho Y_{\alpha}\{v-(da/dt)\} - \rho D_{\alpha}(\partial Y_{\alpha}/\partial r) = 0, \quad \alpha \neq 1 \quad (10)$$

$$\rho\{v-(da/dt)\}L + (a/3)\rho_1 c_1 (dT_s/dt) - k(\partial T/\partial r) = 0 \quad (11)$$

where ρ_1 , c_1 and L are the liquid density, specific heat and heat of vaporization per unit mass. $T_s(t)$ is the liquid droplet temperature that is assumed to be spatially uniform, on the grounds that the characteristic heat conduction time within the droplet is short compared with its heat-up time or its lifetime; in addition the small convective flow within the droplet helps to insure the uniformity of the liquid temperature. Two additional conditions at the interface $r = a$

$$Y_1 = (M_1/M) \cdot \exp\{(LM_1/RT_B)(1-T_B/T_S)\}, \quad T=T_S \quad (12)$$

arise from the condition of thermodynamic equilibrium at the liquid gas interface; the first one is the Clausius-Clapeyron relation. Here T_B is the boiling temperature at the ambient pressure p_∞ . $M^{-1} = \sum(Y_\alpha/M_\alpha)$.

II.- QUASI-STEADY GAS PHASE MODEL

A rough estimate of the droplet lifetime t_v can be obtained from Eqs. (8) and (11). Let v_c be the characteristic value of the radial gas velocity v ; then Eq. (8) yields

$$\rho_\infty v_c \sim \rho_l a_0 / t_v \quad (13)$$

where a_0 is the initial droplet radius and ρ_∞ the ambient gas density. Notice that the characteristic regression velocity of droplet surface a_0/t_v is small compared with the gas velocity for typical values of the ratio $\rho_\infty/\rho_l \ll 1$. We shall then use the fact that $\rho_\infty/\rho_l \ll 1$ to simplify the boundary conditions (8)-(11) to the form

$$\rho v = -\rho_l (da/dt) \quad (15)$$

$$\rho v(1-Y_1) + \rho D_1 (\partial Y_1 / \partial r) = 0 \quad (16)$$

$$\rho v Y_2 - \rho D_2 (\partial Y_2 / \partial r) = 0 \quad (17)$$

$$\rho v L + \frac{2}{3} \rho_l c_e \frac{dT_s}{dt} - k \frac{\partial T}{\partial r} = 0 \quad (18)$$

at $r = a(t)$.

An estimate of the gas velocity can be obtained from Eq. (8) by requiring that the heat reaching the droplet from the gas phase per unit surface, of order $k_{\infty}(T_{\infty}-T_B)/a_0$ is used initially in heating the droplet from its initial temperature T_0 to $T_s \approx T_B$, and afterwards in the liquid vaporization. The typical gas velocity is thus given by

$$\rho_{\infty} v_c L \sim k_{\infty}(T_{\infty}-T_B)/a_0 \quad (19)$$

while the characteristic droplet heat-up time t_h is given by

$$a_0 \rho_l c_l (T_B - T_0) / t_h \sim k_{\infty}(T_{\infty} - T_0) / a_0 \quad (20)$$

Then

$$t_v \sim \frac{a_0^2 \rho_l c_p}{k_{\infty}} \frac{L}{c_l (T_{\infty} - T_B)} \cdot \frac{\rho_l}{\rho_{\infty}} \quad (21)$$

where the first factor in the right handside is the characteristic heat conduction time, or response time of the gas phase $t_g = a_0^2 \rho_{\infty} c_p / R_{\infty}$. This time is short compared with t_v , or with respect to the heat-up time

$$t_h \sim \frac{a_0^2 \rho_l c_p}{k_{\infty}} \frac{T_B - T_0}{T_{\infty} - T_0} \frac{\rho_l c_l}{\rho_{\infty} c_p} \quad (22)$$

Thus the characteristic response time of the gas phase at distances $r \sim a$ from the droplet center is short compared with the times t_h and t_v associated with the changes in the boundary conditions at the moving interface $r = a$. This is the basic for the quasi-steady state approximation in the analysis of the gas phase response, or for neglecting the time deriva-

tive terms in Eqs. (1)-(3), that simplify to

$$\rho v r^2 = \dot{m} \quad (23)$$

$$\frac{\dot{m}}{r^2} \frac{\partial Y_\alpha}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho D_\alpha \frac{\partial Y_\alpha}{\partial r} \right) = w_\alpha \quad (24)$$

$$\frac{\dot{m}}{r^2} c_p \frac{\partial T}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 k \frac{\partial T}{\partial r} \right) = -q w_i = q \rho B Y_1 Y_2^m e^{-E/RT} \quad (25)$$

to be solved for $r > a$, with the boundary conditions (7) at $r \rightarrow \infty$, and (12) together with (16) and (17) at $r = a$.

These conditions suffice to determine the liquid rate of vaporization, $4\pi \dot{m}$, and the heat, $k \left(\frac{\partial T}{\partial r} \right)_{r=a} 4\pi a^2$, coming to the liquid by conduction from the gas, together with the gas phase temperature and concentration distributions, in terms of the instantaneous value of the droplet radius a and its temperature T_s . The time evolution of a and T_s can be calculated afterwards by using the conservation equations (15) and (18) at the interface that take the form:

$$\frac{da}{dt} = - \frac{\dot{m}}{a^2 \rho_l} \quad (26a)$$

$$\frac{a}{3} \rho_l c_l \frac{dT_s}{dt} = \left(k \frac{\partial T}{\partial r} \right)_{r=a} - \frac{\dot{m}}{a^2} L \quad (26b)$$

to be solved with the initial conditions $T_s = T_0$, $a = a_0$ at $t = 0$.

Notice that unsteady effects appear here only in the conservation equations (26) for the droplet. The possibility of neglecting the unsteady terms in the gas phase, resulting in this

quasi-steady' gas phase model is associated, as mentioned before, to the fact that the ratio of densities ρ_∞/ρ_1 is small compared with unity, as it was recognized early by Williams (1960). Unsteady effects are important in determining the variations of order $T_\infty\sqrt{\rho_\infty/\rho_1}$ and $\sqrt{\rho_\infty/\rho_1}$ of temperature and concentrations, from its values at infinity, in the far field, at distances from the droplet of order $a\sqrt{\rho_\infty/\rho_1}$. These variations are such, see Waldman (1975) and Crespo and Liñán (1975), that corrections of order ρ_∞/ρ_1 must be introduced in the boundary conditions at infinity when solving Eqs. (24)-(25), if we want to account for unsteady effects in the gas phase; the resulting change in the vaporization rate is of order in $\sqrt{\rho_\infty/\rho_1}$.

It should be noticed that the quasi-steady gas phase approximation, and therefore the model given above, can also be used in cases where the ambient conditions Y_∞ and T_∞ change with time, if the changes occur in times of order t_h or t_v , that are large compared with the gaseous response time $t_g = a^2 \rho_\infty c_p / k_\infty$. Thus it is possible to use this model to describe the droplet evolution when its environment is changing, due to collective effects from the other droplets in a spray, or to the droplet motion to a different environment.

The relative importance of the reaction term in the conservation equations is measured by the Damköhler number t_g/t_c , where t_c is the chemical reaction time

$$t_c^{-1} = B \exp(-E/RT)$$

when $t_g/t_c \ll 1$, that is for small Damköhler numbers, we have frozen

flow; and the reaction terms can be neglected when analysing the droplet vaporization process. When $t_g/t_c \gg 1$ the reaction is so fast that the reactants do not coexist if the reaction is modelled by the irreversible Arrhenius law of Eq. (5), and thus

$$Y_1 Y_2 = 0. \quad (27)$$

It is a characteristic of combustion, see Frank-Kamenetskii (1969), that in combustion processes

$$E/RT \gg 1 \quad \text{and} \quad q/c_p T \sim 1.$$

That is, the reactions are exothermic with a heat release per unit mass of fuel of the order of, or even larger, than the thermal energy and, in addition, the reaction rates are very sensitively dependent on temperature. Thus the chemical reaction may be very slow in regions of low temperature and very fast in regions of high temperature; if the reaction takes place the temperature will be increased significantly due to the reaction. Multiple solutions of the quasi-steady gas phase equations are possible due to the large sensitivity with T of the exothermic reactions; the process may be chemically frozen if the ambient temperature is low enough, or we may have equilibrium flow, so that Eq. (27) can be used to replace one of the conservation equations (24)-(25) if there is a large enough region of high temperature.

If we plot, for example, the vaporization rate m in terms of T_∞ , the result, when E/RT is large, is an S shaped curve

as sketched in figure 1.

There is a lower branch of the S curve with small departures from a frozen vaporization curve, and an upper branch showing also small departures from a chemical equilibrium curve. There is an extinction value $T_{\infty E}$ of T_{∞} , below which there is only the nearly frozen solution, and an ignition value $T_{\infty I}$ of T_{∞} above which there is only the chemical near-equilibrium solution; the middle branch between the bending points is clearly unstable.

In the following, we begin by giving an analysis of the droplet vaporization process with negligible effects of the chemical reaction. We shall show afterwards how the vaporization process can be analyzed in the limiting case of infinite Damköhler numbers, when we can use the assumption of chemical equilibrium leading to Eq. (27). Finally, we shall indicate how to calculate the ignition and extinction conditions.

III.- FUEL DROPLET VAPORIZATION WITHOUT REACTION

The conservation equations for Y_1 and T in the case $B = 0$ can be integrated once to yield

$$\dot{m} Y_1 - r^2 \rho D_1 \frac{\partial Y_1}{\partial r} = \dot{m} \quad (28)$$

$$\dot{m} T - r^2 \frac{k}{c_p} \frac{\partial T}{\partial r} = \dot{m} \left(T_s - \frac{L}{c_p} \right) - \frac{a^3}{3} \frac{\rho c_l}{c_p} \frac{dT_s}{dt} \quad (29)$$

if we make use of the boundary conditions (16) and (18). These equations must be solved with the conditions

$$Y_1 = Y_{1\infty}, \quad T = T_\infty \quad \text{at } r \rightarrow \infty \quad (30)$$

where we now allow for non-zero concentration of Y_1 at infinity.

Also

$$T = T_s, \quad Y_1 = Y_{1s} \quad \text{at } r = a. \quad (31)$$

If we want to calculate the oxygen concentration we must solve the equation

$$\dot{m} Y_2 - r^2 \rho D_2 \frac{\partial Y_2}{\partial r} = 0 \quad (32)$$

with the boundary condition $Y_2 = Y_{2\infty}$ at $r \rightarrow \infty$.

Obviously we must add the Clausius-Clapeyron relation

$$Y_{1s} = \frac{M_1}{M} \exp\left\{\frac{LM_1}{RT_s} \left(1 - \frac{T_\infty}{T_s}\right)\right\} \quad (33)$$

and Eq. (26) to generate a system of equations to describe the evolution with time of a and T_s .

Even though it is simple to solve Eqs. (28) and (29), as it was done by Kassoy and Williams (1968) for variable k and ρD_1 we shall give only, for simplicity in the presentation, the results for constant k and ρD_1 . Then

$$Y_1 = 1 - (1 - Y_{1\infty}) \exp(-\dot{m} / \rho D_1 r) \quad (34)$$

$$T = T_s - L' / c_p + (T_\infty - T_s + L' / c_p) \exp(-\dot{m} c_p / k r) \quad (35)$$

where

$$L' = L + \frac{\partial^3 \rho c_p}{\partial m} \frac{dT_s}{dt} \quad (36)$$

For Y_2 or any other inert species we have

$$Y_2 = Y_{2\infty} \exp(-\dot{m}/\rho D_2 r). \quad (37)$$

Notice that we can use Eq. (35) for $r = a$ to obtain a relation

$$\dot{m} c_p / ka = \lambda = \ln\{1 + c_p(T_\infty - T_s)/L'\} \quad (38)$$

for the nondimensional vaporization rate constant λ , that can be used with Eq. (26) to give

$$\frac{da^2}{dt} = -\frac{2\dot{m}}{\rho c_p} = -\frac{2k}{\rho c_p} \lambda \quad (39)$$

the evolution of the square of the droplet radius with time.

Notice also that Y_{1s} and Y_{2s} can also be written in terms of λ and the Lewis numbers $L_{e1} = k/\rho D_1 c_p$ and $L_{e2} = k/\rho D_2 c_p$; namely

$$\alpha \neq 1; \quad Y_{1s} = 1 - (1 - Y_{1\infty}) \exp(-\lambda/L_{e1}), \quad Y_{2s} = Y_{2\infty} \exp(-\lambda/L_{e2}) \quad (40)$$

The factor M_1/M appearing in the relation (33) can also be calculated in terms of λ .

We can now summarize the results in the system of equations

$$\frac{da^2}{dt} = -\frac{2k}{\rho c_p} \lambda \quad (41)$$

$$\frac{a^2 \rho c_p}{3 \lambda k} c_1 \frac{dT_s}{dt} = L' - L \quad (42)$$

together with the algebraic equations

$$\lambda = \ln\{1 + c_p(T_\infty - T_s)/L'\} \quad (43)$$

$$1 - (1 - Y_{1\infty}) \exp(-\lambda/L_{e1}) = \frac{M_1}{M} \exp\left\{\frac{L - M_1}{R T_s} \left(1 - \frac{T_s}{T_\infty}\right)\right\} \quad (47)$$

and an additional relation to calculate the factor M_1/M as a function of λ .

We must add the initial conditions

$$T_s = T_o, \quad a = a_o \quad \text{at} \quad t = 0 \quad (45)$$

to generate a complete system of equations for T_s , a , λ and L' (It should be noticed that T_s is necessarily $T_s < T_B$).

We shall give an approximate description of the vaporization process obtained by an asymptotic analysis for large values of the ratio LM_1/RT_B that appears in the Clausius-Clapeyron relation (44). The ratio LM_1/RT_B typically ranges from 10 to 15, see for example Landau and Lifchitz (1959).

If $LM_1/RT_B \gg 1$ and T_s is such that

$$(LM_1/RT_B)(T_B - T_s)/T_s \gg 1 \quad (47)$$

then according to Eq. (33), $Y_{1s} \ll 1$ and then (44) gives

$$\lambda L_{e1} = \ln(1 - Y_{1\infty}) \quad (48)$$

and we have a condensation of the ambient fuel on the droplet if $Y_{1\infty} \neq 0$. In this case the droplet radius grows with time in the form

$$\frac{da^2}{dt} = -\frac{2k}{\rho_l c_p} Le_1^{-1} \ln(1 - Y_{1\infty}) \quad (49)$$

so that a^2 grows linearly with time if $Y_{1\infty}$ is constant.

On the other hand, if $Y_{1\infty}$ is zero, we find two stages in the vaporization process. In a first stage when $T_s < T_B$ is such that (47) holds, $Y_{1s} \ll 1$ and according to Eq. (44), $\lambda L_{e1} = Y_{1s} \ll 1$. The vaporization rate, λ , during this stage is very

small; L' is, according to (42), large compared with L and $c_p(T - T_s)$. From Eqs. (42) and (43) we then obtain the equations

$$\frac{4}{3} \pi a^3 \rho_l c_l \frac{dT_s}{dt} = k \frac{(T_\infty - T_s)}{a} 4\pi a^2 \quad (50)$$

$$\frac{da}{dt} = 0 \rightarrow a = a_0 \quad (51)$$

that describe the droplet heating, without significant vaporization, during the first stage.

When Eq. (50) is integrated with the condition $T_s = T_0$ at $t = 0$, we obtain

$$T_\infty - T_s = (T_\infty - T_0) \exp(3kt/a_0^2 \rho_l c_l) \quad (52)$$

involving a characteristic droplet heat-up time $a_0^2 \rho_l c_l / 3k$.

This analysis fails at times close to t_1 such that

$$T_\infty - T_B = (T_\infty - T_0) \exp(3kt_1/a_0^2 \rho_l c_l) \quad (53)$$

when T_s approaches T_B ; then λ grows rapidly to values of order unity. After a short transition stage T_s stops growing, and it maintains afterwards a, nearly constant, value close to T_B . Then according to (42),

$$L' \approx L$$

and the vaporization rate, λ , is given by

$$\lambda = \lambda_0 = \ln\{1 + c_p(T_\infty - T_B)/L\} \quad (54)$$

a result obtained by Godsave (1953).

Notice that during the second stage the temperature of the liquid droplet keeps a constant value, so that the heat

that comes from the gas is used only in vaporizing the droplet, in contrast with the first stage when it is used only in heating the droplet.

If the ambient temperature T_∞ is constant the nondimensional vaporization rate λ will also be constant during the second stage, and the droplet radius will follow the "square law":

$$a^2 = a_0^2 - (t-t_1)\lambda_0 2k/\rho_1 c_p . \quad (55)$$

The droplet temperature is in this stage the "wet-bulb" temperature slightly different from the boiling temperature T_B , as it results from Eq. (44) with $Y_{1\infty} = 0$ and λ given by (54).

Notice that the droplet lifetime t_L is given by

$$t_L - t_1 = a_0^2 \rho_1 c_p / 2k\lambda_0 \quad (56)$$

where the right hand side is the vaporization time.

Notice also that during the first stage the droplet acts as a heat sink of strength $4\pi a_0(T_\infty - T_s)$. During the second stage it acts as a source of fuel of strength $4\pi a_0^3 k\lambda_0 / c_p$ and as a sink of heat of strength $4\pi a_0^3 k\lambda_0 L / c_p$.

IV.- DIFFUSION CONTROLLED COMBUSTION

We have found that the fuel concentration in the first stage of the vaporization process, when the droplet temperature is not yet close to the boiling temperature, is $Y_1 \ll 1$; no significant heat release effects will occur during this stage due to combustion. In the second stage, the chemical heat release will enhance the vaporization rate. We shall here describe the

vaporization process in the limit of infinite Damköhler numbers, for large values of the ratio $LM_1/RT_B \gg 1$, in the second stage when significant vaporization takes place because T_s is close to T_B .

In the limit $Bt_g \rightarrow \infty$, the reactants do not coexist, as indicated by Eq. (27). There is a thin flame at $r = r_f$ separating an interior region without oxidizer

$$Y_2 = 0 \quad \text{for } r < r_f \quad (57a)$$

and an outer region, free from fuel

$$Y_1 = 0 \quad \text{for } r > r_f \quad (57b)$$

the chemical reaction is confined to an infinitely thin reaction zone, or flame sheet, at $r = r_f$.

The reactants reach, by diffusion from both sides, the reaction zone, at stoichiometric proportions; the heat released at the thin reaction zone is conducted away toward the droplet or toward the surrounding atmosphere. The production terms in Eqs. (24) and (25) are Dirac delta functions and the conservation equations (24) and (25) can be written for $r > a$, in the form

$$\frac{\dot{m}}{r^2} \frac{\partial Y_1}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho D_1 \frac{\partial Y_1}{\partial r} \right) = -\frac{\dot{m}}{r_f^2} \delta(r-r_f) \quad (58)$$

$$\frac{\dot{m}}{r^2} \frac{\partial Y_2}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \rho D_2 \frac{\partial Y_2}{\partial r} \right) = -\nu \frac{\dot{m}}{r_f^2} \delta(r-r_f) \quad (59)$$

$$\frac{\dot{m}}{r^2} c_p \frac{\partial T}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 k \frac{\partial T}{\partial r} \right) = q \frac{\dot{m}}{r_f^2} \delta(r-r_f) \quad (60)$$

and similar equations for the other species.

The boundary conditions for (58)-(60) are

$$r=a : \quad \dot{m} Y_1 - r^2 \rho D_1 \frac{\partial Y_1}{\partial r} = \dot{m} \quad (61)$$

consistent with $Y_1 = 0$ for $r > r_f$ and $Y_2 = 0$ for $r < r_f$.

$$r=a : \quad \frac{\dot{m}}{a^2} L - k \frac{\partial T}{\partial r} = 0, \quad T = T_s \quad (62)$$

with $T_s = T_B$ resulting from (12), in first approximation for $LM_1/RT_B \gg 1$, and

$$T = T_\infty, \quad Y_2 = Y_{2\infty} \quad \text{for } r \rightarrow \infty. \quad (63)$$

The solution to this problem determines the flame location r_f and the flame temperature, T_f , at $r = r_f$, and the vaporization rate \dot{m} as a function of T_∞, T_s , and other parameters in the problem. It is easy to show that r_f/a , and \dot{m}/a , as well as T_f are independent of a ; again the droplet radius follows the "square law" $a^2 = a_0^2 - K(t-t_1)$ if T_∞ is constant.

This diffusion controlled model of droplet vaporization with combustion results from a direct use of the Burke-Schumann (1928) analysis of diffusion flames for infinitely fast reactions.

The problem is easy to solve under the assumption that the Lewis numbers of the reactants are unity, namely for $k/\rho D_1 c_p = k/\rho D_2 c_p = 1$. In this case the Shvab - Zeldovich coupling functions $\hat{Y}_2 = Y_2 - \nu Y_1$ and $\hat{T} = T + q Y_1 / c_p$ follow the same conservation equation as a passive scalar \tilde{Y}

$$\frac{\dot{m}}{r^2} \frac{\partial \tilde{Y}}{\partial r} - \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{k}{c_p} \frac{\partial \tilde{Y}}{\partial r} \right) = 0 \quad (69)$$

and can be solved with the boundary conditions given above to yield

$$Y_2 - \tilde{Y}_1 = -\nu + (Y_{2\infty} + \nu) \exp(-\dot{m} c_p / k r) \quad (70)$$

$$T + q Y_1 / c_p = T_s + (q - L) / c_p + \{T_\infty - T_s - (q - L) / c_p\} \exp(-\dot{m} c_p / k r) \quad (71)$$

These two relations together with the relations $Y_1 = 0$ for $r > r_f$, $Y_2 = 0$ for $r < r_f$, resulting from the chemical equilibrium condition (27), provide us with the temperature and concentration distributions, and the vaporization rate \dot{m} . This is given by (70) and (71) at $r = a$

$$\dot{m} c_p / k a = \lambda \ln \{ 1 + \{ c_p (T_\infty - T_s) + q Y_{2\infty} / \nu \} / L \} \quad (78)$$

an expression similar to Eq. (54) that gives the vaporization rate with frozen chemistry.

The flame location r_f is given by Eq. (70) for $Y_1 = Y_2 = 0$,

$$r_f / a = \lambda / \ln(1 + Y_{2\infty} / \nu) \quad (79)$$

and the flame temperature T_f by

$$T_f = T_s + (q - L) / c_p + \{T_\infty - T_s - (q - L) / c_p\} \nu / (Y_{2\infty} + \nu) \quad (80)$$

V.- IGNITION REGIME

We shall describe the process of liquid droplet vaporization with combustion using the quasi-steady gas phase model and the approximation $T_s = T_B$, resulting from the assumption that

$$LM_1/RT_B \gg 1.$$

It is convenient to write the equations (24)-(25) in nondimensional form, using $\theta = T/T_\infty$ together with Y_α as dependent variables and $x = r/a$ as independent variable. The conservation equations become

$$\frac{\lambda}{x^2} \frac{\partial Y_\alpha}{\partial x} - \frac{1}{x^2} \frac{\partial}{\partial x} \left(\frac{x^2}{Le_\alpha} \frac{\partial Y_\alpha}{\partial x} \right) = -v_\alpha D_\alpha Y_1^n Y_2^m \exp(-\beta/\theta) \quad (81)$$

$$\frac{\lambda}{x^2} \frac{\partial \theta}{\partial x} - \frac{1}{x^2} \frac{\partial}{\partial x} \left(x^2 \frac{\partial \theta}{\partial x} \right) = \frac{q}{c_p T_\infty} D_\alpha Y_1^n Y_2^m \exp(-\beta/\theta) \quad (82)$$

where $v_1 = 1$, $v_2 = v$, $v_{\text{prod}} = v(1+v)$, $\beta = E/RT_\infty$, $D_a = \rho_\infty c_p a^2 B/k$ and $\lambda = \dot{m} c_p / ka$; Le_α is the Lewis number of the species α .

These equations must be solved for $x > 1$ with the boundary conditions

$$\theta = 1, \quad Y_1 = 0, \quad Y_\alpha = Y_{\alpha\infty} \quad \text{for } x \rightarrow \infty \quad (83)$$

$$\theta = \theta_B, \quad \frac{\partial \theta}{\partial x} = \lambda \frac{L}{c_p T_\infty} \quad (84a)$$

$$\text{and } \lambda Y_\alpha L_\alpha = \frac{\partial Y_\alpha}{\partial x}, \quad \lambda(1-Y_1)L_1 = -\frac{\partial Y_1}{\partial x} \quad \text{at } x = 1 \quad (84b)$$

for $\alpha \neq 1$.

The solution of this problem provides in particular, the nondimensional vaporization rate λ as a function of the Damkohler number D_a and other parameters as $L/c_p T_\infty$, $q/c_p T_\infty$, θ_B and β .

The pure vaporization corresponds to the limiting case $D_a \rightarrow 0$, analysed in Section III, leading to the following expressions for λ , θ and Y_1 :

$$\lambda = \lambda_o = \ln\{1 + (1-\theta_B)c_p T_\infty/L\} \quad (85)$$

$$\theta = \theta_B - L/c_p T_\infty + (1-\theta_B + L/c_p T_\infty)\exp(-\lambda_o/x) \quad (86a)$$

$$Y_1 = 1 - \exp(-\lambda_o L_{e1}/x) . \quad (86b)$$

For large values of β , as indicated before, the curve giving λ as a function of $D_a \exp(-\beta)$ has an s shaped form, with an upper diffusion controlled branch and a lower nearly-frozen branch that ends in an, "ignition" bending point.

The deviations from the frozen solution, for $\beta \gg 1$, are small even at the bending, ignition, point. We shall describe the vaporization process for near-ignition conditions.

We note that in this regime the reaction will only occur far from the droplet where θ is largest, close to 1. We will find that for lower values of θ such that $\beta(1-\theta) \gg 1$ the reaction is frozen; this is the case close to the droplet, for $x \sim 1$, where in first approximation, for large β , θ and Y_1 are given by (86).

This frozen solution yields for large $x \gg 1$

$$\theta = 1 - (1-\theta_s + L/c_p T_\infty)\lambda_o/x, \quad Y_1 = \lambda_o L_{e1}/x, \quad Y_2 = Y_{2\infty} . \quad (87)$$

The chemical reaction can not be neglected in the ignition regime for large values of x where $\theta-1$ and Y_1 become of order $1/\beta$. This will occur for $x \sim \beta$; in this outer region the three terms of the energy equation (82) are of order

$$1/\beta^4 \quad 1/\beta^3 \quad \text{and} \quad \frac{q}{c_p T_\infty} D_a e^{-1/\beta} \beta^{-n}$$

respectively. The convective term can then be neglected in first

approximation compared with the heat conduction term, that must be balanced by the reaction term.

Thus in the ignition regime

$$\Delta = D_a e^{-\beta} (q/c_p T_\infty) Y_{20}^m \beta^{3-n}$$

will be of order unity.

In the inner region $x \sim 1$ the reaction term is exponentially small, and thus θ and Y_1 are given by

$$\theta = \theta_s - L/c_p T_\infty + (\theta_a - \theta_s + L/c_p T_\infty) \exp(-\lambda/x) \quad (88a)$$

$$Y_1 = 1 - (1 - Y_{1a}) \exp(-\lambda/x) \quad (88b)$$

where θ_a and Y_{1a} are apparent values of θ and Y_1 far from the droplet as seen from the inner region; they are given as λ by the expansions

$$\theta_a = 1 + \beta^{-1} \phi_a + \dots, \quad Y_{1a} = \beta^{-1} y_{1a} + \dots, \quad \lambda = \lambda_0 + \beta^{-1} \lambda_1 + \dots \quad (89)$$

where, as a result of the boundary conditions (84),

$$\lambda_1 = \phi_a / (1 - \epsilon_s + L/c_p T_\infty). \quad (90)$$

In the outer reacting zone, for $x/\beta = z$ of order unity we shall describe the temperature and concentrations in the form of the expansions

$$\theta = 1 + \beta^{-1} \phi + \dots, \quad Y_1 = \beta^{-1} y_1 + \dots, \quad Y_2 = 1 - \beta^{-1} y_2 + \dots \quad (91)$$

where ϕ , y , and y_2 are functions of z .

When these expansions are used together with Eqs. (81) and (82) we obtain the system

$$z^{-2} \frac{d}{dz} \left(z^2 \frac{d\phi}{dz} \right) = -\frac{q}{c_p T_\infty L_e} z^{-2} \frac{d}{dz} \left(z^2 \frac{dy_1}{dz} \right) = -\Delta y_1^2 e^\phi \quad (92)$$

so that

$$\phi + y_1 q / c_p T_\infty L_{e1} = -\mu_0 / z \quad (92b)$$

where $\mu_0 = \lambda_0 \{1 - \theta_s + (L - q) / c_p T_\infty\}$ is obtained from the matching conditions with the first approximation to the solution for the inner region. We can now use Eq. (92) to generate a single equation for ϕ , that can be written in the form

$$\xi^{-2} \frac{d}{d\xi} \left(\xi^2 \frac{d\phi}{d\xi} \right) = -\delta \left(-\psi - \frac{x}{\xi} \right)^n e^\phi \quad (93)$$

to be solved with the boundary conditions

$$\phi + \alpha \xi \rightarrow 0 \quad \text{for } \xi \rightarrow \infty \quad (94a)$$

$$\phi + 1/\xi \rightarrow \phi_a \quad \text{for } \xi \rightarrow 0 \quad (94b)$$

obtained from the matching conditions with the inner expansion.

In this equation we use as independent variable ξ , given by

$$(1 - \theta_s + L / c_p T_\infty) \beta \lambda_0 / x = 1 / \xi$$

and

$$\alpha = \frac{1 - \theta_s + (L - q) / c_p T_\infty}{1 - \theta_s + L / c_p T_\infty}, \quad \delta = \left(1 - \theta_s + \frac{L}{c_p T_\infty} \right)^2 \lambda_0^2 \left(\frac{c_p T_\infty}{q} \right)^{n-1} L_{e1}^n D_{a12}^m \beta^{3-n} e^{-\beta}$$

The solution of the problem (93)-(94) provides in particular the apparent increment in ambient temperature ϕ_a as a function of δ and α , for a given reaction order n , together with the temperature profile $\phi(\xi)$ in the outer region. The fuel concentration is given in term of ϕ by the relation

$$y_1 = -(\phi + \alpha / \xi) c_p T_\infty L_{e1} / q \quad (95)$$

Notice also that ϕ_a is proportional to the increment $\beta^{-1}\lambda$, of the vaporization rate above the frozen value λ_0 .

The problem (93)-(94) has two solutions for negative values of α if $\delta < \delta_I(\alpha, n)$, and no solutions for $\delta > \delta_I$. The solution is unique for positive values of α . An asymptotic solution of the problem (93)-(94) can be obtained for small values of α , when the reaction zone becomes thin, separating an inner frozen region from an outer equilibrium region. The resulting relation between δ and ϕ_a is

$$2\Gamma_{n+1}\delta = \phi_a^4 \exp(\alpha\phi_a) \quad (96)$$

$$\delta < \delta_I = (4/\epsilon\alpha)^4 / 2\Gamma_{n+1} \quad \text{if } \alpha < 0 \quad (97)$$

Here Γ_{n-1} is the Gamma function of order $(n+1)$. The numerical

The numerical solutions of the problem (93)-(94) was obtained by Liñán (1975) for the case $n=1$; the results for the value of the ignition Damkohler number δ_I in terms of α , for negative values of α can be correlated by the expression

$$\delta_I \alpha^4 = \{16e^{-2}/\sqrt{2} - \alpha\}^2 \quad (98)$$

which has the correct asymptotic form for small values of $-\alpha$ and the exact asymptotic form for large $-\alpha$ if the term $-\alpha$ in the right hand side is replaced by -0.998α .

The two valued expression $\phi_a(\delta)$ obtained from the solution of (93)-(94) for negative values of α provides us with a description of the s shaped curves, giving for example the vaporization rate in terms of the square of the droplet radius,

close to the ignition bending point.

The existence of these multiple solutions in the droplet vaporization problem with combustion was found early both experimentally and theoretically. See, for example, Tarifa et al. (1962), who obtained numerical and approximate solutions to the conservation equations with finite reaction rates. Later, Law (1975) showed how the analysis of Liñán (1974) for the structure of diffusion flames at large activation energies can be used to provide for a description of the extinction process.

We shall not give here the details of the analysis of the diffusion flame structure near extinction for which we refer to Liñán (1974) and Law (1975).

The solution for large values of β is given in first approximation by the Burke-Schumann solution of the problem, corresponding to the limit $D_a e^{-\beta} \rightarrow \infty$, described in Section IV in dimensional form.

Finite rate effects, important under extinction conditions, are responsible for a finite, even though thin, reaction zone, where the temperature drops below the adiabatic flame temperature T_f , given by (80), by an amount RT_f^2/E enough to reduce the exponential Arrhenius factor by an a factor of order unity.

The reaction zone is still thin, typically RT_f/E times r_f , so that convective effects and non planar effects are not important there in first approximation. The temperature and concentration distributions are given by a balance of conduction and

diffusion in the radial direction together with the reaction term, with boundary conditions determined by matching with the outer Burke-Schumann solution. The resulting problem, involving a reduced number of parameters, has no solutions below an extinction value, D_{aE} , of the Damkohler number, and two solutions corresponding to the left bend of the S shaped curves for larger values of D_a .

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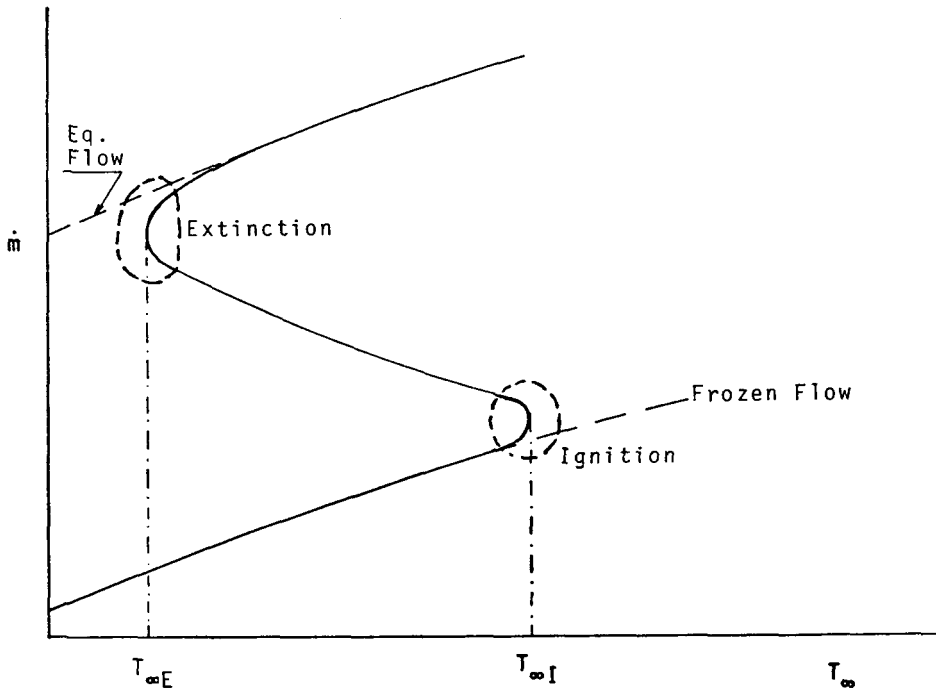


Figure 1