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SOME PROBLEMS IN THE UNSTEADY BURNING OF SOLID PROPELLANTS

by

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ABSTRACT: The model of uniformly distributed combustion in the gas phase is used as the basis for examining several features of unsteady burning. As a zeroth approximation the flame zone begins at the solid-gas interface and responds linearly and quasi-statically to changes of pressure only. Three deviations from this behavior are then examined: the combustion zone is displaced from the surface, the energy release responds to fluctuations of temperature and the response is not quasi-static. It appears that the assumption that the burning begins immediately at the surface can lead to significant changes, more important than the assumption that the energy release responds to changes of pressure only, especially in the interpretation of experimental data. The problem of nonquasi-static behavior, i.e., processes in the gas phase do not follow precisely impressed changes of pressure, is formulated as an expansion in frequency. Approximate results seem to be consistent with existing information.

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FOREWORD

This report was prepared as part of a continuing program of research on combustion instability. This program has been sponsored by the Naval Ordnance Systems Command and the Office of Advanced Research and Technology, a division of National Aeronautics and Space Administration. This particular investigation was funded by Naval Ordnance Systems Command, NOSC Task Assignment ORD-331-001/200-1/UR001-06-01, WU No. 1. The objective of the work was to examine certain assumptions common to published theories of solid propellant combustion perturbation theories, assumptions that were suspected of seriously limiting the relevance to experimental measurements of perturbation behavior.

The author of this report is employed as a personal services contractor to the Naval Weapons Center (NWC), and the present work was conducted in that capacity.

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NOMENCLATURE

A	Parameter, $A = E\ (1 - T_c/\overline{T}_s$
a	Coefficient in burning rate law, $m = ap^n$
B	Parameter, Eq. 70, 82, or 108
b	Coefficient in Eq. 62
c, c_p	Specific heat of solid, gas
E	$E = E_s/R_o\overline{T}_s$
E_s	Activation energy for surface reaction
H	$H = Q_s/c\ (\overline{T}_s - T_c)$
H_β	See Eq. 85
$H_{\beta T}$	See Eq. 109
h	Enthalpy
h_{fr}, h_{sr}	Reference values of enthalpy
K	Eq. 128
k_c, k_g	Thermal conductivity of solid, gas
k_ℓ	$k_\ell = \ell\pi/L$

L_s	$L_s = h_{s+} - h_{s-}$, positive for endothermic reaction
ℓ	$\ell = k_g/mc_p$
M_b	Mach number of product gases
m	Mass flux (gm/cm^2 - sec)
n	Index in $m = ap^n$
n_s	Index in Eq. 16
p	Pressure
Q_f	Heat release per unit mass in gas phase
Q_s	$Q_s = - L_s$
q	Heat transfer (ergs/cm-sec)
q_f'	Heat transfer fluctuation at instantaneous position of flame (outer edge)
$q_{\bar{f}}'$	Heat transfer fluctuation at mean position of flame (outer edge)
R	Response function, $R = (m'/\bar{m})/(p'/\bar{p})$
R_o	Universal gas constant
r	Linear burning rate (cm/sec)
s	Index in Eq. 64, s - A in linear calculations
T	Temperature
T_c	Temperature of cold solid ($x \rightarrow - \infty$)
t	Dimensionless time in Section 5; scale is $\bar{\rho}_s\ k_g/\bar{m}^2 c_p$
T_f'	Temperature fluctuation at instantaneous position of flame (outer edge)
$T_{\bar{f}}'$	Temperature fluctuation at mean position of flame (outer edge)

V	Coefficient in Eq. 100
V_T	Eq. 104
W	Eq. 65
W_β	Value of W for $\beta \neq 0$, Eq. 81
$W_{\beta T}$	Eq. 103
X	Eq. 105
x_f'	Fluctuation of the outer boundary of the flame
x_i, x_f	Inner (upstream) and outer (downstream) boundaries of the flame zone
x_s	Position of solid-gas interface
Z	Eq. 80
α_s	Index in Eq. 16
α_1	$\alpha_1 = \tau_{\bar{f}}'^{(o)} - \frac{q_{\bar{f}}'^{(o)}}{\bar{m} c_p \bar{T}_s} + \Lambda^2 \frac{w'^{(o)}}{\bar{w}} (1 + \bar{\xi}_f) - (2 - \bar{\xi}_f) \Lambda^2 \mu'^{(o)}$, see Eq. 133
β	Exponent in $\xi_i = \xi_f^{\beta}$, Eq. 73
β_1	$\beta_1 = e^{-\bar{\xi}_f} \left[\frac{q_{\bar{f}}'^{(o)}}{\bar{m} c_p \bar{T}_s} - \Lambda^2 \frac{w'^{(o)}}{\bar{w}} + (2 + \bar{\xi}_f) \Lambda^2 \mu'^{(o)} \right]$, see Eq. 133
γ	Ratio of specific heats, $\gamma = c_p/c_{Re}$
γ_1	$\gamma_1 = -\mu'^{(o)} \Lambda^2 e^{-\bar{\xi}_f}$, see Eq. 133

δ_1	$\delta_1 = -\Lambda^2 \frac{w'(o)}{\bar{w}} - \Lambda^2 \mu'(o)$, see Eq. 1[illegible]
ε	Small parameter, Eq. 122
$\dot{\varepsilon}$	Reaction rate, Eq. 19
ζ	$\zeta = \exp\left[\int (mc_p/k_g)dx\right]$
θ	$\theta = (\bar{x}_f - x)/\ell$
κ_c	Thermal diffusivity of solid, $\kappa_c = k_c/\rho_c c$
κ_g	Thermal diffusivity of gas, $\kappa_g = k_g/\bar{\rho}_{gs} c_p$
Λ^2	Eigenvalue, Eq. 34
λ	Complex function of Ω, Eq. 4
λ_r, λ_i	Real and imaginary parts of λ
λ_+, λ_-	Defined in Eq. 125
μ	Normalized mass flux, $\mu = m/\bar{m}$
ξ	$\xi = \int \left(mc_p/k_g\right)dx$
ξ_c	$\xi_c = (\bar{m}_c/k_c)x = (\bar{r}/\kappa_c)x$
ρ_c, ρ_g	Density of solid, gas
τ	Normalized temperature, $\tau = T/\bar{T}_s$
$\bar{\tau}_a$	Average temperature introduced in Eq. 134
Ω	Normalized frequency, $\Omega = \kappa_c \omega/\bar{r}^2$
ω	Real angular frequency

SUBSCRIPTS

$o\pm$	Evaluated on the solid (-) or gas (+) side of the origin
$s\pm$	Evaluated on the solid (-) or gas (+) side of the interface
i	Imaginary part
r	Real part
f	Evaluated at the instantaneous position of the flame
$\bar{f}$	Evaluated at the mean position of the flame

SUPERSCRIPTS

$\bar{\ }$	Mean value
$'$	Fluctuation value
(o), (1)	Zeroth and first order terms in the expansions (Eq. 132)

I. INTRODUCTION

It appears at the present time that much of the unsteady behavior in solid-propellant rocket chambers can be traced to the interaction between pressure fluctuations and the burning in the neighborhood of the solid surface. Thus, an understanding of the processes in this region when the pressure is varying in time is essential. Ultimately, experimental results are required, but their interpretation rests on analysis, which is necessarily approximate.

The various analyses which have appeared differ substantially only in the model adopted for the gas phase. But even in that respect, the basic assumptions involved are so strong that almost all the final results for the response to harmonic pressure oscillations have the same form, a two-parameter function of dimensionless frequency (Ref. 1), shown as Eq. 71 here. In fact, only one of those parameters (denoted by B) depends on the model chosen for the gas phase; the other, A, being associated with the thermal wave in the solid and the surface reaction for transformation of solid to gas. This conclusion is a consequence of four basic assumptions: (1) the problem is linear and treated in one-dimensional form; (2) the solid is homogeneous and nonreacting, with constant properties; (3) the surface reaction is independent of pressure; and (4) the gas phase responds quasi-statically to changes of pressure. It is easy to relax assumption (3), in which case the form of the response function is changed only slightly and one additional parameter is introduced, the index n_s in Eq. 71.

Differences among the explicit formulas for the parameter B, expressed in terms of the properties of the gas phase, arise from differences in the models chosen for the structure of the flame in the gas phase. Hence, the detailed interpretation of numerical results and experimental data must differ among the calculations. So long as the assumption of quasi-static behavior is correctly interpreted and used, disagreement of this sort is in fact associated with dissimilarities among the models used for steady burning. The major portion of the discussion here is grounded on one simple, yet fairly realistic, model, based on uniformly distributed combustion, which has been solved approximately and used previously in a calculation of the response function (Ref. 2). The approximations used in Ref. 2, in addition to those noted above, imply that the fluctuations of total energy release in the gas phase are always in phase with the pressure fluctuations, a result which is open to question. Alternatives are possible, leading, of course, to different expressions for the parameter B. Although part of the approximate nature of the analysis of Ref. 2 can be eliminated, as already noted in Ref. 3 and explicitly shown here, it is not possible to obtain a complete and unique solution for the quasi-static problem.

There are two purposes of the work reported here. (1) within the four crucial assumptions noted above, several of the additional approximations used in Ref. 2 are examined, and means of correcting them are offered. (2) Two of the four basic assumptions listed are relaxed somewhat and the corresponding response functions are computed: a separate calculation is given for nonquasi-static behavior which does not contain a pressure dependence in the surface reaction. In view of the length of this report, it is perhaps helpful to summarize the main points to be made.

As in Ref. 1, the calculation of the response function is split into three parts: analysis of the solid phase, construction of important conservation relations for the solid-gas interface, and analysis of the gas phase. The conservation of energy applied to the interface provides an important equation which is used as the primary relation into which the other results are substituted. In particular, it is the heat transfer at the surface which must be found from the calculations for the solid and gas phases.

The simplest model for the solid will be used: homogeneous, nonreacting material having constant properties (see Ref. 4 and 5 for analyses in which chemical activity in the solid phase is examined). Thus, the treatment of both the solid phase and the interfacial region is essentially the same as in most other researches on this problem. The necessary information is collected in Section II.

By far the most difficult part of the problem is analysis of the gas phase; even the steady-state problem remains at the present time essentially unsolved. As a basis for all the work covered here, the model of uniformly distributed combustion is used. This can be solved "exactly," although certain minor assumptions must be made. For example, if the combustion zone extends from some plane removed from the interface to the outer boundary, an additional condition, such as an "ignition temperature" or a minimum reaction rate, must be used to define the inner boundary. However, the solution can be found, culminating in a transcendental equation (Eq. 49) for the flame thickness. The formal solution to the steady-state problem is covered in Section III. The linear burning rate can be found according to the procedure outlined there, although detailed results are not included here.[1]

Perhaps the most important assumption of those noted above is that the gas phase responds in a quasi-static manner. This effects enormous

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[1] "Linear Burning Rates for Solid Propellants Having Uniformly Distributed Combustion," by F. E. C. Culick and G. H. Dehority (in preparation).

simplifications in the analysis. With this assumption, the necessary results can be obtained in two ways: either directly, from a steady-state solution, as discussed in Section IV-A; or by solving the relevant linearized differential equations, as covered in Section IV-D. The first approach is simpler, but the second must be used in calculations not based on the assumption of quasi-static behavior, a matter treated in Section V.

In order to complete the calculation of the response function, even with the model of uniform combustion and quasi-static behavior of the gas phase, some further approximations are required. There are three principal ones used in Ref. 3 and discussed here in Section IV-A:

1. Combustion in the gas phase begins immediately at the solid-gas interface

2. The flame zone is relatively thick, in a sense made precise in Sect ⌐ IV-A

3. The fluctuations of energy release in the gas phase are uniform and hence always in phase with the pressure

It is generally true that the flame thickness, x_f, measured from the interface to the outer boundary, can be assumed to be large, in the sense that $\exp(mc_p x_f/k_g)$ is very much greater than unity. This implies (Eq. 58 and also remarks following Eq. 164) that a relatively small fraction of the heat released in the gas phase is transferred to the solid by heat conduction. It also implies, as remarks following Eq. 58 clarify, that if in addition one assumes that combustion begins at the surface, the results are in practice restricted to exothermic surface reactions (i.e., the parameter His positive). It appears that endothermic reactions can be included in this model only if simultaneously one allows the combustion zone to be displaced from the surface.

In Section IV-C, a displaced combustion zone is treated, although an approximation used in Eq. 78 implies that the results are still valid only for $H > 0$. Nevertheless, the results indicate that significant numerical differences exist between the calculations of Ref. 2 and those for the more realistic case of a displaced combustion zone. The distinction is interpreted by means of the parameter H in Eq. 85. Use of the results of Ref. 2 in the interpretation of data can therefore be done only with (possibly serious) reservations.

The fourth approximation listed above is examined briefly in Section IV-E. It is supposed that part of the fluctuation of reaction rate is in phase with the flame temperature. The final response function appears as Eq. 106, and once again the results can be interpreted in terms of H, Eq. 111. This crude approach indicates that the influence of temperature dependence of the reaction rate is less significant than that of a displaced combustion zone.

All of these results for the response function necessarily have the same form, Eq. 71. But analysis of data in Ref. 7 indicates that this form--irrespective of the particular formula used for B--does not accommodate all data. Hence, it is necessary to alter one or more of the basic assumptions enumerated earlier. In Section V, the assumption of quasi-static behavior is relaxed--weakly. An expansion in the relevant frequency parameter ($\Omega\varepsilon$, Eq. 122) is carried out to first order, so the results are restricted to relatively low frequencies. For simplicity, the calculations are based on the model of uniform combustion extending from the solid surface. With a number of fairly reasonable numerical approximations, the response function eventually takes the form shown in Eq. 165. The major correction associated with nonquasi-static behavior appears in the imaginary part of the response function and can be numerically significant. This conclusion appears to be consistent with observations, but satisfactory proof will be found only by use of Eq. 165 to analyze data in a manner such as that used, for example, in the work of Ref. 7.

There evidently remain a number of significant problems in respect to the unsteady behavior of a burning solid. Modeling of the combustion zone in the gas phase is still in a very primitive state, and this is probably the severest restriction on all the special cases treated. Moreover, even for the simple model used as a basis here, there are several permutations not yet considered: for example, nonlinear behavior is an important aspect which has not been covered here; the effect of a displaced combustion zone on nonquasi-static and of course on nonlinear results has not been treated; and the combination of nonquasi-static and nonlinear behavior is bound to be of interest, particularly for propellants exhibiting thick combustion zones. In any case, the possibilities for obtaining analytical results relatively easily should not be too quickly passed over in favor of specific numerical calculations.

II. SOLID PHASE AND INTERFACIAL MATCHING CONDITIONS

Since only the gas phase will differ from most earlier works, the other pieces of the problem will be covered first and but briefly; expanded presentations may be found in Ref. 1 and 8 and sources cited there. The model used is sketched in Fig. 1, with coordinate system chosen so that the origin is fixed always to the mean position of the burning surface; the latter fluctuates with amplitude x_s. In this picture, then, cold solid flows in from the left with speed equal to $\bar{r}$,[2] the average linear burning rate, and the unsteady heat conduction (energy) equation for the solid is

$$\rho_c\, c\, \frac{\partial T}{\partial t} + \rho_c\, \bar{r} c\, \frac{\partial T}{\partial x} = k_c\, \frac{\partial^2 T}{\partial x^2} \tag{1}$$

For the case of small amplitude motions, the temperature is split into its average value (independent of time) $\bar{T}$, and the fluctuation T'; $T = \bar{T}(x) + T'(x,t)$. The corresponding solutions of Eq. 1 are

$$(\bar{T}-T_c)/(\bar{T}_s-T_c) = e^{\xi_c} \tag{2}$$

$$T' = T'_o\, e^{\lambda \xi_c} \tag{3}$$

where T_c is the temperature of the cold propellant, $\bar{T}_s$ is the average surface temperature, T'_o is the temperature fluctuation evaluated at $x = \xi_c = 0$, and $\xi_c = \bar{r}x/\kappa_c$, κ_c being the thermal diffusivity, $\kappa_c = k_c/\rho_c c$. The complex function, λ, of dimensionless frequency $\Omega = \kappa_c \omega/\bar{r}^2$ has real and imaginary parts given by

$$\lambda_r = \tfrac{1}{2}\left\{1 + \frac{1}{\sqrt{2}}\left[(1 + 16\Omega^2)^{\frac{1}{2}} + 1\right]^{\frac{1}{2}}\right\} \tag{4}$$

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[2] This definition of the coordinate system will be altered slightly in Section V-B.

$$\lambda_i = \frac{1}{2\sqrt{2}} \left[(1 + 16\Omega^2)^{\frac{1}{2}} - 1 \right]^{\frac{1}{2}} \tag{5}$$

It will be necessary later to match the solutions for the solid and gas phases at the interface, and in particular the fluctuations of temperature and heat transfer at $x = x_s$ must be computed. For small motions of the surface, these can be expanded in Taylor series about the origin, and to first order

$$T_s' = T'_{o\pm} + x_s \left(\frac{d\bar{T}}{dx}\right)_{o\pm} \tag{6}$$

$$\left(\frac{\partial T'}{\partial x}\right)_{s\pm} = \left(\frac{\partial T'}{\partial x}\right)_{o\pm} + x_s \left(\frac{d^2\bar{T}}{dx^2}\right)_{o\pm} \tag{7}$$

where $(\ )_{o\pm}$, $(\ )_{s\pm}$ refer to the gas (+) or solid (-) side of the origin (o) or interface (s). By use of Eq. 2, 3, 6, and 7, one can then find the formula for the fluctuation of heat transfer from the interface to the solid,

$$\left(k_c \frac{\partial T'}{\partial x}\right)_{s-} = \bar{m}c \left[\lambda T_s' + \left(\frac{\bar{T}_s - T_c}{\lambda}\right) \frac{m_s'}{\bar{m}} \right] \tag{8}$$

after Eq. 13, deduced below, is inserted for x_s. As shown later in Section VI the same result can be found in a different way.

There are two important matching conditions which can be deduced by applying conservation of mass and energy to a small control volume including the interface:

$$\rho_c (\bar{r} - \dot{x}_s) = \rho_{gs} (u_{gs} - \dot{x}_s) \tag{9}$$

$$\rho_c (\bar{r} - \dot{x}_s) h_{s-} + \left(k_g \frac{\partial T}{\partial x}\right)_{s+} = \rho_g (u_g - \dot{x}_s) h_{s+} + \left(k_c \frac{\partial T}{\partial x}\right)_{s-} \tag{10}$$

The steady-state form of Eq. 9 is simply an identity, while that for Eq. 10, after use of Eq. 2, is

$$\left(k_g \frac{d\bar{T}}{dx}\right)_{s+} = \bar{m} \left[c (\bar{T}_s - T_c) + L_s \right] \tag{11}$$

where $L_s = h_{s+} - h_{s-}$ is the latent heat for the surface reaction, L_s being positive for an endothermic reaction.

The fluctuating part of Eq. 9 gives a formula for the surface oscillations:

$$\frac{\rho_c \dot{x}_s}{\overline{m}} = -\frac{1}{1 - \rho_{gs}/\rho_c} \frac{m_s'}{\overline{m}} \tag{12}$$

or, for harmonic oscillations, $x_s \sim e^{i\omega t}$, with $\omega \neq 0$, and $\rho_{gs}\ \rho_c \ll 1$,

$$\frac{\rho_c x_s}{\overline{m}} = -\frac{1}{i\omega} \frac{m_s'}{\overline{m}} \tag{13}$$

The unsteady part of the energy balance in Eq. 10 leads to

$$\left(k_g \frac{\partial T'}{\partial x}\right)_{s+} = \left(k_c \frac{\partial T'}{\partial x}\right)_{s-} + m_s' \overline{L}_s + \overline{m}\,(c_p - c)\ T_s' \tag{14}$$

and with Eq. 8,

$$\left(k_g \frac{\partial T'}{\partial x}\right)_{s+} = \overline{m}c \left[\lambda T_s' + \left(\frac{c_p}{c} - 1\right) T_s' \left\{\frac{\overline{T}_s - T_c}{\lambda} + \frac{\overline{L}_s}{c}\right\} \frac{m_s'}{\overline{m}}\right] \tag{15}$$

Finally, an assumption must be made in respect to the rate of conversion of solid to gas at the interface. It is common practice to use a pyrolysis law of the Arrhenius form

$$m_s = B_s p^{n_s}\ T_s^{\alpha_s}\ e^{-E_s/R_o T_s} \tag{16}$$

for which the fluctuation in mass flux is given by

$$\frac{m_s'}{\overline{m}} = E\,\frac{T_s'}{\overline{T}_s} + m_s\,\frac{\rho'}{\rho} \tag{17}$$

where $E = \alpha_s + E_s/R_o\overline{T}_s$. Combination of Eq. 15 and 17 yields

$$\frac{1}{\overline{m}cT_s} k_g \left(\frac{\partial T'}{\partial x}\right)_{s+} = \left(\lambda + \frac{A}{\lambda}\right) \frac{T_s'}{\overline{T}_s} + \left(\frac{c_p}{c} - 1 + \frac{LA}{1 - T_c/\overline{T}_s}\right) \frac{T_s'}{\overline{T}_s}$$
$$+ n_s \left(L + \frac{1 - T_c/\overline{T}_s}{\lambda}\right) \frac{p'}{p} \tag{18}$$

where $A = (1-T_c/\overline{T}_s)\ (\alpha_s + E_s/R_o\overline{T}_s)$ is the parameter mentioned in the introduction and $L = \overline{L}_s/c\overline{T}_s$. This completes the information required of the solid and interface regions.

III. STEADY-STATE SOLUTION FOR THE GAS PHASE WITH UNIFORMLY DISTRIBUTED COMBUSTION

The solution to the problem was summarized in Ref. 3, but not in a form immediately suitable to the analysis of unsteady behavior. Thus, a more complete discussion is necessary here. As formulated in both Ref. 2 and 3, the analysis of the burning solid is carried out within a wholly "thermal theory"--the influence of diffusion appears nowhere explicitly. The conditions under which this idealized limit is attained are discussed in Ref. 8 and in the literature of flame theory; the pressure may be taken to be uniform in space and the problem then consists in solution of the energy equation for the gas phase

$$mc_\rho \frac{dT}{dx} - \frac{d}{dx}\left(k_g \frac{dT}{dx}\right) = Q_f \rho_g \dot{\varepsilon} \qquad (19)$$

where, to conform with the notation of Ref. 2, Q_f is the heat release per unit mass and $\dot{\varepsilon}$ is the reaction rate ($\sec^{-1}$). For convenience, and also in accord with Ref. 2, k_g and c_p will be taken to be constant; however (contrary to Ref. 2), $c_p \neq c$. The calculations to be carried out here are based on the case when $\rho_g \dot{\varepsilon} = w$ is constant for $x_i \leq x \leq x_f$ and zero elsewhere, as shown in Fig. 1.

At the downstream side of the flame ($x \geq x_f$), the boundary conditions are

$$\left.\begin{array}{l} T = T_f \\ \\ \dfrac{dT}{dx} = 0 \end{array}\right\} \quad (x \geq x_f) \qquad (20a,b)$$

and at the surface, Eq. 11 holds: note that in this section T and $\bar{T}$ are identical, as are m and $\bar{m}$ since only strict steady conditions are considered. It should be noted that L_s here is $-Q_s$ of Ref. 2, and x here is -x of Ref. 2. For the purposes of this section, then, the boundary condition Eq. 11 can be written

$$\left(k_g \frac{dT}{dx}\right)_{s+} = m\,[c\,(T_s - T_c) - Q_s] \qquad (21)$$

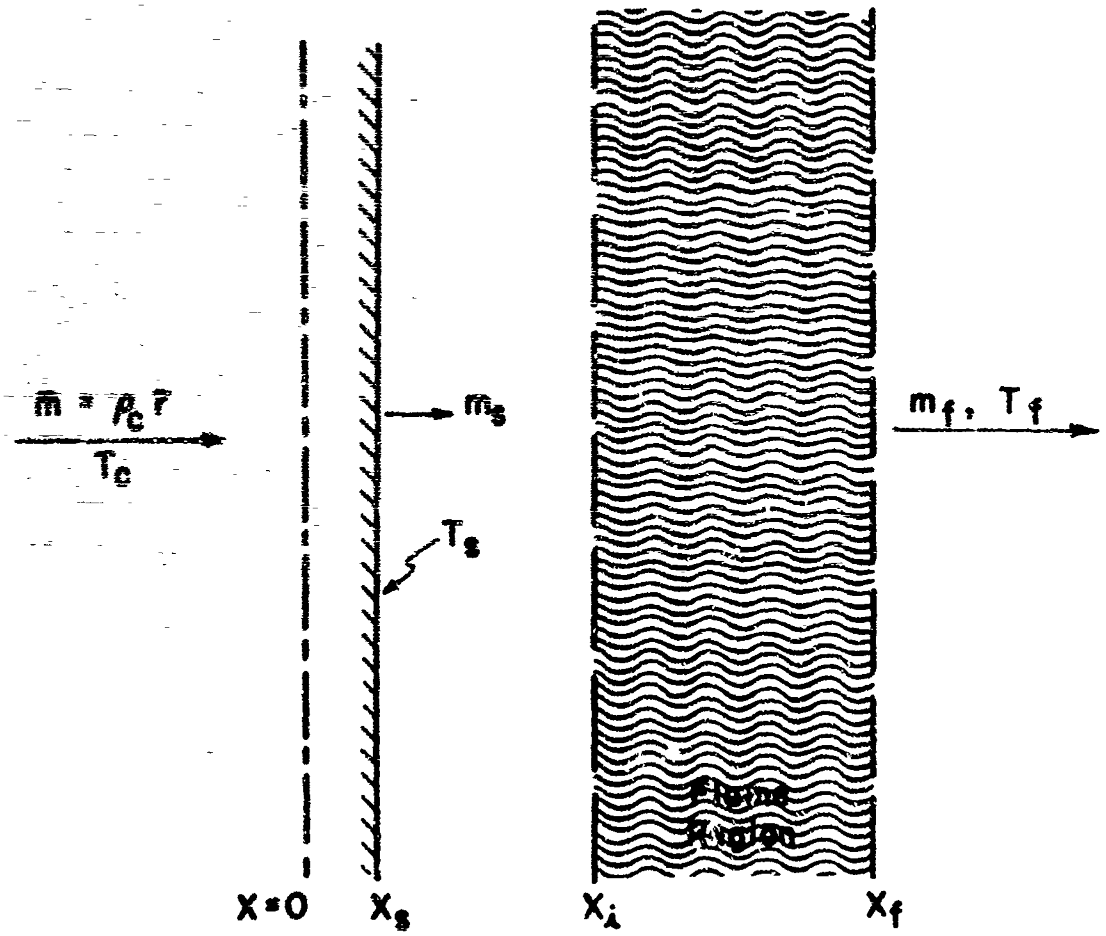

FIG. 1. Sketch of the Model Used for a Solid Propellant Burning With Uniform Combustion in the Gas Phase.

Energy conservation applied to the gas phase only gives

$$\left(k_g \frac{dT}{dx}\right)_{s+} = m\ (h_{s+} - h_f) \tag{22}$$

Thus, Eq. 21 and 22 lead to

$$m\ (h_{s+} - h_f) = m\ [c\ (T_s - T_c) - Q_s] \tag{23}$$

Now, by definition of the enthalpy function,

$$h_f - h_{s+} = h_{fr} + \int_{T_r}^{T_f} c_{pf}\ dT - h_{sr} - \int_{T_r}^{T_s} c_{ps}\ dT$$

where h_{fr}, h_{sr} are the enthalpies of formation at the reference conditions T_r and some pressure which may be taken to be the pressure at which the burning occurs. For constant specific heats, and $c_{pf} = c_{ps}$, the definition therefore yields

$$h_f - h_{s+} = -\ Q_f + c_p\ (T_f - T_s) \tag{24}$$

in which the heat release Q_f in the gas phase is by definition $H_{sr} - h_{fr}$ and is the same as Q_f in the energy equation, Eq. 19. The overall energy balance for the gas phase can be written, by combining Eq. 22 and 24, as

$$\left(k_g \frac{dT}{dx}\right)_{s+} = m\ [Q_f - c_p\ (T_f - T_s)] \tag{25}$$

The last equation is, of course, simply an expression for the overall energy balance of the process; a simple limiting form arises when $c_p = c$, so that when equated, Eq. 21 and 25 give

$$c_p\ (T_f - T_c) = Q_f + Q_s \tag{26}$$

which is practically obvious, since $Q_f + Q_s$ is the net heat release due to chemical reactions. But the point of this exercise is to compare Eq. 25 with the corresponding expression deduced directly from the energy Eq. 19. Integrate that equation from the surface to $x \rightarrow \infty$ downstream of the flame and apply the boundary conditions Eq. 20a,b:

$$m c_p (T_f - T_s) + \left(k_g \frac{dT}{dx}\right)_{o+} = Q_f \int_0^\infty \rho_g \dot{\varepsilon}\, dx \tag{27}$$

Equation 27 is consistent with Eq. 25 only if the reaction rate satisfies the normalization condition

$$\int_0^\infty \rho_g \dot{\varepsilon}\, dx = m \tag{28}$$

In Ref. 3 it was shown that this condition is redundant for the case of a very thin flame front (combustion concentrated at a plane)--it gives no information that cannot be obtained by other means. This is also true for the case of uniform combustion. However, it is a convenient relation to have at hand, particularly for the problem of quasi-static burning in response to pressure changes. It is worthwhile noting that Eq. 28 also leads directly to a relation which was introduced on an approximation in Ref. 2 but which is in fact exact. For suppose that the combustion occurs only in the region $x_i \leq x \leq x_f$ and is uniform there; then Eq. 28 gives

$$\rho_g \dot{\varepsilon} (x_f - x_i) = m \tag{29}$$

In the extreme case $x_i = 0$,

$$w \equiv \rho_g \dot{\varepsilon} = \frac{m}{x_f} \tag{30}$$

a relationship which was adopted as an approximation just before Eq. 8 of Ref. 2.

As a convenience[3] for integrating Eq. 19, it is helpful to define a new variable ζ (called ξ in Ref. 3):

$$\zeta = e^{\frac{mc_p}{k_g} dx} \tag{31}$$

so that

$$\frac{d}{dx} = \frac{d\zeta}{dx} \frac{d}{d\zeta} = \frac{mc_p}{k_g} \zeta \frac{d}{d\zeta} \tag{32}$$

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[3] And only a convenience--the steps leading to Eq. 32 are obviously unnecessary, but using ζ instead of x does simplify writing subsequent manipulations.

With this transformation, the left-hand side of Eq. 19 is

$$\frac{mc_p}{k_g} \zeta \frac{dT}{d\zeta} - \frac{mc_p}{k_g} \zeta \frac{d}{d\zeta} \left(mc_p \; \zeta \frac{dT}{d\zeta} \right) \equiv - \frac{(mc_p)^2}{k_g} \zeta^2 \frac{d^2T}{d\zeta^2}$$

and the whole equation can be written

$$- \zeta^2 \frac{d^2\tau}{d\zeta^2} = \Lambda^2 \tag{33}$$

with $\tau = T/T_s$, and the eigenvalue Λ^2 is defined as

$$\Lambda^2 = \frac{Q_f \, k_g \, w}{m^2 c_p{}^2 T_s} \tag{34}$$

Since $w = \rho_g \dot{\varepsilon}$ is here taken to be nonzero only in the interval $x_i \leq x \leq x_f$ where it is constant, Λ^2 is also nonzero and constant only in the same region. Thus, Eq. 33 is easily integrated to give $d\tau/d\zeta$ and τ as follows:

$$\frac{d\tau}{d\zeta} = c_1 \tag{35}$$

$$\zeta_s \leq \zeta \leq \zeta_i$$

$$\tau = c_1 \zeta + c_2 \tag{36}$$

$$\frac{d\tau}{d\zeta} = \frac{\Lambda^2}{\zeta} + c_3 \tag{37}$$

$$\zeta_i \leq \zeta \leq \zeta$$

$$\tau = \Lambda^2 \ln \zeta + c_3 \zeta + c_4 \tag{38}$$

$$\frac{d\tau}{d\zeta} = c_5$$

$$\zeta_f \leq \zeta_f \tag{39a,b}$$

$$\tau = c_5 \zeta + c_6$$

There are eight conditions to be satisfied: the boundary values $T = T_s$ and Eq. 21 at the surface, Eq. 20a,b downstream of the flame $(x \to \infty)$, and continuity of temperature and heat transfer at the boundaries of the flame, $x = x_i$ and $x = x_f$. As part of the given information defining the problem, w and an "ignition temperature," T_i, at which combustion begins, must be specified. From T_i, x_i, or ζ_i can be calculated from Eq. 36. There are therefore eight quantities determined by the eight conditions listed above: the six constants of integration, $c_1, \ldots, c_6$, the flame thickness x_f, and finally the eigenvalue Λ^2. The end result is an expression for Λ^2 which can be solved to give the mass flux or linear burning rate as a function of pressure and other variables in the problem.

A direct way to apply the various conditions is to find the temperature profiles for the upstream and downstream sides of $x = x_i$ and match there. First, the surface condition on heat transfer, Eq. 21, is in the normalized variables:

$$\zeta_s \left(\frac{d\tau}{d\zeta}\right)_{s+} = \frac{1}{c_p T_s} \left[c(T_s - T_c) - Q_s\right] \tag{40}$$

The correct values are taken by the profile Eq. 35-36 at the surface if

$$c_1 = \left(\frac{d\tau}{d\zeta}\right)_{s+}$$

$$c_2 = \tau_s - \zeta_s \left(\frac{d\tau}{d\zeta}\right)_{s+} \tag{41}$$

so that between the interface and the upstream edge of the flame,

$$\tau = \tau_s + (\zeta - \zeta_s) \left(\frac{d\tau}{d\zeta}\right)_{s+} \qquad (\zeta_s \leq \zeta \leq \zeta) \tag{42}$$

where $(d\tau/d\zeta)_{s+}$ is given by Eq. 40.

The conditions Eq. 20a,b far downstream require

$$c_5 = 0$$

$$c_6 = \tau_f \tag{43a,b}$$

Hence, continuity of temperature and heat transfer at $\zeta = \zeta_f$ are satisfied if in Eq. 38 and 39

$$c_3 = -\frac{\Lambda^2}{\zeta_f}$$

$$c_4 = \tau_f + \Lambda^2 (1 - \ln \zeta_f) \tag{44a,b}$$

and the temperature profile in the flame zone is

$$\tau = \tau_f - \Lambda^2 \left[\ell n\left(\frac{\zeta_f}{\zeta}\right) - \left(1 - \frac{\zeta_f}{\zeta}\right)\right](\zeta_i \leq \zeta \leq \zeta_f) \tag{45}$$

which must be matched to Eq. 42 at $\zeta = \zeta_i$. Continuity of temperature there gives

$$\Lambda^2 = \frac{\tau_f - \tau_i}{\ell n \frac{\zeta_f}{\zeta_i} - (1 - \frac{\zeta_i}{\zeta_f})} \tag{46}$$

and continuity of heat transfer implies

$$\Lambda^2 = \frac{\zeta_f \, \zeta_i}{\zeta_f - \zeta_i}\left(\frac{d\tau}{d\zeta}\right)_{s+} \tag{47}$$

It is easily verified, when due account is taken of the definitions involved, that Eq. 47 is exactly Eq. 7 of Ref. 2.

For completeness, it may be noted that the normalization condition, Eq. 29, in dimensionless variables is

$$\Lambda^2 = \frac{(Q_f/c_p\overline{T}_s)}{\ell n(\zeta_f/\zeta_i)} \tag{48}$$

Any two of Eq. 46-48 give an equation which must be solved to find ζ_f:

$$\frac{\zeta_f}{\zeta_i} = 1 + \left(\frac{c_pT_s}{Q_f}\right) \zeta_f \left(\frac{d\tau}{d\zeta}\right)_{s+} \ell n\left(\frac{\zeta_f}{\zeta_i}\right) \tag{49}$$

(Observe for use in Eq. 46 that according to Eq. 25, $\tau_f - \tau_i = \frac{Q_f}{c_pT_s} - (d\tau/d\zeta)_{s+}$.)

Numerical calculation of the steady-state problem proceeds as follows:

(1) The quantities T_f, Q_s, Q_f are assumed known by thermodynamics or otherwise, while the temperature of the cold propellant T_c can be chosen as desired. The reaction rate w must be specified as a function, say, of pressure and average temperature.

(2) Choose a value for T_s; consider the case $n_s = 0$ so that m can be computed from the pyrolysis law, Eq. 16

$$m = B_s T_s^{\alpha_s} e^{-E_s/R_oT_s}$$

where B_s, α_s, E_s are assumed known.

(3) In the purely steady-state problem, the interface can be fixed at the origin, so $x_s = 0$ and $\zeta_s = 1$; $(d\tau/d\zeta)_{s+}$ can then be computed from Eq. 40 with $\zeta_s = 1$:

$$\left(\frac{d\tau}{d\zeta}\right)_{s+} = \frac{1}{c_pT_s}\left[c(T_s - T_c) - Q_s\right] \tag{50}$$

(4) The position of the inner edge of the flame follows from the specified value of T_i used in Eq. 42, again with $\zeta_s = 1$:

$$\zeta_i = 1 + \frac{\tau_i - \tau_s}{(d\tau/d\zeta)_{s+}} \tag{51}$$

(5) The position of the outer edge of the flame can be found from the transcendental Eq. 49.

(6) The value of the eigenvalue can then be calculated from, say, Eq. 48:

$$\Lambda^2 \equiv \frac{Q_f k_g w}{m^2 c_p^2 T_s} = \frac{1}{\ln(\zeta_f/\zeta_i)}$$

from which, since m was already calculated in step (2), a number is found for w. Since the reaction rate is generally a function of pressure (specified), this last step gives the pressure.

Thus, the net result is the variation of mass flux or linear burning rate with pressure. If $n_s \neq 0$ in Eq. 16, then an iterative calculation is required: a value of pressure must be assumed initially to compute m from the pyrolysis law. The value computed in step (6) must then be compared with the value assumed in step (1) and the calculations repeated until satisfactory agreement is achieved.

This is an interesting problem to complete, for comparison with experimental data for the burning of composite propellants: a thorough discussion, with numerical results, is being prepared. The point of including the discussion here is to clarify just what must be known to solve the problem, and just what can be calculated, for a clear understanding of steady-state burning in this respect is helpful in analysis of the quasi-static behavior.

IV. QUASI-STATIC BEHAVIOR OF THE GAS PHASE

When the pressure varies in time, but is still assumed to be uniform in space, the equations of motion for the gas phase are

$$\frac{\partial \rho}{\partial t} + \frac{\partial m}{\partial x} = 0 \tag{52}$$

$$\rho c_p \frac{\partial T}{\partial g} + m c_p \frac{\partial T}{\partial x} = \frac{\partial}{\partial x}\left(k_g \frac{\partial T}{\partial x}\right) + \frac{\partial p}{\partial t} + Q_f \rho_g \dot{\varepsilon} \tag{53}$$

It is a formal implication of the physical assumption of quasi-static behavior that the time derivatives in the differential equations of motion can be neglected. This is based on the idea (perhaps supported, but not necessarily wholly justified, by order of magnitude estimates) that the processes within the gas phase respond essentially instantaneously to changes of pressure and of boundary values--temperature, heat transfer, and positions of the boundaries.

The origin of the time dependence is of course the pressure field external to the flame, but the assumption of instantaneous response does not mean that other variables in the problem change in phase with the pressure. In fact, a significant lag is introduced by the behavior of the thermal wave in the solid; this influences the behavior of the gas phase mainly through the heat transfer from the interface to the solid, expressed in Eq. 8. But in consequence, the fluctuations of surface temperature, flame temperature, flame thickness, and reaction rate also cannot generally take place in phase with the pressure except in the limit $\omega = 0$. Nevertheless, the solution to the strictly steady-state solution can still be used if the influence of fluctuations is properly accounted for.

An example may serve to clarify what is meant by "quasi-static" when time lags are present. Consider the simple case of heat conduction between two planes at different temperatures T_1, T_2 and separated by a distance L. The temperature profile in the steady state is linear,

$$T(x) = T_1 + (T_2 - T_1)\frac{x}{L} \tag{54}$$

where the temperature $T = T_1$ at $x = 0$. The assumption of quasi-static behavior when small fluctuation T_1', T_2', L' occur implies that the fluctuation of the local temperature profile can be calculated simply by computing its perturbation according to the steady-state solution Eq. 54. Write $T = \overline{T} + T'$, $T_1 = \overline{T}_1 + T_1'$, etc., and since the barred quantities obey Eq. 54, one finds easily a formula for T':

$$T'(x) = T_1' + (T_2' - T_1')\frac{x}{L} - (\overline{T}_2 - \overline{T}_1)\frac{x}{L}\left(\frac{L'}{L}\right) \tag{55}$$

The total temperature profile $T = \overline{T} + T'$ as well as $\overline{T}$ and T' separately are still linear functions of the distance from the colder (T_1) plate but the ends of the profile need not jiggle back and forth in phase. It is entirely permissible, for example, that $T_1' = \hat{T}_1' \cos \omega t$ and $T_2' = \hat{T}_2' \cos (\omega t - \phi)$ so that the fluctuation in temperature of the hotter plate lags in time by the constant angle ϕ. Moreover, $\hat{T}_1'$, $\hat{T}_2'$ could be specified as functions of the frequency, ω. Similar statements apply to the variable separation of the planes. The point is that the steady-state solution fixes the functional form of the solution--in this case linear--while phase lags and dependence on frequency can be introduced through the boundary values. A truly nonsteady calculation would of course lead to a thermal wave in the region between the planes. The distinction between these cases is illustrated in Fig. 2.

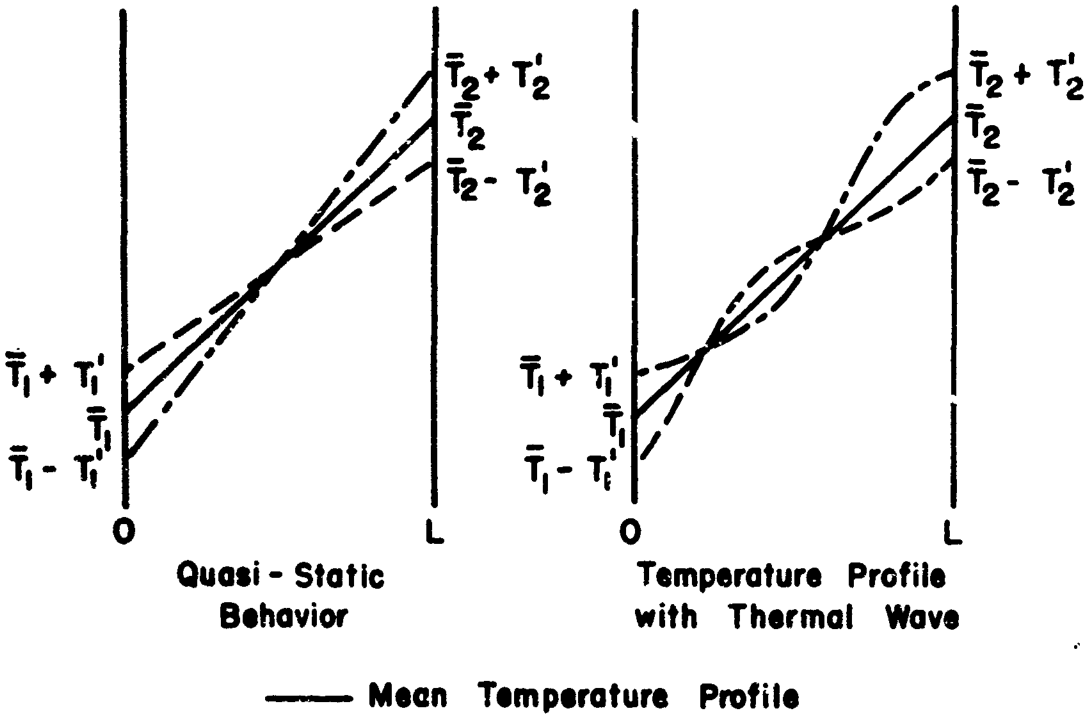

FIG. 2. An Illustration of the Difference Between Quasi-Static and Truly Unsteady Temperature Profiles in a Solid Slab.

The steady-state solution for the gas phase found in the preceding section can be handled in exactly the same way. Although the form of the profile is fixed by Eq. 42 and 45 within the gas phase, the effect of the thermal wave in the solid is to force both a phase lag and a dependence on frequency for practically all variables in the problem. In particular, and this point will be mentioned again, the fluctuation of the flame thickness x_f forces a fluctuation in the ratio $w/m = 1/x_f$ which is not in phase with the pressure.

It is perhaps best to begin the following manipulations by summarizing the results already obtained and required here. What is needed is a formula for the fluctuation of heat transfer from the gas phase to the interface, for use in the energy matching condition, Eq. 18. If $(k_g \partial T/\partial x)'_{s+}$ is expressed as a linear combination of contributions due to m_s', T_s', and p', then with Eq. 17, there is sufficient information to give a formula for the response ratio m_s'/p'. Thus, one already has

$$\frac{m_s'}{\overline{m}} = \frac{A}{1-T_c/\overline{T}_s} \frac{T_s'}{\overline{T}_s} + n_s \frac{p'}{\overline{p}} \tag{17}$$

and

$$\frac{1}{\overline{m}c\overline{T}_s} \left(k \frac{\partial T'}{\partial x}\right)_{s+} = \left(\lambda + \frac{A}{\lambda}\right) \frac{T_s'}{\overline{T}_s} + \left(\frac{c_p}{c} - 1 - HA\right) +$$

$$\frac{T_s'}{\overline{T}_s} + n_s \left(1 - \frac{T_c}{\overline{T}_s}\right)\left(\frac{1}{\lambda} - H\right) \frac{p'}{\overline{p}} \tag{18}$$

For easier comparison with Ref. 2, $L = \overline{L}_s/c\overline{T}_s$ has been replaced by $H = Q_s/c\ (\overline{T}_s - T_c) = - L/(1 - T_c/\overline{T}_s)$.

When the assumption of quasi-static behavior is made, there are two ways of arriving at a formula for $k_g(\partial T'/\partial x)_{s+}$, both giving the same result. One way is to linearize the two equations, Eq. 52 and 53, by writing $T = \overline{T} + T'$ and solving for the fluctuations with the time derivatives ignored. This, as shown later in Section V, is really the first term of an expansion in powers of a frequency parameter. The second way is simply to linearize the steady-state solution, as done above for heat flow between two plates. The equivalence of these two methods is readily demonstrated and is shown below.

A. Quasi-Static Results from the Steady-State Solution

This means of finding the quasi-static formulas has been widely used, sometimes incorrectly; any steady-state solution for the gas phase can be used as a basis. The steady-state problem of Section III from which the quasi-static formula for $(k_g\ \partial T'/\partial x)_{s+}$ may be found has yielded the following:

(1) The temperature profile

$$\tau = \tau_s + (\zeta - \zeta_s)\left(\frac{d\tau}{d\zeta}\right)_{s+} \qquad (\zeta_s \leq \zeta \leq \zeta_i) \tag{42}$$

$$\tau = \tau_f - \Lambda^2\left[\ell n\,\frac{\zeta_f}{\zeta} - 1 - \frac{\zeta}{\zeta_f}\right] \qquad (\zeta_i \leq \zeta \leq \zeta_f) \tag{45}$$

(2) Three expressions for the eigenvalue $\Lambda^2 \equiv k_g\ wQ_f/m^2 c_p{}^2 \bar{T}_s$

$$\Lambda^2 = \frac{\tau_f - \tau_i}{\ell n\,\dfrac{\zeta_f}{\zeta_i} - \left(1 - \dfrac{\zeta_f}{\zeta_i}\right)} \tag{46}$$

$$\Lambda^2 = \frac{\zeta_f \zeta_i}{\zeta_f - \zeta_i}\left(\frac{d\tau}{d\zeta}\right)_{s+} \tag{47}$$

$$\Lambda^2 = \frac{1}{\ell n(\zeta_f/\zeta_i)} \tag{48}$$

(3) A transcendental equation for the flame thickness, found by equating Eq. 47 and 48

$$\frac{\zeta_f}{\zeta_i} = 1 + \left(\frac{d\tau}{d\zeta}\right)_{s+} \zeta_f\ \ell n\left(\frac{\zeta_f}{\zeta_i}\right) \tag{49}$$

(4) The overall energy balance for the gas phase Eq. 25

$$k_g\left(\frac{dT}{dx}\right)_{s+} = m\,[Q_f - c_p\,(T_f - T_s)] \tag{25}$$

which in dimensionless variables is

$$\zeta_s\left(\frac{d\tau}{d\zeta}\right)_{s+} = 1 - (\tau_f - \tau_s) \tag{25a}$$

(5) A normalization conditions, Eq. 29, on the reaction rate w, which is implied by Eq. 25 and which, rewritten, is Eq. 48:

$$w = \frac{m}{x_f - x_i} \quad . \tag{29}$$

Not all of the above are independent results: by use of the expressions for the temperature profile and the energy balance Eq. 25a, any two of the formulas for the eigenvalue can be derived from the third. The model has been solved exactly, but numerical results cannot be obtained until the reaction rate w is independently specified.

The fluctuation of heat transfer is found by linearizing in the usual way: $T = \overline{T} + T'$, $m = \overline{m} + m'$, etc. A question arises whether the values indicated as $(\ )_{s+}$ here are really those at the interface or whether, as for the solution, Eq. 3, to the solid phase, they represent values on $x = 0$, from which the correct values at the interface must be found by using the linearized boundary conditions Eq. 6 and 7. The answer is that within the quasi-static assumption, the terms involving x_s must be ignored. The reason for this is that in the limit $\omega = 0$, the fluctuating part, $\dot{x}_s$, of the surface motion is zero, so that x_s is a constant which, by free choice of the origin, may be taken to be zero. In other words, it is inappropriate to use the formula $\dot{x}_s = i\omega x_s$ and hence in the limit $\omega = 0$, x_s cannot be found from Eq. 13. Incidentally, it is easily verified that the correct quasi-static limit (i.e., $\omega = 0$) does follow from, for example, Eq. 18 for the solid phase, even though that relation involves use of Eq. 13. The reason for this is that the material of the solid phase is assumed to move always at the uniform speed $\overline{r} = \overline{m}/\rho_c$. In the treatment of the gas phase, fluctuations of the speed of the gas are necessarily included, and hence neglect of the terms in x_s must be enforced as a separate statement, part of the quasi-static analysis. A more careful handling of the surface boundary conditions is required if the assumption of quasi-static behavior is relaxed, or if nonharmonic changes of pressure are considered. The point here is that so far as periodic quasi-static variations are concerned, values denoted by $(\ )_{o+}$ are identical with values $(\ )_{s+}$. More detailed considerations appear in Section V.

To find an expression for $(dT'/dx)_{s+}$, consider Eq. 47, with the definition of Λ^2:

$$\left(\frac{d\tau}{d\zeta}\right)_{s+} \equiv \frac{k_g}{mc_pT_s}\ \frac{1}{\zeta_s}\left(\frac{dT}{dx}\right)_{s+} = \frac{Q_f\ k_g\ w}{m^2c_p{}^2T_s}\ \left(\frac{1}{\zeta_i} - \frac{1}{\zeta_f}\right)$$

which gives, since $\zeta_s = 1$ for $x_s = 0$,

$$\left(k_g \frac{dT}{dx}\right)_{s+} = \frac{Q_f k_g}{c_p}\left(\frac{1}{\zeta_i} - \frac{1}{\zeta_f}\right)\frac{w}{m} \tag{56}$$

As already remarked, this can be found by direct integration of the energy equation and appears as Eq. 7 of Ref. 2 and Eq. 87 below. Before considering refinements, the simple case of uniform combustion beginning at the surface will be discussed, the problem handled in Ref. 2; thus, $\zeta_i = 1$. Moreover, the approximation that $\zeta_f >> 1$ will be introduced at this point, and used in almost all the remaining discussion. Hence, Eq. 56 becomes

$$\left(k_g \frac{dT}{dx}\right)_{s+} = \frac{Q_f k_g}{c_p}\left(\frac{w}{m}\right) \tag{57}$$

A useful interpretation of "large $\bar{\zeta}_f$" follows from Eq. 47 and 48, which show that for large $\bar{\zeta}_f$,

$$\left(\frac{d\tau}{d\zeta}\right)_{s+} \equiv \frac{k_g}{\bar{m} c_p \bar{T}_s}\left(\frac{dT}{dx}\right)_{s+} \simeq \frac{1}{\ln\zeta_f}$$

and hence the heat transfer to the solid from the gas is small compared to the energy transferred by convection. Moreover, with Eq. 40, this relation becomes

$$\frac{c}{c_p}\left(1 - \frac{T_c}{\bar{T}_s}\right)(1 - H) \simeq \frac{1}{\ln\zeta_f} = \frac{k_g}{\bar{m} c_p \bar{T}_f} \tag{58}$$

where $H = Q_s/c\,(\bar{T}_s - T_c)$ will appear as an important parameter in the final results for the response function. For practical cases, $T_c/\bar{T}_s \sim 1/3$, so that one must choose H near unity to be consistent with the approximation that ζ_f is large. However, this is not especially troublesome, providing $H > 0$, since $\bar{\zeta}_f$ can be considered large when $\ln\zeta \gtrsim 2$. Then since $c_p/c \sim ½$, $1-H \sim (1/2)/(2/3) = 0.75$ and $H = 0.25$; in other words, the approximation $\zeta_f >> 1$ accommodates positive values of H in the range 0.25 – 1.

A rough numerical estimate indicates that ζ_f may indeed be quite large. Since $\bar{m} c_p \bar{x}_f/k_g = (\rho_c c_p/k_g)\,\overline{r x_f} = (c_p k_c/c k_g)\,\overline{r x_f}/\kappa_c$, and $c_p k_c/c k_g \simeq 1$, one has $\bar{\zeta}_f \sim \exp\,(\overline{r x_f}/\kappa_c)$. A representative value of κ_c is 10^{-3} cm^2/sec and for $r = 1$ cm/sec, $\bar{\zeta}_f \sim \exp\,(1000\,\bar{x}_f)$ with $\bar{x}_f$ in centimeters. Even for $\bar{x}_f$ as unrealistically small as 20 microns = 0.002 centimeters, $\bar{\zeta}_f \sim e^2$.

If, on the other hand, H is negative, then instead of Eq. 58 one has

$$\frac{c}{c_p}\left(1-\frac{T_c}{\bar{T}_s}\right)(1+(-H)) = \frac{1}{\ln(\bar{\zeta}_f/\bar{\zeta}_i)}\left(\frac{1}{\bar{\zeta}_i}-\frac{1}{\bar{\zeta}_f}\right) \tag{58a}$$

and it is apparent that $H < 0$ and large values of ζ_f are not compatible unless ζ_i is also fairly large. The treatment of Ref. 2 is therefore restricted to positive values of H (i.e., exothermic surface reactions) because the assumptions are made that ζ_f is large and that $\zeta_i = 1$ (i.e., combustion begins at the surface).

The quasi-static formula for the fluctuation of heat transfer is found simply by linearizing, Eq. 57:

$$\left(k_g\frac{\partial T}{\partial x}\right)_{s+}' = \frac{Q_f k_g \bar{w}}{c_p \bar{m}}\left(\frac{w'}{\bar{w}}-\frac{m'}{\bar{m}}\right)$$

$$= \bar{m}\, c_p\, \bar{T}_s\, \Lambda^2\left(\frac{w'}{\bar{w}}-\frac{m'}{\bar{m}}\right) \tag{59}$$

where, as usual, $\Lambda^2 = Q_f k_g \bar{w}/(c_p\bar{m})^2\, \bar{T}_s = (Q_f/c_p\bar{T}_s)\,(\ln \bar{\zeta}_f)^{-1}$ by Eq. 48. The fluctuation of mass flux is related to the pressure and surface temperature fluctuations by the linearized pyrolysis law, Eq. 17.

A second formula for the heat transfer at the surface may be deduced by linearizing Eq. 25 for the overall energy balance of the gas phase:

$$\left(k_g\frac{\partial T}{\partial x}\right)_{s+}' = m'\,[Q_f - c_p\,(\bar{T}_f - \bar{T}_s)] - \bar{m}c_p\,(T_f' - T_s') \tag{60}$$

As later calculations will show, this relationship is used to compute the fluctuations of flame temperature, T_f', once the remainder of the problem has been solved.

The preliminary calculations are completed by finding a formula for the fluctuation, w', of reaction rate. First, following Ref. 2, it may be most simply supposed that w' is the fluctuation of the steady-state reaction rate w(p) which was computed in the work in the following way. The average heat transfer from the solid to the gas is given by the results above as Eq. 58. But from the matching condition at the interface and the temperature distribution in the solid Eq. 11,

$$\left(k_g\frac{dT}{dx}\right)_{s+} = m\,[c\,(T_s - T_c) - Q_s]$$

Equating these expressions gives

$$\frac{Q_f k_g}{c_p} w = m^2 \left[c \left(T_s - T_c\right) - Q_s\right] \tag{61}$$

It is supposed that the right-hand side can be expressed as a function of pressure only. This is accomplished by assuming that the linear burning rate may be approximated by the law $m = ap^n$ and that an approximation to the Arrhenius law is

$$m = b(T_s - T_c)^s p^{n_s} \tag{62}$$[4]

so that $T_s - T_c = (p^{n-n_s} a/b)^{1/s}$; hence, Eq. 61 reads

$$\frac{Q_f k_g}{c_p} w = (ap^n)^2 \left[c \left(\frac{a}{b} p^{n-n_s}\right)^{1/s} - Q_s\right] \tag{63}$$

In this way w is given by Eq. 63 as a function of pressure only, and has been so constructed as to satisfy the equations for steady burning for the case $m = ap^n$ and a pyrolysis law approximated by Eq. 62.

The assumption is now made that even during oscillations, the reaction rate is given instantaneously by the same function of pressure, so that w' is computed by setting $p = \bar{p} + p'$ in Eq. 63 and retaining only terms linear in p'. This implies that the local fluctuation of energy release, $Q_f w'$, is also always in phase with the pressure fluctuations.

Now the temperature fluctuations are not in phase with the pressure. For example, the fluctuation of temperature at the edge of the flame, T_f', follows from Eq. 60:

$$T_f' = T_s' + \frac{m'}{\bar{m}} \left[\frac{Q_f}{c_p} - (\bar{T}_f - \bar{T}_s)\right] - \frac{1}{\bar{m} c_p} \left(k_g \frac{\partial T}{\partial x}\right)'_{s+}$$

where $(k_g \, \partial T'/\partial x)_{s+}$ is given here by Eq. 59. Thus, if the local reaction rate is at all sensitive to temperature, there must be a part of w' which (contrary to the assumptions used in Ref. 2 and expressed above) is not in phase with pressure oscillations. The result that w' is entirely in

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[4] Note that $n_s = 0$ in Ref. 2.

phase with pressure is a consequence of two assumptions: (1) that the reaction rate is everywhere in the gas phase a function of pressure only, and (2) quasi-static behavior.

Even though one may represent the steady-state global combustion process as a function of pressure, uniform in space, this does not mean that locally, within the gas-phase flame, one can always simply ignore other variables. In the analysis, the influences of those variables are smeared out in the steady state to give the uniform function of pressure w(p) derived above. In fact, it may be better to regard the calculation as a means of relating the overall rate of energy release, $Q_f w x_f$, to the linear burning rate. Treatment of this as a function of pressure only amounts to finding what the (perhaps fictitious) uniform function of pressure would have to be in order to match an observed burning rate, which is here approximated as ap^n.

The view that the reaction rate depends almost entirely on pressure is based on the idea that the overall burning rate is diffusion controlled. Nevertheless, locally the homogeneous gas-phase reactions must respond to temperature changes, and of course the process of diffusion is itself dependent on temperature. Moreover, under unsteady conditions, there are likely to be fluctuations of mixture ratio. Both of these will affect the local reaction rate and will therefore contribute pieces to w' which are not in phase with the pressure change, even though the processes are treated in a quasi-static manner. Such a possibility is not strictly consistent with the assumption that the reaction rate is uniform in space. That assumption necessarily precludes consideration of dependence of w on variables which exhibit spatial variations.

Thus, one concludes that if one requires the fluctuating, as well as the steady-state, reaction rate to be uniform in space, one is seriously restricted; the energy release is in phase with the pressure. In fact, that conclusion holds even if the assumption of quasi-static behavior is relaxed as shown in Section V. The calculation of Ref. 2 therefore cannot be usefully extended without altering certain approximations on which that work is based.

In summary, the three main approximations which qualify the results of Ref. 2 are:

(1) That combustion in the gas phase begins immediately at the interface

(2) That the flame zone is relatively thick, thus, as the remarks following Eq. 58a show, excluding consideration of endothermic surface reactions unless approximation (1) is relaxed

(3) That the fluctuations of energy release are uniform in the gas phase and hence always in phase with the pressure fluctuations.

The analysis of Ref. 2 is incomplete so that the approximation (1) could not be corrected; a procedure for doing so is developed below in Section IV-C. Correction of (2) to include endothermic reactions can be done without essential difficulty, but it is lengthy and will not be included here. Alternatives to the approximation (3) are more difficult to construct but may be more important than relaxation of (1). Some remarks on this question are given in Section IV-D.

If Eq. 63 is used, the fluctuation of reaction rate is

$$\frac{c_p}{c}\left(\frac{\Lambda^2}{1-T_c/\bar{T}_s}\right)\frac{w'}{\bar{w}} = W\,\frac{p'}{\bar{p}} \tag{64}$$

where

$$W = n\left[2(1-H) + \frac{c_p}{c}\,\frac{(1-n_s/n)}{s}\right] \tag{65}$$

and $H = Q_s/c(\bar{T}_s-T_c)$. The factor c_p/c in $c_p/c)(n-n_s)/s$ arises from the fluctuation of Q_s, $Q_s' = -(c_p-c)T_s'$, which appeared also in Eq. 14.

B. Calculation of the Response Function for the Simplest Case

Before further consideration of the approximations noted above, it is useful to compute the response function for the simplest case, thereby showing most clearly how the various pieces of the problem fit together. For this purpose, all that is required is contained in Eq. 17, 18, 59, and 64. Substitution of Eq. 64 into Eq. 59 gives what amounts to the net result of analysis of the gas phase:

$$\frac{1}{\bar{m}c\bar{T}_s}\left(\frac{\partial T}{\partial x}\right)'_{s+} = \left(1-\frac{T_c}{\bar{T}_s}\right)W\,\frac{p'}{\bar{p}} - \frac{c_p}{c}\,\Lambda^2\,\frac{m'}{\bar{m}} \tag{66}$$

Now substitution of this formula into the energy matching Eq. 18, and use of the pyrolysis law, Eq. 17, to eliminate $T_s'/\bar{T}_s$, leads to

$$\frac{m'/\bar{m}}{p'/\bar{p}} = \frac{\left(AW + \frac{c_p}{c}\,n_s\right) + n_s\,(\lambda-1)}{\lambda + \frac{A}{\lambda} + \frac{c_p}{c}\,E\Lambda^2 - HA + \frac{c_p}{c} - 1} \tag{67}$$

Now for the assumed form of the mean burning rate, $m = ap^n$, the limiting value of the above ratio for $w = 0$ must be n (everything changes in phase with the pressure). Hence, since $\lambda \to 1$, this condition requires

$$n \left[A(1-H) + \frac{c_p}{c} E\Lambda^2 + \frac{c_p}{c} \right] = AW + \frac{c_p}{c} n_s \tag{68}$$

and with W given by Eq. 65,

$$\frac{c_p}{c} E\Lambda^2 + \frac{c_p}{c} \left(1 - \frac{n_s}{n}\right) = A\ (1-H) + \frac{A}{s}\ \frac{c_p}{c} \left(1 - \frac{n_s}{r}\right)$$

That this is identically satisfied can be shown in the following way. First, the index s (called m(!) in Ref. 2) is in fact exactly equal to the more commonly used A. To see this, write the linearized form of the pyrolysis law, Eq. 62,

$$\frac{m'}{\overline{m}} = \frac{}{1-T_c/T_s}\ \frac{T_s'}{\overline{T}_s} + n_s \frac{p'}{\overline{p}}$$

By comparison with Eq. 17, obviously $s = E(1-T_c/\overline{T}_s) = A$. Thus, Eq. 68 requires $A(1-H) = c_p E\Lambda^2/c$. But this is merely the mean energy balance at the surface, written in a slightly obscure form. For with Eq. 57 substituted into Eq. 11 with $L_s = -Q_s$, one has

$$\frac{Q_f\ k_g\ \overline{w}}{\overline{m}\ c_p} = mc\ (\overline{T}_s - T_c)\ (1-H)$$

and by use of the definitions of A and Λ^2, multiplication by E gives

$$\frac{c_p}{c} E\Lambda^2 = A\ (1-H) \tag{69}$$

Thus, Eq. 67 has the correct limit "built-in"; this will always be the case if correct use is made of the steady-state energy balance (see Ref. 5, Eq. 40, for example). This formula can be put into the standard form suggested in Ref. 1 by defining

$$B = \frac{W}{n} + \frac{c_p}{c}\ \frac{n_s}{An} = 2(1-H) + \frac{c_p}{c}\ \frac{1}{A} \tag{70}$$

and in view of Eq. 68, Eq. 67 can be written

$$\frac{m'/\bar{m}}{p'/\bar{p}} = \frac{nAB + n_s\,(\lambda - 1)}{\lambda + \frac{A}{\lambda} - (1+A) + AB} \tag{71}$$

It is clear, therefore, that there are only two significant parameters, A and B, other than $\kappa_c/\bar{r}^2$ in λ and, of course, n. Note also that it is really H that is changed to change B, and that by the steady-state energy balance at the surface, Eq. 69, 1-H can also be interpreted as a combination of gas phase properties:

$$1-H = \frac{c_p}{c}\;\frac{1}{1-T_c/\bar{T}_s}\;\frac{Q_f\,k_g\,\bar{w}}{(mc_p)^2\bar{T}_s} \tag{72}$$

The limit H = 1 probably cannot be reached in practical cases because this implies, according to Eq. 11, that no heat is transferred from the gas to the solid—the heating of the solid is then due entirely to the exothermic reaction at the surface: $\bar{T}_s = \bar{T}_c + Q_s/c$. Although this does not violate conservation of energy, it is an unlikely dynamical state. The fact that Eq. 72 requires $m \to \infty$ is not significant because that result explicitly requires a nonzero value of heat transfer from the gas to the solid. Negative values of H (endothermic surface reactions) are of course accommodated by Eq. 72.

C. Combustion Zone Not Extending to the Surface

Since the assumption that the gas begins burning immediately upon issuing from the surface appears to be a serious restriction, it is useful to examine a simple way of avoiding it. Suppose, for simplicity, that the ratio of the mean position of the inner edge to that of the outer edge is a constant, $\beta : \bar{x}_i = \beta\bar{x}_f$. Then $\bar{\zeta}_i = \zeta_f^{\beta}$ and $\zeta_i'/\bar{\zeta}_i = \beta(\zeta_f'/\bar{\zeta}_f)$. If $\bar{\zeta}_f^{1-\beta} >> 1$ still, then Eq. 57 is replaced by

$$\left(k_g\,\frac{dT}{dx}\right)_{s+} = \frac{Q_f\,k_g}{c_p}\;\frac{1}{m}\;\frac{w}{\zeta_f^{\beta}} \tag{73}$$

and the quasi-static formula, Eq. 59, is replaced by

$$\left(k_g\,\frac{\partial T}{\partial x}\right)'_{s+} = \bar{m}c_p\,\bar{T}_s\,\Lambda^2\,\frac{1}{\bar{\zeta}_f^{\beta}}\left(\frac{w'}{\bar{w}} - \frac{m'}{\bar{m}} - \beta\,\frac{\zeta_f'}{\bar{\zeta}_f}\right) \tag{74}$$

The argument leading to Eq. 63 now provides

$$\left(\frac{Q_f\,k_g}{c_p}\right)w = \zeta_f^{\beta}\,(ap^n)^2\left[c\left(\frac{a}{b}\,p^{n-n_s}\right)^{1/s} - Q_s\right] \tag{75}$$

in which ζ_f must be expressed as a function of pressure from the steady-state problem (see Eq. 78 below).

If one follows the argument used in Ref. 2 and in Section IV-B, only the fluctuation w' in Eq. 74 should be computed from the steady-state relation w(p) given by Eq. 75. Observe that if one assumes that the flame thickness also responds instantaneously and in phase with the pressure, then w/ζ_f^{β} in Eq. 73 could be linearized by use of Eq. 75 and the final result for the response function would be the same as Eq. 71. Thus, in Eq. 74, ζ_f' must be replaced by use of the quasi-static linearization of Eq. 29:

$$w = \frac{m}{x_f - x_i} = \frac{c_p m^2}{k_g} \frac{1}{\left(\frac{mc_p x_f}{k_g}\right) - \left(\frac{mc_p x_i}{k_g}\right)}$$

$$= \frac{m^2 c_p}{k_g} \frac{1}{\ell n(\zeta_f/\zeta_i)}$$

$$= \frac{m^2 c_p}{k_g} \frac{1}{(1-\beta)\ell n\ \zeta_f}$$

which gives

$$\frac{\zeta_f'}{\bar{\zeta}_f} = \ell n\ \bar{\zeta}_f \left[2 \frac{m'}{\bar{m}} - \frac{w'}{\bar{w}}\right] \tag{76}$$

so that Eq. 74 is

$$\left(k_g \frac{\partial T}{\partial x}\right)'_{s+} = \bar{m}\ c_p\ \bar{T}_s\ \Lambda^2 \frac{1}{\bar{\zeta}_f^{\ \beta}} \left[(1 + \beta\ \ell n\ \bar{\zeta}_f) \frac{w'}{\bar{w}} - (1 + 2\ \beta\ \ell n\ \bar{\zeta}_f) \frac{m'}{\bar{m}}\right] \tag{77}$$

Now the relation for $\zeta_f(p)$, the true steady-state flame thickness, is easily obtained from Eq. 49 and 50, with Eq. 62 used for $T_s - T_c$. One finds, with $\zeta_i = \zeta_f^{\beta}$:

$$\zeta_f^{1-\beta} = 1 + \frac{1-\beta}{Q_f} \zeta_f\ \ell n\zeta_f \left[\left(c\ \frac{a}{b}\ p^{n-n_s}\right)^{1/s} - Q_s\right]$$

and again for large ζ_f (a circumstance which is likely always to be true),

$$\zeta_f^{\beta} \ln \zeta_f \approx \frac{Q_f/(1-\beta)}{\left[c\left(\frac{a}{b}\, p^{n-n_s}\right)^{1/s} - Q_s\right]} \tag{78}$$[5]

This gives, for use in Eq. 75,

$$\frac{\zeta_f'}{\bar{\zeta}_f} = -\mathcal{Z}\, \frac{p'}{\bar{p}} \tag{79}$$

where

$$\mathcal{Z} = \frac{c_p/c}{1-H} \left(\frac{n-n_s}{A}\right) \frac{\ln \bar{\zeta}_f}{(1+\beta \ln \bar{\zeta}_f)} \tag{80}$$

The factor c_p/c once again comes from the fluctuation of Q_s. Note that Eq. 79 shows that a decrease of flame thickness accompanies an increase of pressure, which is true for any value of β. Linearization of Eq. 75 gives now

$$\frac{1}{\bar{\zeta}_f^{\beta}} \left(\frac{c_p}{c}\right) \left(\frac{A^2}{1-T_c/\bar{T}_s}\right) \frac{w'}{\bar{w}} = [W-\beta\mathcal{Z}\ (1-H)]\ \frac{p'}{\bar{p}} = W_\beta\, \frac{p'}{\bar{p}} \tag{81}$$

Construction of the response function now proceeds exactly as in Section IV-B, except that Eq. 77 is used in place of Eq. 59, and W_β replaces W. The final result is:

$$\frac{m'/\bar{m}}{p'/\bar{p}} = \frac{AW_\beta\ (1 + \beta \ln \bar{\zeta}_f) + \frac{c_p}{c}\, n_s + n_s\ (\lambda-1)}{\lambda + \frac{A}{\lambda} + \frac{c_p}{c}\, E\, \frac{A^2}{\zeta_f^{\beta}}\ (1+2\beta \ln \bar{\zeta}_f)\ - HA + \frac{c_p}{c} - 1}$$

which again has the form of Eq. 71, with B now defined as

$$B = \frac{W_\beta}{n}\ (1 + \beta \ln \bar{\zeta}_f) + \frac{c_p}{c}\, \frac{n_s}{An} \tag{82}$$

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[5] Note that because the "1" has been dropped, the heat transfer to the solid is a small fraction of the heat released in the gas phase, and also one is still restricted to $H > 0$ - of remarks following Eq. 58.

The correct limit for $\omega = 0$ implies now

$$AW_\beta (1 + \beta \ln \bar{\zeta}_f) + \frac{c_p}{c} n_s = n \left[A(1-H) + \frac{c_p}{c} E \frac{A^2}{\bar{\zeta}_f^{\,\beta}} \cdot (1 + 2\beta \ln \bar{\zeta}_f) + \frac{c_p}{c} \right] \tag{83}$$

To check that this is identically satisfied, substitute Eq. 73 into Eq. 11 to find

$$\frac{c_p}{c} E \frac{A^2}{\bar{\zeta}_f^{\,\beta}} = A\ (1-H) \tag{84}$$

It is a simple matter of substitution to verify that the condition Eq. 83 is in fact satisfied.

For $\beta = 0$, the results of Ref. 2 are of course recovered. Moreover, since the response function, as a function of frequency, has always the same form, Eq. 71, the same numerical results are found, although with different values of some of the parameters if different formulas B are used. A convenient way of comparing is to define a new quantity H_β so that B here has the same dependence on H_β as B of Eq. 72 has on H, i.e.,

$$\frac{1}{n} W_\beta\ (1 + \beta \ln \bar{\zeta}_f) + \frac{c_p}{c} \frac{n_s}{An} = \frac{1}{n} \left[2n\ (1-H_\beta) + \frac{c_p}{c} \left(\frac{n-n_s}{A}\right) \right]$$

Since W_β is given by Eq. 81, one finds the relation between H and H_β:

$$1-H_\beta = (1-H)\ (1 + \beta \ln \bar{\zeta}_f)$$

or

$$H_\beta = H\ (1 + \beta \ln \bar{\zeta}_f) - \beta \ln \bar{\zeta}_f \tag{85}$$

The meaning of H_β is that where H is used in Ref. 2, H_β should appear to account for a combustion zone displaced from the surface. That is, the same numerical results are obtained here for a value of $H_\beta = H_o$, say, as are obtained in Ref. 2 for $H = H_o$. But if the calculations are interpreted in terms of Q_s, different results are obtained. In Ref. 2, $Q_s = c(\bar{T}_s - T_c)H$, whereas if H_β is used, Eq. 85 shows that

$$Q_s = c\,(\bar{T}_s - T_c)\left[\frac{H_\beta + \beta\,\ell n\,\bar{\zeta}_f}{1 + \beta\,\ell n\,\bar{\zeta}_f}\right] \tag{86}$$

Now $\bar{\zeta}_f$ is large, so that it is quite possible that $\beta\,\ell n\,\bar{\zeta}_f$ is a significant correction. Indeed, if $\beta\,\ell n\,\bar{\zeta}_f >> 1$, then $H_\beta = \beta\,\ell n\,\bar{\zeta}_f(H-1)$ which is not only large, but H_β has a different sign from H. Since it is clearly not precise to assume that combustion really begins exactly at the interface, it appears that the modification suggested here may be important not only quantitatively but also qualitatively.

For example, in Ref. 9, the effect of aluminum on the transient response has been examined by relating changes of H to the heat capacity of molten aluminum collecting on the surface. Thus, an increase of H, due to the heat absorbed by the aluminum, accompanies greater accumulation of the metal on the surface. It was then found by numerical results that increasing H (i.e., more aluminum on the surface) is a stabilizing effect: it tends to reduce the peak in the response function. This was offered as an explanation for the observed effect that the addition of aluminum to a propellant tends to suppress pressure oscillations in a chamber.

On the other hand, in both Ref. 4 and 5, an increase in H, or the corresponding parameter, was found to enhance the peak in the response function.[6] The reason for the difference between these conclusions seems to be that account is taken, in the last two works, of the thermal wave in the surface layer. This tends to enhance the peak in the same way that the thermal wave in the solid causes the peak originally for a pure solid. If the thickness of aluminum is comparable to the wavelength of the thermal wave--a likely circumstance--this influence cannot be ignored. Hence, the conclusions of Ref. 9 must, for the present at least, remain qualified in an important respect.

A remark should be made in regard to the interpretation of the influence of H (i.e., Q_s). By combining Eq. 21 and 25, the overall energy balance for the conversion of cold solid to gaseous reaction products is, in the steady state,

$$c\,(\bar{T}_s - \bar{T}_c) - Q_s = Q_f - c_p\,(\bar{T}_f - \bar{T}_s)$$

so that H is related to other parameters in the problem according to

$$H = 1 - \frac{Q_f}{c(\bar{T}_s - T_c)}\left[1 - \frac{c_p\,(\bar{T}_f - \bar{T}_s)}{Q_f}\right] \tag{87}$$

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[6] The work there was not, however, directed to studying the effects of aluminum.

Thus, when different numerical results for the admittance function are obtained by changing $H = Q_s/c(\overline{T}_s - T_c)$, this is necessarily accompanied by changes in the values of Q_f, T_f, or T_s so that Eq. 87 is satisfied. This is not troublesome, but must be recognized in the interpretation of parametric studies.

D. Quasi-Static Results by Integration of the Linearized Differential Equations

Certain aspects of the problem are clarified by finding the perturbations by integrating the differential equations. It is also necessary to follow this route if one wishes to relax the assumption of quasi-static behavior. When the time derivatives are ignored in Eq. 52 and 53, and the variables are written as $T = \overline{T} + T'$, etc., one finds

$$\partial m'/\partial x = 0 \tag{88}$$

$$k_g \frac{\partial^2 T'}{\partial x^2} - \overline{m} c_p \frac{\partial T'}{\partial x} = - Q_f w' + m' c_p \frac{d\overline{T}}{dx} \tag{89}$$

In order to simplify the computations, only the limiting case of combustion extending to the solid-gas interface will be considered here. It is also perhaps less confusing for the time being to use the position variable x rather than the dimensionless ζ. The solutions for the steady-state heat transfer and temperature are

$$q(x) = k_g \frac{dT}{dx} = Q_f \, \overline{w} \, \ell \, [1 - e^{-\theta}] \tag{90}$$

$$T(x) = T_f - \frac{Q_f w \ell^2}{k_g} [\theta - 1 + e^{-\theta}] \tag{91}$$

where $\ell = k_g/\overline{m}c_p$ and $\theta = (\overline{x}_f - x)/\ell$ and the boundary condition $q_f = 0$ at $x = x_f$ has been used.

Equation 88 of course gives the result that m' is constant through the gas phase. It is a straightforward matter to integrate Eq. 89 to find, for the case when w' is uniform in the gas phase:

$$q' = k_g \frac{dT'}{dx} = q_{\overline{f}}' \, e^{-\overline{\theta}} + Q_f \, w' \ell (1 - e^{-\theta}) + \frac{m'}{\overline{m}} Q_f \, \overline{w} \ell \, [(\theta + 1) e^{-\theta} - 1] \tag{92}$$

$$T' = T_{\bar{f}}' - q_{\bar{f}}' \frac{\ell}{k_g} (1-e^{-\bar{\theta}}) - Q_f w' \frac{\ell^2}{k_g} [\bar{\theta}-1 + e^{-\bar{\theta}}]$$

$$- \frac{m'}{\bar{m}} Q_f \bar{w} \frac{\ell^2}{k_g} [2(1-e^{-\bar{\theta}}) - \bar{\theta} (1+e^{-\bar{\theta}})] \qquad (93)$$

where $\bar{\theta} = \bar{m}c_p(\bar{x}_f - x)/k_g$, and ℓ has $\bar{m}$ in place of m. The integrations have been performed from the surface ($x = 0$) to the mean position of the outer edge of the flame ($\bar{x}_f$); extension of the range to $x = \bar{x}_f + x_f'$, the instantaneous position, merely adds terms of higher order in small quantities. Thus, $T_{\bar{f}}'$ and $q_{\bar{f}}'$ denote fluctuations at the mean positio ; at the instantaneous position, $T_f' \neq T_{\bar{f}}'$ and $q_f' = 0$, but note that $q_{\bar{f}}'$ is nonzero. The situation is sketched in Fig. 3.

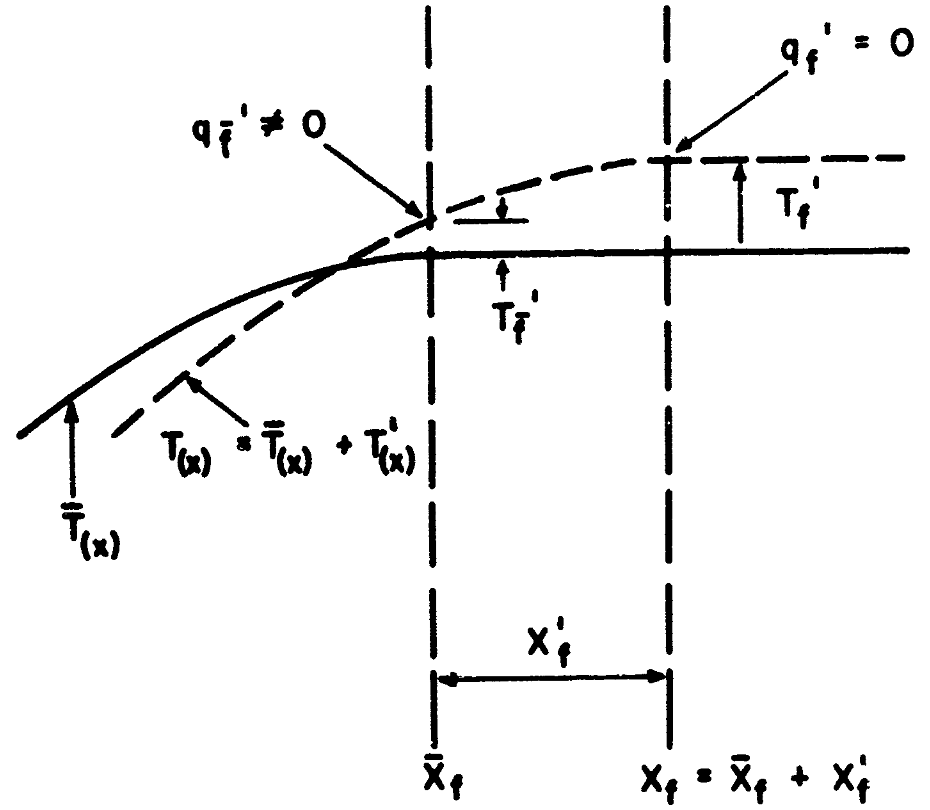

FIG. 3. Conditions at the Downstream Boundary of the Flame for Quasi-Static Behavior.

Now if Eq. 90 and 91 are linearized, the perturbations must be exactly Eq. 92 and 93, but two points should be noted. First, there are contributions from fluctuations in θ, for

$$\theta = \frac{mc_p}{k_g}(x_f - x) = \frac{\bar{m}c_p\,\bar{x}_f}{k_g}\left(1 + \frac{m'}{\bar{m}}\right)\left(1 + \frac{x_f'}{\bar{x}_f} - \frac{x}{\bar{x}_f}\right)$$

$$\approx \frac{\bar{m}c_p\,\bar{x}_f}{k_g}\left[1 + \frac{m'}{\bar{m}} + \frac{x_f'}{\bar{x}_f}\right] - \frac{\bar{m}c_p\,x}{k_g}$$

$$\bar{\theta} + \theta' = \frac{\bar{m}c_p}{k_g}(\bar{x}_f - x) + \frac{\bar{m}c_p\,\bar{x}_f}{k_g}\left(\frac{m'}{\bar{m}} + \frac{x_f'}{\bar{x}_f}\right)$$

Hence, in the exponentials,

$$e^{\theta} = e^{\bar{\theta}+\theta'} = e^{\bar{\theta}}e^{\theta'} \approx e^{\bar{\theta}}\,(1+\theta')$$

Second, since Eq. 90 and 91 extend all the way to the edge of the flame, perturbations at the downstream edge refer to the instantaneous and not the mean position of the flame. Thus, T_f in Eq. 91 gives T_f', not $T_{\bar{f}}'$, and no term in $q_{\bar{f}}'$, which appears in Eq. 92 and 93, is found. By expansion in Taylor series, similar to Eq. 6 and 7, one has

$$T_f' = T_{\bar{f}}' + x_f'\left(\frac{d\bar{T}}{dx}\right)_{\bar{f}} \tag{94}$$

$$\left(\frac{\partial T'}{\partial x}\right)_f = \left(\frac{\partial T'}{\partial x}\right)_{\bar{f}} + x_f'\left(\frac{d^2\bar{T}}{dx^2}\right)_{\bar{f}} \tag{95}$$

and the second gives

$$q_f' \equiv 0 = q_{\bar{f}}' + x_f'\,k_g\left(\frac{d^2\bar{T}}{dx^2}\right)_{\bar{f}} \tag{96}$$

When these relations are taken into account, the desired agreement is found. It may be noted that since the mean heat transfer vanishes at the flame edge, $(d\bar{T}/dx)_{\bar{f}} = 0$ and to first order $T_f' = T_{\bar{f}}'$, but the heat transfer fluctuation at the mean position does not vanish to first order. The fluctuation of heat $q_{\bar{f}}'$ is therefore due to the fluctuation of energy release associated with the change of flame thickness.

The last point merits closer attention. Consider the energy balance for the fluctuations in the region $\bar{x}_f \le x \le x_f'$ shown in Fig. 3. One has for c_p constant

$$Q_f w x_f' + c_p\,(\bar{m}T_f' + m'T_{\bar{f}}') - c_p(\bar{m}T_f' - m'\bar{T}_f) = q_f' - q_{\bar{f}}'$$

or

$$q_{\bar{f}}' = q_f' + \bar{m}c_p\,(T_{\bar{f}}' - T_f') + m'c_p\,(\bar{T}_f - \bar{T}_{\bar{f}})$$

$$+ Q_f\bar{w}x_f' = Q_f\bar{w}x_f \qquad (97)$$

since both temperature differences vanish, to first order, and $q_f' = 0$ by definition of the outer edge of the flame. It follows from the differential equation for the mean temperature that $k_g(d^2\bar{T}/dx^2)_{\bar{f}} = -Q_f\bar{w}$ and Eq. 96 and 97 are the same. This exercise shows how it is that even though conditions downstream of the flame are uniform, the flame temperature can fluctuate, giving rise to the entropy waves discussed in Ref. 2.

When evaluated at x = 0, Eq. 92 gives the expression for heat transfer to the surface, identical to that discussed in Section IV-A. By use of Eq. 92, many of the terms in Eq. 93 can be identified as q'(x) so that Eq. 93 may be written

$$T' = T_{\bar{f}}' - q_{\bar{f}}'\,\frac{\ell}{k_g} + q'\,\frac{\ell}{k} - w'Q_f\,\frac{\ell^2}{k_g}\,\bar{\theta}$$

$$+ \frac{m'}{\bar{m}}\,Q_f\,\bar{w}\,\frac{\ell^2}{k_g}\,[(\bar{\theta}-1) + e^{-\bar{\theta}}] \qquad (98)$$

Evaluated at x = 0, this gives the perturbation of the overall energy balance for the gas phase:

$$q_{o+}' \equiv q_{s+}' = Q_f\,(\bar{w}x_f' + \bar{w}x_f) - m'c_p(\bar{T}_f - \bar{T}_s)$$

$$- \bar{m}c_p\,(T_f' - T_s') \qquad (99)$$

Another way of arriving at the same formula is to consider a control volume extending from $x = 0$ to $x = \bar{x}_f + x_f'$ or integrating the energy equation directly over the entire gas phase:

$$\int_{0}^{\bar{x}_f+x_f'} \frac{d}{dx}\left[mc_pT - k_g\frac{dT}{dx}\right] dx = \int_{0}^{\bar{x}_f+x_f'} Q_f\, w\, dx$$

With $T = \bar{T} + T'$, etc., and retaining only terms of first order in fluctuations, this gives

$$\left[\bar{m}c_p T' - k_g\frac{dT'}{dx}\right]_{0}^{\bar{x}_f+x_f'} + \left[m'c_p\bar{T}\right]_{0}^{\bar{x}_f+x_f'}$$

$$= Q_f\,(\bar{x}_f w' + \bar{w}x_f')$$

and since again $(dT'/dx)_{x\,=\,\bar{x}_f+x_f'} = 0$, Eq. 99 is recovered.

The computations above serve to verify explicitly for the special model of uniform combustion that the procedure of Section IV-A based on the steady-state solution does indeed give exactly the same results obtained by integrating the differential equations for the perturbations.

E. Perturbations of Reaction Rate Depending on Temperature

It was argued in Section IV-A that a potentially important failing of the model proposed by the Princeton group is that the energy release is forced to be in phase with the pressure everywhere in the gas phase. This is obviously an approximation, but on the other hand, treatment of nonuniformities is very difficult, not only because the calculations rapidly become very involved, but also because it is not clear what constitutes a realistic representation.

At the present time, it does not seem worthwhile to give up *a priori* specification of the combustion distribution for the steady state. And in particular, if viewed in the way suggested after Eq. 63, the assumption of a uniform distribution may be accepted as a reasonable first approximation. However, this does not, as argued previously, irrevocably lead one to use a uniform distribution of energy release fluctuations which unavoidably forces the energy release, or reaction rate, and pressure to be in phase.

The point is that in fact one has a great deal of freedom in respect to choice of w'; a very simple alternative is offered here. suppose first that the reaction rate is locally sensitive to temperature, such that the steady-state value is approximated by a uniform distribution, but the fluctuations are

$$\frac{1}{\bar{\zeta}_f^{\,\beta}}\,\frac{c_p}{c}\left(\frac{\Lambda^2}{1-\bar{T}_s/T_c}\right)\frac{w'}{\bar{w}} = W_\beta\,\frac{p'}{\bar{p}} + V\,\frac{T'}{\bar{T}_s} \tag{100}$$

where T' is the local temperature fluctuation and V is a coefficient which can be determined only by a more careful examination of the processes in the flame. It does not appear possible to find V as W was found, by appeal to calculations and experimental results for the steady state. This expression simply adds a term to the formula Eq. 81 already used. However, the formulas for T' and q' in the gas phase cannot now be deduced from the steady-state solution which is supposed to be for a uniform distribution of combustion. Rather, Eq. 100 must be substituted in Eq. 89 and the differential equation solved. Incidentally, it must be emphasized that the arbitrary addition of the term in Eq. 100 in no way violates the assumption of quasi-static behavior, so that Eq. 88 and 89 are still valid.

The computations required are straightforward but involved, and will not be covered here. A quick estimate of the influence of this modification can be obtained in the following way. Suppose that the influence of temperature changes is most important in the hotter regions of the flame, and is therefore roughly represented by setting $T' \approx T_f'$ in Eq. 100. Calculation of the response function now proceeds as in Section IV-C. Substitution of Eq. 100 in Eq. 77 gives

$$\left(k_g\,\frac{\partial T'}{\partial x}\right)_{s+} = \left(1-\frac{T_c}{\bar{T}_s}\right)\left(1+\beta\,\ln\bar{\zeta}_f\right)\left(W_\beta\,\frac{p'}{\bar{p}} + V\,\frac{T_f'}{\bar{T}_s}\right) - \frac{c_p}{c}\,\frac{\Lambda^2}{\bar{\zeta}_f^{\,\beta}}\left(1+2\beta\,\ln\bar{\zeta}_f\right)\frac{m'}{\bar{m}} \tag{101}$$

By use of the perturbation of the over-all energy balance, Eq. 60, $T_f'/\bar{T}_s$ can be eliminated from this equation, leading to

$$\frac{1}{\bar{m}c\bar{T}_s}\left(k_g\,\frac{\partial T}{\partial x}\right)'_{s+} = \left(1-\frac{T_c}{\bar{T}_s}\right)\left(1+\beta\,\ln\bar{\zeta}_f\right)W_{\beta T}\,\frac{p'}{\bar{p}} + V_T\,\frac{T_s'}{\bar{T}_s} - \frac{c_p}{c}\,X\,\frac{m'}{\bar{m}} \tag{102}$$

where

$$W_{Tb} = \frac{W_\beta}{1 + \frac{c}{c_p} V \left(1 - \frac{T_c}{\bar{T}_s}\right)\left(1 + \beta \ln \bar{\zeta}_f\right)} \tag{103}$$

$$V_T = \frac{(1 \quad T_c/\bar{T}_s)\ (1 + \beta \ln \bar{\zeta}_f)\ V}{1 + \frac{c}{c_p} V \left(1 - \frac{T_c}{\bar{T}_s}\right)\left(1 + \beta \ln \bar{\zeta}_f\right)} \tag{104}$$

$$X = \frac{\frac{\Lambda^2}{\bar{\zeta}_f^{\,\beta}} (1+2\beta \ln \bar{\zeta}_f) - V\left(1 - \frac{T_c}{\bar{T}_s}\right)(1-H)\ (1+\beta \ln \bar{\zeta}_f)}{1 + \frac{c}{c_p} V\left(1 - \frac{T_c}{\bar{T}_s}\right)(1+\beta \ln \bar{\zeta}_f)} \tag{105}$$

The response function is once again found by using the energy matching condition, Eq. 18, the pyrolysis law, Eq. 17, and now Eq. 102 for the heat transfer from the gas phase; the result is:

$$\frac{m'/\bar{m}}{p'/\bar{p}} = \frac{AW_{\beta T}\ (1+\beta \ln \bar{\zeta}_f) + n_s\left(\frac{c_p}{c} - V_T\right) + n_s(\lambda-1)}{\lambda + \frac{A}{\lambda} + \frac{c_p}{c} EX - HA + \left(\frac{c_p}{c} - V_T\right) - 1} \tag{106}$$

First checking the limit $\omega = 0$, one has the requirement

$$AW_{\beta T}\ (1+\beta \ln \bar{\zeta}_f) + n_s\left(\frac{c_p}{c} - V_T\right) = n\left[A(1-H) + \frac{c_p}{c} X + \left(\frac{c_p}{c} - V_T\right)\right]$$

which can be rewritten, after substitution of Eq. 103 and 105 as

$$AW_\beta\ (1+\beta \ln \bar{\zeta}_f) = n\left[A(1-H) + \frac{c_p}{c} E \frac{\Lambda^2}{\bar{\zeta}_f^{\,\beta}} (1+2\beta \ln \bar{\zeta}_f) + (n-n_s)\ D\left(\frac{c_p}{c} - V_T\right)\right] \tag{107}$$

where D stands for the denominator of Eq. 103-105. It follows, after use of Eq. 104, that $D(c_p/c - V_T) = c_p/c$ and the condition, Eq. 107, is exactly Eq. 83, which has already been found to be satisfied identically; thus, the limit for $\omega = 0$ is correctly met for any value of V.

Naturally, the response function again has the form of Eq. 71, but with B given by

$$B = \frac{1}{n} W_{\beta T} \; (1 + \beta \; \ell n \; \bar{\zeta}_f) + \frac{c_p}{c} \frac{n_s}{n_A} \left(1 - \frac{c}{c_p} V_T\right) \tag{108}$$

As in Eq. 82 and 85, the results can be interpreted in terms of a new quantity $H_{\beta T}$, defined so that B here has the same dependence on $H_{\beta T}$ as B had on H in Eq. 70; by use of the definitions of Eq. 103, 82, and 81, $H_{\beta T}$ is defined by the equality

$$\frac{1}{n} (1+\beta \; \ell n \; \bar{\zeta}_f) \frac{\left[2n(1-H) + \frac{c_p}{c}\left(\frac{n-n_s}{A}\right)\right] - \beta Z(1-H)}{1 + \frac{c}{c_p} V\left(1 - \frac{T_c}{T_s}\right) (1 + \beta \; \ell n \; \bar{\zeta}_f)}$$

$$+ \frac{c_p}{c} \frac{n_s}{n_A} \left(1 - \frac{c}{c_p} V_T\right) = \frac{1}{n} \left[2n\left(1-H_{\beta T}\right) + \frac{c_p}{c}\left(\frac{n-n_s}{\;}\right)\right] + \frac{c_p}{c} \frac{n_s}{An}$$

which eventually gives

$$H_{\beta T} = H \frac{1+\beta \; \ell n \; \bar{\zeta}_f}{1 + \alpha} + \frac{\alpha - \beta \; \ell n \; \bar{\zeta}_f}{1 + \alpha} \tag{109}$$

where

$$\alpha = \frac{c}{c_p} V \left(1 - \frac{T_c}{T_s}\right) (1+\beta \; \ell n \; \bar{\zeta}_f) \tag{110}$$

Equation 85 is of course recovered for $V = 0$.

It is not obvious how to evaluate V--although for a genuine diffusion flame one might try using the temperature dependence of the diffusion coefficient--so to assess the influence of this correction, suppose once again that $\beta \; \ell n \; \bar{\zeta}_f \gg 1$. Then Eq. 109 becomes

$$H_{\beta T} = \frac{H-1}{\frac{c}{c_p}\left(1 - \frac{T_c}{\bar{T}_s}\right) V} + 1 \tag{111}$$

It appears that this correction is not quite so large as that shown in Eq. 85 for $\beta \ln \bar{\zeta}_f \gg 1$. When V is nonzero, the influence of a displaced combustion zone is in effect blunted. That this is the case may be interpreted roughly as follows. If combustion begins downstream of the interface, then the heat transfer to the surface is less than that when combustion begins at the surface. On the other hand, for $V \neq 0$, the heat transfer is increased according to Eq. 102. Hence, the two effects tend to compensate one another.

Admittedly, the preceding calculations are approximate. The intent is to demonstrate that the results of Ref. 2 and subsequent applications in Ref. 9 and 10 must be qualified in respects which are not considered in those works.

V. DEVIATIONS FROM QUASI-STATIC BEHAVIOR-- SMALL AMPLITUDE HARMONIC OSCILLATIONS

In spite of the assumptions involved, the model based on uniform combustion is nevertheless a most convenient one, and seems to be an acceptable zeroth approximation to the real problem. It is particularly useful for studying an aspect of the general problem which has received too little attention, namely, behavior when the assumption of strict quasi-static is not enforced. The only treatment of this question is Ref. 6, which is not only complicated but is based on a very thin flame front as the model for the gas phase.

The two conservation equations, Eq. 52 and 53, will be solved with the time derivative terms nonzero. By use of Eq. 52, the left-hand side of Eq. 53 can be written

$$c_p \frac{\partial}{\partial t} (\rho T) + c_p \frac{\partial}{\partial x} (mT)$$

and with the perfect gas law, the first term is $(c_p/R)\ \partial p/\partial t$ which can be grouped with $\partial p/\partial t$ on the right-hand side. It is best to work with dimensionless variables; in terms of ζ defined earlier, the equations can be written

$$\frac{\partial}{\partial t}\left(\frac{p/\bar{p}}{\tau}\right) + \zeta\frac{\partial \mu}{\partial \zeta} = 0 \qquad (112)$$

$$\zeta^2 \frac{\partial^2 \tau}{\partial \zeta^2} + \zeta \frac{\partial}{\partial \zeta} [\tau(1-\mu)] = \frac{1}{\gamma} \frac{d}{dt} \left(\frac{p}{\bar{p}}\right) - \Lambda^2 \left(\frac{w}{\bar{w}}\right) \qquad (113)$$

where $\tau = T/\bar{T}_s$, $\mu = m/\bar{m}$, and t now stands for the dimensionless time, the scale being $\bar{\rho}_s\ k_g/\bar{m}^2 c_p$, i.e., t stands for $(\bar{m}^2 c_p/\bar{\rho}_s k_g)t$. In order to get results without excessive complications, it will be supposed that w is always independent of position. This means that dependence of reaction rate on temperature can be accounted for only in the somewhat artificial manner discussed in Section IV-D.

When the quasi-static assumption is dropped, it is no longer possible to obtain neat closed solutions. Several methods of approaching the problem will be indicated, but eventually only the first-order correction for the case of harmonic oscillations will be treated. With w dependent only on time, Eq. 113 can be integrated once--divide by ζ and integrate from an arbitrary position to the flame edge where in all cases $d\tau/d\zeta = 0$. After integration of the first term by parts and combination of the various pieces, one finds

$$\zeta \frac{\partial\tau}{\partial\zeta} - \mu\tau = -\mu_f\tau_f - \ln\left(\frac{\zeta_f}{\zeta}\right)\left[\frac{1}{\gamma}\frac{d}{dt}\left(\frac{p}{\bar{p}}\right) - \Lambda^2\frac{w}{\bar{w}}\right] \tag{114}$$

Thus, Eq. 112 and 114 constitute a coupled pair of first-order equations not yet restricted to small motions. For the purposes of this section only small fluctuations are considered; the linearized forms of Eq. 112, 113, and 114 are

$$\zeta \frac{\partial\mu'}{\partial\zeta} = -\frac{1}{\bar{\tau}}\frac{d}{dt}\left(\frac{p'}{\bar{p}}\right) + \frac{1}{\bar{\tau}^2}\frac{\partial\tau'}{\partial t} \tag{115}$$

$$\zeta^2\frac{\partial^2\tau'}{\partial\zeta^2} - \zeta\frac{\partial}{\partial\zeta}(\bar{\tau}\mu') = \frac{1}{\gamma}\frac{d}{dt}\left(\frac{p'}{\bar{p}}\right) - \Lambda^2\frac{w'}{\bar{w}} \tag{116}$$

$$\zeta\frac{\partial\tau'}{\partial\zeta} - \tau' - \bar{\tau}\mu' = -(\bar{\tau}_f\mu_f' + \tau_f') - \ln\left(\frac{\bar{\zeta}_f}{\zeta}\right) \times$$

$$\times\left[\frac{1}{\gamma}\frac{d}{dt}\left(\frac{p'}{\bar{p}}\right) - \Lambda^2\frac{w'}{\bar{w}}\right] - \Lambda^2(\ln\bar{\zeta}_f)\frac{\zeta_f'}{\bar{\zeta}_f} \tag{117}$$

The fact that $\bar{\mu} = 1$ has been used, and μ_f' stands for the fluctuation of mass flux at the flame edge. As before, the fluctuation of flame thickness in Eq. 117 must be found from its connection, Eq. 29, with w and m. Equation 117 is identical with Eq. 98; to show this it is necessary to replace q_f' by use of Eq. 96, and to use $\bar{\theta}$ rather than ζ as the independent variable.

Second-order equations for both μ' and τ' separately can be constructed. For example, solve Eq. 117 for μ' and substitute into Eq. 115 to find

$$\zeta^2 \frac{\partial}{\partial\zeta}\left(\frac{1}{\bar{\tau}}\frac{\partial\tau'}{\partial\zeta}\right) - \frac{1}{\bar{\tau}^2}\frac{\partial\tau'}{\partial t} + \left(\frac{\zeta}{\bar{\tau}^2}\frac{d\bar{\tau}}{d\zeta}\right)\tau'$$

$$=\left(\frac{\zeta}{\bar{\tau}^2}\frac{d\bar{\tau}}{d\zeta}\right)\left[\bar{\tau}_f\ \mu_f' + \tau_f' + (\Lambda^2\ \ell n\ \bar{\zeta}_f)\frac{\zeta_f'}{\bar{\zeta}_f}\right]$$

$$-\ \zeta\frac{d}{d\zeta}\left(\frac{1}{\bar{\tau}}\ \ell n\ \frac{\bar{\zeta}_f}{\zeta}\right)\left[\frac{1}{\bar{\gamma}}\ \frac{d}{dt}\ \left(\frac{p'}{\bar{p}}\right) - \Lambda^2\frac{w'}{\bar{w}}\right] \tag{118}$$

Solution of this equation will give τ' and then Eq. 108 can be used to find μ' by direct integration from the surface outwards:

$$\mu' = \mu_s' + \int_{\zeta_s}^{\zeta}\left[\frac{1}{\zeta\bar{\tau}^2}\ \frac{\partial\tau'}{\partial t} - \frac{1}{\zeta\bar{\tau}}\ \frac{d}{dt}\ \left(\frac{p'}{\bar{p}}\right)\right]\ d\zeta \tag{119}$$

Note that μ_s' is connected to the surface temperature fluctuation τ_s' by the pyrolysis law, so that Eq. 119 can be used as a formula for μ_f', the quantity ultimately required. Since $\bar{\tau}$ is an awkward function of ζ, analytical solution to Eq. 118 does not seem possible, although Eq. 118 and 119 could be used as the basis for numerical calculations. An interesting approximation--at first sight extremely crude--is to set the mean temperature equal to some average value $\bar{\tau}_a$ everywhere so that $d\bar{\tau}/d\zeta = 0$ as well; then Eq. 118 is

$$\zeta^2\frac{\partial^2\tau'}{\partial\zeta^2} - \frac{1}{\bar{\tau}_a}\frac{\partial\tau'}{\partial t} = \frac{1}{\gamma}\ \frac{d}{dt}\ \left(\frac{p'}{\bar{p}}\right) - \Lambda^2\frac{w'}{\bar{w}} \tag{120}$$

Since $\zeta^2\ \partial^2\tau'/\partial\zeta^2 = (k_g/\bar{m}c_p)^2\ [\bar{m}c_p\ \partial\tau'/\partial x - k_g\ \partial^2\tau'/\partial x^2]$ (of Eq. 32), the left-hand side is the ordinary heat conduction equation with convection of energy by the mean flow. The right-hand side is of course the fluctuation of energy release associated with the chemical reactions.

To see how close Eq. 120 is to a more acceptable approximation, substitute Eq. 115 for $\zeta\partial\mu'/\partial\zeta$ in Eq. 116:

$$\zeta^2\frac{\partial^2\tau'}{\partial\zeta^2} = \frac{1}{\bar{\tau}}\frac{\partial\tau'}{\partial t} - \left[\left(\frac{\gamma-1}{\gamma}\right)\frac{d}{dt}\left(\frac{p'}{\bar{p}}\right) + \Lambda^2\ \frac{w'}{\bar{w}}\right] + \left(\zeta\ \frac{d\bar{\tau}}{d\zeta}\right)\mu' \tag{121}$$

This is exact within the initial assumptions used, but the difficulty of course is that μ' on the right-hand side is not known. In addition to the terms in Eq. 120, there appears on the right-hand side a term

$d/dt(p'/\bar{p})$ which is a consequence of compressibility, and the term with μ' in it, which represents transport of mean energy by fluctuations of the mass flux. A more reasonable approximation is to set $\bar{\tau} = \bar{\tau}_a$ on the left-hand side of Eq. 121, retain $d\bar{\tau}/d\zeta$ on the right, and then worry about what to do with μ'. This procedure will be followed for the case of harmonic oscillations whose frequencies are not too high.

A. Solution to the Gas Phase for "Low" Frequency Oscillations

The time derivatives are now replaced according to

$$\frac{\partial}{\partial t} \rightarrow i\omega \left(\frac{\bar{\rho}_{gs}}{\bar{m}^2} \frac{k_g}{c_p} \right) = i\Omega\varepsilon$$

where $\Omega = \kappa_c \omega / \bar{r}^2$ is the dimensionless frequency arising in the problem of unsteady heat conduction in the gas phase, and

$$\varepsilon = \frac{\rho_{gs}}{\rho_c} \left(\frac{k_g c}{k_c c_p} \right) = \left(\frac{\rho_{gs}}{\rho_c} \right)^2 \left(\frac{\kappa_g}{\kappa_c} \right) \tag{122}$$

is a small quantity of order 10^{-2}. It is a little easier to work with the variable $\xi : \zeta = e^{\xi}$ so that Eq. (121) and (116) are

$$\frac{d^2\tau'}{d\xi^2} - \frac{d\tau'}{d\xi} - \frac{i\Omega\varepsilon}{\bar{\tau}} \tau' = - \left[i\Omega\varepsilon \left(\frac{\gamma - 1}{\gamma} \right) \frac{p'}{\bar{p}} + \Lambda^2 \frac{w'}{\bar{w}} \right] + \mu' \frac{d\tau'}{d\xi} \tag{123}$$

$$\frac{d\mu'}{d\xi} = \frac{i\Omega\varepsilon}{\bar{\tau}} \left(\frac{\tau'}{\bar{\tau}} - \frac{p'}{\bar{p}} \right) \tag{124}$$

In later calculations it will be assumed that $\Omega\varepsilon$ is small, the quasi-static assumption of course corresponding to $\Omega\varepsilon = 0$. To interpret the meaning of this parameter, rewrite it in the form

$$\begin{aligned} \Omega\varepsilon &= \frac{\rho_{gs}}{\bar{m}^2} \frac{k_g}{c_p} \omega \\ &= \frac{k_g (\bar{T}_f - \bar{T}_s)/\bar{x}_f}{\bar{m}\, c_p (\bar{T}_f - \bar{T}_s)} \; \frac{\omega}{(\bar{m}/\bar{\rho}_g)/\bar{x}_f} \\ &\approx \left[\frac{\bar{q}_{s+}}{\bar{m}\, c_p (\bar{T}_f - \bar{T}_s)} \right] \frac{\omega}{(u_g/x_f)} \end{aligned}$$

The ratio of mean heat transfer rate at the surface to the rate of energy transferred by convection appears in brackets; this is multiplied by the ratio of the average transit time $(\overline{x}_f/\overline{\mu}_g)$ for an element of gas, to the period $1/\omega$ of the oscillations. Thus, it is the product of these which matters, not the relative importance of heat conduction or transit time alone.

The relatively weak assumption will be made that $\overline{\tau} = \overline{\tau}_a$ in $i\Omega\varepsilon/\tau$; in any case, $\overline{\tau}$ varies smoothly from unity at the surface to a value of three or four at the flame, which is a very small change relative to ζ^2 in the second derivative of Eq. 121, i.e., compared with the first two terms in Eq. 17. It appears that the gross behavior of the results should not be greatly affected by this approximation, and by doing so one has the opportunity to obtain results quite easily. For now Eq. 123 is formally integrable. The solutions to the homogeneous equation are $\exp(\lambda_+\xi)$, $\exp(\lambda_-\xi)$, where

$$\lambda \pm = \tfrac{1}{2}\left[1 \pm \sqrt{1 + i\,\frac{4\Omega\varepsilon}{\tau_a}}\right] \tag{125}$$

These of course represent merely the thermal wave solutions. The complete formal solution to Eq. 123 is therefore

$$\tau' = e^{\lambda_+\xi}\left[C_1 + \int_{\xi}^{\overline{\xi}_f} \frac{h}{\Delta}\, e^{\lambda_-\xi}\, d\xi\right] + e^{\lambda_-\xi}\left[C_2 - \int_{\xi}^{\overline{\xi}_f} \frac{h}{\Delta}\, e^{\lambda_+\xi}\, d\xi\right]$$

where

$$h = -\left[i\Omega\varepsilon\left(\frac{\gamma-1}{\gamma}\right)\frac{p'}{p} + \Lambda^2\,\frac{w'}{w}\right] + \mu'\,\frac{d\overline{\tau}}{d\xi} \tag{126}$$

and the Wronskian is

$$\Delta = \begin{vmatrix} e^{\lambda_+\xi} & e^{\lambda_-\xi} \\ \frac{d}{d\xi}\left(e^{\lambda_+\xi}\right) & \frac{d}{d\xi}\left(e^{\lambda_-\xi}\right) \end{vmatrix} = (\lambda_- - \lambda_+)\, e^{(\lambda_+ + \lambda_-)\,\xi} \tag{127}$$

Thus, some of the integrals in the solution for τ' can be carried out and τ' is

$$\tau' = C_1 e^{\lambda_+ \xi} + C_2 e^{\lambda_- \xi} + K - \frac{1}{\lambda_+ - \lambda_-} \left\{ e^{\lambda_+ \xi} \int_{\xi}^{\bar{\xi}_f} \mu' \frac{d\bar{\tau}}{d\xi} e^{-\lambda_+ \xi} d\xi \right.$$

$$\left. - e^{\lambda_- \xi} \int_{\xi}^{\bar{\xi}_f} \mu' \frac{d\bar{\tau}}{d\xi} e^{-\lambda_- \xi} d\xi \right\}$$

with

$$K = \frac{-1}{\lambda_+ \lambda_-} \left[i\Omega\varepsilon \left(\frac{\gamma - 1}{\gamma} \right) \frac{p'}{\bar{p}} + \Lambda^2 \frac{w'}{\bar{w}} \right] \tag{128}$$

The constants C_1 and C_2 are set by requiring that $\tau' = \tau_{\bar{f}}' \simeq \tau_f'$ and $(\partial\tau'/\partial\xi)_{\bar{f}} = q_{\bar{f}}'/\bar{m}c_p\bar{T}_s = (Q_f\bar{w}/\bar{m}c_p\bar{T}_s)x_f'$ (see Eq. 96 and 97) one finds:

$$C_1 = \frac{e^{-\lambda_+ \bar{\xi}_f}}{\lambda_- - \lambda_+} \left[\lambda_- (\tau_f' - K) + \frac{q_{\bar{f}}'}{\bar{m}c_p\bar{T}_s} \right] \tag{129}$$

$$C_2 = \frac{e^{-\lambda_- \bar{\xi}_f}}{\lambda_- - \lambda_+} \left[- \lambda_+ (\tau_f' - K) + \frac{q_{\bar{f}}'}{\bar{m}c_p\bar{T}_s} \right] \tag{130}$$

With these expressions for the constants, the solution for τ' can be written

$$\tau' = K + \frac{\tau_f' - K}{\lambda_- - \lambda_+}\left[\lambda_- e^{\lambda_+(\xi-\bar{\xi}_f)} - \lambda_+ e^{\lambda_-(\xi-\bar{\xi}_f)}\right]$$

$$- \frac{q_f'}{\bar{m} c_p \bar{T}_s (\lambda_- - \lambda_+)}\left[e^{\lambda_+(\xi-\bar{\xi}_f)} - e^{\lambda_-(\xi-\bar{\xi}_f)}\right] \tag{131}$$

$$- \frac{1}{\lambda_+ - \lambda_-}\left\{ e^{\lambda_+\xi}\int_{\xi}^{\bar{\xi}_f} \mu' \frac{d\bar{\tau}}{d\xi} e^{-\lambda_+\xi} d\xi - e^{\lambda_-\xi}\int_{\xi}^{\bar{\xi}_f} \mu' \frac{d\bar{\tau}}{d\xi} e^{-\lambda_-\xi} d\xi \right\}$$

Since μ' is given by Eq. 119, Eq. 131 is still not an explicit solution, but it is a convenient form for iterative approximations, from which both τ' and $(d\tau'/d\xi)_{0+}$ can be computed.

The zeroth approximation is found by setting $\Omega\varepsilon = 0$ in both Eq. 119 and 131; this is of course the quasi-static case considered in Section IV: Eq. 119 gives μ' a constant through the gas phase, and Eq. 131 reduces to Eq. 93 or 98 if proper account is taken of the variables, $\theta = \bar{\xi}_f - \xi$. The next and higher approximations may be found by expanding in powers of $i\Omega\varepsilon$. Only the correction or order $\Omega\varepsilon$ will be considered here, so that the results are valid only for relatively low frequencies. From the definitions, Eq. 125 and 128, it follows that $\lambda_+\lambda_- = -i\Omega\varepsilon/\bar{\tau}_a$ exactly, so that

$$K = \frac{\bar{\tau}_a}{i\Omega\varepsilon}\left[i\Omega\varepsilon\left(\frac{\gamma-1}{\gamma}\right)\frac{p'}{\bar{p}} + \Lambda^2 \frac{w'}{\bar{w}}\right]$$

and to first order

$$\lambda_+ = 1 + i\,\frac{\Omega\varepsilon}{\bar{\tau}_a}$$

$$\lambda_- = -\,i\,\frac{\Omega\varepsilon}{\bar{\tau}_a}$$

With these used in Eq. 131, one eventually finds

$$\mu' = \mu'^{(o)} + i\Omega\epsilon\, \mu'^{(1)}$$

$$\tau' = \tau'^{(o)} + i\Omega\epsilon\, \tau'^{(1)} \tag{132}$$

where the quasi-static fluctuations are $\mu'^{(o)}$ equal to the surface value, $\mu'^{(o)} = \mu_s'^{(o)}$ and

$$\tau'^{(o)} = \tau_{\bar{f}}'^{(o)} + \frac{q_{\bar{f}}'^{(o)}}{\bar{m}c_p\bar{T}_s}\left(e^{\xi-\xi_f}-1\right)$$

$$-\Lambda^2 \frac{w'^{(o)}}{\bar{w}}\left[e^{(\xi-\bar{\xi}_f)}-1+\xi-\bar{\xi}_f\right]$$

$$-\mu'^{(o)}\left[e^{\xi}\int_{\xi}^{\bar{\xi}_f}\frac{d\bar{\tau}}{d\xi}e^{-\xi}d\xi-\int_{\xi}^{\bar{\xi}_f}\frac{d\bar{\tau}}{d\xi}\,d\xi\right]$$

Since the mean temperature profile is given by Eq. 45 and $\zeta = \exp(\xi)$,

$$\frac{d\bar{\tau}}{d\xi} \equiv e^{\xi}\frac{d\bar{\tau}}{d\zeta} = \Lambda^2 e^{\xi}\left(\frac{1}{\zeta}-\frac{1}{\zeta_f}\right) = \Lambda^2\left(1-e^{\xi-\bar{\xi}_f}\right)$$

The integrals above can be easily carried out and $\tau'^{(o)}$ is

$$\tau'^{(o)} = \tau_{\bar{f}}'^{(o)} + \frac{q_{\bar{f}}'^{(o)}}{\bar{m}c_p\bar{T}_s}\left(e^{\xi-\bar{\xi}_f}-1\right) - \Lambda^2\frac{w'^{(o)}}{\bar{w}}\left[(\xi-\bar{\xi}_f)-1+e^{\xi-\bar{\xi}_f}\right]$$

$$-\mu'^{(o)}\Lambda^2\left[2\left(1-e^{\xi-\bar{\xi}_f}\right)-(\bar{\xi}_f-\xi)\left(1+e^{\xi-\bar{\xi}_f}\right)\right] \tag{133}$$

$$= \alpha_1 + \beta_1 e^{\xi} + \gamma_1 \xi e^{\xi} + \delta_1 \xi$$

which agrees exactly with Eq. 93 since $\Lambda^2 = (Q_f k_g \overline{w}/\overline{m}^2 c_p{}^2 \overline{T}_s)$; the constants $\alpha_1, \ldots, \delta_1$, are defined in the nomenclature.

The first-order correction to the fluctuation of mass flux is given by the integral of Eq. 124:

$$\mu'^{(1)} = \mu_s'^{(1)} + \left\{ \int_{\xi}^{\overline{\xi}_f} \frac{\tau'^{(o)}}{\overline{\tau}^2} d\xi - \frac{p'}{\overline{p}} \int_{\xi}^{\overline{\xi}_f} \frac{d\xi}{\overline{\tau}} \right\}$$

The mean temperature profile as a function of ξ is

$$\overline{\tau} = \overline{\tau}_f - \Lambda^2 \left[(\overline{\xi}_f - \xi) - (1 - e^{-\xi - \xi_f}) \right]$$

which prohibits easy evaluation of the integrals. Hence, in the interests again of obtaining results, $\overline{\tau}$ is approximated by $\overline{\tau}_a$. With Eq. 126, one eventually finds for $\mu'^{(1)}$:

$$\mu'^{(1)} \approx \mu_s'^{(1)} + \frac{1}{\overline{\tau}_\alpha{}^2} \left(\alpha_1 - \tau_a \frac{p'}{\overline{p}}\right) \xi + \beta_1 e^{\xi}$$

$$+ \gamma_1 \left\{ e^{\xi} (\xi - 1) - 1 \right\} + \delta_1 \frac{\xi^2}{2} \qquad (134)$$

Note that the mean value $\xi_s = 0$ has been used since fluctuations ξ_s' contribute terms of second order only. Equation 134 is required in the first-order correction $\tau'^{(1)}$ for the temperature fluctuations. However, the entire profile $\tau'^{(1)}$ is not required for the computation of the response function. One needs only the gradient at the surface, which from Eq. 131 is

$$\left(\frac{\partial \tau}{\partial \xi}\right)'_{o+} = (\tau_f' - K) \frac{\lambda_+ \lambda_-}{\lambda_+ - \lambda_-} \left[e^{-\lambda_- \overline{\xi}_f} - e^{-\lambda_+ \overline{\xi}_f} \right]$$

$$+ \frac{q_f'}{\overline{m} c_p \overline{T}_s} \frac{1}{(\lambda_+ - \lambda_-)} \left[\lambda_+ e^{-\lambda_+ \overline{\xi}_f} - \lambda_- e^{-\lambda_- \overline{\xi}_f} \right]$$

$$- \frac{1}{\lambda_+ - \lambda_-} \left\{ \int_0^{\overline{\xi}_f} \mu' \frac{d\overline{\tau}}{d\xi} \left(\lambda_+ e^{-\lambda_+ \xi} - \lambda_- e^{-\lambda_- \xi} \right) \right\} \qquad (135)$$

Henceforth, only first-order terms in $\Omega\varepsilon$ will be considered, and evaluation of Eq. 135 is relatively easy. First in the integral

$$\int_0^{\bar{\xi}_f} \mu' \frac{d\bar{\tau}}{d\xi} \left(\lambda_+ e^{-\lambda_+ \xi} - \lambda_- e^{-\lambda_- \xi}\right) d\xi \simeq \int_0^{\bar{\xi}_f} \left(\mu'^{(o)} + i\Omega\varepsilon\mu'^{(1)}\right) \frac{d\bar{\tau}}{d\xi} \left(e^{-\xi} + \frac{i\Omega\varepsilon}{\bar{\tau}_a}\right) d\xi \simeq$$

$$\mu'^{(o)} \Lambda^2 \left[1 - (\bar{\xi}_f + 1) e^{-\bar{\xi}_f}\right] + i\Omega\varepsilon \left[\frac{\mu'^{(o)}}{\bar{\tau}_a} (\bar{\tau}_f - \bar{\tau}_s) + \int_0^{\bar{\xi}_f} \mu'^{(1)} \frac{d\bar{\tau}}{d\xi} e^{-\xi} \, d\xi\right]$$

With the very good approximation that terms containing $\exp(-\bar{\xi}_f)$ can be neglected, one finds

$$\int_0^{\bar{\xi}_f} \mu' \frac{d\bar{\tau}}{d\xi} \left(\lambda_+ e^{-\lambda_+ \xi} - \lambda_- e^{-\lambda_- \xi}\right) d\xi \simeq \Lambda^2 \mu'^{(o)} + i\Omega\varepsilon \left[\frac{\mu'^{(o)}}{\bar{\tau}_a} (\bar{\tau}_f - \bar{\tau}_s) + \Lambda^2 \mu_s'^{(1)} - \frac{\Lambda^2}{\bar{\tau}_a} \left(\alpha_1 - \bar{\tau}_a \frac{p'}{\bar{p}}\right) + \Lambda^2 \frac{\hat{\delta}_1}{\bar{\tau}_a^2}\right]$$

If the remaining terms are expanded to order $i\Omega\varepsilon$ and terms of order $\exp(-\bar{\xi}_f)$ are dropped, Eq. 128 becomes

$$\left(\frac{\partial\tau'}{\partial\xi}\right)'_{o+} = \left\{\tau_f'^{(o)} + i\Omega\varepsilon\tau_{\bar{f}}'^{(1)} - \frac{\bar{\tau}_a}{i\Omega\varepsilon}\left[i\Omega\varepsilon\,\frac{\gamma-1}{\gamma}\,\frac{p'}{\bar{p}} + \Lambda^2\left(\frac{w'^{(o)}}{\bar{w}} + i\Omega\varepsilon\,\frac{w'^{(1)}}{\bar{w}}\right)\right]\right\}$$

$$\times\left(\frac{-i\Omega\varepsilon}{\bar{\tau}_a}\right)\left(1-2\,\frac{i\Omega\varepsilon}{\bar{\tau}_a}\right)\left(1 + \frac{i\Omega\varepsilon}{\bar{\tau}_a}\,\bar{\xi}_f\right) + \frac{q_{\bar{f}}'^{(o)} + i\Omega\varepsilon\, q_{\bar{f}}'^{(o)}}{\bar{m}c_p\bar{T}_s}\left(1-2\,\frac{i\Omega\varepsilon}{\bar{\tau}_a}\right)\left(\frac{i\Omega\varepsilon}{\bar{\tau}_a}\right)$$

$$-\left(1-2\,\frac{i\Omega\varepsilon}{\bar{\tau}_a}\right)\left\{\Lambda^2\mu'^{(o)} + i\Omega\varepsilon\left[\frac{\mu'^{(o)}}{\bar{\tau}_a}\,(\bar{\tau}_f - \bar{\tau}_a) + \Lambda^2\mu_s'^{(1)} - \Lambda^2\,\frac{1}{\bar{\tau}_a}\left(\alpha_1 - \bar{\tau}_a\frac{p'}{\bar{p}}\right)\right.\right.$$

$$\left.\left. + \Lambda^2\,\frac{\delta_1}{\bar{\tau}_a^{\,2}}\right]\right\}$$

The zeroth order terms give

$$\left(\frac{\partial\tau}{\partial\xi}\right)'^{(o)}_{o+} = \Lambda^{2}\left[\frac{w'^{(o)}}{\bar{w}} - \mu'^{(o)}\right] \tag{136}$$

which is exactly the quasi-static results, Eq. 92, evaluated at the surface. The first-order terms give

$$\left(\frac{\partial\tau}{\partial\xi}\right)'^{(1)}_{o+} = \Lambda^2\left[\frac{w'^{(1)}}{\bar{w}} - \mu_s'^{(1)} + \frac{1}{\bar{\tau}_a^{\,2}}\left(\alpha_1 - \bar{\tau}_a\,\frac{p'}{\bar{p}}\right) - \frac{\delta_1}{\bar{\tau}_a^{\,2}}\right]$$

$$+ \frac{q_{\bar{f}}'^{(o)}}{\bar{\tau}_a\bar{m}c_p\bar{T}_s} - \frac{\tau_{\bar{f}}'^{(o)}}{\bar{\tau}_a} + \frac{\gamma-1}{\gamma}\frac{p'}{\bar{p}} - \frac{\mu'^{(o)}}{\bar{\tau}_a}\,(\bar{\tau}_f - \bar{\tau}_s) \tag{137}$$

The definitions of α_1 and δ_1 can be substituted to give a more explicit form. In doing so, further terms are neglected, based on the fact that $\Lambda^2 = (Q_f/c_p\bar{T}_s)(\ln\bar{\zeta}_f)^{-1} = Q_f/(c_p\bar{T}_s\zeta_f)$ is much larger than unity (cf. remarks following Eq. 57. Although $\bar{\zeta}_f = \bar{m}c_px_f/k_g$ may be 10 - 20, $Q_f/c_p\bar{T}_s$ may be as large as 5 - 6, so that the assumption $\Lambda^2 << 1$ may sometimes be borderline--it should be checked in individual cases. However, it is true that the terms containing Λ^2 will make a smaller contribution in any case, and if they are retained, subsequent manipulations become very tedious indeed. It appears that the major behavior will be accounted for, and hence the essential implications of the calculation will be found, if the approximation is made. The relative simplicity of the final formula Eq. 155 partly justifies this step. Then Eq. 137 reduces to

$$\left(\frac{\partial\tau}{\partial\xi}\right)_{o+}^{\prime(1)} = \Lambda^2 \left[\left(\frac{w^{\prime(1)}}{\bar{w}} - \mu_s^{\prime(1)}\right) + \frac{1}{\bar{\tau}_a^{\,2}}\left(\frac{w^{\prime(o)}}{\bar{w}} - \mu^{\prime(o)}\right)\right]$$

$$+ \frac{1}{\bar{\tau}_a}\left[\frac{q_{\bar{f}}^{\prime(o)}}{\bar{m}c_p\bar{T}_s} - \tau_{\bar{f}}^{\prime(o)} - \mu^{\prime(o)}\left(\bar{\tau}_f - \bar{\tau}_s\right)\right]$$

$$+ \frac{\gamma-1}{\gamma}\frac{p'}{\bar{p}} \qquad (138)$$

Some terms containing Λ^2 have been retained to show the only first-order contributions on the right-hand side. However, these may be neglected compared with the remaining terms, and one is left with the quite simple result

$$\left(\frac{\partial\tau}{\partial\xi}\right)_{o+}^{\prime(1)} = \frac{\gamma-1}{\gamma}\frac{p'}{\bar{p}} + \frac{1}{\bar{\tau}_a}\left[\frac{q_{\bar{f}}^{\prime(o)}}{\bar{m}c_p\bar{T}_s} - \tau_{\bar{f}}^{\prime(o)} - \mu^{\prime(o)}\left(\bar{\tau}_f - \bar{\tau}_s\right)\right] \qquad (139)$$

There are no first-order fluctuations appearing on the right-hand side, which makes later calculations very much shorter than if Eq. 138 is used. This is of course a consequence of the fact that the normalized thickness $\bar{\zeta}_f$ is very much greater than unity. It is by no means obvious that all of the labor following Eq. 131 would culminate in such a result as Eq. 139.

A second necessary relation among the heat transfer, surface temperature, and flame temperature fluctuations can be deduced by direct integration of the linearized energy equation, Eq. 116. After change of variable from ζ to ξ and integrating over $0 \leq \xi \leq \bar{\xi}_f$, one finds

$$\left(\frac{\partial\tau}{\partial\xi}\right)_{o+}^{\prime} = \left(\tau_o' + \bar{\tau}_s\,\mu_o'\right) - \left(\tau_{\bar{f}}' + \bar{\tau}_f\,\mu_{\bar{f}}'\right) + \frac{q_{\bar{f}}'}{\bar{m}c_p\bar{T}_s} - \Lambda^2\,\bar{\xi}_f\,\frac{w'}{\bar{w}}$$

$$+ \frac{i\Omega\varepsilon}{\gamma}\frac{p'}{\bar{p}}\,\bar{\xi}_f \qquad (140)$$

Once again expand all quantities to first order in $\Omega\varepsilon$ to find the zeroth and first order pieces of Eq. 140:

$$\left(\frac{\partial\tau}{\partial\xi}\right)_{o+}^{\prime(o)} = (\tau_o^{\prime(o)} + \bar{\tau}_o\mu_o^{\prime(o)}) - (\tau_{\bar{f}}^{\prime(o)} + \bar{\tau}_{\bar{f}}\,\mu_{\bar{f}}^{\prime(o)})$$

$$+ \frac{q_{\bar{f}}^{\prime(o)}}{\bar{m}c_p\bar{T}_s} - \Lambda^2\,\frac{w^{\prime(o)}}{\bar{w}}\,\bar{\xi}_f \qquad (141)$$

$$\left(\frac{\partial\tau}{\partial\xi}\right)_{o+}^{\prime(1)} = (\tau_o^{\prime(1)} + \bar{\tau}_s\,\mu_o^{\prime(1)}) - (\tau_{\bar{f}}^{\prime(1)} + \bar{\tau}_f\,\mu_{\bar{f}}^{\prime(1)})$$

$$+ \frac{q_{\bar{f}}^{\prime(1)}}{\bar{m}c_p\bar{T}_s} - \Lambda^2\,\frac{w^{\prime(1)}}{\bar{w}}\,\bar{\xi}_f + \frac{1}{\gamma}\,\frac{p^{\prime}}{\bar{p}}\,\bar{\xi}_f \qquad (142)$$

Note that because of the factor $\bar{\xi}_f$, the last term is dominant. Equation 141 is of course the same as Eq. 99; to show this explicitly requires use of Eq. 30 which, when linearized and expanded, gives

$$\frac{w^{\prime(o)}}{\bar{w}} = \mu^{\prime(o)} - \frac{\xi_f^{\prime}}{\bar{\xi}_f} \qquad (143)$$

for the quasi-static contributions. Recall also from Eq. 97 (which is valid for all linear motions) that

$$\frac{q_{\bar{f}}^{\prime(o)}}{\bar{m}c_p\bar{T}_s} = -\,\xi^{\prime(o)}\left(\frac{d^2\bar{\tau}}{d\xi^2}\right)_{\bar{f}} \qquad (144a)$$

$$\frac{q_{\bar{f}}^{\prime(1)}}{\bar{m}c_p\bar{T}_s} = -\,\xi_f^{\prime(1)}\left(\frac{d^2\bar{\tau}}{d\xi^2}\right)_{\bar{f}} \qquad (144b)$$

Equations 141 and 135 may be used to find the fluctuations of flame temperature, $\tau_{\bar{f}}^{\prime}$.

The only task remaining so far as the gas phase is concerned is to find formulas for the fluctuations of the flame thickness. For the quasi-static problem, Eq. 30, $w = m/x_f$, can be used; thus giving Eq. 142 but in the case of nonquasi-static motions, one must return to the origin of the term $Q_f w$. If one examines the balance of enthalpy for the gas phase, by use of a control volume, one finds by comparison with the integral of the unsteady energy equation that the following equality must be met:

$$Q_f \int_0^{x_f} w \, dx = m_s h_{sr} - m_f h_{fr}$$

$$= m_s (h_{sr} - h_{fr}) - (m_f - m_s) h_{fr}$$

$$= m_s Q_f - (m_f - m_s) h_{fr}$$

where h_{fr}, h_{sr} are reference enthalpies. This corresponds to Eq. 28 for steady flow; but here, instead of Eq. 30, one has

$$w = \frac{1}{x_f} \left[m_s - (m_f - m_s) \frac{h_{fr}}{Q_f} \right]$$

Hence, the perturbations are related by

$$\frac{w'}{\bar{w}} = - \frac{x_f'}{\bar{x}_f} + \frac{m_s'}{\bar{m}} - \left(\frac{m_f'}{\bar{m}} - \frac{m_s'}{\bar{m}} \right) \frac{h_{fr}}{Q_f}$$

or, for the first-order terms:

$$\frac{w'^{(1)}}{\bar{w}} = \mu_s'^{(1)} - \frac{\xi_f'^{(1)}}{\bar{\xi}_f} - (\mu_f'^{(1)} - \mu_s'^{(1)}) \frac{h_{fr}}{Q_f} \tag{145}$$

The value of h_{fr} depends on the thermodynamics of the reactions in the gas phase. If the gases leaving the surface burn completely to form the elementary products in terms of which the reference enthalpy is defined, then $h_{fr} = 0$ and $Q_f = h_{sr}$. Rather than complicate the results with another parameter, this limiting case will be supposed here so that

$$\frac{w'^{(1)}}{\bar{w}} = \mu_s'^{(1)} - \frac{\xi_f'^{(1)}}{\bar{\xi}_f} \tag{146}$$

which gives $\xi_f'^{(1)}$ for use in Eq. 144b. Because of the form of Eq. 139, this result is not required for calculation of the response function; however, it must be used in Eq. 142 to find the fluctuations of flame temperature. Note that if one assumes w to be given by the simplest form (Eq. 64), then $w'^{(1)}$ vanishes, and $\xi_f'^{(1)}/\bar{\xi}_f = \mu_s'^{(1)}$

B. <u>First-Order Interfacial Conditions</u>

The linearized forms of the pyrolysis law, Eq. 17, and conservation of energy at the interface are easily expanded in powers of $\Omega\epsilon$ to give the following relations:

<u>Quasi-static approximation</u>:

$$\mu_s'^{(o)} = \left(\alpha_s + \frac{E_s}{R_o\bar{T}_s}\right)\tau_s'^{(o)} + n_s\frac{p'}{\bar{p}} \tag{147}$$

$$\left(\frac{\partial\tau}{\partial\xi}\right)_{s+}'^{(o)} = \left(\lambda + \frac{A}{\lambda}\right)\tau_s'^{(o)} + \left(\frac{LA}{1-T_c/\bar{T}_s} + \frac{c_p}{c} - 1\right)\tau_s'^{(o)}$$
$$+ n_s\left(L + \frac{1-T_c/\bar{T}_s}{\lambda}\right)\frac{p'}{\bar{p}} \tag{148}$$

<u>First-order contribution</u>:

$$\mu_s'^{(1)} = \left(\alpha_s + \frac{E_s}{R_o\bar{T}_s}\right)\tau_s'^{(1)} \tag{149}$$

$$\left(\frac{\partial\tau}{\partial\xi}\right)_{s+}'^{(1)} = \left(\lambda + \frac{A}{\lambda}\right)\tau_s'^{(1)} + \left(\frac{c_p}{c} - 1 - HA\right)\tau_s' \tag{150}$$

Also, the linearized boundary conditions at the surface must be expanded in powers of $i\Omega\epsilon$. Just as in the analysis of the solid phase outlined in Section II, the solutions to the sets of equations above in Section V-A will yield the values at the origin $x = \xi = 0$, but one requires the values at the interface located at x_s; again, for small motions one has Eq. 6 and 7, and the corresponding formula for μ_s':

$$T_s' = T_{o+}' + x_s \left(\frac{d\bar{T}}{dx}\right)_{o+} \tag{6}$$

$$\left(\frac{\partial T}{\partial x}\right)'_{s+} = \left(\frac{\partial T}{\partial x}\right)'_{o+} + x_s \left(\frac{d^2\bar{T}}{dx^2}\right)_{o+} \tag{7}$$

$$\mu_s' = \mu_o' + x_s \left(\frac{d\bar{\mu}}{dx}\right)_{o+} \tag{151}$$

However, since for the one-dimensional problem $d\bar{\mu}/dx = 0$, Eq. 151 shows that $\mu_s' = \mu_o'$ and no correction is required for surface motion. In order to obtain the correct quasi-static limit for $\omega = 0$, it is necessary to adopt a coordinate system such that the solid moves at uniform speed $\bar{r}$ plus a correction u_o; then conservation of mass applied to the interface gives, instead of Eq. 9:

$$\rho_c (\bar{r} + u_o - \dot{x}_s) = \rho_{gs} (u_{gs} - \dot{x}_s)$$

and since $\bar{\rho}_{gs} << \rho_c$,

$$\dot{x}_s = \frac{\bar{m}}{\rho_c}\left(\frac{\rho_c u_o}{\bar{m}} - \frac{m_s'}{\bar{m}}\right) = \frac{\bar{m}}{\rho_c}\left(\frac{\rho_c u_o}{\bar{m}} - \mu_s'\right) \tag{152}$$

Expand the surface speed in powers of $i\Omega\varepsilon$,

$$\dot{x}_s = \dot{x}_s^{(o)} + (i\Omega\varepsilon)\, \dot{x}_s^{(1)} + \ldots$$

and collection of terms according to powers of $i\Omega\varepsilon$ in Eq. 152 gives the formulas for the successive terms in $\dot{x}_s$:

$$\dot{x}_s^{(o)} = \frac{\bar{m}}{\rho_c}\left(\frac{\rho_c u_o}{\bar{m}} - \mu_s'^{(o)}\right)$$

$$\dot{x}_s^{(1)} = -\frac{\bar{m}}{\rho_c}\,\mu_s'^{(1)} \tag{153}$$

In order to have the origin and the interface coincide at all times in the limit $\omega = 0$, it is necessary that $\dot{x}_s^{(o)}$ vanish, as argued in Section III. Hence, the correct choice for u_o is

$$u_o = \frac{\bar{m}}{\rho_c} \mu_s{}'^{(o)} = \frac{m'^{(o)}}{\rho_c} \tag{154}$$

which has the reasonable interpretation that for a small D.C. ($\omega = 0$) change of pressure, the solid is taken to move from the left at a speed equal to the original linear burning rate plus the change of linear burning rate corresponding to the change of pressure. Incidentally, this value for u_o appears nowhere explicitly; its presence here merely serves as a formal justification for the assertion in Section III that values at the interface coincide with those at the origin in the quasi-static limit--$T'_{o+} = T_g$, etc. It therefore follows by expansion of Eq. 6 and 7, and by use of Eq. 153, that the correct boundary values for the various approximations are:

$$T_s{}'^{(o)} = T_{o+}{}'^{(o)} \tag{155a}$$

$$\left(\frac{\partial T}{\partial x}\right)_{s+}'^{(o)} = \left(\frac{\partial T}{\partial x}\right)_{o+}'^{(o)} \tag{155b}$$

$$\begin{aligned}\tau_s{}'^{(1)} &= \tau_{o+}{}'^{(1)} + x_s{}'^{(1)} \left(\frac{d\bar{\tau}}{dx}\right)_{o+} = \tau_{o+}{}'^{(1)} \\ &\quad + x_s{}'^{(1)} \left(\frac{\bar{m}c_p}{k_g}\right) \left(\frac{d\bar{\tau}}{d\xi}\right)_{o+}\end{aligned} \tag{156a}$$

$$\left(\frac{\partial \tau}{\partial \xi}\right)_{s+}'^{(1)} = \left(\frac{\partial \tau}{\partial \xi}\right)_{o+}'^{(1)} + x_s{}'^{(1)} \left(\frac{\bar{m}c_p}{k_g}\right) \left(\frac{d^2\bar{\tau}}{d\xi^2}\right)_{o+} \tag{156b}$$

and similar equations for the higher order terms.

C. First-Order Correction to the Response Function

By definition, the response function R is the ratio evaluated at the downstream edge of the flame

$$R = \frac{(m'/\overline{m})_f}{p'/\overline{p}} = \frac{\mu_f{}'}{p'/\overline{p}}$$

$$= R^{(o)} + i\Omega\varepsilon\, R^{(1)}$$

when carried out to first order in $i\Omega\varepsilon$. The calculations of Section IV give $R^{(o)}$, the response function subject to the quasi-static approximation, $R^{(o)} = \mu'^{(o)}/(p'/\overline{p})$; the task here is to compute

$$R^{(1)} = \frac{\mu_f{}'^{(1)}}{p'/\overline{p}} \tag{157}$$

Equation 134, evaluated at the edge of the flame and with terms of order $\exp(-\overline{\xi}_f)$ and $1/\overline{\xi}_f$ dropped where appropriate, leads to the formula for $\mu_f{}'^{(1)}$:

$$\mu_f{}'^{(1)} = \mu_s{}'^{(1)} + \frac{\overline{\xi}_f}{\overline{\tau}_a}\left[\tau_{\overline{f}}{}'^{(o)} - \frac{q_{\overline{f}}{}'^{(o)}}{\overline{m}c_p\overline{T}_s} + \tfrac{1}{2}\left(\frac{w'^{(o)}}{\overline{w}} + \mu'^{(o)}\right) - \overline{\tau}_a\frac{p'}{\overline{p}}\right]$$

Thus, with Eq. 142, the first-order correction to the response function is

$$R^{(1)} = \frac{A}{(1-T_c/\overline{T}_s)}\left(\frac{\tau_s{}'^{(1)}}{p'/\overline{p}}\right) + \frac{\overline{\xi}_f}{\overline{\tau}_a(p'/\overline{p})} \times\left[\tau_{\overline{f}}{}'^{(o)} - \frac{q_{\overline{f}}{}'^{(o)}}{\overline{m}c_p\overline{T}_s} + \tfrac{1}{2}\left(\frac{w'^{(o)}}{\overline{w}} + \mu'^{(o)}\right) - \overline{\tau}_a\left(\frac{p'}{\overline{p}}\right)\right] \tag{158}$$

Equations 139, 150, 153, and 156a-b combine to give

$$\frac{\tau_s{}'^{(1)}}{p'/\overline{p}} = \frac{\dfrac{\gamma-1}{\gamma} + \dfrac{1}{\overline{\tau}_c(p'/\overline{p})}\left[\dfrac{q_f{}'^{(o)}}{\overline{m}c_p\overline{T}_s} - \tau_{\overline{f}}{}'^{(o)} - \mu'^{(o)}\,(\overline{\tau}_f-\overline{\tau}_s)\right]}{\lambda + \dfrac{A}{\lambda} + \dfrac{\sigma E/\overline{\xi}_f}{\Omega} - HA + \left(\dfrac{c_p}{c} - 1\right)} \tag{159}$$

where $\sigma = (k_c c_p / k_g c)$. Note that $\tau_s{}'^{(1)}$ vanishes in the limit of zero frequency, while the fluctuation of heat transfer is finite.

From Eq. 141,

$$\frac{q_{\overline{f}}{}'^{(o)}}{\overline{m}c_p\overline{T}_s} - \tau_{\overline{f}}{}'^{(o)} - \mu'^{(o)}\ (\overline{\tau}_f - \overline{\tau}_s) = \left(\frac{\partial \tau}{\partial \xi}\right)_{o+}^{'(o)} - \left(\tau_o{}'^{(o)} - \frac{w'^{(o)}}{\overline{w}}\right)$$

$$= (\Lambda^2+1)\ \frac{w'^{(o)}}{\overline{w}} - \left(1 + \frac{A}{1-T_c/\overline{T}_s}\Lambda^2\right)\tau_s{}'^{(o)}$$

$$\simeq \frac{w'^{(o)}}{\overline{w}} - \tau_s{}'^{(o)} \qquad (160)$$

and Eq. 159 becomes

$$\frac{\tau_s{}'^{(1)}}{p'/\overline{p}} = \frac{\dfrac{\gamma-1}{\gamma} + \dfrac{1}{\tau_a}\left[\dfrac{w'^{(o)}/\overline{w}}{p'/\overline{p}} - \dfrac{\tau_s{}'^{(o)}}{p'/\overline{p}}\right]}{\lambda + \dfrac{A}{\lambda} + \dfrac{\sigma E/\overline{\xi}_f}{i\Omega} - HA + \left(\dfrac{c_p}{c} - 1\right)} \qquad (161)$$

The term $(\sigma E/\overline{\xi}_f)/i\Omega$ is, because of the large size of $\overline{\xi}_f$, negligible except for very small values of $i\Omega$. Its effect on the response function is therefore insignificant, because Eq. 161 is multiplied by $i\Omega\varepsilon$ in the response function. Now with Eq. 160, Eq. 158 becomes

$$R^{(1)} = \frac{A}{1-T_c/\overline{T}_s}\ \left(\frac{\tau_s{}'^{(1)}}{p'/\overline{p}}\right)$$

$$+ \frac{\overline{\xi}_f}{\overline{\tau}_a}\ \left[\frac{3}{2}\,\frac{w'^{(o)}/\overline{w}}{p'/\overline{p}} + \left(\frac{A}{2\left(1 - \dfrac{T_c}{\overline{T}_s}\right)} - 1\right)\ \frac{\tau_s{}'^{(o)}}{p'/\overline{p}} - \overline{\tau}_a\right] \qquad (162)$$

Now from Eq. 64, with

$$\frac{w'^{(o)}}{\bar{w}} = W \frac{1}{\Lambda^2} \frac{c}{c_p} \left(1 - \frac{T_c}{\bar{T}_s}\right) \frac{p'}{\bar{p}}$$

$$= W \bar{\xi}_f \frac{c}{c_p} \left(1 - \frac{T_c}{\bar{T}_s}\right) \frac{p'}{\bar{p}}$$

Thus, because of the factor $\bar{\xi}_f{}^2$, the dominant terms in Eq. 162 are the terms involving $w'^{(o)}$; approximately, then:

$$R^{(1)} \simeq \frac{W}{\bar{\tau}_a} \bar{\xi}_f \frac{c}{c_p} \left(1 - \frac{T_c}{\bar{T}_s}\right)$$

$$\times \left[\frac{1}{\lambda + \frac{A}{\lambda} + \frac{\sigma E}{i\Omega\bar{\xi}_f} - 1 + A + \frac{c_p}{c} - 1} + \frac{3}{2} \bar{\xi}_f\right] \qquad (163)$$

and with all these simplifications, the response function to first order in $\Omega\varepsilon$ is

$$R = \frac{nAB + n_s\,(\lambda - 1)}{\lambda + \frac{A}{\lambda} - (1 + A) + AB} + i\Omega\varepsilon \frac{W}{\bar{\tau}_a} \bar{\xi}_f \frac{c}{c_p} \left(1 - \frac{T_c}{\bar{T}_s}\right) \times$$

$$\left[\frac{3}{2} \bar{\xi}_f + \frac{1}{\lambda + \frac{A}{\lambda} + \frac{\sigma E}{i\Omega\bar{\xi}_f} + \left(\frac{c_p}{c} - 1\right) - 1 + A}\right] \qquad (164)$$

It is somewhat surprising that such a simple result can finally be obtained. The primary reason for this, as remarked above, is formally that the normalized thickness of the flame, $\bar{\xi}_f$, is large, and it is possible to throw away many contributions. The physical meaning of this dimensionless quantity may be seen by writing it as

$$\bar{\xi}_f = \frac{\bar{m} c_p}{k_g} x_f$$

$$= \frac{\bar{m} c_p\, (\bar{T}_f - \bar{T}_s)}{k_g \left(\frac{\bar{T}_f - \bar{T}_s}{x_f}\right)}$$

The numerator is roughly the net flow of enthalpy through the gas phase, and the denominator is an estimate of the heat transfer from the gas phase to the solid. Thus, large $\bar{\xi}_f$ implies a relatively small heat transfer loss from the gas phase to the solid, a connection which also follows from Eq. 49 and other results arising in the solution to the steady-state problem.

Now the second term in the square brackets of Eq. 164 is, under practical conditions, not huge, whereas the first term is; hence, finally, the response function can be written approximately as

$$R \simeq \frac{nAB + n_s\ (\lambda-1)}{\lambda + \frac{A}{\lambda} - (1+A) + AB} + (i\Omega\varepsilon)\ \frac{3}{2}\,\bar{\xi}_f^{\,2}\,\frac{c}{c_p}\,\frac{\bar{w}}{\bar{\tau}_a}\left(1 - \frac{T_c}{\bar{T}_s}\right) \qquad (165)$$

The first-order correction in frequency, therefore, mainly affects the imaginary part of the response function.

The term proportional to $\bar{\xi}_f^2$ in Eq. 165 arises from the formula for $\mu_f'^{(1)}$, just before Eq. 158. It is effectively the integrated effect of the energy release fluctuation over the gas phase: note that $w'^{(o)}/\bar{w}$-$\bar{\xi}_f p'/\bar{p}$ according to Eq. 71 and the manipulations following Eq. 162. The energy release enters through its effect on the temperature profile in the conservation of mass, for example, as shown in Eq. 124 and subsequent integrations.

One therefore has the following interpretation for the major correction shown in Eq. 165. According to the conservation of mass, Eq. 52, the fluctuation of mass flux depends on the rate of change of temperature with time, and hence m' (i.e., μ') is out of phase with T'. When the entire gas phase is considered, the dominant contribution to the fluctuation of temperature is the fluctuation of energy release, which is, in the simplest case, <u>in</u> phase with the pressure fluctuations. Hence, since the energy release and associated temperature fluctuations are in phase, the net result is a contribution to the mass flux which is out of phase with the pressure changes, the last term of Eq. 165.

The fact that the correction term in Eq. 165 can be quite large, and yet the real part of the response function is not much affected, is a significant result. Earlier numerical calculations in Ref. 11 indicated that the real part of the response function given by the strictly quasi-static analysis did not differ much from that given by the results of Ref. 6 which did account for nonquasi-static behavior of the gas phase. Moreover, the real part of the quasi-static formula, the first ratio in Eq. 165, does give remarkably good (at least qualitatively) agreement with experimental results. However, the analysis of Ref. 7, in which both the real <u>and</u> imaginary parts of the quasi-static formula were used to interpret data, showed very serious discrepancies. These earlier observations are all accommodated qualitatively by the result shown in Eq. 165, that nonquasi-static behavior contributes overwhelmingly to the "out of phase" component of the response function.

It must be emphasized, however, that this does not mean that one is free to determine numerical values of A and B by comparing only the real part of the response with experimental results. In this respect, the discussion of Ref. 7 is of course valid. The charts developed in that work will, however, be substantially changed if the correction term of Eq. 165 is taken into account. This will be treated in a subsequent publication.

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13 ABSTRACT

The model of uniformly distributed combustion in the gas phase is used as the basis for examining several features of unsteady burning. As a zeroth approximation the flame zone begins at the solid-gas interface and responds linearly and quasi-statically to changes of pressure only. Three deviations from this behavior are then examined: the combustion zone is displaced from the surface, the energy release responds to fluctuations of temperature and the response is not quasi-static. It appears that the assumption that the burning begins immediately at the surface can lead to significant changes, more important than the assumption that the energy release responds to changes of pressure only, especially in the interpretation of experimental data. The problem of nonquasistatic behavior, i.e., processes in the gas phase do not follow precisely impressed changes of pressure, is formulated as an expansion in frequency. Approximate results seem to be consistent with existing information.

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