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Linear Burning Rate Dynamics of Solids Subjected to Pressure or External Radiant Heat Flux Oscillations

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Linearized analysis of burning solids subjected to pressure or external radiant heat flux oscillations results in relatively simple expressions for the burning rate response, particularly when combined with quasisteady gas and surface zone assumptions. In this article, a linear expression for the radiant heat flux response function R_q as a function of primary experimental parameters is obtained. The implications of this expression and its relationship to the pressure coupled response function R_p are examined. In particular, the linearized effects of the mean radiant heat flux level and in-depth absorption on R_q are investigated. The linear response to a series of radiant pulses is also presented to suggest an alternate method of experimentally measuring R_q. The effects of nonlinearities are investigated using numerical calculations. It is shown that, in general, the relationship between R_p and R_q is more complicated than a constant scaling factor. The results demonstrate that in-depth absorption of the thermal radiant energy in the solid significantly affects R_q for many practical conditions. Furthermore, even when the equivalence principle holds in the steady case, an equivalent change in the initial temperature does not have the same effect on R_q, in general, as an equivalent mean radiant flux. Also, the mean radiant flux is seen to have a significant effect on R_q. An attempt is made throughout this article to further clarify the relationship and differences between the flame modeling (FM) and the Zeldovich-Novozhilov (ZN) phenomenological approaches in the prediction of R_q and R_p.

Nomenclature

A = most general form is $(\bar{T}_s - T_0)(\partial \ln \bar{r}_b/\partial \bar{T}_S)_{p,q=0} = k/r$
A^* = most general form is $(\bar{T}_s - T_0)(\partial \ln \bar{r}_b/\partial \bar{T}_s)_{p,q} = k^*/r^*$
B = most general form is $1/[(\bar{T}_s - T_0)(\partial \ln \bar{r}_b/\partial T_0)_{p,q=0}] = 1/k$
B^* = most general form is $1/[(\bar{T}_s - T_0)(\partial \ln \bar{r}_b/\partial T_0)_{p,q}] = 1/k^*$
C = specific heat of condensed phase
C_p = specific heat of gas phase
E = activation energy
f_g = $(\bar{r}_b/\alpha_c)[T_s(p, r_b) - T_0(p, r_b)]$
f_r = fraction of q absorbed below surface reaction zone
f_s = $(\mathrm{d}T/\mathrm{d}x)_{s-}$
H = dimensionless heat release, $Q_s/C(\bar{T}_s - T_0)$
i = $\sqrt{-1}$
J = dimensionless mean radiant flux, $\bar{q}/\rho\bar{r}_b C(\bar{T}_S - T_0)$
K_a = absorption coefficient of condensed phase material
k = $(\bar{T}_s - T_0)(\partial \ln \bar{r}_b/\partial T_0)_{p,q=0} = (\bar{T}_S - T_0)\sigma_p$; also thermal conductivity
k^* = $(\bar{T}_S - T_0)(\partial \ln \bar{r}_b/\partial T_0)_{p,q} = (\bar{T}_S - T_0)\sigma_p^*$
n = pressure exponent in $u \sim p^n$ expression; parameter in Eq. (2)
n_s = pressure exponent in pyrolysis expression, parameter in Eq. (2)
p = pressure
Q = heat release
q = absorbed radiant heat flux, $(1 - r_\lambda)\tau_\lambda q_r$
q_r = external radiant flux
R_p = pressure frequency response function, $(\Delta r_b/\bar{r}_b)/(\Delta p/\bar{p}) = \Delta\upsilon/\Delta\eta$
R_q = radiant heat flux frequency response function, $(\Delta r_b/\bar{r}_b)/(\Delta q/\bar{q}) = \Delta\upsilon/\Delta\psi$
r = $(\partial\bar{T}_s/\partial T_0)_{p,q=0}$
r_b = burning rate
r_λ = reflectivity of condensed phase material
r^* = $(\partial\bar{T}_s/\partial T_0)_{p,q}$
S = $(\bar{T}_s - T_0)/[\bar{T}_s - T_0 - (\bar{q}f_I/\rho\bar{r}_b C)]$
T = temperature
TF = R_p/R_q
t = time
X = length scale
x = spatial coordinate
z = $-\frac{1}{2} + \frac{1}{2}(1 + 4i\Omega)^{1/2}$
α = thermal diffusivity
α_s = temperature exponent in surface pyrolysis expression
β_r = ratio of thermal to radiant length scale $(K_a\alpha_c/\bar{r}_b)$
$\Delta(\ )$ = denotes an oscillatory value
δ = $\nu r - \mu k$
δ_q = $\nu_q r^* - \mu_q k^*$
δ^* = $\nu^* r^* - \mu^* k^*$
η = dimensionless pressure, $p/\bar{p}$
θ = dimensionless temperature, $(T - T_0)/(\bar{T}_s - T_0)$
λ = $\frac{1}{2} + \frac{1}{2}(1 + 4i\Omega)^{1/2}$
μ = $1/(\bar{T}_s - T_0)(\partial\bar{T}_s/\partial \ln \bar{p})_{T_0,q=0}$
μ_q = $1/(\bar{T}_s - T_0)(\partial\bar{T}_s/\partial \ln \bar{q})_{T_0,p}$
μ^* = $1/(\bar{T}_S - T_0)(\partial\bar{T}_S/\partial \ln \bar{p})_{T_0,q}$
ν = $(\partial \ln \bar{r}_b/\partial \ln \bar{p})_{T_0,q=0}$
ν_q = $(\partial \ln \bar{r}_b/\partial \ln \bar{q})_{T_0,p}$
ν^* = $(\partial \ln \bar{r}_b/\partial \ln \bar{p})_{T_0,q}$
ξ = $x(\bar{r}_b/\alpha_c)$
ρ = density
σ_p = $(\partial \ln \bar{r}_b/\partial T_0)_{p,q=0}$
σ_p^* = $(\partial \ln \bar{r}_b/\partial T_0)_{p,q}$
τ = $t(\bar{r}_b^2/\alpha_c)$ or characteristic time
τ_λ = transmittance through plume above propellant
υ = dimensionless burning rate $r_b/\bar{r}_b$
ϕ_s = dimensionless surface gradient $f_s/\bar{f}_s$
ψ = dimensionless radiant heat flux $q/\bar{q}$, $\bar{q} \neq 0$
Ω = dimensionless frequency $\omega(\alpha_c/\bar{r}_b^2)$
ω = angular frequency, rad/s

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Superscripts and Subscripts

$*$ = $\bar{q} > 0$ included
c = condensed phase or convective-diffusive
g = gas phase
p = pulse
R = reaction layer
r = radiant heat flux
s = surface
$s-$ = just below the surface
$s+$ = just above the surface
$-$ = denotes the steady condition
$'$ = denotes differentiation w.r.t.x
0 = deep into the propellant ($x = -\infty$)

Introduction

THERE is a resurgent and growing recognition in the U.S. of the advantages of studying the combustion of solids, particularly solid propellants, using unsteady external radiant energy sources such as lasers. Through this technique, information can be obtained about the dynamic burning of solids including the characteristics of the thermal relaxation and the material's response to high heating rates (up to 10^5 K/s). Furthermore, it is easier to control a radiant flux and to simultaneously measure the instantaneous burning rate than to perform a similar experiment with an unsteady pressure field. Measuring the recoil using a microforce transducer during an irradiation transient is one way to obtain experimental values of R_q. Using this technique, several researchers have measured R_q.[1-5] R_q has also been measured recently using a microwave technique.[6] Thus, it is worthwhile to thoroughly examine the theory used to predict R_q to gain a better understanding of experimental results. The objective of this work is to derive a general result for R_q by using the Zeldovich–Novozhilov (ZN) approach, and based on this result, investigate the usefulness of a radiant flux to study the unsteady burning of solid propellants.

R_p Prediction

Zeldovich first introduced the ZN approach in an analysis of the burning rate response to pressure oscillations that assumed a constant surface temperature.[7] The ZN approach assumes a quasisteady gas phase and surface reaction zone wherein only the nonreacting zone below the surface reaction zone is allowed to be completely unsteady. Although there are conditions where this assumption is probably violated, there are important applications where it is justifiably applied, and it will always serve as a first approximation. The most complete description of the ZN method is found in the books by Zeldovich et al.[8] and Novozhilov.[9] A similar phenomenological approach, which included the temperature sensitivity as an explicit parameter, was also taken in the U.S. by Hart and co-workers in a series of papers (e.g., Ref. 10). However, U.S. literature has been dominated by the flame modeling (FM) approach which models the processes in the gas phase and surface reaction instead of relying directly on measured steady burning parameters.

The connection between the ZN and FM approaches has been addressed many times in the open literature (cf. Refs. 11–14), yet a full understanding of this issue is still not common. The ZN approach has the advantage of not requiring detailed knowledge of the flame structure. Instead it is based on knowledge of the steady burning [specifically $\bar{r}_b(T_0, \bar{p})$ and $\bar{T}_s(T_0, \bar{p})$]. From this information, the unsteady heat feedback function to the condensed phase from the surface and gas phase reaction zones can be deduced within the quasisteady surface and gas phase approximation. It can be shown that this heat feedback function deduced using the ZN method must be the same as that deduced in FM approaches (assuming the FM accurately models the steady burning).[14] Flame modeling requires hypotheses to be made concerning the actual mechanisms involved in the surface and gas phase reaction zones. Although much more information is required (including kinetic parameters that may be uncertain), the advantage of the FM approach is that more detailed information is also predicted that may be helpful to propellant formulators. Thus, the potential for the appropriate use of the ZN theory is still great since a wide variety of propellants will satisfy the assumptions of the method under many conditions. In short, both ZN and FM approaches are useful and should be fully utilized to gain the most understanding.

Using an FM approach, Denison and Baum[15] were among the first to relax the assumption of a constant surface temperature. Later, Novozhilov[16] relaxed the assumption of a constant surface termperature to further generalize the ZN description. Novozhilov obtained the following expression for the pressure coupled response function:

$$R_p \equiv \frac{\Delta r_b/\bar{r}_b}{\Delta p/\bar{p}} \equiv \frac{\Delta v}{\Delta \eta} = \frac{\nu + \delta z}{1 - k + (r - ik/\Omega)z} \tag{1}$$

In system dynamics terminology, Eq. (1) is a transfer function relating the input oscillating pressure and output burning rate. Because of the assumed linearity, this expression is limited to applications where the amplitude of the burning rate oscillations is small with respect to the steady burning rate. To apply this equation to a specific case, the parameters (ν, δ, k, r) need only be known at the corresponding stable (stationary) burning condition. However, to consider the changes in R_p subject to different environments (i.e., various $\bar{p}$ and T_0 conditions), the change in these parameters should be accounted for. The k and r parameters are dimensionless sensitivities of the burning rate and surface temperature, respectively, to the initial temperature. Likewise, ν is the dimensionless sensitivity of the burning rate to pressure. If the burning rate can be expressed as $\bar{r}_b \sim \bar{p}^n$, ν is then equal to the exponent n, but other relationships between the burning rate and pressure can be considered within the assumptions of this expression. The Jacobian parameter δ (see nomenclature) is identically zero if the surface temperature is a function of the burning rate only. R_p then reduces to a complex valued function of only three parameters.

Using the FM approach, various gas phase descriptions will result in different final expressions for R_p which may appear quite different from each other, even though the basic assumptions used in the derivations are identical (except for the gas phase description). In a classic work, Culick[17] showed that various proposed FM models for R_p, all based on similar assumptions, could be expressed in the following common form:

$$R_p \equiv \frac{\Delta r_b/\bar{r}_b}{\Delta p/\bar{p}} \equiv \frac{\Delta v}{\Delta \eta} = \frac{nAB + n_s(\lambda - 1)}{\lambda + (A/\lambda) - (1 + A) + AB} \tag{2}$$

Likewise, one would expect that the result of the ZN analysis, Eq. (1), should be of the same form. In the original form shown in Eq. (1) it is not immediately possible to show this. The difference originates in the way the energy equation is linearized. Novozhilov[16] linearized using the dimensionless temperature expressed as $\theta = \bar{\theta}(1 + \Delta\theta e^{i\Omega\tau})$ instead of the equally appropriate alternative of $\theta = \bar{\theta} + \Delta\theta e^{i\Omega\tau}$. Using the latter linearization in the ZN approach, an expression entirely equivalent to Eq. (1) is

$$R_p \equiv \frac{\Delta r_b/\bar{r}_b}{\Delta p/\bar{p}} \equiv \frac{\Delta v}{\Delta \eta} = \frac{\nu + \delta(\lambda - 1)}{1 + (r - k/\lambda)(\lambda - 1)}$$

$$= \frac{\nu/r + (\delta/r)(\lambda - 1)}{\lambda + (k/r)/\lambda - (1 + k/r) + 1/r} \tag{3}$$

Equation (3) shows that the ZN result for R_p can be written in the same form as that from the FM approach, Eq. (2), with $A \equiv k/r$, $B \equiv 1/k$, $n \equiv \nu$, and $n_s \equiv \delta/r$. The equivalence of the first three parameters (A, B, and n) has also been noted elsewhere.[11-13] If A, B, n, and n_s are defined based on Eq. (3), the two expressions are identically equivalent. However, typically these parameters (A, B, n, and n_s) have been based on proposed models (gas phase descriptions, pyrolysis expressions, etc.). In this sense, the ZN formulation is somewhat more general because these dependencies are not specified a priori. In the following sections, an analysis of the unsteady burning with an external radiant heat flux is presented.

Governing Equation and Boundary Conditions

The model considered is illustrated schematically in Fig. 1. The coordinate origin is located just below the surface reaction zone of the condensed phase. The external radiant flux is attenuated by the gas phase combustion products and a certain amount of the incident radiative flux is reflected by the condensed phase to the surroundings. The activation energy of the surface reaction is assumed high enough so that a reactive-diffusive zone (X_R) can be identified, and the reaction zone is thin relative to the thermal layer thickness ($X_c >> X_R$; see Fig. 1). In this approximation, the condensed phase is nonreacting below the surface reaction layer. The density, thermal conductivity, and heat capacity of the solid are considered constant and the natural radiant feedback to the solid is neglected. It has been assumed that scattering is negligible, the absorption coefficient K_a is spatially constant, thermal emission by the condensed phase is negligible, and the incident flux is collimated. The simplified unsteady energy equation and boundary conditions are then

$$\frac{\partial T}{\partial t} = -r_b \frac{\partial T}{\partial x} + \alpha_c \frac{\partial^2 T}{\partial x^2} + (1 - r_\lambda)\tau_\lambda f_r \frac{q_r}{\rho C} K_a \exp(K_a x) \tag{4a}$$

$$T(-\infty, t) = T_0 \tag{4b}$$

$$f_s \equiv \left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)_{s-} = (1 - r_\lambda)\tau_\lambda(1 - f_r)q_r/k_c + \left[k_g\left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)_{s+} + \rho r_b Q_s\right]\Big/k_c \tag{4c}$$

The surface gradient condition (Eq. 4c) includes the radiant energy that is absorbed in the surface reaction zone and the effect of the conductive feedback and heat release (or endothermic reaction) at the surface. An interesting feature of the posted problem is the coupling of this surface boundary condition, Eq. (4c), with Eq. (4a) through the burning rate. For the FM approach in unsteady analyses, the second term on the right side of Eq. (4c) is obtained by modeling the surface pyrolysis and flame processes. Ideally, this modeling will predict all the experimentally observed characteristics of the steady burning propellant. In the ZN approach this term is obtained from a direct appeal to steady burning results.

The fraction of radiant flux that is absorbed below the reaction layer f_r needs to be specified. If $f_r = 0$, then all the radiation is absorbed in the surface reaction layer and the radiant flux term will only appear in the surface boundary condition. As f_r approaches 1.0, the length scale for absorption (X_r) is much larger than the thickness of the actual reaction layer so that the majority of the radiant energy is absorbed below the surface reaction layer. The scale of the reaction layer thickness X_R is $(\alpha_c/\bar{r}_b)(R\bar{T}_s/E_s)$. Using this scaling as the actual reaction layer thickness and Beer's law, the fraction absorbed below the surface layer is $f_r = \exp(-X_R/X_r) = \exp(-\beta_r R\bar{T}_s/E_s)$. Since the thickness of the actual reaction layer is not known accurately, the value of f_r is only an approximation and is assumed constant for a given calculation. It is evaluated using the above approximation at the corresponding steady burning conditions. Note that the assumption of a thin surface reaction layer could be violated by a very large absorbed radiant flux with $f_r > 0$ which could alter the temperature profile sufficiently to create a thicker reaction zone.

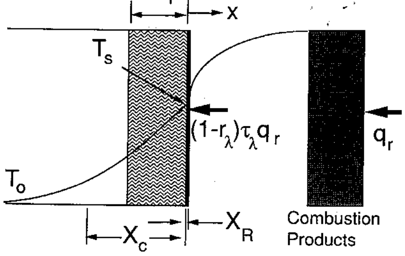

Fig. 1 Schematic of problem considered.

Since steady burning is a subset of transient burning, it is considered in the Appendix. The characteristics of the primary experimental parameters of surface temperature and burning rate of noncatalyzed double-base propellants are also briefly reviewed in the Appendix.

Radiant Heat Flux Frequency Response

The majority of analytical studies of the unsteady burning of solids have only addressed the effect of unsteady pressure oscillations. However, a few studies have also considered effects of an external radiant flux. The results of these studies are discussed briefly and then a somewhat more general expression for the linearized radiant heat flux response function is presented.

If the propellant is sufficiently opaque at the wavelength of the incident radiant flux, the energy is absorbed in a thin region near the surface and acts as an external factor to the fully unsteady condensed phase; that is, the absorption length scale X_r is small compared to the condensed phase reaction length scale X_R. However, if $X_r >> X_R$, most of the radiant flux is absorbed below the surface reaction layer and appears as a source in the energy equation in the condensed phase. In the former case, the energy deposited in the surface zone is transported upstream by way of conduction only into the condensed phase, whereas, in the latter case the energy is transported radiatively until it is absorbed. Therefore, it can be expected that the absorption coefficient of the propellant will play an important role in the prediction of R_q.

Assovskii and Istratov[18] assumed that $X_r >> X_R$ (in-depth absorption only) and considered conditions somewhat beyond linear by also including second-order terms to obtain an expression for the burning rate response to an oscillatory radiant flux. Assuming $X_r << X_R$, Zarko et al.[3] proposed a modification of two of the parameters in Eq. (1) to obtain an expression for R_q in the same form as Eq. (1). To obtain this expression Zarko et al. defined $\nu_q \equiv (\partial \ln \bar{r}_b/\partial \ln \bar{q})_{p,T_0}$ and $\delta_q \equiv \nu_q r - \mu_q k$ to replace ν and δ respectively, where $\mu_q \equiv [1/(\bar{T}_s - T_0)](\partial \bar{T}_s/\partial \ln \bar{q})_{p,T_0}$ (note that δ_q is defined as shown in the nomenclature for the remainder of the article). This is an extremely convenient form because if it is further assumed that $\delta_q = \delta = 0$, R_p is then related to R_q by the vactor ν/ν_q at every frequency. Earlier, De Luca[19] proposed a similar relationship between R_q and R_p using a distributed FM approach. An analysis presented by Strand et al.[6] recently showed a similar result, without considering the relaxation in the condensed phase; thus, their analysis represents a verification that at the zero frequency point (steady burning), $R_p/R_q = \nu/\nu_q$. It is emphasized here, and shown below, that the approach described above is only valid if the k and r (or the A and B) parameters are not changed by the mean value of the oscillating external radiant flux, in addition to surface absorption prevailing ($X_R << X_R$). References 3, 6, and 19 did not clearly identify or justify this assumption. Therefore, one objective of this article is to address this issue because in at least some cases this assumption could be violated. Mihlfeith[1] and Prevish and Brewster[20] also developed expressions for R_q within the FM approach assuming a thin flame. In the following section, a linear expression based on experimental parameters is obtained for the effects of an oscillating pressure field and external radiant flux on the instantaneous burning rate.

Analysis

The derivation presented in this section closely follows the steps taken in the original work by Novozhilov[9,16] in his derivation of an expression for R_p. This work is viewed as a generalization of the result obtained by Novozhilov. In the case of an external radiant flux, if the gas phase reaction zone does not absorb thermal radiation, the functional dependence of the burning rate on the surface temperature gradient and pressure takes the same form as in the absence of external irradiation.[8] The external irradiation is accounted for as an additional thermal source in the energy equation and in the surface energy balance (Eq. 4). This means that the effect of an external radiant heat flux can be straightforwardly included in any quasisteady description of the gas phase and interface. For example, invoking the quasisteady gas phase and interface assumption and using Eq. (A6), the surface boundary condition, Eq. (4c), becomes

$$f_s \equiv \left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)_{s-} = (1 - r_\lambda)\tau_\lambda(1 - f_r)q_r/k_c + \frac{r_b}{\alpha_c}$$

$$\cdot\,[T_s(p,\, r_b) - T_0(p,\, r_b)] = (1 - f_r)q/k_c + f_g(p,\, r_b) \tag{5}$$

where $f_g(p, r_b)$ is the same feedback function as the case without a radiant flux present. In the unsteady case $r_b = r_b(p, q, f_s)$ follows from Eq. (5) and, correspondingly, $T_s = T_s(p, q, f_s)$, where for convenience, $q \equiv (1 - r_\lambda)\tau_\lambda q_r$. The latter dependence $[T_s = T_s(p, q, f_s)]$ can be shown by leaving the T_s dependence in the function f_g and eliminating the r_b dependence. In dimensionless form, these expressions are $\upsilon = \upsilon(\eta, \Psi, \phi_s)$ and $\theta_s = \theta_s(\eta, \Psi, \phi_s)$. The dimensionless variables used are scaled in reference to the steady burning condition (fixed by the $\bar{p}$, $\bar{q}$, and T_0 environment) around which the unsteady solution oscillates. Specifically, $\upsilon \equiv r_b/\bar{r}_b$, $\eta \equiv p/\bar{p}$, $\psi \equiv q/\bar{q}$, $\phi_s \equiv f_s/\bar{f}_s$, and $\theta_s \equiv (T_s - T_0)/(\bar{T}_s - T_0)$. An important parameter that follows from this choice of dimensionless variables is the dimensionless mean radiant heat flux level $J \equiv \bar{q}/[\rho\bar{r}_b C(\bar{T}_s - T_0)]$. Based on the above functional dependencies of υ and θ_s, the following expressions for the oscillatory dimensionless buring rate ($\Delta\upsilon$) and surface temperature ($\Delta\theta_s$) are obtained:

$$\Delta\upsilon = \Delta\phi_s\left(\frac{\partial\upsilon}{\partial\phi_s}\right)_{\eta,\psi} + \Delta\eta\left(\frac{\partial\upsilon}{\partial\eta}\right)_{\phi_s,\psi} + \Delta\psi\left(\frac{\partial\upsilon}{\partial\psi}\right)_{\phi_s,\eta} \tag{6}$$

$$\Delta\theta_s = \Delta\phi_s\left(\frac{\partial\theta_s}{\partial\phi_s}\right)_{\eta,\psi} + \Delta\eta\left(\frac{\partial\theta_s}{\partial\eta}\right)_{\phi_s,\psi} + \Delta\psi\left(\frac{\partial\theta_s}{\partial\psi}\right)_{\phi_s,\eta} \tag{7}$$

The partial derivatives in Eqs. (6) and (7) can be evaluated directly from the expressions $\upsilon = \upsilon(\eta, \psi, \phi_s)$ and $\theta_s = \theta_s(\eta, \psi, \phi_s)$. It is somewhat more convenient, however, to derive the relationships of these partials to the primary parameters from steady burning experimental data, similar to what was done by Novozhilov[16] in the derivation of an expression for R_p. Using the energy balance

$$\bar{f}_s = (\bar{r}_b/\alpha_c)[\bar{T}_s - T_0 - (\bar{q}f_r/\rho\bar{r}_b C)] \tag{8}$$

the following relationship is found:

$$\frac{\bar{T}_s - T_0}{S}\left(\frac{\partial \ln \bar{f}_s}{\partial T_0}\right)_{p,q} = (\bar{T}_s - T_0)\left(\frac{\partial \ln \bar{r}_b}{\partial T_0}\right)_{p,q}$$

$$+ \left(\frac{\partial\bar{T}_s}{\partial T_0}\right)_{p,q} - 1 = k^* + r^* - 1 \tag{9}$$

Similarly, two more relationships are also found

$$\frac{1}{S}\left(\frac{\partial \ln \bar{f}_s}{\partial \ln \bar{p}}\right)_{T_0,q} \nu^* + \mu^*; \qquad \frac{1}{S}\left(\frac{\partial \ln \bar{f}_s}{\partial \ln \bar{q}}\right)_{T_0,p} = \nu_q + \mu_q - f_r J \tag{10}$$

Using these expressions, relationships between the primary experimental parameters and the partial derivatives appearing in Eqs. (6) and (7) can be obtained. The most straightforward way to derive these relationships is through Jacobian transformations.[16] For example

$$\left(\frac{\partial\upsilon}{\partial\eta}\right)_{\phi_s,\psi} = \left(\frac{\partial \ln \bar{r}_b}{\partial \ln \bar{p}}\right)_{f_s,q} \equiv \frac{\partial(\ln \bar{r}_b,\ \ln \bar{f}_s,\ \bar{q})}{\partial(\ln \bar{p},\ \ln \bar{f}_s,\ \bar{q})}$$

$$= \frac{\partial(\ln \bar{r}_b,\ \ln \bar{f}_s,\ \bar{q})/\partial(\ln \bar{p},\ T_0,\ \bar{q})}{\partial(\ln \bar{p},\ \ln \bar{f}_s,\ \bar{q})/\partial(\ln \bar{p},\ T_0,\ \bar{q})}$$

$$= \frac{\partial(\ln \bar{r}_b,\ \ln \bar{f}_s,\ \bar{q})/\partial(\ln \bar{p},\ T_0,\ \bar{q})}{\partial(\ln \bar{f}_s,\ \ln \bar{p},\ \bar{q})/\partial(T_0,\ \ln \bar{p},\ \bar{q})}$$

$$= \frac{\left(\frac{\partial \ln \bar{r}_b}{\partial \ln \bar{p}}\right)_{T_0,q}\left(\frac{\partial \ln \bar{f}_s}{\partial T_0}\right)_{p,q} - \left(\frac{\partial \ln \bar{r}_b}{\partial T_0}\right)_{p,q}\left(\frac{\partial \ln \bar{f}_s}{\partial \ln \bar{p}}\right)_{T_0,q}}{\left(\frac{\partial \ln \bar{f}_s}{\partial T_0}\right)_{p,q}}$$

$$= \frac{\nu^*(r^* - 1) - k^*\mu^*}{k^* + r^* - 1} = \frac{\delta^* - \nu^*}{k^* + r^* - 1} \tag{11}$$

In a similar manner, the appropriate expressions for the remaining partial derivatives appearing in Eqs. (6) and (7) can be found. Substituting these relations into Eqs. (6) and (7), the following expressions are obtained:

$$\Delta\upsilon = \Delta\phi_s\left(\frac{k^*/S}{k^* + r^* - 1}\right) + \Delta\eta\left(\frac{\delta^* - \nu^*}{k^* + r^* - 1}\right)$$

$$+ \Delta\psi\left(\frac{\delta_q - \nu_q + k^* f_r J}{k^* + r^* - 1}\right) \tag{12}$$

$$\Delta\theta_s = \Delta\phi_s\left(\frac{r^*/S}{k^* + r^* - 1}\right) - \Delta\eta\left(\frac{\delta^* + \mu^*}{k^* + r^* - 1}\right)$$

$$- \Delta\psi\left(\frac{\delta_q + \mu_q - r^* f_r J}{k^* + r^* - 1}\right) \tag{13}$$

In addition to Eqs. (12) and (13), two more equations are needed to complete the solution. These will come from the solution of the linearized Eq. (4a). In other words, Eqs. (12) and (13) will provide a surface boundary condition for Eq. (4a). To obtain this solution, Eq. (4a) is linearized using: $\theta = \bar{\theta} + \Delta\theta e^{i\Omega\tau}$; $\upsilon = \bar{\upsilon} + \Delta\upsilon e^{i\Omega\tau}$; $\eta = \bar{\eta} + \Delta\eta e^{i\Omega\tau}$; $\psi = \bar{\psi} + \Delta\psi e^{i\Omega\tau}$. The resulting expression is

$$\Delta\theta'' - \Delta\theta' - \Delta\theta i\Omega = \Delta\upsilon\bar{\theta}' - \Delta\psi f_r J\beta_r \exp(\beta_r\xi) \tag{14}$$

The characteristic equation is $\lambda^2 - \lambda - i\Omega = 0$, which has a physical root of $\lambda = \frac{1}{2} + \frac{1}{2}(1 + 4i\Omega)^{1/2}$. The second root of the characteristic equation is rejected, since it corresponds to an infinitely increasing solution when $\xi \rightarrow -\infty$. Using the steady solution of the temperature profile, Eq. (A1), to eliminate $\bar{\theta}'$, Eq. (14) becomes

$$\Delta\theta'' - \Delta\theta' - \Delta\theta i\Omega = \Delta\upsilon\left(\frac{1 - \beta_r - f_r J}{1 - \beta_r}\right)\exp(\xi)$$

$$+ \left[\frac{\Delta\upsilon - \Delta\psi(1 - \beta_r)}{(1 - \beta_r)}\right] f_r\beta_r \exp(\beta_r\xi) \tag{15}$$

From the general solution of Eq. (15) the final two equations that are necessary for a solution are obtained

$$\Delta\theta_s = A_1 + \Delta v\left[\frac{\beta_r + f_rJ - 1}{\lambda(\lambda - 1)(1 - \beta_r)}\right] + \left\{\frac{\Delta v - \Delta\psi(1 - \beta_r)}{[\beta_r^2 - \beta_r - \lambda(\lambda - 1)](1 - \beta_r)}\right\} f_r\beta_r \tag{16}$$

$$\frac{\Delta\phi_s}{S} = A_1\lambda + \Delta v\left(\frac{\beta_r + f_rJ - 1}{\lambda(\lambda - 1)(1 - \beta_r)}\right) + \left\{\frac{\Delta v - \Delta\psi(1 - \beta_r)}{[\beta_r^2 - \beta_r - \lambda(\lambda - 1)](1 - \beta_r)}\right\} f_rJ\beta_r^2 \tag{17}$$

where A_1 is an integration constant. From Eqs. (12), (13), (16) and (17) expressions can be obtained for Δv, $\Delta\phi_s$, and $\Delta\theta_s$ as functions of $\Delta\eta$ and $\Delta\psi$. Eliminating A_1 and solving for Δv it is found that

$$\Delta v = \Delta\psi\left\{\frac{\nu_q + \delta_q(\lambda - 1) - \dfrac{k^*f_rJ(\lambda - 1)}{\beta_r + \lambda - 1}}{1 + \left[r^* - \dfrac{k^*(\beta_r + \lambda - 1 + f_rJ)}{\lambda(\beta_r + \lambda - 1)}\right\}(\lambda - 1)}\right\} + \Delta\eta\left\{\frac{\nu^* + \delta^*(\lambda - 1)}{1 + \left[r^* - \dfrac{k^*(\beta_r + \lambda - 1 + f_rJ)}{\lambda(\beta_r + \lambda - 1)}\right](\lambda - 1)}\right\} \tag{18}$$

For no pressure oscillations

$$R_q \equiv \frac{\Delta v}{\Delta\psi} = \frac{\nu_q + \delta_q(\lambda - 1) - \dfrac{k^*f_rJ(\lambda - 1)}{\beta_r + \lambda - 1}}{1 + \left[r^* - \dfrac{k^*(\beta_r + \lambda - 1 + f_rJ)}{\lambda(\beta_r + \lambda - 1)}\right\}(\lambda - 1)} \tag{19}$$

It is readily seen that for surface absorption ($f_r = 0$, $\beta_r = \infty$) Eq. (19) takes on the same form as Eq. (3). Similarly, for a constant radiant flux

$$R_p^* \equiv \frac{\Delta v}{\Delta\eta} = \frac{\nu^* + \delta^*(\lambda - 1)}{1 + \left[r^* - \dfrac{k^*(\beta_r + \lambda - 1 + f_rJ)}{\lambda(\beta_r + \lambda - 1)}\right](\lambda - 1)} \tag{20}$$

Equation (20) could be used to investigate the effect of a constant flux on R_p. For the case of no constant external radiant heat flux $J = 0$, Eq. (20) simplifies to Eq. (3). For surface absorption ($f_r = 0$, $\beta_r = \infty$), Eq. (20) assumes the same form as Eq. (3), but its parameters are, in general, modified by the external radiant flux. At $\Omega = 0$ ($\lambda = 1$), $R_p = \nu$ and $R_q = \nu_q$, as they must. If the equivalence principle holds [see Eq. (A10)] it can be shown that $\nu_q = Jk^*$ and assuming further that $\delta_q = 0$ [e.g., $\bar{r}_b = \bar{r}_b(\bar{T}_s)$] Eq. (19) becomes

$$R_q \equiv \frac{\Delta v}{\Delta\psi} = \frac{k^*J\left[1 - \dfrac{f_r(\lambda - 1)}{\beta_r + \lambda - 1}\right]}{1 + \left[r^* - \dfrac{k^*(\beta_r + \lambda - 1 + f_rJ)}{\lambda(\beta_r + \lambda - 1)}\right](\lambda - 1)} \tag{21}$$

Note that f_r and β_r affect the unsteady burning response, whereas, they have no effect on the steady burning rate in cases where the gas phase is the controlling layer and the equivalence principle holds. For a constant absorption coefficient in the reaction zone and the nonreacting condensed phase, the value of f_r will depend directly on β_r. However, the absorption coefficients and subsurface condensed phase may be different due to changes in chemical species and phase. Thus, f_r could change independently of β_r. From examination of Eq. (21), it can be seen that considering the limits of $f_r \to 0$ or $\beta_r \to \infty$ (surface absorption) the same expression is obtained.

Comparison with Flame Models

It is also possible to recast R_q in a form which is more similar to that based on the FM approach. Defining $A^* \equiv k^*/r^*$ and $B^* \equiv 1/k^*$, an alternative form of Eq. (21) is

$$R_q = \frac{A^*J\left[1 - \dfrac{f_r(\lambda - 1)}{\beta_r + \lambda - 1}\right]}{\lambda + A^*/\lambda - (1 + A^*) + A^*B^* - \dfrac{f_rJA^*(\lambda - 1)}{\lambda(\beta_r + \lambda - 1)}} \tag{22a}$$

A form of Eq. (22a) has been previously derived.[4] For example, assuming a distributed flame structure, $A^* = (\alpha_s + E_s/R\bar{T}_s)(1 - T_0/\bar{T}_s)$ and $B^* = 2(1 - H) - J + C_p/(CA^*)$. To evaluate this model, several additional parameters need to be specified. However, assuming the flame description adequately models the actual flame and interface reaction, the effect of parameters such as H, E_s, etc. could be investigated using this approach. Through comparisons with unsteady and steady experimental data the model which most accurately represents the trends exhibited may, in principle, be determined. If several models are considered, this is most easily accomplished by evaluating k^* and r^* (or their equivalents) directly from the proposed steady model without going through the tedious process of deriving a new unsteady expression for each proposed model. These parameters could also be evaluated by numerically solving the steady problem. This would be particularly useful when considering complex gas phase flame structures.

De Luca[19] obtained the following expression for R_q assuming a distributed flame, and no surface absorption ($f_r = 1$):

$$R_q = \frac{\dfrac{\bar{q}E}{\rho\bar{r}_bC\bar{T}_s}\left[1 - \dfrac{(\lambda - 1)}{\beta_r + \lambda - 1}\right]}{\lambda + A/\lambda + \dfrac{E\Lambda^2C_p}{C} - AH + C_p/C - 1} \tag{22b}$$

where $E \equiv \alpha_s + E_s/(R\bar{T}_s)$, $A \equiv (1 - T_0/\bar{T}_s)E$, and $\Lambda^2 \equiv (Q_fW_dk_g)/[(\rho\bar{r}_b)^2\bar{T}_sC_p{}^2)]$. It can be shown from a steady energy balance extending from $s+$ to $-\infty$ and using the relation $k_g(\mathrm{d}T/\mathrm{d}x)_{s+} = Q_fk_gW_d/(\rho\bar{r}_bC_p)$ that without a radiant flux ($q = 0$), $(C_p/C)E\Lambda^2 = A(1 - H)$. Similarly, with a nonvanishing radiant flux ($q > 0$), $(C_p/C)E^*\Lambda^2 = A^*(1 - H) - JA^*$ (the * indicates the parameter is evaluated with external radiant heat flux present). In De Luca's analysis[19] of the relationship of R_q with R_p, it was implicitly assumed that the mean flux was small so that these types of effects were not considered, in addition to assuming that $A = A^*$ and $B = B^*$, as discussed at the beginning of this section. If the mean flux is assumed small enough so these types of effects are small and the other parameters are not changed significantly, the results obtained by De Luca are valid. However, as shown below, these effects of the mean radiant flux on R_q can be important in the relationship between R_q and R_p.

Relationship of R_q to R_p

There is substantial interest in relating measurements of R_q to R_p. To accomplish this a transfer function is defined as TF

$\equiv R_p/R_q$. Performing the complex multiplication of TF with R_q will result in the desired R_p. Typically, the case of no external radiant flux, $J = 0$, is of interest for R_p. Therefore, from Eqs. (3) and (19) the transfer function is obtained:

$$TF \equiv \frac{R_p}{R_q} \equiv \frac{\Delta\psi}{\Delta\eta} = \frac{\nu + \delta(\lambda - 1)}{\nu_q + \delta_q(\lambda - 1) - \dfrac{k^* f_r J(\lambda - 1)}{\beta_r + \lambda - 1}}$$

$$\cdot \left\{ \frac{1 + \left[r^* - \dfrac{k^*(\beta_r + \lambda - 1 + f_r J)}{\lambda(\beta_r + \lambda - 1)} \right] (\lambda - 1)}{1 + (r - k/\lambda)(\lambda - 1)} \right\} \tag{23}$$

Assuming that $X_r << X_R(f_r \rightarrow 0)$ and assuming $\delta = \delta_q = 0$

$$TF = \frac{\nu}{\nu_q} \left\{ \frac{1 + [r^* - (k^*/\lambda)](\lambda - 1)}{1 + [r - (k/\lambda)](\lambda - 1)} \right\} \tag{24}$$

If it is now further assumed that $r^* = r$ and $k^* = k$, as may be approximately true for small J, the transfer function is

$$TF = (\nu/\nu_q) \tag{25}$$

and further, if the equivalence principle holds

$$TF = (\nu/k^*J) \tag{26}$$

If the conditions for Eqs. (25) and (26) are met, then R_q and R_p are related at every frequency by a single scaling factor. From these results, it is seen that ideally to obtain R_p from R_q measurements it is desirable to perform the experiments at the same pressure as the desired $R_p(k^* \sim k,\ r^* \sim r)$, use a small mean radiant flux (if possible, such that $k^* \sim k$, $r^* \sim r$), and have a high absorption coefficient (if possible, such that $f_r \rightarrow 0$).

Parametric Effects of Primary Parameters

Equation (21) is evaluated to investigate the effects of the primary parameters on R_q. A baseline condition based on Zenin and Nefedova's data[21] for double-base propellant at 2.03 MPa (20 atm) is chosen as: $T_0 = 273$ K, $\bar{q} = 2$ cal/cm²-s ($J = 0.05$), $k^* = 1.34$, $r^* = 0.43$, $f_r = 0.74$, $\beta_r = 1.73$, $\delta_q = 0$. The equivalence principle was applied to obtain the radiation augmented burning rate. $K_a = 500$ cm^{-1} was assumed and using the burning rate and surface temperature ($\bar{r}_b = 0.29$ cm/s, $\bar{T}_s = 583$ K) corresponding to $T_0 = 273$ K and $\bar{q} = 2$ cal/cm²-s for propellant N, f_r, and β_r are estimated.

Figure 2 shows both the phase and magnitude of R_q as a function of dimensionless frequency. The dimensionless sensitivity of the burning rate to initial temperature in the presence of a radiant flux (a dimensionless "temperature sensitivity") k^* is varied ±20%. Because of experimental difficulties, directly measured values for k^* could involve uncertainties of 20% or more. It can be seen that at $\Omega = 0$ the phase is zero as it must be. When a resonance peak is present a phase lead will occur followed by a lag approaching 45 deg at high frequencies. As k^* is increased from the baseline condition (holding all other parameters fixed), the maximum and minimum of the phase increase. The magnitude of R_q is also seen to increase with k^*. Increasing k^* tends to increase the resonance frequency. If k^* were decreased sufficiently, no resonance peak would be evidenced. The peak magnitude would then be at $\Omega = 0$ and the phase would never be positive. This condition corresponds to an overdamped dynamical system.

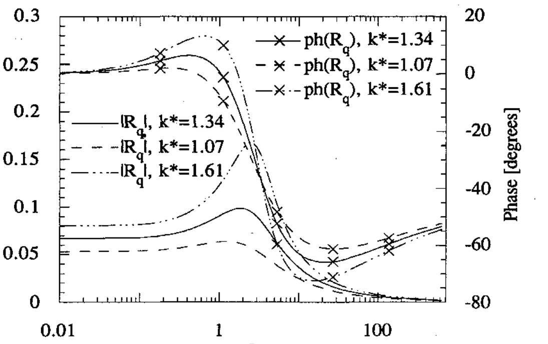

Fig. 2 Effect of k^* on R_q (k^* is varied ±20% from $\bar{q} = 2$ cal/cm²-s, $T_0 = 273$ K, $J = 0.05$, $r^* = 0.43$, $k^* = 1.34$, $\beta_r = 1.73$, $f_r = 0.74$). Parameters correspond to Ref. 21.

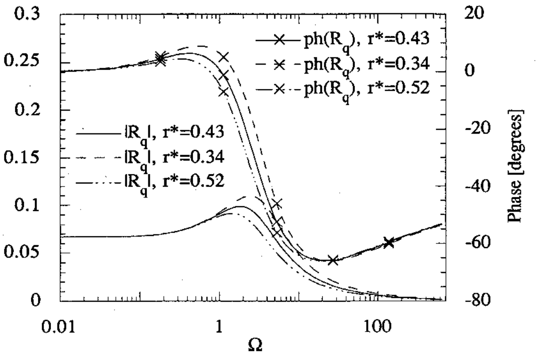

Fig. 3 Effect of r^* on R_q (r^* is varied ±20% from $\bar{q} = 2$ cal/cm²-s, $T_0 = 273$ K, $J = 0.05$, $r^* = 0.43$, $k^* = 1.34$, $\beta_r = 1.73$, $f_r = 0.74$). Parameters correspond to Ref. 21.

Similarly, the parametric effect of varying the dimensionless sensitivity of the surface temperature to initial temperature in the presence of a radiant flux r^* is shown in Fig. 3. The parameter r^* is also varied ±20% from the base case. With increasing r^*, the magnitude of R_q is seen to decrease and the value of the maximum phase lead decreases. Also, the resonance peak frequency is seen to increase with decreasing r^*. The trends presented in Figs. 2 and 3 are similar to those of R_p. One reason for presenting these results to illustrate the sensitivity of R_q to k^* and r^* parameters. This sensitivity can make the comparison with unsteady measurements based on measured steady burning parameters difficult due to the uncertainty of the steady measurements. This raises the possibility, however, that this sensitivity may be exploited by using unsteady measurements of dynamic burning rate to deduce k^* and r^* and, thereby, reduce the uncertainty of these important parameters. Also, these parametric calculations are presented because realistic radiant flux levels can significantly change k^* and r^* parameters. Therefore, the level of the mean flux can be expected to greatly affect R_q by changing the k^* and r^* parameters. Pressure can also affect R_q through k^* and r^* parameters since these parameters are typically dependent on pressure.

In addition to the implicit dependence through r^* and k^*, there is also an explicit dependence of the response function on the dimensionless radiant flux J in Eq. (19). For surface absorption ($f_r = 0$), this explicit dependence vanishes and the mean radiant flux only affects R_q through changes in the other parameters (k^*, r^*, ν_q, δ_q). Experimentally, it is impossible to independently change J without changing the other parameters. Therefore, Fig. 4 shows the effect of increasing the mean flux based on Zenin and Nefedova's steady burning data[21] and includes both explicit and implicit effects of J. The magnitude of R_q at $\Omega = 0$ increases with J since in the case of the equivalence principle holding, $\nu_q = k^*J$. The magnitude of R_q at other frequencies also increases and the maximum phase lead increases with increasing J. This change in R_q is due to both the explicit effect of J and the change in k^*, r^*, β_r, and f_r with $\bar{q}$. Experimental measurements by Finlinson et al.[4] support the conclusion that the mean flux can significantly affect R_q. Recently, measurements have shown that the natural radiant feedback in metallized composite propellants can be significant.[22] Equation (20) shows that with a constant external radiant flux R_p is also affected by an explicit effect of J and by implicit changes in the other parameters due to the radiant flux. Thus, R_p of metallized propellants may be affected by the natural radiant feedback in some conditions; however, the analysis in this article does not include the coupling of the natural feedback with the oscillating pressure or the heterogeneous nature of composite propellants which may also affect R_p.

Figure 4 illustrates differences that can occur in dynamic response between raising the initial temperature vs applying an equivalent mean radiant flux. Case 1 is the baseline condition. Cases 2 and 3 are variations obtained by increasing $\bar{q}$ and T_0, respectively. In cases 2 and 3, $T_0 + \bar{q}/\rho\bar{r}_b C = 334$ K is specified which gives the same steady burning rate since the equivalence principle is assumed to hold. The resulting parameters using calculations based on Zenin and Nefedova's data[21] are shown in the caption. J, k^*, and r^* are seen to differ significantly between the two cases considered. This results in significant differences in the responses for the two cases, as shown in Fig. 4. This effect has also been observed experimentally. For example, Simonenko et al.[2] observed distinct differences in the response between experiments that used either an elevated initial temperature or a higher mean radiant flux primarily to achieve the same burning rate. Simonenko et al.[2] attributed the observed differences to nonequivalence effects; however, the analysis presented in this article shows that even if the equivalence principle holds in the steady state, R_q can significantly differ depending on the specific values of T_0 and $\bar{q}$. Since T_0 and $\bar{q}$ values were not reported by Simonenko et al.[2] a detailed comparison could not be made.

Figure 5 shows the effect of the absorption coefficient (K_a) of the solid on R_q. As X_r decreases (increasing K_a), $|R_q|$ can be expected to increase, as shown in Fig. 5. It can also be seen that the maximum phase lead increases and the maximum phase lag decreases with decreasing X_r for the cases considered. Note that even with a very high absorption coefficient of 1000 cm^{-1} ($X_r = 10$ μm), there is a difference observed in the predicted R_q from the case of surface absorption ($X_r = 0$). This is because even though the characteristic scale of absorption is very small, the thermal and reaction layers are also very thin. As an additional example, if properties more typical of ammonium perchlorate (AP) are assumed: $\bar{r}_b = 0.1$ cm/s, $\alpha_c = 0.001$ cm^2/s, $\bar{T}_s = 850$ K, $E_s = 20$ kcal/mole, then $X_c = 100$ μm and $X_R = 8.4$ μm. For $K_a = 1000$ cm^{-1} (a value estimated from measured optical properties[23] of AP at 10.6 μm), it follows that $\beta_r = 10$ and $f_r = 0.43$. At even higher burning rates β_r will decrease and f_r will increase further, making the assumption of surface absorption even less justifiable. Adding opacifier powders such as carbon black will increase the absorption coefficient in the spaces between AP particles; however, absorption coefficients of 1000 cm^{-1} or higher are difficult to obtain with the addition of small amounts of opacifier. In composite propellants it should be noted that only the opacity of the binder is increased by the addition of an opacifier. Thus, the effect of adding an opacifier to a composite propellant should be interpreted accordingly. Ideally, the absorption coefficient of the binder and oxidizer should be matched to avoid selective absorption of the radiation in one of the components. The limiting case of surface absorption appears to be the exception rather than the rule.

Even in the limiting case of surface absorption, the transfer function TF can be affected by parameter changes due to the mean radiative flux. To illustrate this, two cases are considered in Fig. 6. The first case (solid lines) shows TF for the ideal case of Eq. (26) where R_q and R_p are related by a constant scaling factor (ν/k^*J). The phase difference between R_p and R_q is then zero. The second case also assumes surface absorption, but due to the effect of the mean flux, r^* and k^* are assumed to increase by an amount corresponding to the case based on Zenin and Nefedova's steady data[21] of $\bar{q} = 10$ cal/cm^2-s. Figure 6 shows that significant changes in the TF can then occur. If the change in TF due to the changes in k^* and r^* with the mean level of the radiant flux were not appropriately accounted for, the resulting $|R_p|$ obtained would have errors due to the transformation of about 10% and phase errors as high as 8 deg for the case considered (see Fig. 6). These effects will vary with propellant formulation, pressure, and mean radiant flux level. Under certain conditions these effects may be small, but in general, they should be included if experimental results are to be properly interpreted. If the effect of in-depth absorption were included, the TF could have differed from the ideal case of Eq. (26) even more significantly.

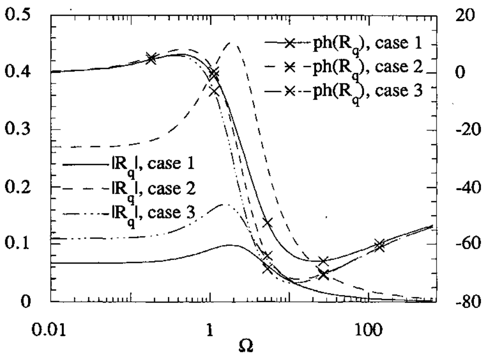

Fig. 4 Effect of T_0 and J on R_q. Case 1: $\bar{q} = 2$ cal/cm^2-s, $T_0 = 273$ K, $J = 0.05$, $r^* = 0.43$, $k^* = 1.34$, $\beta_r = 1.73$, $f_r = 0.74$; Case 2: $\bar{q} = 10$ cal/cm^2-s, $T_0 = 273$ K, $J = 0.18$, $r^* = 0.48$, $k^* = 1.50$, $\beta_r = 1.36$, $f_r = 0.78$; Case 3: $\bar{q} = 2$ cal/cm^2-s, $T_0 = 321$ K, $J = 0.07$, $r^* = 0.62$, $k^* = 1.64$, $\beta_r = 1.36$, $f_r = 0.78$. Parameters correspond to Ref. 21.

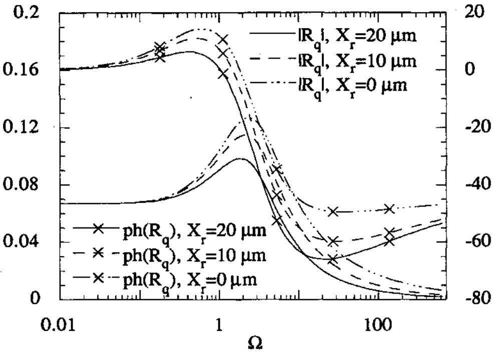

Fig. 5 Effect of X_r on R_q. $X_r = 20$ μm: $\bar{q} = 2$ cal/cm^2-s, $T_0 = 273$ K, $J = 0.05$, $r^* = 0.43$, $k^* = 1.34$; $X_r = 10$ μm: $\bar{q} = 2$ cal/cm^2-s, $T_0 = 273$ K, $J = 0.05$, $r^* = 0.43$, $k^* = 1.34$; $X_r = 0$ μm: $\bar{q} = 2$ cal/cm^2-s, $T_0 = 273$ K, $J = 0.05$, $r^* = 0.43$, $k^* = 1.34$. Parameters correspond to Ref. 21.

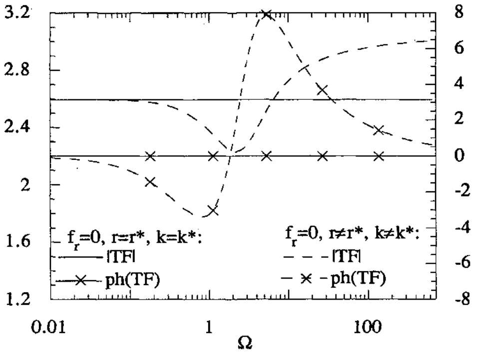

Fig. 6 TF for the case of $f_r = 0$ (surface absorption) with $k = 1.28$ and $r = 0.41$ for the $\bar{q} = 0$ case and $k^* = 1.50$ and $r^* = 0.48$ corresponding to $\bar{q} = 10$ cal/cm^2-s (except for the case where $r = r^*$, $k = k^*$ is assumed). Parameters correspond to Ref. 21.

Linear Pulse Response

A Fourier series can be obtained for any practical periodic dimensionless radiant flux ψ, and R_q can be applied to this expression resulting in the dimensionless burning rate. For example, consider a series of periodic radiant heat flux pulses of width $2c$

$$\psi(\tau) = 1 + \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin(\Omega_p nc)\cos(\Omega_p n\tau) \tag{27}$$

where Ω_p is the dimensionless frequency of the pulse. Since the Fourier series decomposes the signal into discrete frequencies, R_q can be applied to Eq. (27) to obtain

$$v(\tau) = 1 + \sum_{n=1}^{\infty} |R_q(\Omega_p n)| \frac{(-1)^n}{n} \sin(\Omega_p nc)\cos\{\Omega_p n\tau - ph[R_q(\Omega_p n)]\} \tag{28}$$

Equation (28) is the linear burning rate response to a series of pulses. This analysis suggests an alternative approach for obtaining R_q experimentally. R_q can be measured by pulsing a sample instead of applying a sinusoidal waveform. In this way R_q at several harmonics can be determined during a single test which reduces testing time. Of course, as with sinusoidal experiments, one should carefully verify that the response is linear by repeating tests at different amplitudes. Also, at higher frequencies the amplitude of the input harmonic may be too small to produce a measurable output (low S/N). In this case, the pulsing of the radiative source may be modified so it is composed of a higher amplitude harmonic at those higher frequencies of interest so a measurable output signal will result. This pulsing approach would be particularly useful if a radiative source is used which cannot operate in a continuous mode. Figure 7 shows the linear responses to two series of pulses. Equation (28) was evaluated with enough terms such that the effect of additional terms would have been negligible. The parameters used are again based on Zenin and Nefedova's steady data.[21] Qualitatively similar results were obtained experimentally by Son et al.[31]

Comparison of the Linearized Result with Nonlinear Calculations

As the burning rate and corresponding temperature oscillations become large, the higher-order terms due to the convective term that were neglected in the linearized analysis become significant. The equations were numerically integrated in order to compare the linear calculations to nonlinear solutions. Assuming a steady burning relationship of $\bar{r}_b = b/(T_e - T_0)^w$ and considering the pressure constant, Eq. (5) (in dimensionless form) becomes

$$\phi_s \equiv \left(\frac{d\Theta}{d\xi}\right)_{s-} = (1 - f_r)J + v[\theta_s + \theta_e(v^{-1/w} - 1) - Jv^{-1/w}] \tag{29}$$

where a zeroth-order Arrhenius relationship can be used to relate the surface temperature to burning rate to eliminate θ_s. Basing parameters on Zenin and Nefedova's steady data[21] with $\bar{q} = 2$ and 10 cal/cm²-s, the nonlinear burning response to a sinusoidal radiant flux at $\Omega = 2$ is considered. For more details concerning the numerical integration see Ref. 31. In Fig. 8, the dimensionless burning rate is normalized so that if the response is linear it will vary from between 1 and -1. For the cases of a) a large mean flux, small oscillatory amplitude ($J = 0.18$, $\Delta\psi = 0.01$), and b) a small mean flux, large oscillatory amplitude ($J = 0.05$, $\Delta\psi = 1.0$) the magnitude and phase [-23 deg for a) and -18 deg for b)] of both of the responses match the linearized results well. There is about 2% difference in the amplitude of case b), and case a) is within 0.5% of the linearized result. Note that even if $\Delta q \sim \bar{q}$ [case b)], a linear burning response can result if $\bar{q}$ is sufficiently small so the burning rate oscillations are small, as Fig. 8 shows. In other words, for the burning rate oscillations to be small does not necessarily require $\Delta q << \bar{q}$ or $\Delta p << \bar{p}$, only that their respective effects produce small burning rate oscillations with respect to the mean burning rate. The other two cases, both with large mean flux and large oscillatory amplitude, show clear evidence of nonlinear behavior. The shapes are more triangular and the amplitudes differ from the linear values. Of the two, the surface absorption case ($f_r = 0$), appears to be more in phase with the driving signal. The linearized analysis also predicts that this case should be more in phase. Linearity should always be verified experimentally by varying the amplitude of the driving radiant heat flux if comparisons are to be made with linear theory.

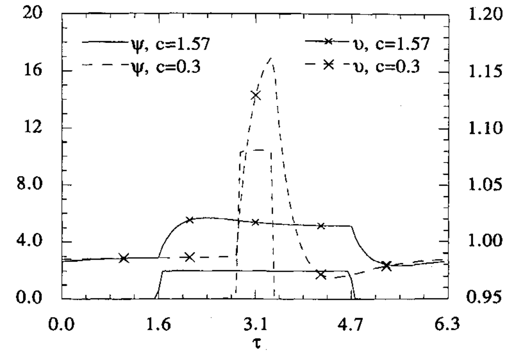

Fig. 7 **Linear response to a series of radiant flux pulses with $\bar{q}$ = 0.4 cal/cm²-s, Ω = 3.0, T_0 = 273 K, J = 0.01, r^* = 0.42, k^* = 1.30, β_r = 1.82, f_r = 0.73. Parameters correspond to Ref. 21.**

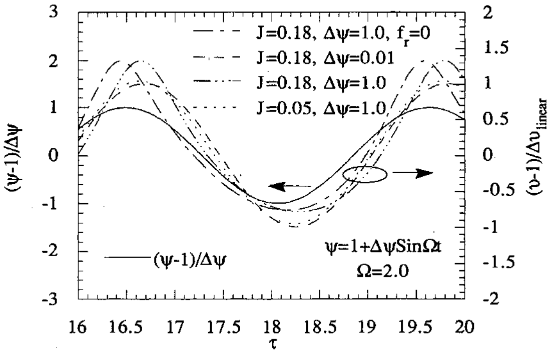

Fig. 8 **Nonlinear response to a sinusoidal radiant flux. Parameters correspond to Ref. 21.**

Conclusions

The results of a linearized analysis of the unsteady burning of energetic solids provides a useful first approximation because of the simplicity of the resulting expression. In this article an expression for burning rate response to an external radiant flux as a function of primary experimental parameters has been presented. The phenomenological approach (ZN) was used to obtain the linear burning response of solids subjected to radiant heat flux oscillations. The advantage of taking this approach is that the resulting expression is more general than if additional modeling approximations of the gas phase flame structure and interface had been made. The ZN expression was compared to the corresponding result based on the FM approach. The form of the two expressions was found to be the same. The linearized effects of the mean heat flux level and in-depth absorption on R_q and the relationship between R_q and R_p were presented. In-depth absorption can play a significant role in the burning rate response to a radiant heat flux. Due to in-depth absorption and the effect of the mean flux, it was shown that in general the relationship between R_p and R_q is somewhat more complicated than a constant scaling factor. Even for surface absorption, a constant scaling factor may be inadequate. From these results, it is seen that ideally to obtain R_p from R_q measurements, it is desirable to perform the experiments at the same pressure as the desired R_p ($k^* \sim k$, $r^* \sim r$), use a small mean radiant flux (if possible, such that $k^* \sim k$, $r^* \sim r$), and have a high absorption coefficient (if possible, such that $f_r \rightarrow 0$). It appears, however, that the limiting case of surface absorption appears to be the exception rather than the rule for typically used wavelength-materal combinations, and the effect of in-depth absorption should be included. It was also shown that the classical ZN result for R_p can be written in the same form as the "*AB*" expression from FM analyses. This is viewed as a further extension of the idea presented in the classic work by Culick[17] and it clarifies the relationship between the ZN and FM approaches. This connection between the ZN and FM approaches has been addressed many times in the open literature (e.g., 11–14); yet a full understanding of this issue is still not common. The linear burning rate response to a series of radiant heat flux pulses was presented as an alternate method of experimentally measuring R_q instead of using sinusoidal oscillations. This approach is especially useful if a pulsed radiant source is used and it could decrease testing time. The effects of large amplitude oscillations of the radiant flux were investigated by conducting a few nonlinear numerical calculations.

Appendix A: Steady State Analysis

The dimensionless steady state temperature profile in the condensed phase, with an external radiant flux, is

$$\bar{\theta}(\xi) = [1 - (f_r J/1 - \beta_r)]\exp(\xi) + (f_r J/1 - \beta_r)\exp(\beta_r \xi) \tag{A1}$$

For sufficiently opaque propellant, all the radiant energy is absorbed near the surface reaction zone ($f_r = 0$) and, similar to the case without an external radiant flux, the Michelson temperature profile (exponential) is obtained in the condensed phase.

Experimentally it is observed in steadily burning solids that changing the initial temperature, pressure, and radiant flux will affect the values of the burning rate and surface temperatures. That is, $\bar{r}_b = \bar{r}_b(\bar{p}, \bar{q}, T_0)$ and $\bar{T} = \bar{T}_s(\bar{p}, \bar{q}, T_0)$. From these relationships the following dimensionless parameters are defined in the nomenclature: k^*, r^*, ν^*, ν_q, u^*, μ_q. In the steady state, with $q = 0$, the burning rate and surface temperature commonly have the dependencies $\bar{r}_b = \bar{r}_b(\bar{p}, T_0)$ and $\bar{T}_s = \bar{T}_s(\bar{p}, T_0)$ with the corresponding primary dimensionless parameters: k, r, ν, μ. Inverting the former expression gives $T_0 = T_0(\bar{p}, \bar{r}_b)$ and it then follows that $\bar{T}_s = \bar{T}_s(\bar{p}, \bar{r}_b)$.

From a surface energy balance for the case of no external radiant flux

$$\left(\frac{\mathrm{d}\bar{T}}{\mathrm{d}x}\right)_{s-} = \left[\rho\bar{r}_b Q_s + k_g\left(\frac{\mathrm{d}\bar{T}}{\mathrm{d}x}\right)_{s+}\right] \Big/ k_c \tag{A2}$$

or

$$\bar{f}_s = \bar{f}_g \tag{A3}$$

where $\bar{f}_s$ is the gradient on the condensed side of the surface and $\bar{f}_s$ is defined as the right side of Eq. (A2). From an energy balance extending from s- to $x \rightarrow -\infty$ it follows that

$$\bar{f}_s = (\bar{r}_b/\alpha_c)(\bar{T}_s - T_0) \tag{A4}$$

From the discussion preceding Eq. (A2) it was shown that $\bar{T}_s$ and T_0 are functionally dependent on the pressure and burning rate; therefore it follows from Eq. (A4) that $\bar{f}_s$ is a function of $\bar{r}_b$ and $\bar{p}$. That is

$$\bar{f}_s = \bar{f}_s(\bar{p}, \bar{r}_b) = (\bar{r}/\alpha_c)[\bar{T}_s(\bar{p}, \bar{r}_b) - T_0(\bar{p}, \bar{r}_b)] \tag{A5}$$

This is a very useful result. Given experimental data of $\bar{r}_b = \bar{r}_b(\bar{p}, T_0)$ and $\bar{T}_s = \bar{T}_s(\bar{p}, T_0)$, the gradient condition is a function of only pressure and burning rate. Or, inverting this expression, the burning rate is a function of the gradient and pressure $[(\bar{r}_b = \bar{r}_b(\bar{f}_s, \bar{p})]$. Thus, the experimentally observed $\bar{r}_b = \bar{r}_b(\bar{p}, T_0)$ can be transformed to $\bar{r}_b = \bar{r}_b(\bar{p}, \bar{f}_s)$ using Eq. (A4). The reason for using this transformation is that the latter expression $[\bar{r}_b(\bar{p}, \bar{f}_s)]$ holds in the unsteady case within the quasisteady approximation [that is, $r_b = r_b(p, f_s)$]; whereas, the former does not $[r_b \neq r_b(p, T_0)]$.[9,16]

In the steady burning case with $q = 0$, $\bar{r}_b$ is changed by changing $\bar{p}$ or T_0. However, $\bar{r}_b$ could also be changed by the thermal effect of an external radiant heat flux. Likewise, in the unsteady case the instantaneous burning rate will vary due to the thermally relaxing condensed phase. In both of these cases, assuming no absorption of the radiation in the gas phase reaction zone, the functionality $r_b = r_b(p, f_s)$ remains unchanged. In the analysis presented in this article, the transmission loss τ_λ is the attenuation of the external radiant flux through the combustion products above the gas phase reaction zone. It is unlikely that the small amount of radiative energy that is absorbed in the gas pahse reaction zone near the surface at commonly considered wavelengths has any significant effect on the conductive feedback to the condensed phase. In the unsteady form of Eq. (A5), $T_0(p, r_b)$, is often called an apparent initial temperature. It is equal to the actual initial temperature only in the steady case with $q = 0$. This is seen from an energy balance from $s-$ to $x = -\infty$ in the unsteady case with $q = 0$

$$f_s = \frac{r_b}{\alpha_c}\left[t_s(p, r_b) - T_0 + \frac{1}{r_b}\frac{\partial}{\partial t}\int_{-\infty}^{0} T\,\mathrm{d}x\right]$$

$$= \frac{r_b}{\alpha_c}[T_s(p, r_b) - T_0(p, r_b)] = f_g(p, r_b) \tag{A6}$$

It is also worthwhile to examine the differential form of the gas phase energy equation. The following simplifying assumptions are also often made in formulating the FM quasisteady gas phase[24]: 1) the gas phase density is spatially uniform; 2) all gas phase specific heats are equal; 3) Fick's law of diffusion is valid and all diffusivities are equal ($D_i = D$), and 4) gas phase reactions are represented by a single-step forward reaction. Applying these assumptions, the steady form of the gas phase energy equation for a single-step reaction becomes

$$k_g\frac{\mathrm{d}^2\bar{T}}{\mathrm{d}x^2} - \rho\bar{r}_b C_p\frac{\mathrm{d}\bar{T}}{\mathrm{d}x} + \rho_g Q_f \dot{\varepsilon} = 0 \tag{A7}$$

Integrating once between $x \rightarrow \infty$ (far downstream in the gas phase) and $x = 0$ (the propellant surface), it can be shown that

$$k_g\left(\frac{\mathrm{d}\bar{T}}{\mathrm{d}x}\right)_{s+} = \int_0^\infty \rho_g Q_f \dot{\varepsilon}\exp\left(-\frac{\rho\bar{r}_b C_p x}{k_g}\right)\mathrm{d}x \tag{A8}$$

Using this expression, Eq. (A2) can be written as

$$\bar{f}_g = \left[\rho\bar{r}_b Q_s + \int_0^\infty \rho_g Q_f \dot{\xi}\exp\left(-\frac{\rho\bar{r}_b C_p x}{k_g}\right)\mathrm{d}x\right] \Big/ k_c \tag{A9}$$

To simplify this further, the nature of the species generation term $\dot{\varepsilon}$ must be specified. Typically this approach results in an expression for $\bar{f}_g$ which is a function of only pressure and burning rate as variables along with various physical parameters, in agreement with Eq. (A6). This approach has the advantage that the individual effect of various gas phase and interface parameters can be isolated.

Table A1 Steady burning data for propellant N at 2.03 MPa (20 atm) from Ref. 21 and curve fits

Variable	T_0, K 123	173	223	273	323	373	413	$\sigma_{\max}$
r_b, cm/s	0.18	0.19	0.22	0.26	0.35	0.49	0.76	±0.01
r_b, cm/s; c-fit[a]	0.17	0.19	0.22	0.27	0.34	0.49	0.76	
k (from c-fit)	0.97	1.02	1.08	1.28	1.65	2.24	3.51	
T_s, K	523	533	543	573	613	633	653	±20
T_s, K; from c-fit[a]	534	544	556	572	602	641	698	
r (from c-fit)	0.20	0.24	0.29	0.41	0.63	1.01	1.76	

[a] $b = 22.5951$, $T_e = 470.1708$, $w = 0.8382$, $A = 98.7$ cm/s, $E_s/R = 3400$ K.

Thermal Effect of an External Radiant Flux

The equivalence principle is expressed by the following relation[27]:

$$T_0 + \frac{(1 - r_\lambda)\tau_\lambda \bar{q}_r}{\rho C \bar{r}_b} \equiv T_0^* = T_0(\bar{p}, \bar{r}_b) \qquad \text{(A10)}$$

Given the functional dependence of initial temperature on pressure and burning rate without an external radiant heat flux, $T_0(\bar{p}, \bar{r}_b)$, the equivalence principle can be used to predict the radiation augmented steady burning rate. The equivalence principle is rigorously applicable whenever the amount of radiation absorbed within and above the zone controlling the burning rate is negligible.[27]

Experimental Data

Only a few studies have been made of the burning rate as a function of both initial temperature and radiant flux. Figure A1 shows the effect of initial temperature on the burning rate for two noncatalyzed double-base propellants. The data for the Princeton propellant 1041 obtained by Caveny et al.[25] was originally analyzed assuming a constant temperature sensitivity. The data obtained by Konev[26] using propellant N was obtained over a much broader range of initial temperatures, and the temperature sensitivity (σ_p) exhibits a strong dependency on initial temperature. This is in agreement with other studies which have shown a similar dependence on initial temperature.[21,28] Both data sets have been fit according to $\bar{r}_b = b/(T_e - T_0)^w$ (b, T_e, and w are fitting parameters) which has been previously applied to such propellants (cf. Ref. 27). Because of the narrow range of initial temperatures considered by Caveny et al. a dependence of σ_p on T_0 cannot be clearly established or ruled out. Using the above empirical expression, the temperature sensitivity is then $\sigma_p = w/(T_e - T_0)$. The range of variation of the temperature sensitivity appears to be higher for propellant N than propellant 1041. The reasons for these differences may be the different operating pressures and propellant compositions. From other data[28] it appears that as pressure increases, σ_p becomes less sensitive to the initial temperature. In contrast, one study reported that σ_p decreased with initial temperature for NC/NG cast propellant.[29] More recently, Kubota and Ishihara[30] studied two double-base propellant formulations. They found that the higher energy propellant had a lower σ_p. Three initial temperatures over a relatively narrow temperature range from −30 to 70°C were considered. Consequently, the dependence of σ_p on initial temperature could not be clearly established.

If the equivalence principle is applied according to Eq. (A10) and the empirical expression above, the required absorbed radiant flux to produce a given $\bar{r}_b$ is $\bar{q} = \rho C \bar{r}_b[T_e - T_0 - (b/\bar{r}_b)^{1/w}]$. Figure A2 shows that the results of these calculations agree well with the experimental data indicating that in these cases the equivalence principle appears to hold. Assuming a constant σ_p, the original analysis significantly underpredicted the radiant flux data.[25] Fig. A2 also shows the corresponding values for ν_q.

Data were not reported of the corresponding surface temperature by Caveny et al.[25] or Konev.[26] However, Zenin and Nefedova[21] have measured burning rate and surface temperatures of N propellant over a very broad range of parameters. Table A1 shows the effect of initial temperature on the steady burning parameters from Zenin and Nefedova's data. The k and r parameters are seen to be sensitive to initial temperature and consequently to the mean radiant flux also. This has significance in interpreting the unsteady experimental results and in relating the measured R_q to R_p as discussed in connection with Figs. 3, 4, and 6.

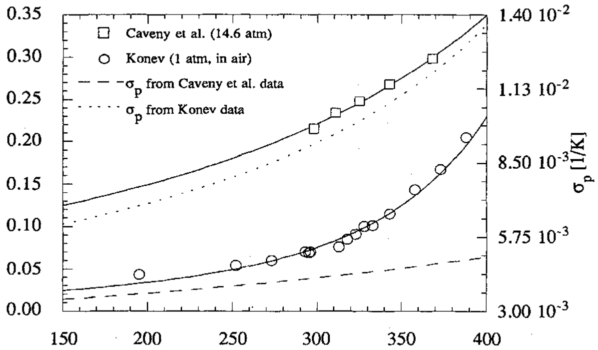

Fig. A1 Burning rate dependence on T_0 for double-base propellant at $\bar{p}$ = 101 kPa (1 atm) (Konev[23]) and 1.48 MPa (14.6 atm) (Caveny et al.[22]).

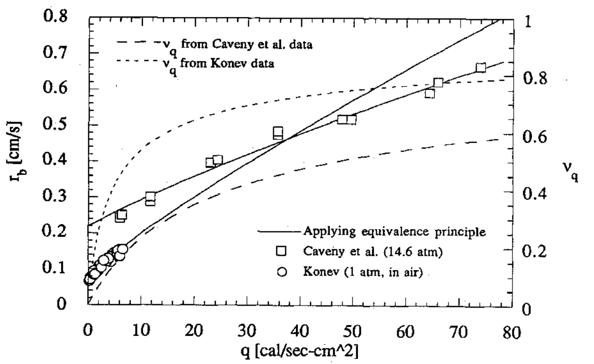

Fig. A2 Measured radiation-augmented burning rate for double base propellant at $\bar{p}$ = 101 kPa (1 atm) (Konev[23]) and 1.48 MPa (14.6 atm) (Caveny et al.[22]).

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