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Response Function Theories That Account for Size Distribution Effects—A Review

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Nomenclature

A,A',B,B',C = constants appearing in response function expressions
c_g = specific heat of gas
c_s = specific heat of solid propellant
D_i = ith particle diameter in a multimodal distribution
D_1-D_4 = constants appearing in Zeldovich response function expression
E_1-E_4 = constants appearing in Cohen-Bowyer response function expression
E_g = activation energy of gas-phase reactions
E_s = activation energy of surface decomposition
f = frequency of oscillations
f_i = peak response frequency, or preferred frequency, for the ith particle size
$\bar{m},m'$ = mean and fluctuating components of propellant mass flux
n = burning rate pressure exponent
p = pressure
$\bar{p},p'$ = mean and fluctuating components of pressure
R = universal gas constant
R_b = pressure-coupled response function
r = propellant burning rate
T_F = flame temperature
T_o = initial propellant temperature
T_w = propellant surface temperature
α_i = weight fraction of ith particle size
κ = propellant thermal diffusivity
λ,λ' = complex variables defined in text
Ω = dimensionless frequency of oscillations
σ_p = burning rate temperature sensitivity

Introduction

THE tendency of solid propellants to drive pressure oscillations in a combustor is characterized by their response functions. The pressure-coupled response function, which is the subject of this paper, is defined as:

$$R_b = (m'/\bar{m})/(p'/\bar{p}) \tag{1}$$

It is basically the burning rate response to imposed pressure perturbations, a propellant property that appears in the stability analysis of motors.[1] It is a useful quantity for comparing propellants, as well as motor analysis. A number of laboratory devices have been developed for response function measurement, the most notable being the T-burner.[2] This paper deals with theoretical developments.

Motor firing experience and data acquired in a variety of laboratory burners have established that the combustion instability of composite propellants is significantly affected by ammonium perchlorate particle size distribution.[3-13] Classical theories of the response function, which were reviewed by Culick,[14] are unable to explain these particle size effects because they assume that the solid propellant is a homogeneous medium and the response mechanism is essentially the thermal lag in the solid phase. Another phenomenon is being revealed by recent experimental work with devices that have improved the capability to conveniently acquire response function data at many oscillatory frequencies.[15,16] Results suggest that much of what has been thought to be data scatter may be manifestations of multipeaked response function curves. Classical theory predicts a single-peaked response function curve. Another relevant phenomenon may be the detection of pressure, temperature, and compositional fluctuations in devices burning composite propellants under steady-state conditions[17-19]; the frequencies of the pressure fluctuations were associated with AP particle sizes, and a strong coupling was noted when the propellant was made to burn unstably at such a frequency. Comprehensive theories of steady-state burning are based on statistical treatment of the composite propellant heterogeneity.[20] Therefore, it appears necessary to consider the heterogeneity of composite propellants in formulating a model of unsteady burning.

Since the time of the Culick review, there have been a number of attempts to address the heterogeneity of composite propellants in developing a model of the pressure-coupled combustion response function. The purpose of this paper is to review those models. It can be stated at the outset that none have successfully resolved the problem. Therefore, the field is fertile for continued work and it is hoped that this paper will stimulate ideas and further effort. It is necessary to resort to considerable experimentation to measure the combustion response function in the course of research and development programs. An understanding of the important mechanisms would serve to guide propellant development and to minimize the amount of testing required to characterize and evaluate new formulations.

Comments on Theory of Homogeneous Propellants

Theory predicated upon a homogeneous propellant leads to an expression for the response function as follows:

$$R_b = \frac{nAB}{AB-(1+A)+(A/\lambda)+\lambda} \tag{2}$$

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Table 1 Values for model input parameters

E_s	= 22,000 cal/mol
R	= 1.987 cal/mol
T_w	= 850 K
T_o	= 298 K
	($A = 8.46$)
c_g	= 0.4 cal/gm-K
T_F	= 2100 K (for JANNAF standard A-13 propellant)
n	= 0.41 (measured for A-13 at 2 MPa)
E_g	= 15,000 cal/mol
c_s	= 0.3 cal/gm-K
	($B = 1.59$)
κ	= 0.0013 cm²/s
r	= 0.43 cm/s (measured for A-13 at 2 MPa)
	($\Omega = 0.0435f$)

where n is the pressure exponent and, by this theory, the value of R_b at zero frequency. λ is a complex quantity given by:

$$\lambda = \tfrac{1}{2} + \tfrac{1}{2}\sqrt{1+4i\Omega} \qquad [\Omega = 2\pi f(\kappa/r^2)] \tag{3}$$

Expressions for A and B depend upon the particular combustion model used. A representative set is given by[21]

$$A = \frac{E_s}{RT_w}\left(1 - \frac{T_o}{T_w}\right) \tag{4}$$

$$B = \frac{c_g T_F}{[1 + n + (E_g/2RT_F)]c_s(T_w - T_o)} \tag{5}$$

Equation (2) would be adequate for composite propellants if it could be said that particle size is of importance only insofar as it affects burning rate, pressure exponent, and the values of A and B. Unfortunately, application of this expression reveals several problems. A list of values for the parameters is given in Table 1. These values are consistent with usage in steady-state theory for ammonium perchlorate propellants.

Recourse to the paper of Denison and Baum[21] reveals that, for a value of A of 8.46 and a value of B (α in the notation of that paper) of 1.59, the peak response function is 1.5 times the pressure exponent. Thus, the peak response would have a value of 0.6 for A-13 propellant at 2 MPa. Experimentally, the peak response has a value of about 4. The significant underprediction of response function behavior is characteristic of the homogeneous theory, and arises because of the relatively low value of A and high value of B. Indeed, the relatively large B will restrict the results to small peaks regardless of A. Thus, the predicted response function vs frequency curve tends to be flat. More pleasing results can be obtained by other combinations of A and B. For example, $A = 14$ and $B = 0.7$ is a combination that produces a strong peak and has been used to fit data; however, those numbers cannot be justified by this model. An early discussion of this point was published by Boggs and Beckstead.[8]

Other deficiencies are found when attempting to account for changes in pressure and propellant ingredients. Values of A and B are essentially independent of pressure and AP particle size. For example, varying pressure over the range 1.4-5.4 MPa changes A by 0.1% and B by 3%. An increase in solids loading or a reduction in the gas phase activation energy (to represent a catalyst) would tend to increase B. For large values of B, however, a further increase in B would not significantly affect the response function curve.[21] Some studies have indicated that catalysts increase the kinetics prefactors rather than reduce the activation energies, in which case A and B would not change. With A and B essentially constant, the response function vs frequency curve should shift only in accordance with the square of the burning rate, and the peak response magnitude should vary only in proportion to the pressure exponent. These trends are not borne out by data. For example, a study of the effects of pressure, particle size, and ballistic modifiers has been performed with A-13 propellant.[6] Results[2,6] show no consistent relation between peak response magnitude and pressure exponent. Peak response frequency appears to vary more closely with the first power of burn rate, and inversely with particle size. Other data show that reducing particle size tends to increase the response function at higher frequencies, even where burning rate is constant (by changing solids loading or using modifiers).[13] More data are required over a sufficient range of frequencies to reveal the true differences in peak response characteristics with particle size.

The conclusion is that particle size cannot be of importance only insofar as it affects burning rate, pressure exponent, and the values of A and B in the framework of homogeneous propellant theory. There is no basis for any consequential change in A or B with particle size. Given realistic values of A and B, experimental response function curves cannot be reproduced. Moreover, the theoretical and experimental effects of burning rate and pressure exponent are in serious disagreement.

Initial Attempts to Account for the Propellant Heterogeneity

Sideways Sandwich Model

The first attempt to account for the heterogeneity of composite propellants in the framework of modern instability theory was made by Lengelle and Williams.[22] They assumed a "sideways sandwich" model in which properties of the propellant varied sinusoidally with depth. The sinusoidal variation was selected for mathematical convenience. Periodic variations that would not be sinusoidal can be envisioned for the average properties of well-mixed propellants. The spatial periodicity could be related to the particle size, and the timewise periodicity could therefore be the ratio of the particle size to the time-averaged burning rate. It was noted that burning rate could not be a constant in such a model, so the "steady-state" value was taken to be a baseline mean or average over the one cycle.* The inverse of the timewise periodicity was denominated the "preferred frequency" of the propellant. The analysis was otherwise equivalent to the homogeneous propellant theory.

The mechanism is intuitively appealing because the oscillating steady-state burning rate reflects true composite propellant behavior on the local microscopic scale. Indeed, the steady-state models seek to represent a statistical average of this process to calculate a burning rate for the aggregate propellant. Furthermore, the "preferred frequency" provides a mechanism whereby the frequency response to the propellant is not solely dependent upon the square of the burning rate; the particle size and the burning rate of the first power also directly enter into the equation. However, these appealing features of the model were also the seeds for criticism. It is argued that the microscopic process is randomly distributed over the propellant surface, so there would be no coherent phase relationship between the geometric oscillations and the pressure oscillations. Furthermore, there could be no synchronization with pressure oscillations at frequencies other than the preferred frequency. Lengelle and Williams developed an argument for phase synchronization at the preferred frequency through a second-order coupling of the local burning rate and the local phase until an equilibrium

*Two of the current steady-state models employ a similar averaging process. They consider burning from the time that an AP particle is first exposed until the time it burns out, and the rate varies as the particle is consumed.[20]

was everywhere achieved. An adjustment of the surface structure from a plane to a random terrain was implied. On the other hand, Boggs and Beckstead[8] argued that coherence is possible over portions of the propellant surface on a statistical basis. Also, synchronization at other frequencies is possible if it is recognized that real propellants do not contain one single particle size and that the compositional periodicities in the structure are not sinusoidal. These complications were beyond the scope of the analysis. The point has remained controversial.

Lengelle and Williams succeeded in deriving a "heterogeneity response function," for two species of heterogeneity, that would be added to the homogeneity or "conventional response function" to achieve the total response. The two cases selected for illustration were thermal conductivity, representing an effect of the bulk solid, and the activation energy of decomposition, E_s, representing an effect of the boundary. The expressions for these heterogeneity response functions are very complicated, but there are several features worth noting: First, they contain the familiar A and B parameters. Second, they do not contain the pressure exponent. Third, frequency is not tied to the square of the burning rate, rather it is tied to the ratio of burning rate to some dimension in the solid characteristic of the heterogeneity (such as particle size or, perhaps more generally, something relatable to particle size or sizes in real propellants). Thus, the response function is determined by features of a homogeneous propellant plus qualitatively different features of a heterogeneous solid.

The authors presented some theoretical results in parametric form. One important result was that the heterogeneity response can be larger than the conventional response for a given A and B. Thus, calculation of small values from the homogeneous theory need not cause concern. Another important result was that the heterogeneity response can change appreciably with changes in burning rate and particle size. The significance is that the change in total response need not follow the change in pressure exponent. These results could provide a basis for explaining differences between the homogeneous propellant theory and experimental trends. However, there was no attempt to apply the model to propellants or to compare theory with data. Two practical difficulties which arise are: 1) the Lengelle-Williams concept of phase synchronization introduces a quasi-nonlinearity (i.e., an amplitude-dependence); and 2) proper determination of the spectrum of characteristic dimensions of a real propellant would require a Fourier decomposition of the actual heterogeneity. In other words, an evaluation of the theory requires raw T-burner data and details of the particle size distribution. The evaluation would be relatively tedious and has never been performed.

A subsequent paper by Law and Williams[23] applied this model to the special case of L^* instability. There were no comparisons with data, but some additional points were made regarding the properties of this theory. First, a size distribution consisting of j modals should exhibit $j+1$ preferred frequencies, the last being associated with homogeneous propellant theory. Second, it was implied that real propellants might have a more continuous distribution of preferred frequencies; the idealized propellants assumed in theoretical models, if they could be manufactured, would have the potential for stronger response but at discrete preferred frequencies. Third, a preferred frequency could appear apart from the L^* instability frequency, but if the two happened to coincide, the heterogeneity would contribute to driving the L^* instability. Fourth, the L^* instability boundary would be shifted to lower values of L^* because of the coherence requirement assumed by this theory. Perhaps the most important point is that of the compositional structure of real propellants. Functional forms for the heterogeneity could be derived for ideal geometries, but sooner or later it may be necessary to obtain measurements from real propellants.

Perturbed BDP Model

Hamann[24] took a completely different approach and performed a perturbation analysis upon the Beckstead-Derr-Price (BDP) steady-state model. The BDP model[25] contains no in-depth solid-phase heterogeneity, but there are important particle size effects on the propellant surface structure and the modeled multiple flame structure of the gas phase. The surface structure is of importance because it relates the burning of the AP particles to the burning of adjacent binder under the constraint of mass continuity. The multiple flame structure is of importance because it defines the roles of the AP monopropellant flame and the AP/binder diffusion flames in supplying heat to sustain combustion consistent with the conservation of energy. These processes depend upon particle size and pressure. Hamann, therefore, was the first to consider gas-phase and surface structure heterogeneities in combustion response modeling.

The expression for the response function was derived to be in the following form:

$$R_b = \frac{(\lambda - 1)C + n(1 + A' + B')}{B' + (A'/\lambda) + \lambda} \tag{6}$$

Except for one term, Eq. (6) is identical in form to Eq. (2). The constants A' and B' are analogous to A and B but involve far more complicated expressions commensurate with the relative complexity of the BDP model. The term involving C is an addition which stems from the time lag in the propellant surface structure. A similar form can be derived if the surface pyrolysis law is given an explicit pressure dependence.[14] However, the combustion models discussed in this paper and those in current use do not employ such a pyrolysis law. The surface structure analysis in effect creates one, and is a unique feature of BDP-type models.

Hamann did not report any theoretical results. However, Condon, Osborn, and Glick subsequently obtained numerical solutions to this form of problem, as one of their approaches to be discussed later, and the results were not encouraging.[26] From the form of Eq. (6), frequency response will still be tied to the square of the burning rate through λ. It appears to be impossible to avoid that result when adopting a conventional perturbation approach.[14]

Layer Frequency Postulates

Cohen[11] revived the layer frequency concept proposed by Boggs and Beckstead[8] to propose a heuristic, semiempirical method to calculate the response function for any given AP size distribution. It was postulated that the peak response frequency varies inversely with the characteristic time for thermal interaction between the solid-phase thermal wave and a particle situated within. This postulate yields the ratio of the burning rate to the particle size as the measure of the frequency response. The result is like that of the sideways sandwich model, but by a different mechanism. It was also postulated that the peak magnitude varies with the surface-to-volume ratio of the particle, a measure of its thermal response. Experimental response function data for A-13 propellant were then used to calibrate a normalized response function curve to these two characteristic parameters. It was then asserted that this calibrated curve would hold true for any AP composite propellant. The rationale was that the A and B parameters are largely fixed by propellant type, as discussed earlier. Finally, it was asserted that the response function curve for a multimodal propellant would be a weighted summation of the constituent curves. Thus,

$$f_i \sim r/D_i \tag{7}$$

$$R_b/n \sim \sum_i (\alpha_i/D_i) \cdot \text{function}\,(f/f_i)_{\text{A-13}} \tag{8}$$

The simplicity of the method has considerable practical appeal, and it has been used to guide propellant development and reduce the scope of T-burner testing.[27] It does not, however, rest upon a firm analytical footing and may have gone too far in completely ignoring the thermal wave response mechanism of homogeneous propellant theory.

As applied,[27] the method tends to underestimate the contributions of coarse sizes and overestimate the contributions of fine sizes (A-13 contains monomodal 90 μ AP, an intermediate size). Thus, an adjustment to the size dependence in Eq. (8) has been made in current work. On the other hand, Eq. (7) appears to be qualitatively correct in showing trends of peak response frequency. Moreover, the implication that multimodal propellants exhibit multipeaked response functions is beginning to be confirmed by data.[16] The precise relationship between the peaks and the size distribution is yet to be established.

A variation of this approach constitutes one of the Condon-Osborn-Glick methods.[26] In this method, the Denison and Baum theory substitutes for A-13 data to obtain the frequency dependence of the response function. For each modal size, Eq. (2) is differentiated with respect to frequency and the result is set to zero. Use of Eqs. (7) and (8) with Eq. (2) and the differential then provides two equations for A and B. The intent is not to deduce effects of particle size on A and B, which would be artificial, but to fit a frequency response curve through values at zero frequency and peak response frequency. The method produces the same qualitative behavior as the Cohen model. Further calculations by Condon and Osborn[28] show that the width of the size distribution for each modal size has a significant effect upon the appearance of the response function curve for the aggregate propellant.

The appearance of a multipeaked response function curve implies the existence of multiple zeros in the phase relation between the burning rate and pressure fluctuations. Multiple zeros associated with multiple peaks are being confirmed experimentally.[16] Multiple regions of phase lead/phase lag behavior would have important consequences regarding velocity-coupled contributions to instability. Some calculations of the velocity-coupled response function based upon application of the Cohen postulates appear in Ref. 28. A more extensive discussion of velocity coupling is beyond the scope of this paper.

Recent Attempts to Account for the Propellant Heterogeneity

BDP Model Developments in Conjunction with Homogeneous Propellant Theory

Recent years have seen refinements of the BDP model and extensions of that model to multimodal propellants.[20] Beckstead[29] and Condon-Osborn-Glick[26] have taken the approach of using their respective steady-state models to calculate values for the combustion constants which are called for by a homogeneous propellant theory of oscillatory combustion. Although the approach avoids the tedious algebra encountered by Hamann, there is a fundamental inconsistency in using two different models in this manner. The constants of Eq. (6), based upon a perturbed BDP model, are not the same as the constants of Eq. (2), based upon a perturbed homogeneous propellant model. Nevertheless, the approach has been pursued and results have been acquired.

The use of a homogeneous propellant to describe oscillatory behavior [i.e., Eq. (2)] raises the limitations already discussed in that context. The problem is to justify a "better" value of B, and a basis for consequential variations in B with particle size and pressure. The authors tentatively accomplish this purpose by using a Hart-McClure definition for B rather than the Denison-Baum definition. As discussed in the Culick review,[14] a suitable manipulation of the boundary conditions can cast the final result in such a way that no parameter of the gas phase appears. The result is Eq. (2) with the same definition for A, but B is now defined as:

$$B = \frac{1}{\sigma_p (T_w - T_o)} \tag{9}$$

where σ_p is the burning rate temperature sensitivity. As pointed out by Culick, this reformulation does not resolve the problem. Indeed, consistency of Eq. (2) requires that

$$\sigma_p = \frac{c_s}{c_g T_F}\left(1 + n + \frac{E_g}{2RT_F}\right) \tag{10}$$

Using the set of numerical values in Table 1, $\sigma_p = 0.0014/°C$, which is a low but plausible result. The value of B is still 1.59. The way to improve upon B is to instead use either BDP-type model calculations or experimental data for σ_p. Experimentally, σ_p can be as high as 0.005/°C, in which case B would fall to 0.36. With $A = 8.5$, such a value of B produces intrinsic instability.[21] A value of B in the neighborhood of 0.7 is adequate to produce sharp peaks in the response function curve. Furthermore, it is known that σ_p varies with particle size and pressure, so B may vary with particle size and pressure as $1/\sigma_p$. For $B = 0.7$, changes in B are very consequential in changing the peak response frequency and magnitude.[21] Thus, the door is opened to more pleasing, though incorrect, calculations of response function behavior.

Beckstead[29] presented some parametric results to show the calculated effects of AP size distribution upon n, σ_p, and the response function. Essentially, effects on the response function stem from the effects on n and σ_p. Propellants with higher exponent and temperature sensitivity would tend to have a higher response. Such results have appeal for practical application, but no applications were discussed and there were no comparisons with data.

BDP Model Developments in Association with the Technique of Zeldovich and Novozhilov

The third approach taken by Condon, Osborn, and Glick[26] is based upon a derivation by Zeldovich and Novozhilov. The derivation recognized the fact that any model of oscillatory combustion must deal with three essential dependent variables (burning rate, surface temperature, and temperature gradient at the surface) and two independent variables (pressure and initial bulk temperature). A particular combustion model was viewed simply as a tool with which to calculate the dependent variables, but those variables constitute the essence of the analysis. Combining total derivatives with an energy balance at the burning surface produced four key parameters:

$$D_1 = (T_w - T_o)\left(\frac{\partial \ln r}{\partial T_o}\right)_p \tag{11}$$

$$D_2 = \left(\frac{\partial T_w}{\partial T_o}\right)_p \tag{12}$$

$$D_3 = \left(\frac{\partial \ln r}{\partial \ln p}\right)_{T_o} \tag{13}$$

$$D_4 = \frac{1}{T_w - T_o}\left(\frac{\partial \ln T_w}{\partial \ln p}\right)_{T_o} \tag{14}$$

The response function was derived to be a function of these four parameters as follows:

$$R_b = \frac{D_3 + (D_2 D_3 - D_1 D_4)(\lambda - 1)}{1 - D_1 + D_2(\lambda - 1) + (D_1/\lambda)} \tag{15}$$

where D_3 is the pressure exponent n by definition and the partial derivative in D_1 is the temperature sensitivity σ_p by definition. The partial derivatives in D_2 and D_4 do not have names, but can be related to σ_p and n through the parameters $(\partial \ell n r/\partial T_w)_p$ and $(\partial \ell n r/\partial T_w)_{T_0}$, respectively. All of these parameters are to be computed from a particular model. The point of the method is that the model is up to the user, and whatever the model, the response function depends upon a set of fundamental quantities as defined by Eq. (15). For the Hart-McClure model, $D_1 = B^{-1}$, $D_2 = (AB)^{-1}$, $D_3 = n$, and $D_4 = n/A$; indeed, Eq. (15) reduces to Eq. (2). For another model, these quantities will be different. The only restriction on the use of models for this purpose appears to be that the solid must be homogeneous. The method does not contemplate heterogeneous preferred-frequency behavior.

Condon, Osborn, and Glick recognized that this method is the proper way to apply the steady-state model. Essentially, by use of their petit ensemble (PEM) steady-state model computer program, they then accomplish numerically what Hamann accomplished by tedious algebra. The numerical capability is useful in view of the additional complexities of the model extensions to multimodal propellants. Only a few results have been obtained, but the comparisons to data were not encouraging. Peak response frequency is still tied to the square of the burning rate in Eq. (15), and peak response magnitude is still restrained by the consequences of a high value of B. Their current thinking is that the method is the correct approach, but an improved model is needed to implement the method.

Stationary Sideways Sandwich Model

Cohen and Bowyer[30] studied the effect of a one-dimensional layered heterogeneity upon the nonsteady thermal profile in the solid phase. The model is similar to that of Lengelle and Williams, but there were several important differences in the approach. First, the layers consisted of alternate layers of AP and binder having different thicknesses; thus, the properties varied in steps rather than as a continuous sinusoidal function. Second, the surface layer of AP contained a finite surface melt layer as a region of temperature-dependent heat release. The usual model approach is to take all of the condensed phase heat release as occurring right at the surface. The finite melt layer introduces another aspect of solid-phase heterogeneity. Third, and perhaps most important, this geometric configuration was held stationary with respect to the surface with the AP layer at the surface. The idea was to eliminate the motion-induced preferred-frequency behavior, and to examine only the interaction between the thermal wave and the layers. Thus, the bases for the Cohen postulates would be evaluated. The energy equation contained the convective transport term to account for actual motion, but the geometry remained fixed (like the statistical average representative of the propellant surface in the BDP model). Fourth, the gas phase was represented by an approximate form of the BDP model so that the boundary condition heat transfer could be a function of particle size. The authors succeeded in deriving a closed-form expression for the response function, but it required a numerical integration through the solid phase to implement.

The response function can be expressed in the following form:

$$R_b = E_1 \Big/ \left[E_2 + i\frac{E_3}{\Omega} + \lambda' \left(1 - i\frac{E_4}{\Omega} \right) \right] \tag{16}$$

It can be shown[30] that this form is equivalent to Eq. (2) for $\lambda' = \lambda$. The quantity λ is a characteristic of the analytic solution for a homogeneous solid. For a heterogeneous solid, such as described by this model, the characteristic becomes a function of the heterogeneity. It is computed through a numerical solution and is referred to here as λ'. A difference between λ' and λ is significant because it reflects an alteration of the frequency response by the heterogeneity. The combustion constants of Eq. (16) are more involved than those of Eq. (2), commensurate with the additional model details. A fourth combustion constant arises because of the finite melt layer. The pressure exponent does not appear explicitly, but can be made to appear through the use of the zero-frequency limit to replace one of the other constants.[24,30] The zero-frequency limit of R_b is the effective pressure exponent. The nondimensional frequency Ω can be set by the thermal diffusivity of the AP or the mean solid, depending upon the structuring of E_3 and E_4. Solutions were obtained and compared with solutions for a homogeneous solid, with and without a finite melt layer.

The results showed that the effects of the heterogeneity as represented by this model were negligible. Replacing the layered bulk solid with a homogeneous solid, or taking the surface heat release to be right at the surface rather than in a finite melt layer, or both, produced only slight changes in the peak response frequency and magnitude. Basically, there was little numerical difference between λ' and λ or between E_3 and E_4. For all practical purposes, then, Eq. (16) has three combustion constants, with $\lambda' = \lambda$, like Eq. (2). Furthermore, parametric variation with particle size and burning rate showed that the properties of the solutions were essentially those of a homogeneous solid. Only changes in burning rate affected the peak response characteristics significantly. Finally, comparisons with experimental data revealed the same deficiencies discussed earlier in the context of the homogeneous propellant theory. The model predicted slight peaks in the response function and at too low a frequency.

There are several implications of this work which bear further discussion. First, the effect of solid-phase heterogeneities upon the thermal wave need not be taken into account. This affords a potential simplification of the problem. Second, and by the same token, the bases for the Cohen postulates have been discredited. Although the frequency response postulate can still be supported by the preferred-frequency concept, the peak response postulate needs to be reexamined. Third, and in connection with the peak response, it is interesting that the use of this model did not improve upon the effective value of the equivalent "B parameter." In other words, the characteristics of the solution were those of Eq. (2) with a high value of B. In view of Eq. (15) and earlier remarks in connection with σ_p, it appears that a small peak in the response function is a characteristic of any model that will predict a small value of σ_p. Referring to Eq. (15), the constants D_1 and D_2 will approach zero as σ_p approaches zero. Under those circumstances, the response function cannot rise significantly above D_3, which is n. The implication is that a particular model should be evaluated for its ability to predict σ_p before it is considered for use in developing a response function. It appears that the Cohen-Bowyer model was deficient in this respect, even though containing elements of heterogeneity in the solid and gas phase. On the other hand, progeny of the formal BDP model seem capable of meeting this test. The ability to predict σ_p appears desirable even though response functions might be augmented by heterogeneous mechanisms such as the preferred-frequency concept.

Concluding Remarks

A number of theoretical models have been developed to account for the heterogeneity of composite propellants in expressing the pressure-coupled combustion response function. There have been few comparisons with data, and these were not encouraging, with the exception of the qualitative aspects of preferred-frequency behavior. It appears that aspects of preferred-frequency behavior can be associated with the oxidizer particle size distribution, but the precise nature of the relationship and its underlying mechanism are yet to be ascertained. More data are required to obtain the frequency dependence of the response function in detail.

It appears that the model of Lengelle and Williams furnishes a viable basis to explain the effects of heterogeneity. In order to implement this model, it will be necessary to better characterize the functional forms of the heterogeneity as they exist in propellants. It appears that idealized models will not be as tolerable in this analysis as they have been in steady-state combustion theories, and it may be necessary to acquire these forms experimentally in order to be able to properly test the theory in relation to experimental behavior. Characteristics of the heterogeneity have also appeared in steady-state burning, and this should be used to provide a supplemental means to develop and validate the theory. The concepts of Boggs and Beckstead, and of Cohen, fall into this general category, but they require a firm analytical foundation that the Lengelle-Williams approach can provide.

Approaches not based on a preferred-frequency concept stemming from the composite propellant structure in the solid phase appear doomed to retain the essential characteristics of homogeneous propellant theory. However, as pointed out by Lengelle and Williams, the thermal lag in the solid phase still has a role to play in the overall scheme. The Cohen-Bowyer work shows that average thermal properties can be assumed to describe the thermal wave, suggesting that preferred-frequency behavior be imposed at the boundary. In expressing the boundary condition, use of one of the advanced steady-state models would be preferred in that they appear more capable of describing σ_p behavior than simpler combustion models. The use of such models would forecast the need for numerical solutions with the loss of some of the insight that closed-form solutions can provide. Ultimately, this may be unavoidable owing to the complexity of the problem.

Fluctuations observed during steady-state burning and the importance of parameters gleaned from steady-state theory suggest that a compositional form of heterogeneity be imposed at the boundary. In other words, there would be fluctuations in the formulation at frequencies determined by the heterogeneity form function. Associated with these would be fluctuations in the surface heat release and in the primary flame temperature. Coherence and synchronization remain unresolved questions. The complication introduced by Lengelle and Williams may not be necessary upon closer examination of the ordering of well-mixed propellants, the statistics of the packed bed, and the spectrum available in the structures of real propellants. It may be that there is always some portion of the structure that can couple to the pressure oscillations.

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