Response of Radiation Driven Transient Burning of AP and HMX Using Flame Modeling

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The radiation driven response function (R_q) for AP and HMX propellant was obtained and compared with experimental results by using a simple $\alpha\beta\gamma$ flame model rather than with detailed chemistry. For an AP propellant, the profile of heat release was assumed by the experimental data. The calculated R_q shows a frequency shift of the peak amplitude to the higher frequency and a decrease in the maximum amplitude as radiation increases. In addition, it was found the increase in the total flux could enhance the mean burning rate $\bar{r}_b$ while the phase differences between the radiation and resulting conduction could consequently reduce the fluctuation amplitude Δr_b. Fortunately, this is the qualitative duplication of the behavior recently observed in the experiments of RDX propellants. For HMX, the response function R_q has been calculated and showed a quite good agreement with the experimental data. Even though the fairly good agreement of R_q with experimental ones, the unsteady behavior of HMX was not reproduced as the radiation input increased. This is due to lack of the material properties of HMX or the physical understanding of HMX burning at high pressure.

Key Words: Radiation, Response Function, Solid Propellant

Nomenclature

A : Pre-exponential factor
C_c : Specific heat of condensed phase
E_c : Activation energy of condensed phase decomposition reaction
$k_{()}$: Thermal conductivity of ( ) phase
m : Mass burning rate, $\rho_c r_b$
Q_c : Chemical heat release of condensed phase
q_0 : Incident radiation heat flux intensity
q_g : Conductive heat flux from a gas region
R_q : Radiation-driven response function
R_u : Universal gas constant
r_b : Burning rate
x : Coordinate normal to a solid surface
x_f : Flame thickness in a gas region
Y : Mass fraction of unreacted solid

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Greek Symbols

α_c : Thermal diffusivity of propellant, $k_c/\rho_c C_c$
a_p : Volumetric absorption coefficient of the condensed phase
ρ_c : Condensed phase density
τ : Characteristic time, $\alpha_c/\bar{r}_b^2$
ω : Chemical reaction rate in a gas region
Ω : Reaction rate in a condensed region

Subscripts

c : Condensed phase
g : Gas phase

1. Introduction

Many studies have been focused on the transient solid propellant burning to understand the general mechanisms of unsteady combustion (Kuo et al., 1984; T'ien et al., 1984; De Luca, 1992). The response function is one of the most crucial measures for evaluation of the energetic materials. In general, the combustion of solid propellants could be accompanied by oscillation of flame temperature, radiance, and velocity of combustion products caused by burning rate oscillations. Pressure and radiation are two main candidates that can perturb the combustion field and frequently lead to instability in a solid motor. Since these are intrinsically coupled in combustion field, it is very difficult to study the isolated influence of the parameter on the combustion of solid propellants. Usually, pressure oscillation in a motor may lead to the flame temperature oscillation in the gas phase. The radiation from reaction zone can be altered by temperature fluctuation as a consequence.

With advancement of the laser technique, the reaction zone in the experiment can be easily perturbed by the external laser signal without producing any unwanted negative side effects. Thus, the problem of understanding unsteady combustion can be successfully investigated by using an external radiation stimuli. Zarko et al. (1992) also did an experimental study of radiation driven response function by using the external laser perturbation. He performed a series of experiments to study radiation-driven response for DB (double base) and composite propellants and revealed many basic understandings of unsteady solid propellant combustion. Also, he investigated a computer simulated response function of solid propellant by using the ZN (Zeldovich-Novozhilov) approach. Recently, Son et al. (1993) suggested a theoretical expression of radiation response function accounting for in-depth absorption in the condensed phase against the external radiation input. Lee et al. (1999) reexamined a response function by directly applying ZN method within quasi-steady approximation. Kudva et al. (1999) conducted a series of experiments to investigate the response of nitramine monopropellants, RDX (cyclotrimethylene-trinitramine) and HMX (cyclotetramethylene-tetranitramine), by varying the magnitude of radiation input. Hence, they found the peak frequency of the response function migrated to a higher frequency, and that amplitude decreased with the increase of the radiation input.

Although the theoretical response function was useful in predicting transient combustion of solid propellant, it had an unavoidable limit that the surface adiabatic condition was implemented in the derivation. This is mainly due to not only the complexity of chemical reaction but also the unavailability of an appropriate reaction model for theoretical treatment in a gas region. Zebrowski et al. (1996) conducted a numerical study in a more realistic way on the response function to radiation perturbations. They implemented a distributed reaction in the condensed phase as well. They, however, did not account for the conduction transfer from the gas phase but used the adiabatic condition as in the theoretical study. Recently, Erikson et al. (1999) performed a numerical study for nitramine monopropellants with detailed chemistry. In addition, they accounted for the effect of propellant decomposition and evaporation at the surface simultaneously to implement real combustion phenomena. Although this approach showed a relatively good agreement with the experimental results of DB, composite and RDX propellants, it should be noted that this approach required a huge amount of computational resources and efforts.

Thus, it would be very efficient to investigate a response function with a simple flame modeling in order to grasp the physically important ingredients at the initial stage of study. In this paper, we suggest a very effective way to obtain the radiation driven response function by using a flame model for gas phase reaction rather by a detailed chemistry. Then the response function (Rq) over various solid propellants has been calculated and compared with the experimental ones.

2. Combustion in Condensed and Gas Phase

For the governing equations of condensed phase, we assumed the distributed reaction with one-step irreversible zeroth order chemical kinetics. The continuity equation is shown in Eq. (1) with one-step chemical kinetics of Eq. (2). The reference frame is selected so that the origin coincides with the propellant surface. Thus, there appears a bulk velocity corresponding to the burning rate in the governing equations. Within this frame, we can write species and energy conservation equations in the condensed phase as

$$\rho_c\frac{\partial Y}{\partial t}+\rho_c r_b\frac{\partial Y}{\partial t}$$

$$=-\Omega,\ Y(-\infty,\ t)=1,\ Y(0,\ t)=0 \qquad (1)$$

$$\Omega=\rho_c A\exp(-E_c/R_u T) \qquad (2)$$

$$\rho_c C_c\frac{\partial T}{\partial t}+\rho_c C_c r_b\frac{\partial T}{\partial t}$$

$$=k_c\frac{\partial^2 T}{\partial x^2}+Q_c\Omega+q_0 a_p\exp(a_p x) \qquad (3)$$

$$m(t)=\rho_c r_b(t)$$

$$=\rho_c A\int_{-\infty}^{0}e^{-E_c/R_u T}dx+\rho_c\int_{-\infty}^{0}\frac{\partial Y}{\partial t}dx \qquad (4)$$

$$T(-\infty,\ t)=T_0,\ k_c\left(\frac{\partial T}{\partial x}\right)_{x=0-}=k_g\left(\frac{\partial T}{\partial x}\right)_{x=0+} \qquad (5)$$

The last term in Eq. (3) is the radiation input to the surface and expressed as an exponential function of Beer's law. The boundary conditions of Eq. (5) are the heat flux balance at the propellant surface and the cold temperature of propellant. And it is worthwhile noting that many theoretical approaches did adopt an adiabatic condition for simplicity of analysis. In this study, however, conductive flux to the propellant surface was taken into account by a flame model suggested by De Luca (De Luca, 1992). Generally one must solve the energy equation in the gas region with detailed chemical kinetics in order to evaluate conductive flux to the propellant surface. Moreover, the reaction kinetics and the material properties of the solid propellants are too complicate to have a proper model for calculation. Thus, it seems quite natural to depend upon experimental data or researcher's experience in determining parameters needed for numerical calculation. It may be effective and reasonable rather to use a flame model for conduction flux to the surface than to directly solve the energy equation with detailed chemistry. In the modeling, the spatial heat release distribution was assumed by the experimental data and then the conductive heat flux to the surface was obtained.

De Luca(1992) proposed a simple way to estimate heat flux from the gas region to the surface by assuming the heat release distribution based on the experimental data known as $\alpha\beta\gamma$ model. By applying the proper QSHOD (Quasi-Steady Homogeneous One Dimension) assumption to the gas region, the integration of Eq. (3) yields the conduction flux from the gas phase. Figure 1 displays a schematic of $\alpha\beta\gamma$ model.

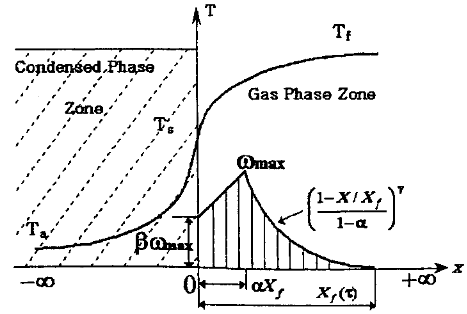

Fig. 1 The schematic of the $\alpha\beta\gamma$ model

$$q_g=k_g\left(\frac{\partial T}{\partial x}\right)_{x=0+}=\int_{0^+}^{x_f}\rho_g Q_g\omega\exp\left(-\frac{c_g}{k_g}r_b x\right)dx \qquad (6)$$

Readers should refer to De Luca(1992) for more details on the flame modeling.

3. Radiation-Driven Response of AP Propellant

The sinusoidal perturbation was used in the calculation to obtain the radiation driven response. The perturbation consists of the mean flux of q=125. 6 J/cm^{2}s (50cal/cm^{2}s) with an amplitude of 5cal/cm^{2}s and the frequency ranging from 30 Hz to 300Hz. This input level seems to be appropriate enough to deal with linear responses in the above frequency range. The response function was then evaluated for each frequency by approximately averaging 20 responded cycles. The calculation results was verified by using the same adiabatic surface condition and the same AP propellant as used in Zebrowski et al. (1996). Figure 2 shows the comparison of the numerical response with previous result in Zebrowski et al. (1996). In Fig. 2, it is seen two results are quite similar except at the peak amplitude of the response. Physically, the peak is analogous to the mathematical concept of the resonance for spring-

Table 1 Parameters adopted for $\alpha\beta\gamma$ model

parameters	α	β	γ	Q_g
Case I	0.5	0.3	3	23.2
Case II	0.3	0.6	2	44.5
Case III	0.1	0.8	1	60.7

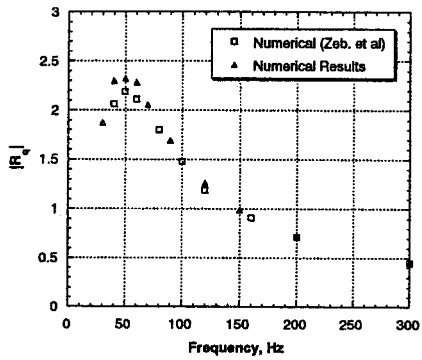

Fig. 2 The response of present study is compared with the previous one [6] when the same adiabatic condition was used at the propellant surface

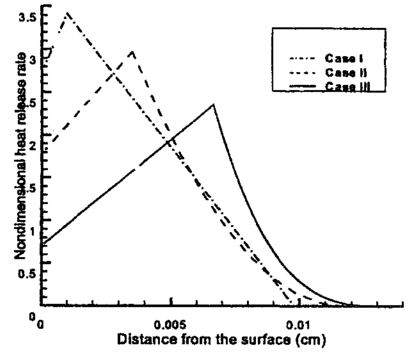

Fig. 3 The profile of heat release of AP propellant in region for $\alpha\beta\gamma$ flame model

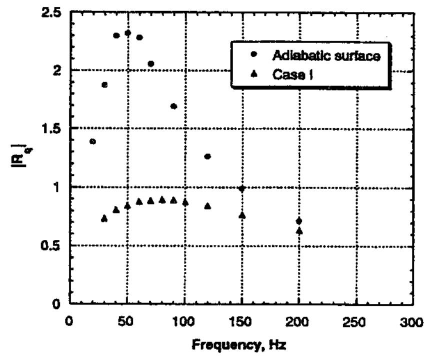

Fig. 4 Comparison of the response function between the adiabatic condition and non-adiabatic condition(Case I)

mass system against an external excitation. The discrepancy may be attributed to different schemes from the previous one and the scaled coordinate in the calculation.

As mentioned earlier, conduction transfer to the surface is of significance in determining the burning rate of the propellant. However, the theoretical approach could not include the conduction because of the complexity of chemical reaction in the gas region and took an assumption of adiabatic surface condition. In this study, we included the conduction transfer by implementing the $\alpha\beta\gamma$ model. From the experiments with a Strand burner, it was revealed that the heat release distribution was quite dependent on the pressure level in the gas region. The low pressure could produce the low amplitude and spatially spread heat release distribution. Whereas the high amplitude and narrow reaction region is attributed to the high-pressure level in the gas. So, we chose arbitrary three different models. Table 1 is the summary of three cases of $\alpha\beta\gamma$ model and the schematic was illustrated in Fig. 3. Case I was designed to model the low amplitude in heat release and wide spread of chemical reaction. Case III was chosen to show the reverse of case I. And case II was a compromise between the two extremes. Also, we can see that case III has the biggest conduction flux contribution to the propellant and case I contributes the smallest heat feed back.

Figure 4 shows the difference between the response with adiabatic surface condition and the response with conduction flux from the gas region. As seen in the Fig. 4, if we do account for the conduction flux from the gas region, the response curve shows a big drop in amplitude (2. 32 to 0.89 for non-adiabatic condition) and a small increase in peak frequency from 50Hz to 80Hz. It should be noted that there are three time scales involved in the solid propellant combus-

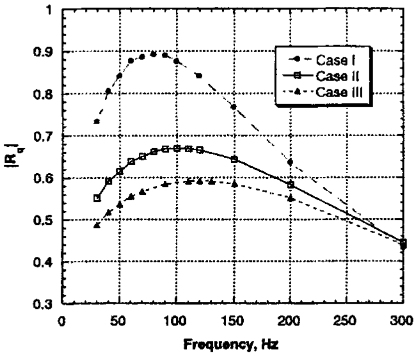

Fig. 5 The behavior of calculated radiation response for AP as the increase in the conduction flux implemented by a simple flame heat distribution modeling

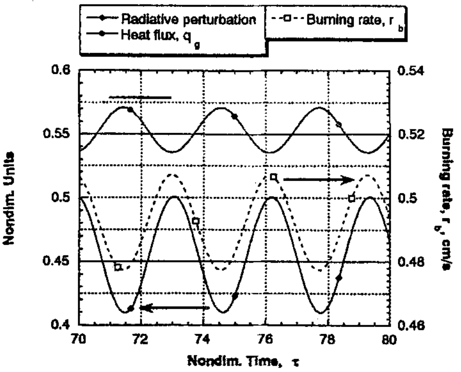

Fig. 6 The radiation input and the calculated conduction flux. The phase lag is observed between two fluxes

tion; condensed time scale, reaction region time scale and gas phase time scale. Here the condensed time scale is the largest one and defined by the ratio of thermal diffusivity to the square of average burning rate, $\tau_c = \alpha_c / \bar{r}_b^2$. In addition we know that the frequency is proportional to the inverse of time. It is, therefore, clear that the peak frequency should show an increase when we consider the conduction flux from the gas region that can trigger the additional increase in burning rate $\bar{r}_b$(for adiabatic condition; τ_c=94Hz, for non-adiabatic condition, Case I ; τ_c=128Hz).

Figure 5 shows the radiation driven responses R_q for three different flame models. Each model was chosen so that the magnitude of conduction was 93.1 J/cm^{2}s (23.2 cal/cm^{2}s) (Case I), 183.6 J/cm^{2}s (44.5 cal/cm^{2}s)(Case II) and 254.1 J/cm^{2}s (60.7cal/cm^{2}s)(Case III) respectively. With three flame models, we can qualitatively explain the behavior of peak magnitude of R_q. The increase in the conduction flux causes the increase in the total heat flux and consequently enhances the mean burning rate of the propellant. It should be noted that the definition of R_q is the ratio of burning rate fluctuation normalized by mean value to the radiation fluctuation divided by mean flux as in Eq. (7)

$$|R_q| = \left| \frac{\Delta r_b / \bar{r}_b}{\Delta q_r / \bar{q}_r} \right| \tag{7}$$

Since the radiation is chosen as an input parameter, we can see the denominator of Eq. (7) is constant and the necessary condition for small magnitude of R_q is the decrease in the burning rate fluctuation Δr_b and/or the increase in the average burning rate $\bar{r}_b$. Bearing this in mind, it is quite interesting to find that the burning rate oscillates in phase with the sinusoidal input whereas the conduction flux shows a phase lag of about 180 degree. Figure 6 shows phase lag between two heat fluxes; radiation input and conduction flux. Although the total heat flux (radiation+conduction) increases, the fluctuation amplitude of total flux is diminished due to the phase difference between radiation input and the resulting conduction. This consequently results in increase in the mean burning rate $\bar{r}_b$ whereas contributes to lower the amplitude of Δr_b. The numerical study revealed that the maximum of total flux fluctuation Δq_{total} decreases from 0.251 J/cm^{2}s (0.060 cal/cm^{2}s)(case I), to 0.209 J/cm^{2}s (0.050 cal/cm^{2}s)(case III). And the corresponding maximum of Δr_b was 0.052cm/sec, 0.041cm/sec, and 0.036cm/sec. This was shown in Fig. 7. Thus, we can simply conclude that the shift of peak frequency and lowering the magnitude of R_q is mainly caused by the phase lag between radiation input and conduction output. Figure 7 shows the influence of conduction flux on the radiation driven responses. It is quite interesting to note that this is a qualitative duplication of the behavior recently observed in the experiments of RDX propellant.

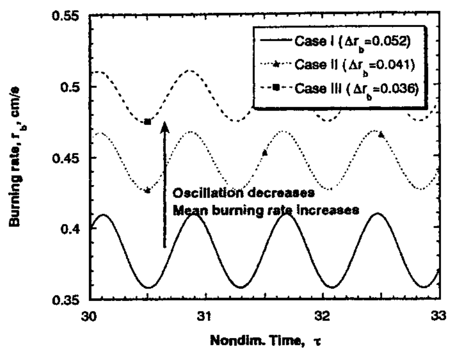

Fig. 7 The increase of radiation causes to reduce the magnitude of burning rate fluctuation and consequently leads to the decrease of amplitude of response function because of phase lag between radiation and conduction flux

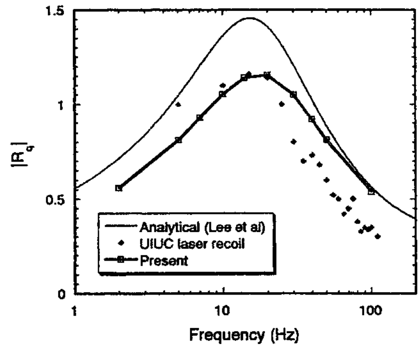

Fig. 8 The radiation driven response function for HMX propellant. The present study shows a good agreement with a theoretical response and experimental response

4. Radiation-Driven Response of HMX Propellant

Although HMX is known as one of good additives used in many solid propellants, the combustion behavior is not still fully understood in many aspects. In addition, it is not surprising to find different material properties for a certain HMX in various literatures. Thus, the lack of understanding of HMX renders the analysis more difficult. In this study, material properties of HMX were mainly excerpted from reference. And, some of data was obtained from as well. The followings are the material properties used in the calculation; $\rho_c=1.8\text{g/cm}^3$, $C_c=1.403\text{J/gK}$(0.335cal/gK), $\alpha_c=0.008\text{cm}^2/\text{s}$, E_c=42.1kcal/mole, $A_c=1.64\times10^{15}$(1/sec), $a_p=5670\text{cm}^{-1}$, Q_c=209.3J/g(50cal/g), Q_g=3273.3J/g(783cal/g), C_g=1.403J/gK(0.335cal/gK), and $k_g=7.0\times10^{-4}\text{J/cm}\cdot\text{s}\,K$($1.67\times10^{-4}\text{cal/cm}\cdot\text{s}\,K$). Then we investigated the radiation response and compared with the experimental data of Loner et al. (1998). A sinusoidal function was chosen with a mean of 34.75J/cm^{2}s (8.3 cal/cm^{2}s) and 1.38 J/cm^{2}s (0.33 cal/cm^{2}s) in amplitude to obtain the response function. Three curves are found in Fig. 8; the radiation response in this study Rq, experimental response, and theoretical response function. As can be seen, it shows a fairly good agreement between the experimental and the calculated Rq. The theory predicts the peak frequency is located about 14~16Hz and the experimental results show that it is about 16~18Hz. The numerical results also coincide with the experimental ones showing its peak frequency around 16~18Hz. Thus, the numerical response with a simple flame modeling can be useful in analyzing the transient behavior of solid propellants even for HMX.

5. Conclusions

In order to find the physically important ingredients for combustion of solid propellants, an effective study was done with a flame modeling of gas phase rather than with a detailed chemistry. And the radiation response function (R_q) for an AP propellant and HMX has been calculated and compared with the experimental results. For AP propellant, the heat release distribution was assumed according to the experimental data of Zenin(1992). The calculated response R_q shows a shift of the peak frequency to a higher frequency and a decrease in the maximum amplitude. In addition, it was found the 180-degree phase difference between the radiation flux and the calculated conductive flux. Thus, an increase in the total flux to the surface could enhance the mean burning rate while the phase differences caused to reduce the fluctuation amplitude. The same phenomena were observed in the experiments of RDX propellants.

For HMX, the response function Rq was calculated to predict the experimental function by Loner(1998) and showed quite a good agreement with the experimental data. Even though the fairly good agreement with the experimental results, it failed to predict the unsteady behavior of HMX when the radiation input increased. This is due to the partial lack of material properties of HMX or the physical understanding of HMX burning at the high pressure.

Acknowledgements

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