A Steady-State Combustion Modelling of Composite Solid Propellants

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By depicting the transfer of heat and combustion reaction to take place within thin gas layers close to the propellant surface burning in a steady-state fashion, a mathematical equation has been deduced to describe the burning rate of solid propellant as a function of initial grain temperature and chamber pressure. It has been also assumed that chemical reaction could take place in premixing-diffusing zone but were carried out mainly in the reaction-flame zone. All these phenomena taken place in each zone of combustion have been assumed to be steady-state. In the present investigation, the equation, $\gamma = k \cdot (1/R(T_i + C))^n \cdot \exp(-E_a/R(T_i + C))(P/z)$ is being presented and it is compared with experimental data. The proposed model has been tested and evaluated vis-a-vis strand burner data for three different propellants based on CTPB, and it has been found that the deviation of the computed burning rates from the measured rates ranged up to 2%.

Key Words : Composite Solid Propellants, Burning Rate Mechanism, Decomposition, Initial Grain Temperature, Combustion Chamber Pressure

Nomenclature

A : Chemical reaction parameter, Pa · sec · cm^{-1}.

a : Constant of combustion characteristics, cm · sec^{-1} · Pa^{-n}.

B : Diffusion parameter, Pa$^{1/3}$ · sec · cm^{-1}.

b : Constant of combustion characteristics, cm · sec^{-1}.

C : Effective temperature difference, K.

C : Mole concentration of i-component gas, mol · L^{-1}.

c : Constant of combustion characteristics, cm · sec^{-1} · Pa^{-m}.

E_a : Activation energy of gas reaction in the reaction zone, Jmol^{-1}.

k : Reaction constant.

M_d : Average molar weight of decomposed gas mixture, grams · mol^{-1}.

N_d : Mole number of decomposed gases.

N_i : Mole number of i-component gas.

N_R : Total mole number of reaction gases.

n : Constant of combustion characteristics.

P : Combustion chamber pressure, Pa.

P_i : Partial pressure of i-component gas, Pa.

q : Heat flux, J · sec^{-1} · cm^{-2}.

R : Universal gas constant, J · mol^{-1} · K^{-1}.

S_p : Burning surface, cm2.

T_i : Initial grain temperature, K

T_o : Constant, K

T_R : Reaction temperature, K

U : Overall heat transfer coefficient, J · sec^{-1} · cm^{-2} · K^{-1}.

V_P : Volume of propellant, cm^3.

W_P : Weight, grams.

r : Burning rate, cm · sec^{-1}.

ρ_P : Solid propellant density, grams · cm^{-3}.

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1. Introduction

The burning rate mechanism of solid propellants has been studied throughout the history of rocketry and predictions of rocket performance have been made possible. Although many theories have been developed to explain the burning rate mechanism, none has been entirely successful. Especially the burning mechanism of solid propellants at high pressures in combustion chamber has not yet been well established (Kishore, 1979 ; Boyers and Klayer, 1969).

Studies on the combustion of composite solid propellants have been proceeded qualitatively and quantitatively on the basis of condensed phase reactions and gas-phase reactions. The condensed phase reactions include surface and subsurface reactions(Cohen *et al.*, 1974) and the gas-phase reactions are generally categorized into three proposed models : i) granular diffusion flame model(Summerfield *et al.*, 1960) , ii) sandwich columnar diffusion model(Nachbar and Narks, 1957) , iii) multiple flame model (Beckstead *et al.*, 1970), and iv) quasi-steady combustion model (Brewster and Son, 1995 ; Yoon, 1993).

Many mathematical models based on the above two reactions have been proposed for the relationship between burning rate and combustion chamber pressure. Typical burning rate relationships are as follows;

i) an empirical equation known as Saint Robert's law

$$r = ap^n \qquad (1)$$

ii) an alternate form of the above equation,

$$r = b + cp^m \qquad (2)$$

iii) a theoretical equation based on the granular diffusion flame model(Summerfield et al., 1960)

$$\frac{1}{r} = \frac{A}{P} + \frac{B}{P^{1/3}} \qquad (3)$$

The theory based on the granular diffusion flame model is the most convenient for deducing expressions for the pressure dependence of burning rate. A wide range of experimental data is available and none of the proposed equations fit the entire range(Rastogi *et al.*, 1978). One of the reasons why the above equations do not fit the measured burning rates reasonably well in certain range of pressure was to neglect the effect of thermal radiation of combustion product gas on the combustion response(Brewster and Son, 1995).

Burning rates of composite solid propellant are influenced by composition factors and environmental factors. Major composition factors are the constituents of the propellant and the weight percent of its ingredients. Environmental factors include surface area of oxidizer, burning catalyst, combustion chamber pressure, reaction temperature of decomposed gases, and velocity of combustion gases passing through the burning surface. Among all these factors combustion chamber pressure and reaction temperature are most important. Much research has been devoted to the dependence of the burning rate on pressure but the research has rarely been carried out for the dependence of the burning rate on reaction temperature.

In the present work burning rates are studied mainly as a function of combustion chamber pressure and reaction temperature. Considering not only the reaction layer of gases, which are produced from the decomposition of solid propellant at its surface, but also the chemical reaction of decomposed gases, we deduced a new burning rate equation of solid propellant. Calculated burning rates by the present model were compared with experimental values. Excellent agreement has been obtained for pressures from 2 MPa to 20 MPa and initial grain temperatures of 233. 15K, 293. 15K and 323. 15K.

2. Combustion Mechanism

Propellant can not be ignited without enough heat, so that the necessary heat igniting the propellant of rocket motors should be supplied by an ignition system. By investigating the autoignition temperature of carboxyl terminated polybucadiene (CTPB) propellant with thermogravimetric analyzer (TGA) and

differential scanning calorimetry (DSC), it was found to be higher than 573 K (Waesche and Wenograd, 1967 ; Morisaki and Komamiya, 1975).

The burning mechanism of composite solid propellants shows several apparent differences compared with homogeneous propellants. Since the oxidizer and the fuel are present in separate phases, they must decompose to give alternately oxygen-rich and fuel-rich gas streams. Sutherland (1958) recommended that there was no evidence of turbulent motion immediately near the regressing surface. Previous investigations (Hightower and Price, 1967; Schultz and Dekker, 1952; Bastress, 1961) suggest that the ammonium perchlorate (AP) propellants surface is dry, and slightly molten surface is not sufficiently mobile to disrupt the burning surface from the standpoint of a combustion mechanism.

Waesche and Wenograd(1969) found the phenomenon of flameless combustion where the combustion wave propagates through the propellant after extinguishment of the gas-phase flame. Many other investigators(Manelis and Strunin, 1975; Rastogi *et al.*, 1975; Kishore *et al.*, 1975) have arrived at the proposition that there are condensed-phase reactions in composite propellants. It assumes that the solid-phase decomposition is the result of heat transfer from the flame zone or from the thermal layer surrounding oxidizer particles and that this flame zone is the result of the gas-phase redox reactions between oxidizer fuel pyrolysis products. Levy and Friedman's investigation(Rastogi *et al.*, 1975) of the gas-phase reaction zone show that the thickness of the zone is less than a few microns, although it varies with combustion pressures. Through the above mentioned results, the following combustion mechanism is assumed.

1) Burning is considered to occur in four distinct phenomena: the decomposition of propellant surface, the premixing-diffusing, the chemical reaction in the reaction-flame zone, and the gas flow through the flame-burnt gas zone, as shown in Fig. 1.

2) The surface decomposition of composite propellants needs certain amount of heat which is
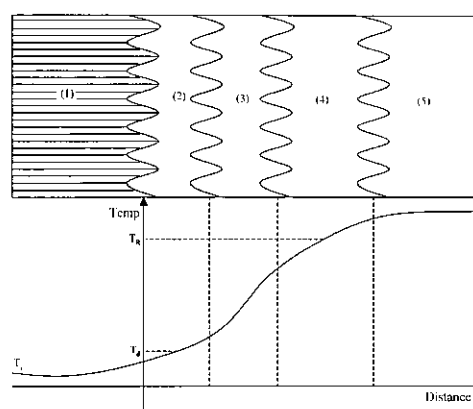
Fig. 1 Combustion Zones and Temperature Profile
(1) Solid propellant, (2) : Decomposition,
(3) : Premixing-Diffusion,
(4) : Reaction-Flame,
(5) : Flame-Burnt Gases
T_i : Initial Grain Temperature, T_d : Decomposition Temperature,
T_R : Average Reaction Temperature

initially provided by the ignition system and then by the heat of chemical reactions in the reaction -flame zone. Even though the gas reaction stops, the solid surface will continue decomposing if enough heat for decomposition is provided by hot burnt gases. Namely, the decomposition temperature of the solid surface is much less than that of gas reactions.

3) The burning surface of composite solid propellants is effectively dry and the subsurface reaction can be ignored. Decomposed gases from the solid surface are premixed and diffused simultaneously to enter the reaction-flame zone.

4) Chemical reactions can take place in the premixing-diffusing zone but are carried out mainly in the reaction-flame zone.

5) All phenomena taken place in each zone of combustion of composite solid propellant are assumed to be steady-state.

3. Mathematical Model of Burning Rate

The burning rate of composite solid propellants can be expressed as a function of various composition factors and environmental factors, as previ-

ously mentioned. Since the state of each zone depicted in Fig. 1 can be assumed steady-state, the mass balance for each zone is established as follows:

decomposition rate of composite propellant at the surface
=mass flow rate from the premixing-diffusion zone into the
reaction-flame zone
=reaction rate of decomposed gases in the reaction-flame zone
=mass flow rate of burnt gases from the reaction-flame zone

Using the propellant weight which is pyrolyzed per unit time, the linear burning rate can be written as

$$\gamma = -\frac{1}{\rho_p S_p}\frac{dW_p}{dt} \qquad (4)$$

where, $W_p = \rho_p V_p$, and W_p, ρ_p, V_p and S_p are weight, density, and volume of propellant, and burning surface respectively. The negative sign implies that the propellant weight is decreased by the decomposition of solid propellant surface. If we introduce average molar weight of decomposed gases, Eq. (4) can be written as,

$$\gamma = \frac{M_d}{\rho_p S_p}\left(\frac{dN_d}{dt}\right)_{pr} \qquad (5)$$

where, Md, and Nd are the average molar weight and the mole number of decomposed gases, and pr means produced gases.

The decomposed gases are premixed and diffused simultaneously in the premixing-diffusion zone and enter the reaction-flame zone. These phenomena take place at steady-state, and the mole number of decomposed gases produced per unit time should be equal to that reacted per unit time in the reaction-flame zone. Therefore, the relationship is established as,

$$\left(\frac{dN_d}{dt}\right)_{pr} = -\left(\frac{dN_d}{dt}\right)_{re} \qquad (6)$$

where, *re* means reaction. From Eqs. (5) and (6), the burning rate can be expressed to be Eq. (7),

$$\gamma = -\frac{M_d}{\rho_p S_p}\left(\frac{dN_d}{dt}\right)_{re} \qquad (7)$$

In the reaction-flame zone, the gas reaction rate is generally expresses as Eq. (8),

$$-\left(\frac{dN_d}{dt}\right)_{re} = kC_A^{\alpha}C_b^{\beta}\cdots C_D^{\delta} \qquad (8)$$

where C_i is the mole concentration of component i, and $\alpha, \beta, \cdots \delta$ are order of reaction with respect to each component. Substituting Eq. (8) into Eq. (7),

$$\gamma = \frac{M_d}{\rho_p S_p}\cdot kC_A^{\alpha}C_B^{\beta}\cdots C_D^{\delta} \qquad (9)$$

Introducing an Arrhenius law for κ,

$$\gamma = \frac{M_d}{\rho_p S_p}\cdot k_0 \exp\left(-\frac{E_a}{R T_R}\right)C_A^{\alpha}C_B^{\beta}\cdots C_D^{\delta} \qquad (10)$$

Since C_A, C_B, $\cdots$, C_D are the mole concentration of gas components in the reaction-flame zone, they can be replaced by their partial pressures according to the following equations.

$$C_i = \frac{P_i}{z_i R T_R}, \; i = A, B, \cdots, D \qquad (11)$$

where z_i is the compressibility of i-component gas and,

$$P_i = \frac{N_i}{N_R}\cdot\frac{z_i}{z}\cdot P, \; i = A, B, \cdots, D \qquad (12)$$

In the above equation, N_i and NR are the mole number of i-component gas and the total number of mole of reaction gases, respectively. From Eq. (10) to (12), Eq. (13) is expressed as follows.

$$\gamma = \frac{M_d}{\rho_p S_p}\cdot k_0 \exp\left(-\frac{E_a}{R T_R}\right)\left(\frac{1}{R T_R}\right)^n \\ \left(\frac{N_A}{N_R}\right)^{\alpha}\left(\frac{N_B}{N_R}\right)^{\beta}\cdots\left(\frac{N_D}{N_R}\right)^{\delta}\left(\frac{P}{z}\right)^n \qquad (13)$$

where, $n = \alpha + \beta + \cdots + \delta$.

The term, $(N_A/N_R)^{\alpha}(N_B/N_R)^{\beta}\cdots(N_D/N_R)^{\delta}$ is constant because the mole fraction of reaction gases is assumed to be fixed in accordance with the propellant composition. Therefore, the burning rate equation becomes,

$$\gamma = K\left(\frac{1}{R T_R}\right)^n \exp\left(-\frac{E_a}{R T_R}\right)\left(\frac{P}{z}\right)^n \qquad (14)$$

Where

$$K = (M_d/\rho_p S_p)(k_0)(N_A/N_R)^{\alpha}(N_B/N_R)^{\beta} \\ \cdots(N_D/N_R)^{\delta}$$

Table 1 Composition of propellant type L, M, and N (wt%)

Propellant	Ammonium Perchlorate	Binder	Aluminium	Total
L	70	14	16	100
M	80	16	4	100
N	82	16	2	100

If gases in the reaction–flame zone behave as ideal gases, z becomes a unit.

4. Effect of Initial Grain Temperature on the Burning Rate

The burning rate varies with intial grain temperature. The variation is expressed in terms of the temperature coefficient of burning rate of constant pressure which is defined as the average rate of variation over some specified temperature range. Although the effect of initial grain temperature on burning rate is considered small, there is a great risk of the bursting of rockets to be operated at extreme temperatures ranging from 219 K to 347 K. A major portion of propellant research is devoted to the search for means of reducing the effect of temperature.

A modified burning rate equation such as Eq. (15) is suggested by introducing the effect of initial grain temperature (Crow and Grimshow, 1932.)

$$\gamma = \frac{BP^n}{T_0 - T_i} \tag{15}$$

Here, T_o is a constant which is dependent upon the composition of the propellant and has the dimension of temperature. T_i is the initial grain temperature. Many investigators and designers, instead of this equation, frequently use the temperature coefficient of burning rate which is calculated from measured data, because Eq. (15) shows some deviation from actual values. If, in the combustion region, the temperature profiles influenced by initial grain temperature is assumed as Fig. 1, and the pyrolyzed gas compositions are mainly dependent on the composition of solid composite propellant, heat flux from the burnt gases zone to the solid propellant is constant at steady–state. Then, the average reaction tempera-

ture of Eq. (10) can be expressed as the function of initial grain temperature as follows,

$$q = U\Delta T = U(T_R - T_i)$$
$$T_R = T_i + \frac{q}{U} = T_i + C \tag{16}$$

where q is heat flux and U is overall heat transfer coefficient. As will be stated later, C is a quantity to be determined. Combining Eqs. (16) and (14) makes the burning rate equation as a function of initial grain temperature and combustion chamber pressure is obtained in Eq. (17).

$$\gamma = K\left(\frac{1}{R(T_i + C)}\right)^n \exp\left(-\frac{E_a}{R(T_i + C)}\right)\left(\frac{P}{z}\right)^n \tag{17}$$

5. Comparison with Experimental Results

5.1 Experiments

In order to test the performance of the present model, we have chosen three different types of AP composite propellants (*L*, *M* and *N*) based on CTPB. Major compositions are shown in Table 1. Slurry preparation, mixing and casting have been done by the typical propellant manufacture processes. A vertical one gallon capacity mixer by Baker Perkins was used. Voids were prevented by utilizing a vacuum system both in mixing and casting processes.

Burning rates were measured by the strand–burning method. Even strands were prepared from slicing sections of casted propellant grain. A surge tank was attached to the pressure chamber to maintain constant pressure during burning. The burning rates of the three different propellants were measured for the pressure range of 3 to 20 MPa at the initial temperatures of 233 K, 293 K, and 323 K. The experimental results are presented on the Figs. 2 and 3. Calculated burn-
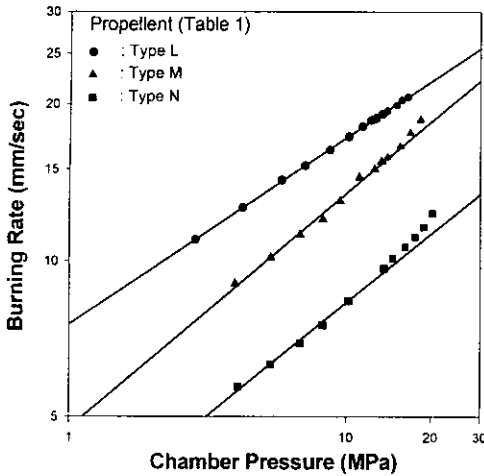
Fig. 2 Relationship between burning rate and chamber pressure according to propellant types
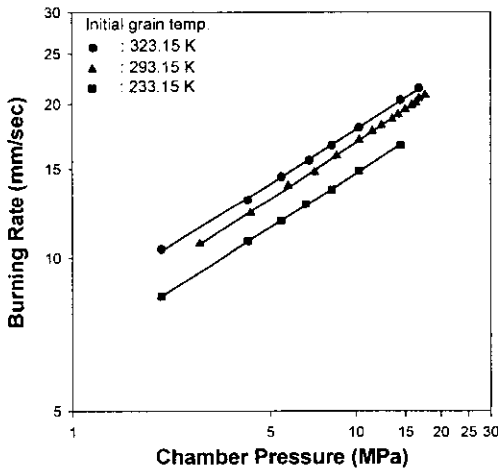

Fig. 3 Relationship between burning rate of propellant L and chamber pressure according to initial grain temperatures. The solid lines show the calculated burning rate by Eq. (15)

ing rates by the present model are discussed below.

5.2 Pressure dependence of the burning rate

If the decomposed gases behave like ideal gases, the compressibility factor of Eq. (17) becomes a unit. Then, the equation is converted into Eq. (18) in a case where the initial grain temperature is fixed

Table 2 Calculated burning characteristics

Propellant	a	n
L	0.0540	0.348
M	0.0155	0.423
N	0.0257	0.407

$$\gamma = aP^n \tag{18}$$

By using the measured burning rates below 10 MPa, the values of a, n for the three different propellants are calculated as Table 2. The calculated burning rates by Eq. (18) are compared with the experimental values in Fig 2. Good agreement between the experiments and the calculated values have been obtained for the pressure range of 2 MPa to 10 MPa. However, the calculated values deviated from the measured ones at pressures higher than 10 MPa, except for propellant L. And the deviation becomes greater as the combustion pressure increases. These results indicate that the decomposed gases participating in the chemical reaction do not behave as ideal gas and the compressibility fact cannot be a unit.

The decomposition mechanism of the same propellants was investigated to obtain the critical temperatures, reduced temperatures, and critical pressures(Hur and Mok, 1984). According to these results and the chart of compressibility factor(Su, 1946), the compressibility decreased and deviated from a unit as the combustion pressure increased between 10 MPa and 39 MPa. Therefore, the deviation of the calculated burning rates from the measured ones in Fig 2. can be explained by more greatly swerving of the compressibility factor from a unit as the increase of chamber pressure.

5.3 Initial grain temperature dependence of the burning rate

Introducing the logarithm on the both sides of Eq. (17) to compute K, C and E_a,

$$\ln\gamma = \ln K + n\ln\left(\frac{1}{R(T_i + C)}\right)$$
$$- E_a\left(\frac{1}{R(T_i + C)}\right) + n\ln\left(\frac{P}{z}\right) \tag{19}$$

Table 3 Combustion characteristics for composite solid propellants L and N

Propellant	K	C(K)	E_a(J/gmol)
L	190	2,040	100,207
N	117	1,530	76,040

Table 4 Radiation temperature according to initial grain temperature

Initial Grain Temperature(K)	Reaction Temperature(K)	
	Propellant L	Propellant M
233.15	2,273	1,763
293.15	2,333	1,823
325.15	2,363	1,853

Applying the initial grain temperatures, 233. 15K, 293. 15K and 323. 15K to Eq. (19),

$$\ln \gamma_{233.15,i} = \ln K + n \ln \left(\frac{1}{R(233.15+C)} \right)$$
$$- E_a \left(\frac{1}{R(233.15+C)} \right)$$
$$+ n \ln \left(\frac{P}{z} \right)_{233.15,i} \qquad (20)$$

$$\ln \gamma_{293.15,i} = \ln K + n \ln \left(\frac{1}{R(293.15+C)} \right)$$
$$- E_a \left(\frac{1}{R(293.15+C)} \right)$$
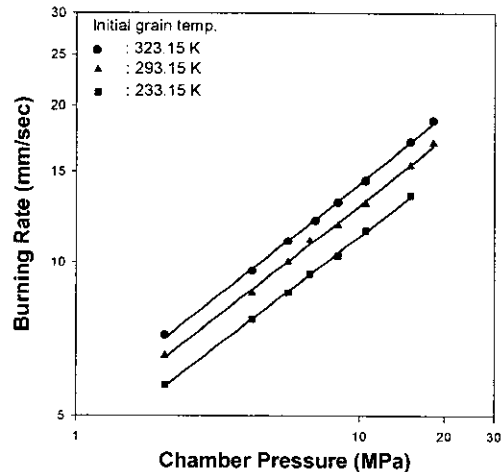$$+ n \ln \left(\frac{P}{x} \right)_{293.15,i} \qquad (21)$$

$$\ln \gamma_{323.15,i} = \ln K + n \ln \left(\frac{1}{R(323.15+C)} \right)$$
$$- E_a \left(\frac{1}{R(323.15+C)} \right)$$
$$+ n \ln \left(\frac{P}{z} \right)_{323.15,i} \qquad (22)$$

Subtracting Eq. (20) from (21) and applying the measured values,

$$l \cdot \left(\frac{E_a}{R} \right) \left(\frac{1}{233.15+C} - \frac{1}{293.15+C} \right)$$
$$= \sum_{i=1}^{l} [\ln \gamma_{293.15,i} - \ln \gamma_{233.15,i}] - l \cdot n \cdot$$
$$\left[\ln \frac{1}{R(293.15+C)} - \ln \frac{1}{R(233.15+C)} \right]$$
$$- n \sum_{i=1}^{l} \left[\ln \left(\frac{P}{z} \right)_{293.15,i} - \ln \left(\frac{P}{z} \right)_{232.15,i} \right] \quad (23)$$

Here, l is a number of experimental data.

Similarly,

Fig. 4 Relationship between burning rate of propellant M and chamber pressure according to initial grain temperatures. The solid lines show the calculated burning rates by Eq. (15)

$$l \cdot \left(\frac{E_a}{R} \right) \left(\frac{1}{293.15+C} - \frac{1}{325.15+C} \right)$$
$$= \sum_{i=1}^{l} [\ln \gamma_{325.15,i} - \ln \gamma_{293.15,i}] - l \cdot n \cdot$$
$$\left[\ln \frac{1}{R(325.15+C)} - \ln \frac{1}{R(293.15+C)} \right]$$
$$- n \sum_{i=1}^{l} \left[\ln \left(\frac{P}{z} \right)_{325.15,i} - \ln \left(\frac{P}{z} \right)_{232.15,i} \right] \quad (24)$$

C and Ea are calculated from Eqs. (23) and (24) by applying the measured burning rates with combustion pressure at the initial grain temperatures 233. 15K, 293.15K and 325.15K. And K is obtained from Eq. (20) by using C and Ea. Table 3 represents the results for propellants

L and N. These results indicate that the average reaction temperatures of propellant L were 2273K, 2333K and 2363K according to the initial grain temperatures and those of the propellant N were 1973K, 1823K and 1853K (Table 4). Figs. 3 and 4 represent the comparison of the experimental burning rates with the calculated data by Eq. (19) for propellants L and N. These figures show a good agreement between the measured burning rates and the calculated ones. In order to test the performance of Eq. (19), the error in the following equation, has been obtained to be with the maximum of 2%.

$$Error(\%)$$
$$=\frac{(measured\ value-calculated\ value)}{measured\ value}\times100$$
$$(25)$$

6. Conclusion

Burning of composite solid propellant has been considered to occur in four distinct phenomena such as decomposition of propellant surface, simultaneous premixing and diffusing, chemical reaction in reaction-flame zone, and gas flow through flame-burnt gas zone. It has been also assumed that chemical reaction could take place in premixing-diffusing zone but were carried out mainly in the reaction-flame zone. All these phenomena taken place in each zone of combustion have been assumed to be steady-state. And a theoretical equation on the burning rate was deduced.

The proposed equation was tested and evaluated vis-a-vis strand burner data for three different types (L, M, N) of solid propellants based on CTPB. The calculated data for the type L containing 16 wt%Al agree very well with the measured ones over the range from 2 MPa to 30 MPa. But the computed data for type M containing 4 wt%Al deviated a little from the measured ones for the range of pressure higher than 10 MPa. The reaction temperature and the activation energy for type L were higher than those for type M. It was found that the deviation of the computed burning rate from the measured

ones ranged within 2%.

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