Defence Science Journal, Vol 48, No 3, July 1998, pp. 287-296

Analysis of Metallised Propellant Ignition Process under Conductive Heating

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ABSTRACT

Ignition of a composite aluminised propellant (AP-HTPB-*Al*) in stagnant hot air is analysed theoretically, based on solid phase and gas phase theories. According to solid phase theory, ignition is due to reaction of the propellant in the solid phase at elevated temperatures. One-dimensional transient solid phase energy equation is solved to obtain the surface temperature profile of the propellant. By gas phase theory, an exothermic gas phase reaction, adjacent to the propellant surface, is considered responsible for the ignition. The changes in temperature and concentrations in the gas phase and the temperature profile below the propellant surface during the pre-ignition induction period are considered. Equations of energy and concentrations of reactants have been solved to obtain the species concentration and temperature profiles in the gas phase. An experimental investigation of the ignition of AP-HTPB-*Al* propellant is also carried out in a shock tube under end-mount conditions. Pressure and temperature ranges were 6-16 bar and 1500-3000 K, respectively. A comparison of the experimental data with predicted results shows that the ignition in an oxidizing atmosphere is by gas phase reaction, whereas in an inert atmosphere, solid phase reaction may be predominant.

NOMENCLATURE

A	Pre-exponential factor for pyrolysis
A_R	Pre-exponential factor for gas phase reaction
C_P	Constant pressure specific heat
C_S	Specific heat of solid
D	Coefficient of diffusion
E	Activation energy
Lv	Latent heat of pyrolysis
$\dot{m}$	Mass flow rate per area
n	Stoichiometric ratio
R	Universal gas constant
T	Temperature
t	Time
Y	Mole fraction
λ	Thermal conductivity
μ	Coefficient of viscosity
v	Volume fraction
ρ	Density

Subscripts

g	Gas phase
s	Solid phase
o	Initial conditions
OX	Oxidizer

Received 18 March 1997, revised 18 November 1997

F Fuel

Al Aluminium

1. INTRODUCTION

Studies on the performance of solid propellants have established that the presence of metal powder in a composite propellant can improve the burning properties considerably[1,2]. On the other hand, metal powder can cause increased ignition resistance due to the increased thermal conductivity and the higher ignition temperatures. It can also cause reduction in evaporation rate of the fuel and oxidiser components due to its shielding effect, causing further resistance. Several ignition theories have been proposed to explain the ignition process in a propellant. Foremost among current theories is the gas phase theory proposed by Mc Alvey[3] which assumes that the propellant ingredients decompose by thermal process and undergo exothermic reaction in the gas phase. The solid phase theory[4] assumes that the rate of ignition process is controlled by the solid phase temperature.

Ignition studies on propellant by convective hot gas have been carried out in shock tube by Bear[5]. Hermance,[6] *et al.* modelled the ignition of an evaporating fuel based on gas phase theory. Kellar,[7] *et al.* studied convective ignition of the propellant, and the results were in agreement with those predicted by thermal ignition theory. The propellant sample ammonium perchlorate-polybutadiene acrylic acid (AP-PBAA) was mounted flush with the wall of a constant area test section of 3 cm length at the end of the shock tube. Kashiwagi[8], *et al.* studied the ignition and flame spread on a solid polymeric fuel in a hot oxidising stream. The problem was analysed theoretically as a boundary layer flow. The ignition was modelled as a gas phase exothermic reaction in boundary layer and a heterogenous reaction on the solid surface. The surface reaction model predicts that the ignition will occur at the leading edge of the flat plate fuel specimen. The gas phase model predicts that the ignition will occur at some downstream position. The experiments were carried out in a shock tube with a flat plate fuel specimen placed parallel to the hot oxidising stream. The results confirmed the predictions of the gas phase model that the ignition always occurs at a distance away from the leading edge. Since a propellant consists of an oxidiser and metal powder apart from the polymeric fuel, the ignition characteristics of the polymeric fuel will not represent that of a propellant. Theoretical and experimental studies on ignition of ammonium perchlorate-polyvinyl chloride (AP-PVC) propellant in stagnant hot gas were carried out by Ramaprabhu and Bhaskaran[9]. Propellant was mounted on the end wall of the shock tube and the results were in agreement with the gas phase theory. Birk[10] developed a model to study the ignition dynamics of homogeneous propellant and discussed ignition delay time and its dependence on pressure, temperature and velocity. Kumar[11], *et al.* developed a comprehensive model and presented numerical solution for the ignition of a propellant in a stagnation region under rapid pressure loading. Reactions in the gas phase and solid phase were considered. Ignition delay variation with pressurisation rate, temperature variation in solid and gas phase, and the effect of particle size were analysed. Huang[12], *et al.* conducted studies on hot fragment conductive ignition of nitramine-based propellants in which stainless steel cylindrical fragment (6.3 mm diameter) heated in a furnace was allowed to fall on the propellant sample kept in a confined enclosure. The effect of ignition temperature and size of the cylindrical fragment, were discussed. The ignition boundaries obtained through experiments were compared with the predicted results.

The purpose of the present study is to model the ignition delay of ammonium perchlorate-hydroxy-terminated polybutadiene-aluminium (AP-HTPB-*Al*) propellant, subjected to conductive heating using gas phase and solid phase theories, and to conduct experiments to confirm their validity. The propellant was mounted on the end flange of the test section flush with the interior so that the stagnant gas behind the reflected shock conductively heated the propellant to ignition. The ignition process in AP-HTPB-*Al* propellant is

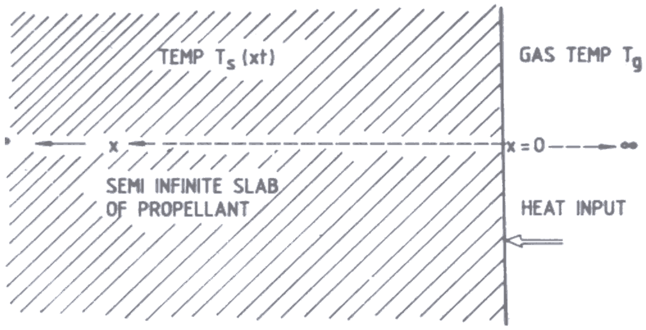

Figure 1. Schematic representation of solid phase model

analysed as a gas phase reaction close to the propellant surface, and the solid phase reaction on the surface of the propellant.

2. IGNITION MODELS

2.1 Solid Phase Ignition Model

According to the solid phase ignition theory, the ignition process is controlled by the temperature in the solid phase. For rapid heating, this would be the temperature of the propellant surface. When the temperature of the solid phase increases by external heat flux, exothermic reaction takes place between the oxidiser and the fuel in the solid phase, causing the temperature to reach a runaway condition. In a composite propellant, the oxidiser particles of random size and very minute particles of *Al* powder were distributed in the fuel binder (HTPB) and it was assumed that the binder, oxidiser and *Al* particles were physically in perfect contact. Because of the complex shape and nonuniform size of the particles, no specific shape was considered for the analysis. The diffusion of the reactants into the gas phase was not considered in this theory and hence, the ignition process was insensitive to the composition of the surrounding gas. The diagrammatic representation of the physical model is shown in Fig. 1.

It was assumed that the propellant surface was uniformly heated by the stagnant hot gases, and the effect of heating penetrates only a small portion of the thickness of the propellant. Hence, the model can be taken as a semi-infinite slab. For further simplification of the model, assumptions are: (i) Thermophysical properties of propellants are not temperature dependent and (ii) Interface temperature of the gas and the solid is the same. The temperature history of the propellant surface can be obtained by solving the energy equation of the solid phase with an additional term for the heat generation due to solid phase reaction as

$$\rho_s C_s \frac{\partial T_s}{\partial t} = \lambda_s \frac{\partial^2 T_s}{\partial x^2} + Q$$

where Q is the heat generated per unit volume of the propellant by condensed phase reaction. The solid phase reaction of AP-HTPB-*Al* propellant is considered as a set of two chemical reactions comprising AP-HTPB and AP-*Al* reactions. It is further assumed that the solid phase reactions are of zero order and are independent of concentrations. Therefore,

$$Q = Q_F A_{SF} \exp(-E_{SF}/R T_S) + Q_{Al} A_{SAl} \exp(-E_{SAl}/R T_S) \qquad (2)$$

Substituting the value of Q in Eqn (1) gives

$$\rho_s C_s \frac{\partial T_s}{\partial t} = \lambda_s \frac{\partial^2 T_s}{\partial x^2} + Q_F A_{SF} \exp\left(-\frac{E_{SF}}{RT_s}\right) + A_{SA1} Q_{A1} \exp\left(-\frac{E_{SA1}}{RT_s}\right) \qquad (3)$$

The boundary conditions are:

At $t \leq 0$,

$$T_S = T_o \text{ for all values of } x \text{ in solid phase} \qquad (4)$$

$$T_g = T_{go} \text{ for all values of } x \text{ in gas phase} \qquad (5)$$

At $t > 0$ and $x = -\infty$ (for solid phase)

$$T_S = T_o \qquad (6)$$

At $t > 0$ *and* $x = \infty$ (in gas phase)

$$T_g = T_{go} \qquad (7)$$

The heat balance at the surface is

$$\lambda_s\left(\frac{\partial T_s}{\partial x}\right)=\lambda_g\left(\frac{\partial T_g}{\partial x}\right) \tag{8}$$

Equation (3) with boundary conditions are non-dimensionalised and solved by the finite difference method. The ignition is assumed to occur when the surface temperature of the propellant reaches a critical value. At this temperature, the removal of the external heat flux does not affect the ignition process since the heat generation rate is greater than the applied heat flux. In the present study, the ignition criterion is taken as 1.5 times the surface temperature, obtained by linear heating as proposed by Price[13], *et al*. It was observed that the ignition delay value was almost independent of the ignition criterion values between 1.15 and 1.5.

2.2 Gas Phase Ignition Model

According to the gas phase ignition model, the propellant vapour evolved from the surface by external heat flux diffuses into the surrounding gas and reacts with it generating heat. The reaction can take place between the vaporised fuel and oxidiser species, either vaporised or present in the test gas. The exothermic reaction in the gas phase leads to the additional heat feedback to the surface, causing further evaporation and reaction which ultimately reaches a runaway condition.

To simplify the analysis, the following approximations are considered:

(a) Within the solid phase, chemical reactions are negligible and the specific heat, thermal conductivity, and density are constant.

(b) The molecular weight and the diffusion coefficient of all the species are same.

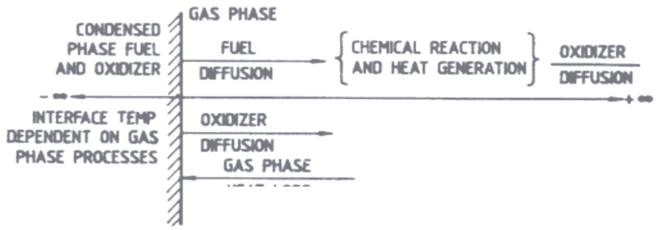

Figure 2. Schematic representation of gas phase model

(c) The gas phase reaction is assumed to be a one-step irreversible chemical reaction of second-order Arrhenius kinetics.

The physical model of the gas phase ignition of the propellant is given in Fig. 2. The mathematical model consists of gas phase and solid phase energy equations coupled by heat balance equation in the interface and species conservation equations.

2.2.1 *Energy Equation*

$$\rho_g c_p\left(\frac{\partial T_g}{\partial t}\right)=\lambda_g\frac{\partial^2 T_g}{\partial x^2}+Q\rho^2 Y_{ox} Y_F A_R \exp\left(-\frac{E_R}{RT_g}\right) \tag{9}$$

The last term in the energy equation accounts for the exothermic reaction of the diffused fuel with the oxidiser.

2.2.2 *Conservation of Fuel Species*

$$\rho_g\frac{\partial Y_F}{\partial t}=\rho_g D\left(\frac{\partial^2 Y_F}{\partial x^2}\right)-\rho_g^2 Y_{ox} Y_F A_R \exp\left(-\frac{E_R}{RT}\right) \tag{10}$$

In addition to the change in concentration due to diffusion, the reduction in the mole fraction of fuel due to consumption by gas phase reaction is considered by the last term in the equation.

2.2.3 *Conservation of Oxidizer Species*

$$\rho_g\frac{\partial Y_{ox}}{\partial t}=\rho_g D\left(\frac{\partial^2 Y_{ox}}{\partial x^2}\right)-n\rho_g^2 Y_{ox} Y_F A_R \exp\left(-\frac{E_R}{RT_g}\right) \tag{11}$$

As in the case of fuel species, the reduction in concentration of the oxidiser species by gas phase reaction is represented by the last term.

2.2.4 *Solid Phase Energy Equation*

It is assumed that there is no exothermic reaction in the solid phase.

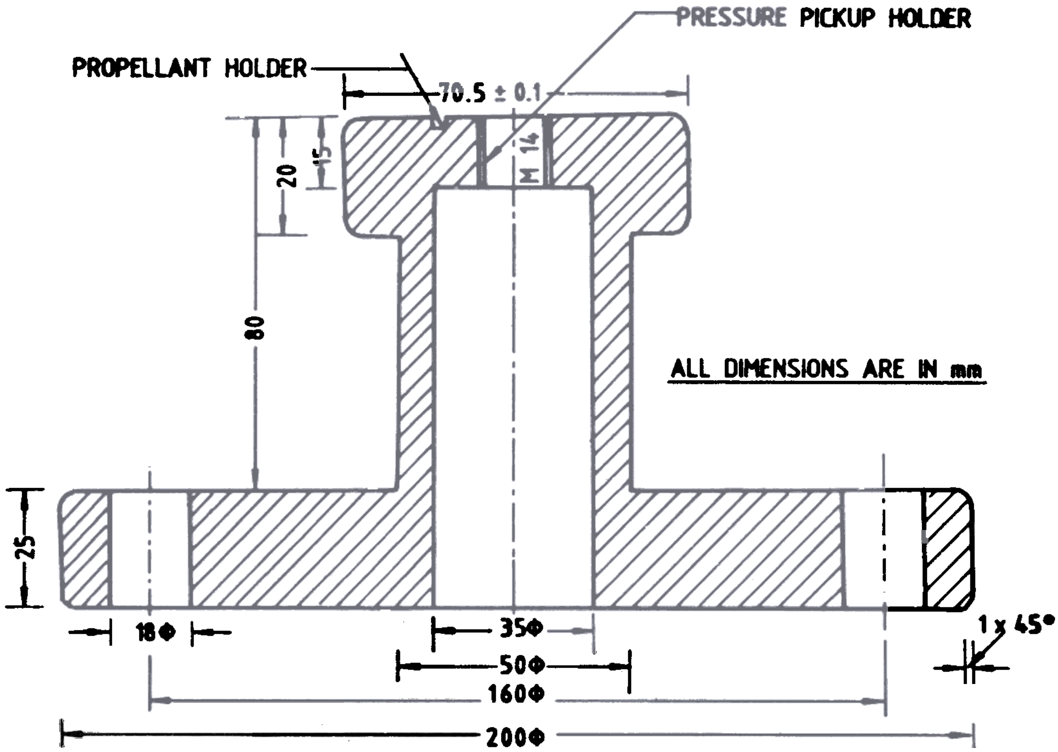

Figure 3. Sectional view of the flange used in shock tube for propellant mounting

$$C_s \rho_s \left(\frac{\partial T_s}{\partial t} \right) + C_s \rho_s V_s \left(\frac{\partial T_s}{\partial x} \right) = \lambda_s \frac{\partial^2 T_s}{\partial x^2} \tag{12}$$

In Eqn (12), the first term describes the rate of accumulation of energy and the second term accounts for the rate of energy convection due to regression of the surface, the sum of which is equated to the energy gain by conduction. The regression velocity $V_S = \dot{m} / \rho_s$, where the mass flux from the surface is approximated by an Arrehenius type pyrolysis law which depends only on the surface temperature. The total mass flux is considered as the sum of independent mass flux from the exposed fraction of oxidiser and binder surface areas.

The gas-solid interface conditions can be represented as

At x = 0

$$\lambda_g \frac{\partial T}{\partial x} = \quad {}_s \frac{\partial T}{\partial x}$$ [illegible]

$$-\rho_g D \left(\frac{\partial Y_{ox}}{\partial x} \right) = \dot{m}_{ox} \tag{14}$$

$$-\rho_g D \left(\frac{\partial Y_F}{\partial x} \right) = \dot{m}_F \tag{15}$$

The boundary conditions are:

At $t \le 0$ and for all values of x

$$T_s = T_{so},\ T = T_{go},\ Y_{OX} = Y_{OXo},\ Y_F = 0 \tag{16}$$

Table 1. Kinetic constants used for the model calculations (AP/HTPB/ *Al*)

Solid Phase Model		Gas Phase Model	
$A_{s,F}$	5×10^{11} 1/s	A_R	8×10^{11} 1/ s
$A_{s,Al}$	$1 - 1.5 \times 10^{11}$ 1/s	E_R	180,000 kJ/kg-mole
$E_{s,F}$	60,000 kJ/kg-mole	$A_{p,F}$	2990 kg/m^2- s
$E_{s,Al}$	9,53,000 kJ/kg-mole	$E_{p,F}$	70,000 kJ/kg-mole
Q_F	2×10^6 J/kg	$A_{p,OX}$	2.925×10^6 kg/m^2-s
Q_{AL}	1×10^7 J/kg	$E_{p,OX}$	132,300KJ/(kg)mole.

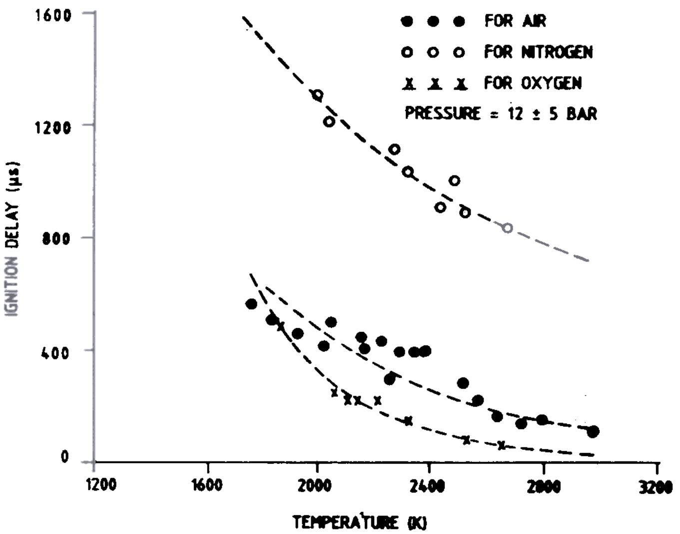

Figure 4. Variation in experimental value of ignition delay with temperature in different environments.

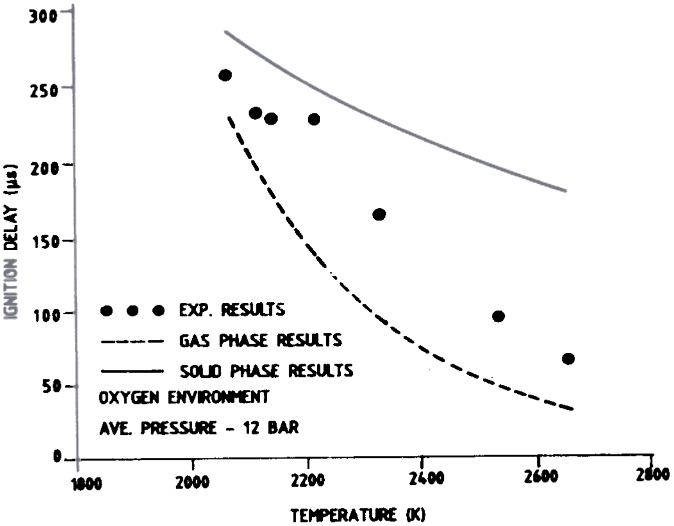

Figure 6. Comparison of experimental and theoretical ignition delay in oxygen environment.

At $t \geq 0, x = +\infty$

$$T_g = T_{go}, \quad Y_{OX} = Y_{OXo}, \quad Y_F = 0 \quad \text{and}$$

$$x = -\infty \quad T_s = T_{so} \tag{17}$$

Above equations are non-dimensionalised and solved by finite difference method.

The theoretical ignition criterion reported by Mc Alvey[3], which states that ignition occurs instantly when the heat loss by conduction is equal to the heat generated by the chemical reaction in the gas phase, is adopted for the present study. The mathematical statement of the ignition criterion is:

$$\frac{\partial T_g}{\partial_t} = 0 \tag{18}$$

The data sets on kinetic parameters used for the theoretical calculation of ignition delay are shown in Table 1. These values were arrived at after trying out several sets suggested in the literature. The

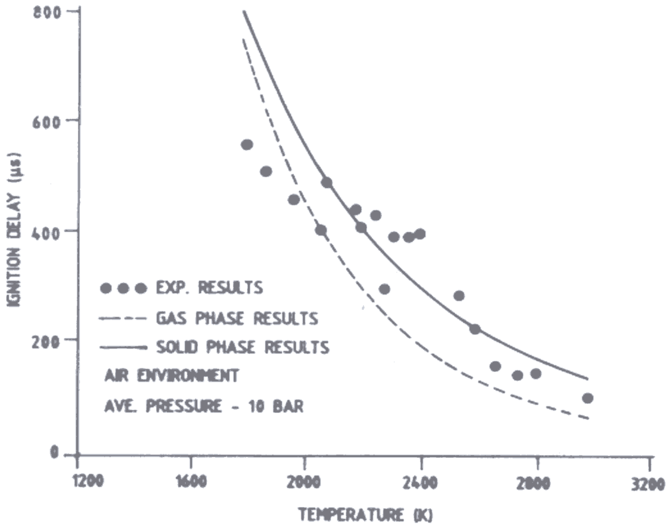

Figure 5. Comparison of experimental and theoretical ignition delay in air environment.

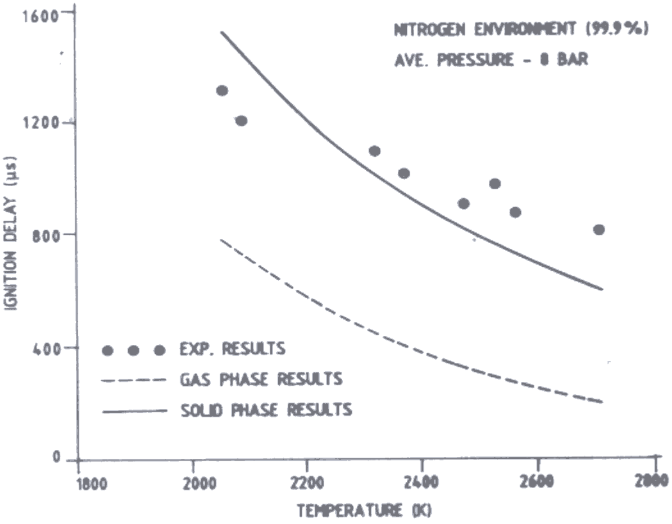

Figure 7. Comparison of experimental and theoretical ignition delay in nitrogen environment.

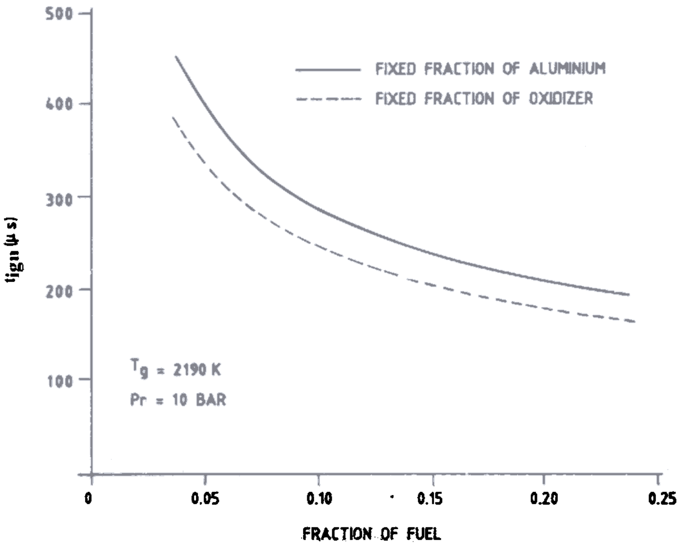

Figure 8. Variation of ignition delay with different fractions of fuel in the propellant in air environment.

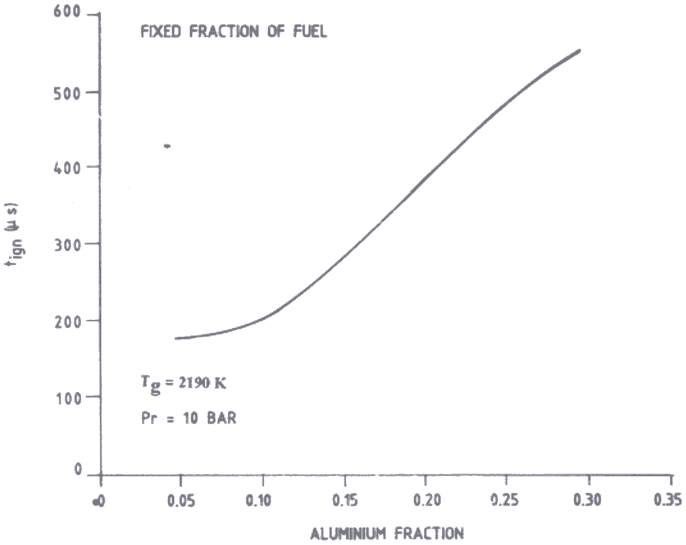

Figure 9. Variation of ignition delay with different fractions of aluminium in the propellant in air environment.

kinetic parameters reported by Cohenm[14], *et al.* and Krishnan[15], *et al* were used for the pyrolysis of HTPB and AP, respectively. The kinetic parameters reported by Longwell and Weiss[16] for the exothermic reaction of oxidiser and the fuel, and by Robert[17], *et al.* for *Al* reactions were taken.

3. EXPERIMENTAL DETAILS

In the present study, shock tube technique was adopted to produce high temperature gas which conductively heated the propellant to ignition. The end flange at shock tube test section was replaced by a stepped aluminium flange, as shown in Fig. 3. The stepped end of the flange protruded 8 cm into the test section. On the surface of the protruded end of the flange, a cubical slot of 10 m x 10 m was provided to fix the propellant sample. There was a provision to fix a piezoelectric pressure transducer at the end of flange. When the shock was reflected at the end flange, the high temperature and pressure stagnant gas behind the reflected shock heated the propellant mounted on the end flange flush with inside to ignition. The light emission due to ignition could be sensed by a photomultiplier through an optical window located 8 cm from the end of the tube.

3.1 Measurement Techniques

Shock velocity was measured with the help of four piezoelectric pressure transducers [PCB Piezotronics, Model-A22] and microsecond electronic counters, using time distance method. Shocked-gas properties were calculated from the shock speed measured, taking into consideration real gas effect. The time interval between the arrival of the shock at the end flange and appearance of visible light due to ignition of the propellant was taken as ignition delay. The arrival of the shock was sensed by the end flange pressure transducer and emission of light due to ignition by a photomultiplier tube (RCA-935A). The signal from the photomultiplier was given to one beam of the CRO and the signal from the pressure transducer was used for triggering the sweep. The ignition delay was measured from the trace of the photomultiplier signal recorded in the CRO. The experiments were conducted in nitrogen, oxygen and air environment. The ranges of test temperature and pressure were 1500 K-2500 K and 8-16 bar, respectively.

4. RESULTS & DISCUSSION

Experiments were conducted to find the ignition delay of the solid propellant by conductive heating in shock tube under end-mount conditions. The effect of various environmental conditions on ignition delay was observed and compared with the corresponding predicted results.

Figure 4 compares the variation of ignition delay obtained from the shock tube experiment with temperature for different environmental conditions. It was concluded that the ignition delay was smaller for higher environmental oxygen concentration. At the temperatures encountered during the ignition process (around 600 K), it was estimated that the rate of decomposition of HTPB was approximately ten times that of AP, and hence, the pre-ignition reaction took place with the help of environmental oxygen which ultimately led to a faster runaway in the gas phase. In inert environment, the ignition would not take place until sufficient concentration of fuel and oxidiser species were reached by pyrolysis. This could be the reason for higher ignition delay observed in nitrogen environment. It was also observed that at lower temperature, the ignition delay in oxygen environment approached that in air environment (Fig. 4). Evaporation rate of the fuel would be lower for lower temperature, causing an oxygen rich condition. The effect of extra oxygen would be to dilute the mixture as in the case of nitrogen in air.

Ignition delay obtained by experiments in air environment was compared with the predicted results in Fig. 5. It is not possible to conclude whether the ignition of solid propellant was achieved by gas phase or solid phase reactions by this comparison. It was clear that at lower temperature range, the experimental values were significantly lower than the predicted values. According to gas phase theory, at lower temperature range the evaporation rate will be lower, reducing the gas phase pre-ignition reaction rate and causing a higher ignition delay. As the temperature is reduced the delay is increased, resulting in longer exposure of the propellant to the hot gas. This leads to thicker thermal layer formation in the propellant and exothermic reaction in the solid phase. The ignition delay variation predicted by gas phase and solid phase theories converge at lower temperature region, indicating that the heat release in the solid phase and the gas phase are identical at lower temperature. Figure 6 compares the experimental ignition delay in oxygen environment with that of predicted values. The plot clearly indicates that the trend of variation of observed ignition delay with temperature is similar to that predicted by gas phase theory, though the absolute values are higher. The results of the ignition experiments conducted in nitrogen environment were compared with the predicted results in Fig. 7. In the oxygen environment, the measured value of ignition delay lies in between the values predicted by gas phase and solid phase theories, whereas in nitrogen environment, the observed ignition delay values were almost in confirmation with the values predicted by solid phase theory. If oxygen was present in the environment, it was possible that the decomposed fuel from the propellant surface reacted with oxygen in gas phase and suppleimented the solid phase reaction.

Figure 8 shows the effect of propellant composition on the predicted ignition delay (gas phase). For the purpose of calculation, it was assumed that the fuel fraction in the propellant was increased (a) by replacing a portion of oxidiser with fuel, and (b) by replacing a portion of *Al* with fuel. In both cases, the plot clearly showed that the ignition delay decreased with increase in fuel concentration. This shows that the decomposition of the fuel and its subsequent reaction with AP is the deciding factor in the ignition of the propellant. Figure 9 indicates that the ignition delay is independent of *Al* concentration up to 10 per cent, and above that, the ignition delay increased with increase in *Al* concentration. At higher *Al* concentrations, the thermal conductivity of the propellant was more, which would delay the attaintment of ignition temperature, resulting in an increased ignition delay. Since the presence of *Al* up to 10 per cent in the propellant does not influence the ignition process of the propellant significantly, it was concluded that AP-*Al* reaction is less significant, compared to AP-HTPB reaction.

5. CONCLUSIONS

The ignition process by conductive heating in a metallised propellant was modelled, based on gas phase and solid phase theories. The ignition experiments were conducted in a shock tube under end-mount conditions in air, oxygen and nitrogen environment. The results were compared with the theoretical results predicted by different ignition theories. The present study concludes that:

(a) In an inert environment, the propellant ignition is controlled by solid phase reaction.

(b) In an environment containing oxygen, the ignition process is also influenced by gas phase reactions. Under such conditions, the exothermic reaction of oxygen with the fuel component is a deciding factor in the ignition of the propellant.

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