J Therm Anal Calorim (2010) 100:331–335
DOI 10.1007/s10973-009-0569-3

Experimental study on ballistic behaviour of an aluminised AP/HTPB propellant during accelerated aging

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Received: 23 July 2009 / Accepted: 13 October 2009 / Published online: 10 November 2009

Abstract The chemical stability of a propellant and its influence on the ballistic properties during aging is a subject of interest. The effect of aging on ballistic properties, viz., ignition delay, burning rate, and heat of combustion for an aluminised ammonium perchlorate–hydroxyl-terminated polybutadiene (AP/HTPB) composite propellant during accelerated aging were investigated. Samples of composite propellants were aged at 60 and 70 °C at relative humidity of 50% in a climatic chamber. The propellant samples were tested with pressurized nitrogen gas environment for ignition delay measurement. Test results indicate that aging does not have any appreciable effect on ignition delay. The change in ignition delay time is less than 3% within the scatter of the data. Experiment results indicate that burn rate do affect with pressure but aging does not have much effect on burn rate. It was also observed that the burning rate at low pressures did not undergo significant changes during the aging period. The most significant of all the ballistic properties of this propellant is the burning rate exponent which increased by about 10% during the aging period.

Keywords AP/HTPB propellant · Ballistic properties · Accelerated aging · Burn rate index

Introduction

Despite the widespread use and long investigation history of ammonium perchlorate (AP)-fuel mixtures, it still can be said that AP/HTPB (hydroxyl-terminated polybutadiene) composites remain among the most confounding materials in research. Thermal analysis is a frequently used tool in propellants' researches [1, 2]. The most important properties that affect the choice of a solid propellant used in different applications are the specific impulse and density. If the application is for a tactical purpose, the significance of burn rate characteristics decides the smoke and flame in the exhaust. Knowledge of the burn rate and a pressure index is greatly helpful in deciding the ability to burn the propellant to be loaded in a volume with minimum free space in a given time. The burn rate–pressure behaviour is many times very complex phenomena best described by Vielle's law. The measurement of the ballistic properties of solid propellant mainly involved the steady-state combustion of the propellant in a vented vessel meant for maintaining the combustion pressure constant. The obtained data are most often correlated by the equation, $r = ap^n$; where a is called the pre-exponent and n is the pressure index of burn rate [3, 4]. In respect of initial temperature dependence, experiments have been conducted by essentially soaking the propellant at a specified temperature (between −40 and +55 °C) for a sufficiently long duration [5]. The procedure using a set of data-analysis definitions specifically treats the effects of variable exponent, non-instantaneous burnout, mean propellant shrinkage, hardware variations, and non-neutral pressure to reduce bias and scatter in measured burning rates was discussed in Ref. [6]. Modern composite propellants consist of inorganic oxidizer, fuel, binder and burn rate modifiers (metal oxides), and other additives (like curing agent). In the propellant used here, oxidizer is AP, the fuel is aluminum (Al) and HTPB as a binder which also acts as fuel. When AP/HTPB-based composite solid propellants are preserved over a long period of time, undergo degradation. The

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degradation of solid propellant is generally the limiting factor in the service life of rocket motor; therefore, it is necessary to understand the mechanisms of the propellant aging process at various storage temperatures. These processes are usually chemical reactions and physical changes in the propellant, and may be different at different temperatures. Attempts made in the past to understand the aging behaviour of composite solid propellants emphasize mainly their mechanical properties [7–9], with only limited understanding of the aging mechanism. A systematic study carried out by Kishore et al. [10] revealed that the rate-controlling process in the aging of composite solid propellants is oxidizer decomposition. There is experimental evidence that the slow chemical reactions inside the propellant cause degradation [11]. As the propellant degrades, its mechanical and ballistic properties may also change over a period of time. Therefore, the degradation and the performance of the propellant can be monitored by measuring the changes in its properties that are either mechanical or ballistic. Since the determination of change in properties of a propellant subjected to natural aging takes a very long time, the propellant is subjected to accelerated aging at elevated temperatures, and the changes in the properties are measured.

The first step in propellant combustion is ignition. Ignition of solid propellant is the process occurring between the application of some external energy stimulus (can be electrical, chemical, or even mechanical) to full-scale combustion. This study focused on the aging behaviour of aluminized AP/HTPB composite propellant by accelerated aging at elevated temperatures of 60 and 70 °C and at 50% RH by observing the changes in the ballistic properties like ignition delay and burn rate variation with pressure. The AP/HTPB composite decomposition and the combustion mechanism were extensively investigated in the last decades and the appearance of advanced methods In this article, the results of experimental investigations carried out on ignition of a composite propellant using thermocouple technique have been presented. Heat of combustion test was conducted for both the aged and unaged propellant samples of 1 g each in a bomb calorimeter. Thermal diffusivity measurements were also carried out using a laser flash apparatus (LFA) at the regular intervals during aging period.

Experimental procedure

Heat of combustion test was conducted for both aged and unaged propellant samples using bomb calorimeter. The sample was pressurized to 25 bar by nitrogen gas in the bomb. Thermal diffusivity measurements were carried out using a laser flash apparatus (LFA) at the regular intervals during aging period. The propellant samples for these measurements were of circular disc-type with 2.5 mm thickness and 12.5 mm diameter. The experiments have been carried to measure the ignition delay period. A new thermocouple technique has been used to measure surface temperature versus time trace during ignition transient. This technique has been demonstrated successfully for the sandwich propellant, Sridhar et al. [12, 13]. In this technique, temperature at propellant surface is directly measured using Pt/13%Rh-Pt (40 μm diam.) thermocouple. In the experimental study, ignition delay, temperatures versus time history during ignition transient of AP-based composite propellant samples have been measured. The samples have been tested at three different pressures, i.e., 1, 5, and 10 bars for measuring the ignition delay. The effect of aging on ignition delay has been investigated.

Under the ballistic testing of the solid propellant, burn rate measurement of the propellant strands has been performed at different pressures. Fuse wire technique has been used to measure burning rate in a nitrogen-purged combustion bomb [14, 15]. Propellant strands of dimension 5 mm × 5 mm × 65 mm have been cut from the composite propellant block. Three nichrome wires of diameter 100 μm were embedded across the propellant strands by keeping the distance between the wires as 15 mm. Strands were coated with resin and titanium-di-oxide on the sides as an inhibitor to ensure linear burning. A propellant holder supported by brass rods and hylum block houses the propellant strands vertically in propellant strand burner. A 24 V auto variable transformer was used for heating the ignition coil. The three fuse wires have been connected to a timer circuit of 2-timer display. Once samples have been ignited, the flame regresses and reaches the first fuse wire; it gets fused and the timer count starts, similarly when flame reaches the second wire, it also gets fused; this stops the first timer count and starts the second timer, i.e., successive breaking of the fuse wires starts and stops the timer count. Thus, the time taken by the flame to reach the fuse wires kept at fixed distance was recorded in the timer circuit. By knowing the distance and the time, the burning rate has been calculated. Burning rate experiments were conducted for the AP/HTPB composite propellant samples drawn at different aging temperatures of 60 and 70 °C at 50% RH.

Results and discussion

Heat of combustion and thermal diffusivity

Experiments have been conducted to find the heat of combustion for both the aged and unaged samples. From all the tests, it was noticed that the change in heat of combustion with respect to aging is insignificant irrespective of the aging conditions. The value varied from 10.121 to 9.845 MJ kg^{-1} of propellant, and these values lie within the error bandwidth of bomb calorimeter experiments conducted on the samples. Thermal diffusivity measurements were also carried out using laser flash apparatus on both aged and unaged samples. But no significant change in diffusivity could be noticed. The diffusivity varied from 0.301 to 0.299 mm^2 s^{-1} during the aging periods.

Ignition delay measurement:

The sample dimensions used in the present investigation are 25 mm length, 6 mm width, and 8 mm depth. For placing the thermocouple, the sample is exactly cut into two-halves and thermocouple is placed on the one half of so that junction exactly lies at the surface. The two wires are kept apart such that included angle between them is more than 90°, Fig. 1. Then the other half is placed on it, retaining its initial shape. Figure 2 shows the surface temperature variation with time for composite propellant sample during ignition transient for surface heat flux, SHF = 7 kW m^{-2} in inert pressurized environment of N_2 gas at 10 bar of gauge pressure. The curve shows a long inert heating time followed by a sudden thermal run-away. Propellant surface temperature increases continuously with time. Marginal increment in temperature with respect to time can be noticed during initial stage of inert heating period. In latter stage, increment in temperature is moderate. At the end of inert heating period, sudden increment in slope indicate thermal run-away. In the present investigation, surface temperature versus time curve for composite propellant consists of three different slopes. The inert heating consists of two slopes. The first slope is of positive value near to zero followed by a slope with moderate value. At the end of inert heating time, slope value becomes near to one as thermal run away take place.

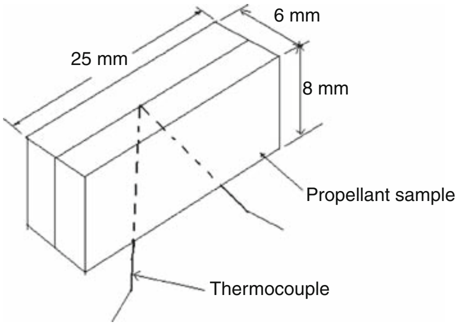

Fig. 1 Positioning of thermocouple in propellant sample for ignition delay measurement

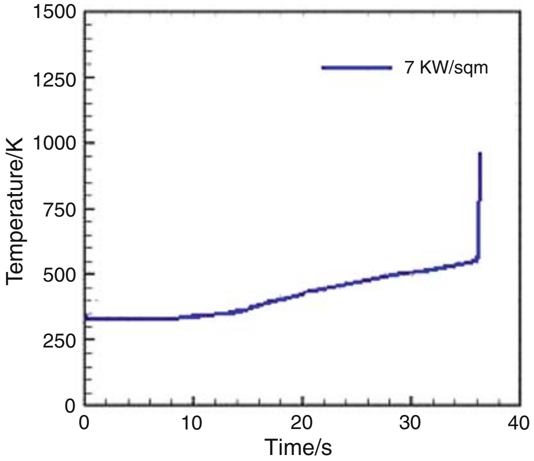

Fig. 2 Surface temperature versus time curve for composite propellant

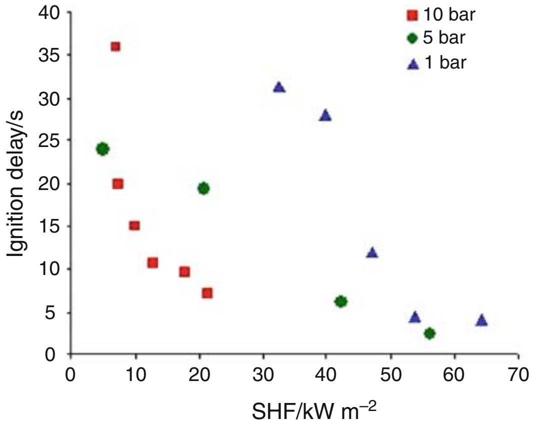

Fig. 3 Effect of pressure on ignition delay of composite propellant

Figure 3 shows measured effect of pressure on ignition delay for various values of SHF. The composite propellant samples were tested at three different pressures, i.e. 1, 5, and 10 bar gauge pressure in inert atmosphere. The SHF values were varied from 7 to 22.67 kW m^{-2} for 10 bar inert pressure, which resulted in ignition delay values of 7–36 s, i.e., 223% in increase of SHF value resulted in decrement of ignition delay by 414%. For 5 bar inert atmosphere SHF value is varied from 5.47 to 56 kW m^{-2}, which resulted in decrement of ignition delay from 24 to 2.5 s (90% decrement). For 1 bar inert atmosphere SHF value is varied from 32.8 to 64.3 kW m^{-2} (95% increment), which resulted in decrement of ignition delay from 31 to 4 s (87% decrement). The chemical stability of a propellant and its influence on the ballistic properties during the period of aging is a subject of interest. The results of ballistic tests during the aging period are presented here. The behaviour of ignition delay with respect to

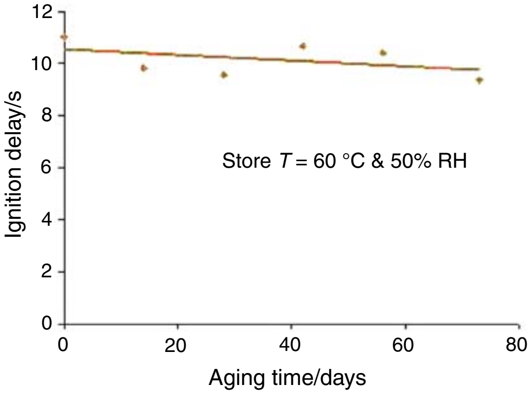

Fig. 4 Effect of aging on ignition delay

aging time is shown in Fig. 4. The incident radiant heat flux to the propellant surface was about 1.3 kW m^{-2}. The propellant sample was tested with pressurized nitrogen gas environment (at 10 bar). Samples of composite propellants were kept at 60 °C and relative humidity of 50% in climatic chamber ignition delay tests were carried out on aged propellant samples using a micro-thermocouple technique. Measured ignition delay for unaged sample was of 10.93 s, which is higher than all aged samples. Ignition delay obtained for 72 days aged sample is 9.1 s. The change in ignition delay time is less than 3% within the scatter of the data. Test results indicate that aging does not have any appreciable effect on ignition delay.

Burning rate measurement

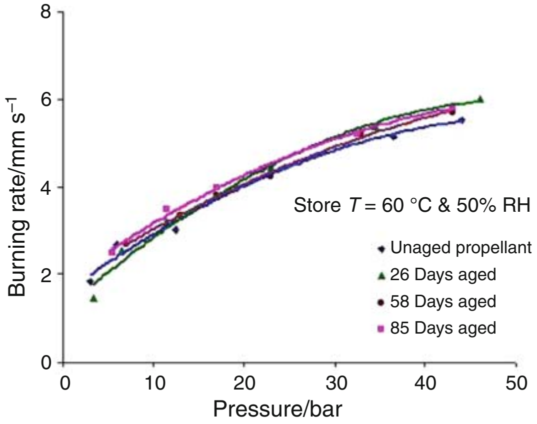

Fig. 5 Effect of pressure on burning rate

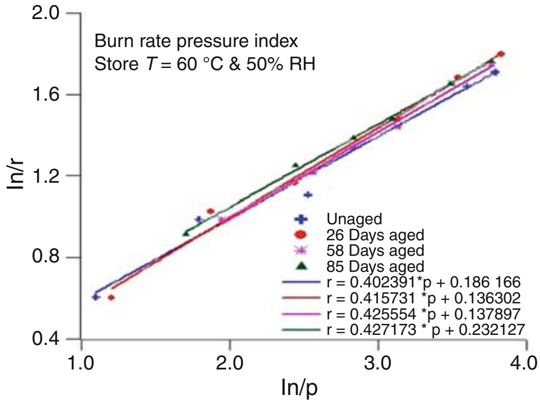

Fig. 6 Effect of pressure on burning rate index at store $T = 60$ °C, 50% RH

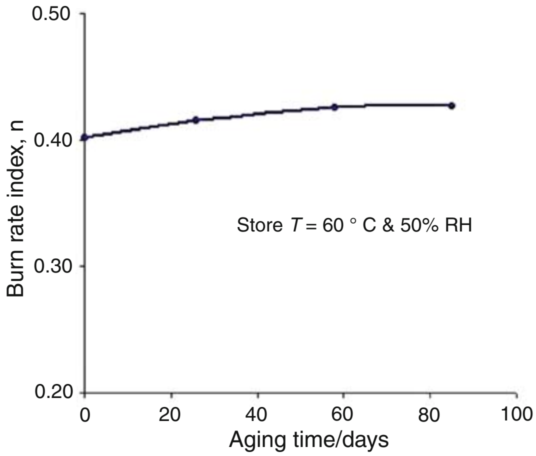

Fig. 7 Effect of aging on burning rate index

Propellant burning rates were determined from strand burner set up. Burn rates depend on the test pressure, initial temperature, and lateral velocity past the surface of the burning propellant (erosive burning effect). The burn rate tests were carried out at 1 atm pressure in nitrogen gas environment. From experimental results, it can be noticed that the change in burning rate due to aging is less than 1%. The pressure dependence was described by Vielle or Saint Robert law, $r = ap^n$ where a is called the pre-exponent and n the pressure index of burn rate. The burn rate dependence on aging was summarized with a single pressure exponent. Pressure–burning rate plot for both aged and unaged propellants is presented in Figs. 5 and 6. Burn rate exponent n values obtained for aged and unaged propellants were 0.386 and 0.424, respectively, for propellant store temperature 60 °C. The burning behaviour is governed by a surface heat flux balance. The heat flux from the gas phase usually increases with pressure because, with increase in pressure, higher density of molecules allows for smaller collision distances, and hence completion of reaction at a shorter distance from the surface. This leads to higher heat flux into the propellant since the flame is closer to the propellant surface. This naturally increases the burning rate with increase in pressure. Burn rate do affect with pressure but aging does not have much effect on burn rate. This is evident from the Fig. 7. From this study, it is observed that the burning rate at low pressures did not undergo significant changes during the aging period.

Conclusions

The effect of aging on ignition delay, burning rate, and heat of combustion was investigated. The following conclusions have been drawn form the present experimental investigation:

- The variation in thermal diffusivity due to accelerated aging was insignificant (<2%).
- The variation of heat of combustion was also less than 2%.
- At low pressures, burning rate did not show any significant change and the change was less than 1%. But at higher pressures (>10 atm), there was some noticeable change.
- An increase of 10% in the burning rate exponent during the aging period was observed.

Acknowledgements The authors wish to thank Mr. Chandel Indrajit Pratapsinh for his assistance in conducting experiments at CAP Laboratories of Department of Aerospace Engineering, Madras Institute of Technology, Chennai, India.

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