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Experimental investigation of the combustion products in an aluminised solid propellant



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ABSTRACT

Aluminium is widely used as an important additive to improve ballistic and energy performance in solid propellants, but the unburned aluminium does not contribute to the specific impulse and has both thermal and momentum two-phase flow losses. So understanding of aluminium combustion behaviour during solid propellant burning is significant when improving internal ballistic performance. Recent developments and experimental results reported on such combustion behaviour are presented in this paper. A variety of experimental techniques ranging from quenching and dynamic measurement, to high-speed CCD video recording, were used to study aluminium combustion behaviour and the size distribution of the initial agglomerates. This experimental investigation also provides the size distribution of the condensed phase products. Results suggest that the addition of an organic fluoride compound to solid propellant will generate smaller diameter condensed phase products due to sublimation of AlF₃. Lastly, a physico-chemical picture of the agglomeration process was also developed based on the results of high-speed CCD video analysis.

1. Introduction

Aluminium particles have been the most commonly used fuel additive in solid propellants [1]. The combustion of aluminium particles produces condensed combustion products (CCP) which are widely distributed within the range of a few microns to hundreds of microns [2]. CCP can be divided into smoke oxide particles (SOP) and agglomerates according to size and source [3]. Sub-micrometric SOP are the products of gas-phase chemical reactions formed around combusting aluminium particles, and SOP can effectively suppress high-frequency oscillation at frequencies of up to 4 kHz [4]. Agglomerates consist of molten aluminium (Al) and alumina (Al₂O₃), whose particle size can reach several hundreds, or even thousands of microns, and agglomerates can effectively inhibit low-frequency oscillations. For example, agglomerate with a particle size ranging from 10 to 30 µm can effectively suppress oscillation at a frequency of 500 Hz [4]. Nevertheless, CCP are responsible for problems such as: slag accumulation, two-phase flow loss, erosion of nozzles, increases in dead mass, and performance loss in motors. Furthermore, those particles without agglomeration at the burning surface, and the agglomerate which detached from the burning surface, will take distributed combustion in the area away from the burning surface. The particle size varies with the extent of this distributed combustion, so the presence of distributed combustion also indicates that the size of CCP released from the burning surface is different from the size of the final combustion products [5,6]. Agglomerates consist of the products which are near the burning surface and the larger final combustion products. The features that differentiate these two types of agglomerates are the degree of oxidation and the particle size [7]. In this paper, the agglomerates which are formed near the burning surface are called initial agglomerates, and the larger final combustion products are called final agglomerates.

The combustion of aluminised solid rocket propellants involves complex physico-chemical processes. Galfetti et al. [8] and Maggi [9] analysed the combustion process of aluminium particles in solid propellants. Babuk et al. [10] proposed the concept of the skeleton layer (SL), and the effect of the SL on the agglomeration of aluminium particles in various propellants was studied. To understand the combustion products of aluminised propellants, many experimental devices have been designed to collect combustion products and measure the size of CCP. In the early work, Dobbins et al. [11] and Sambamurthi [12] used a tank and dart to collect, and measure, plume particle samples: these two methods exerted adverse effects on the size of plume particle samples. Nowadays, a new contact diagnostic tool is used to measure the combustion products [13]; because there is no contact between particles, this optical diagnostic method is widely used

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in the study of particles [14,15]. An actual engine test is expensive, therefore a quenching test was conducted to collect the combustion products [16–18], and the static size distribution of CCP was measured. Maggi et al. [19,20] and Jeenu et al. [21] used high-speed photography and electron microscopy to obtain CCP morphology. All of the aforementioned studies are aimed at investigation of the initial agglomerates after complete combustion. None of them considered the effect of the propellant formulation on the size distribution of the initial agglomerates. However, the size distribution and aggregation / agglomeration process of initial agglomerates affect the combustion process of aluminium in the combustion chamber and exert a more significant effect on the size of the final agglomerates, the distributed combustion process, motor performance, and combustion stability. One of the purposes of this paper is to obtain the size distribution of the initial agglomerates.

Regarding the structure of this paper, firstly the propellant formulation and basic ballistic properties will be discussed. The second part deals with the measurements of the static size distribution and morphology of CCP obtained through the quenching experiment and Scanning Electron Microscopy (SEM). The third part focuses on the dynamic combustion characteristic of the propellants analysed here by use of an optical diagnostic technique. The fourth part presents a physico-chemical picture, of the agglomerate formation at the burning surface and their evolution, obtained by a high-speed video recording system. Finally, a conclusion on current developments closes the paper.

2. Experimental techniques

Several diagnostic techniques have been adopted to study CCP size distribution and aggregation / agglomeration processes, such as burning rate measurement, high-speed photograph experiments, and quenching measurement. Based on *in situ* diagnostic techniques, a special method was chosen to measure the dynamic combustion characteristics of the considered propellants.

2.1. Tested propellants' features

2.1.1. Propellant formulations

The key to solving the problem of two-phase flow loss caused by CCP in the combustion chamber is to improve the ignition characteristics of the aluminium particles and decrease the size of CCP [22]. To reduce the size of CCP, Yavor et al. [23] and Glotov et al. [24] studied the effect of coating on the size of CCP. The research carried out by Deluca et al. [25] and Galfetti et al. [26] showed that the ignition temperature of nano-sized aluminium particles is lower than that of micron-sized aluminium particles, which can improve the ignition characteristics; however, a high content of nano-sized aluminium particles will reduce the specific impulse of a propellant and accelerate its aging, which makes it disadvantageous for application in practice [27,28]. Yavor et al. [29] and Glotov et al. [24] indicated that metalcoating and polymer-coating can shorten the residence time of aluminium particles on the burning surface, but have no effect on the size of CCP.

The ignition characteristics of the aluminium particles can be enhanced, on account of the aluminium fluoride (AlF/AlF₂/AlF₃), which presents a higher enthalpy of formation and whose adiabatic flame temperature can reach 4400 K [30]. For this reason, the propellant used in this experiment consists of: Al, HTPB, AP, and an organic fluoride compound (OF). OF, contenting \geq 60% fluorine, was prepared in our laboratory. This propellant contains a blend of 7– 10 µm sized OF particles, 4–10 µm sized aluminium particles, and two different sizes of AP. In our study, 6% AP in propellant P0 was substituted for the equivalent amount of OF in propellant P1. The propellants P0 and P1 were prepared through homogeneous mixing, vacuum casting, and heat curing. Table 1 summarises the propellant formulations used in this study.

Table 1	
Propellant formulations.	

	Al (wt%)	HTPB (wt%)	AP (wt%)	OF (wt%)
PO	18	15	67	0
P1	18	15	61	6



Fig. 1. Experimental set-up schematic: 1) windowed bomb, 2) high-speed camera, 3) test stand, 4) pressurisation gas inlet valve, 5) pressure regulator, 6) purge gas flow adjusting valve.

2.1.2. Burning rate measurement

The steady burning rate of different propellants was measured in a stainless steel high-pressure combustion vessel similar to that used elsewhere [31] with quartz viewing windows placed opposite each other. The vessel was pressurised with nitrogen, and the pressure was controlled by a pressure feedback system during testing. The steady burning rate was measured within the range of 1–90 atm. The propellant specimen dimensions were, approximately, 30 mm high, 4.5 mm wide, and 4.5 mm thick. A conventional hot-wire igniter technique was used to ensure uniform ignition. The schematic of this experimental set-up is shown in Fig. 1.

2.2. Dynamic combustion experiment

Nowadays, theoretical models are used to predict the size distribution and aggregation / agglomeration process of the initial agglomerates. Four agglomeration models were considered, namely: an empirical model, a pocket model, a physical model, and a pack model. However, the size distribution of the initial agglomerates cannot be predicted by any the four models owing to the processes complexity, such as that contributed by droplet movement on the burning surface and the SL model. To obtain the size distribution of the initial agglomerates, an optical test system was designed. The average distribution data for two different propellants were obtained through use of a Malvern Spectris Analyser. Then the mid-range diameter value (Dv(50)) was obtained. The sketch of the optical test system is shown in Fig. 2, which mainly consists of a Malvern analyser, a protection system, a data acquisition card, a processing system, and a combusting frame. The analyser and the combustion frame were fixed by a steel bar to ensure that the laser beam and burning surface are mutually perpendicular. Before ignition, the distance between the laser beam and the specimen surface was adjusted to 5 mm by the controller, which consists of a synchronous motor and a rail. Upon propellant combustion, the distance between the laser beam and the specimen surface increases, so each data point corresponds to a different

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