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Burning characteristics of ammonium perchlorate-based composite propellant supplemented with diatomaceous earth

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A R T I C L E I N F O

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A B S T R A C T

For composite propellants, solid-phase thermal conduction is one of the dominant processes of propellant combustion and influences their burning characteristics. In this study, diatomaceous earth (DE) was used as a low-thermal-conductivity material, and the influence of DE on the burning characteristics of an ammonium perchlorate (AP)-based composite propellant was investigated.

The ignitability of the propellant was improved by the addition of DE. DE showed both positive and negative effects on the burning rate of the propellant. The negative effect was attributable to the reduction of energy by the addition of DE. The enhancement in the burning characteristics was attributable to the particle shape and size of DE, the catalytic effect of Fe_2O_3, and the physical effect of SiO_2; Fe_2O_3 and SiO_2 are constituents of DE. The mechanism of the physical effect of SiO_2 is as follows. The heat conduction in the solid phase is obstructed by SiO_2 particles in the propellant matrix, and the temperature in the vicinity of these particles becomes higher. Consequently, a hot spot is formed on the burning surface side of the SiO_2 particles, and the burning rate is then increased. Further, the hot spot effect was dependent on the AP interparticle distance in propellant matrix and the specific surface area of AP.

1. Introduction

Solid propellants are commonly used as solid fuels for rockets and missiles. Composite propellants are solid propellants that consist of oxidizer crystals, binder, curing agent, metal fuel, burning catalyst, and other components. Propellants with a high burning rate that generate a large quantity of combustion gases in a short period of time are required to realize high-performance rocket motors that would enable rockets to fly at high speeds. On the other hand, propellants with a low burning rate generate low thrust, and for example, are used as a gas generator for controlling vehicle flight. Therefore, many studies have been directed toward the development of propellants with wide range of burning rates, especially high burning rates.

Ammonium perchlorate (AP) and hydroxyl-terminated polybutadiene (HTPB) have been widely used as an oxidizer and a binder, respectively. This is because AP/HTPB-based propellants have excellent burning and mechanical characteristics. The burning rate of an AP-based composite propellant depends on the AP particle size and AP content; it increases with a decrease in the AP diameter and an increase in the AP content.

The burning process of the AP/HTPB composite propellant begins with the production of the decomposition gases of AP and HTPB at the burning surface with the heat fed back from the flame and the heat generated at the burning surface by AP thermal decomposition. The heat is conducted from the burning surface to the solid propellant and preheats the unreacted solid phase. These decomposition gases diffuse and mix in the gas phase and finally burn.

It is important to study the combustion flame structure of a propellant to control its burning rate. The combustion mechanism of AP-based composite propellants has been investigated for many years and is almost established [1]. However, sufficient experimental data required to describe the combustion mechanism have not yet been collected.

The thermal conduction process in solid propellants is one of the dominant processes of solid propellant combustion and influences the burning characteristics of the propellants. Metal wires embedded in a propellant increase its burning rate, thereby improving the thermal conduction in the solid phase [2–7]. However, SiC powder, which is a fine and high-thermal-conductivity material, does not affect the burning characteristics of propellants [8]. To study the influence of variations in solid-phase thermal conduction on the burning characteristics of a propellant, it is necessary to investigate the burning characteristics of a propellant supplemented by not only high-thermal-conductivity materials but also by low-thermal-conductivity materials.

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Nomenclature

Chemicals

AP	ammonium perchlorate
DE	diatomaceous earth
HTPB	hydroxyl-terminated polybutadiene

Acronyms

CAP	coarse AP
DTA	differential thermal analysis
FAP	fine AP
GDE	ground DE
SEM	scanning electric microscopy
TG	thermogravimetry

Symbols

D_w	weight mean diameter
I_{sp}	specific impulse
L_p	AP interparticle distance in propellant matrix
R	ratio of burning rate of AP/DE propellant to that of corresponding AP propellant
T_0	temperature of unreacted propellant
T_1	final combustion temperature
T_f	adiabatic flame temperature
T_P	exothermic peak temperature on DTA curve
λ	thermal conductivity
ξ	amount of DE added to propellant

A low-thermal-conductivity additive must be unburnable because a burnable material generates heat by combustion, thereby increasing the burning rate. Kevlar fiber is an unburnable and low-thermal-conductivity material. It is used for increasing the burning rate of propellants by addition in small amounts [9]. This is because Kevlar fibers protruding through the propellant surface and into the gas phase have a “flame-holding” effect due to the shape of Kevlar. Reports of this nature have been hardly published to date. In this study, diatomaceous earth (DE) was used as an unburnable and low-thermal-conductivity material. Furthermore, DE is a powder, is cheap, and can be easily handled. The purpose of this study is to investigate the influence of DE on the burning characteristics of AP-based propellants.

The addition of an unburnable material reduces the propellant performance by decreasing the energy density of the propellant. This study is part of a series of studies on the influence of variations in solid-phase thermal conduction on the burning characteristics of propellants, and is not an attempt to obtain high-performance propellants.

2. Experimental

2.1. Sample ingredients

Coarse AP (CAP) and fine AP (FAP) were used as oxidizers in this study. CAP was prepared by grinding commercial AP (Kanto Kagaku) for 5 min in a vibration ball mill. FAP was prepared by the freeze-drying method [10]. The mean particle diameters of CAP and FAP were about 100 and 4 μm, respectively. DE (Kanto Kagaku) was used as a low-thermal-conductivity material. Figure 1 shows an SEM image of DE. The DE sample had a lot of porous particles with a heterogeneous shape. The specific surface area of DE was 27 $m^2 g^{-1}$. The main ingredients of DE are SiO_2, Al_2O_3, and Fe_2O_3. Therefore, these materials were used as additives. SiO_2 was supplied by Denka Kagaku Kogyo, Al_2O_3 by Association of Powder Process Industry & Engineering, and Fe_2O_3 by Kanto Kagaku.

For a propellant with a high AP content, variations in solid-phase thermal conduction due to the addition of DE would not be detected because of a large quantity of heat feedback from the gas phase and heat generated by the exothermic decomposition of AP at the burning surface. In this study, fuel-rich propellant samples with 75%, 70%, and 65% AP were prepared. HTPB was used as a binder and was cured by the addition of 8% isophorone diisocyanate (Tokyo Kasei). DE was added to the propellants in amounts ranging from 0.2% to 12%. The amount of DE added to a propellant is represented by ξ.

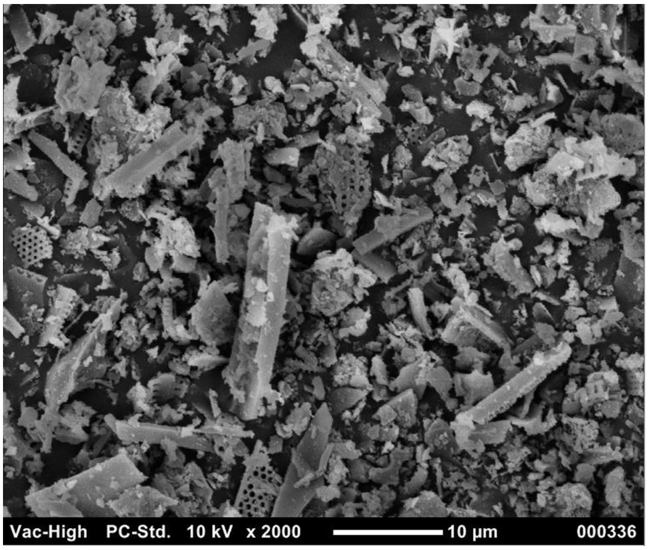

Fig. 1. SEM image of DE.

For preparing propellant samples, HTPB binder and additives were first sufficiently mixed in a polyethylene container. AP was then added to this mixture and mixed manually till a viscous slurry of the uncured propellant was obtained. The mixing temperature was 333 K. The uncured propellants were poured into a steel container and were degassed under vacuum. Subsequently, the degassed samples were cured in an oven at 333 K for one week. The density of AP is larger than that of the binder. If the volume of HTPB is larger than that of AP, AP particles would be precipitated at the bottom of the propellant. Therefore, an uncured propellant with a low AP content was rotated slowly during the curing process to avoid the separation of AP and HTPB. Three batches of propellants were prepared with the same propellant formulation.

2.2. Estimation of theoretical propellant performance

The specific impulse (I_{sp}), adiabatic flame temperature (T_f), and combustion products of the propellants were theoretically estimated in this study. The theoretical performance of the propellants was calculated using the NASA CEA program [11] with a combustion pressure of 7 MPa, an exit pressure of 0.1 MPa, and an initial temperature of 298 K. The theoretical performance was calculated at frozen equilibrium and optimum expansion. The standard en-