



# Thermal decomposition behaviors and burning characteristics of ammonium nitrate/polytetrahydrofuran/glycerin composite propellant

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## ABSTRACT

Ammonium nitrate (AN)-based propellant is plagued by several drawbacks: low burning rate, poor ignitability, and low energy. The application of an energetic binder is an effective approach to the practical use of AN-based propellants but has not been used to date because the synthesis processes are complicated and expensive. Polytetrahydrofuran (PTHF) is a raw material for rubber products and is not an energetic material. The repeating unit of PTHF has one oxygen atom, and although PTHF could not become solid with use of a curing agent alone, PTHF became rubbery with the use of glycerin as a cross-linking modifier. Thus the PTHF/glycerin mixture would be a useful binder to improve the burning characteristics of AN-based propellants, due to the oxygen atoms in the mixture. The use of PTHF/glycerin binder is theoretically effective for enhancing the performance of AN-based propellant, but the combined AN/PTHF/glycerin propellants failed to ignite; they only generated a flashing flame at the ignition event. The ignition failure of the AN/PTHF/glycerin propellant occurred because the melted binder covered the burning surface and interfered with the evolution of the AN decomposition gases and the heat flux feedback from the flame to AN. The AN/PTHF/glycerin propellant was then burned with the addition of a catalyst, and the effect of PTHF/glycerin binder on the improvement of burning characteristics was apparent. The PTHF/glycerin binder was an effective binder material for improving a performance of the AN-based composite propellant.

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## 1. Introduction

Ammonium perchlorate (AP)-based composite propellants are widely used because this type of propellant not only has excellent burning characteristics but also good processability and storability. One of the few serious drawbacks of AP-based propellants is that the products of its combustion, which include HCl, chlorine, and chlorine oxides, cause atmospheric pollution. Because HCl serves as a nucleation site for water vapor, chlorine must either be eliminated from the propellant or scavenged from the exhaust products to eliminate secondary smoke. To overcome these environmental problems and to meet needs for low-hazard, low detectability (smoke-less) propellants, numerous chlorine-free propellant systems have been developed.

Various attempts at eliminating chlorine include the use of nitramine (HMX and RDX) and other oxidizers (HNF ADN, and AN) in place of AP [1–12]. These attempts have led to a rejuvenated interest in AN-based propellants, primarily because of its low cost and easy availability. However, the use of AN as a propellant

ingredient is not without some significant technical challenges that must be overcome before its use is viable. Some of the major problems of AN-based propellants are its low burning rate, poor ignitability, low energy, and phase transition of AN.

The phase-stabilized ANs have been developed to overcome the problem of phase transition [13–16]. Numerous approaches adopted to improve the burning characteristics of AN-based propellants include the use of catalysts [17–20], the addition of metals [21–25], and the application of azide polymer-based energetic binders [26–30].

The application of an energetic binder is one effective approach that could be applied to AN-based propellants. However, the synthesis processes of the energetic binders are complicated, so it is difficult to manufacture these binders industrially. To date, these energetic binders have not been used for practical applications because they are expensive.

Polytetrahydrofuran (PTHF) is a raw material for rubber products that has been produced on a commercial basis. Table 1 shows the chemical properties of PTHF and hydroxyl-terminated polybutadiene (HTPB), a widely used binder. The structure of PTHF is similar to that of HTPB. The specific impulse and burning rate characteristics of the AP/HTPB composite propellant could be improved by the use of PTHF as a binder by more than the use of HTPB because there is oxygen in the repeating unit of PTHF [31–33].

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## Nomenclature

ADC	ammonium dichromate	$n$	pressure exponent
ADN	ammonium dinitramide	$p$	pressure, MPa
AN	ammonium nitrate	PTHF	polytetrahydrofuran
AP	ammonium perchlorate	PTHF1	PTHF of Mn = 650
DTA	differential thermal analysis	PTHF2	PTHF of Mn = 1400
HMX	cyclotetramethylene-tetranitramine	PTHF3	PTHF of Mn = 2900
HNF	hydrazinium nitroformate	$r$	burning rate, mm s <sup>-1</sup>
HTPB	hydroxyl-terminated polybutadiene	RDX	cyclotrimethylene-trinitramine
IPDI	isophorone diisocyanate	SEM	scanning electric microscopy
$I_{sp}$	specific impulse, Ns kg <sup>-1</sup>	$T_f$	adiabatic flame temperature, K
$k$	constant	TG	thermogravimetry

PTHF is not an energetic material, although PTHF improved the performance of the AP-based propellant.

It was expected that PTHF would be a useful binder ingredient to improve the burning characteristics of AN-based composite propellants. This study investigated the thermal decomposition behaviors and burning characteristics of AN-based composite propellants with the use of PTHF as a binder ingredient and compared these results with those of the AN/HTPB composite propellant.

## 2. Experimental

### 2.1. Sample ingredients

PTHF and HTPB (R45M) were used as binder materials. HTPB is a widely used binder, used in this study to compare with the burning characteristics of the PTHF-based propellant. PTHF is produced in several different molecular weights. Three kinds of PTHF were used in this study; the molecular weights of the PTHF samples were 650, 1400, and 2900, designated as PTHF1, PTHF2, and PTHF3, respectively. The number of the symbol increased with the increasing molecular weight. PTHF was supplied by DuPont.

Isophorone diisocyanate (IPDI, Tokyo Kasei) was used as the curing agent. PTHF did not become solid with the use of isophorone diisocyanate alone as the curing agent [33], but did become sufficiently rubbery for use as a binder with the addition of glycerin as a cross-linking modifier. Glycerin was supplied by Kanto Chemical. Table 2 shows the binder formulations. The PTHF/IPDI/glycerin mole ratio of these systems was 1.00/1.50/0.33 [33]. The binder mass formulations varied with the molecular weight of PTHF.

AN (Kanto Chemical) was used as the oxidizer; it was ground with a vibration ball mill for 5 min. Ammonium dichromate (ADC, Junsei Chemical) was used as a burning catalyst and was added to the propellant at 6% [17]. The weight mean diameters of AN and ADC were 125 and 61  $\mu\text{m}$ , respectively.

### 2.2. Measurement of thermal decomposition

The thermal decomposition of the propellants was measured by differential thermal analysis (DTA) and thermogravimetry (TG)

**Table 1**  
Chemical properties of binder ingredient.

Binder ingredient	PTHF	HTPB
Molecular structure	HO-( $-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{O}-$ ) $_n$ -H	HO-( $-\text{CH}_2-\text{CH}=\text{CH}-\text{CH}_2-$ ) $_n$ -OH
Density (g cm <sup>-3</sup> )	0.970–0.981	0.902
Heat of formulation (kJ mol <sup>-1</sup> )	-219.2	-21.1

**Table 2**  
Binder formulations.

Binder	Mass fraction (%)			
	HTPB	PTHF	Glycerin	IPDI
HTPB	99.26	0	0	0.74
PTHF1/glycerin	0.0	64.1	3.0	32.9
PTHF2/glycerin	0.0	79.4	1.7	18.9
PTHF3/glycerin	0.0	88.9	0.9	10.2

using Rigaku Thermo plus TG-DTA8120. The equipment was operated under flowing nitrogen (0.5 dm<sup>3</sup> min<sup>-1</sup>) at atmospheric pressure. DTA and TG were carried out with a heating rate of 20 K min<sup>-1</sup>. The sample weight was 1–2 mg. The TG-DTA measurements were conducted more than four times for each sample. The averages of the TG-DTA curves from the data were used in this experiment.

Decomposition phenomena of AN, binders, and propellants were observed with a hot plate heated from room temperature to 636 K at a heating rate of about 12 K min<sup>-1</sup> under atmospheric pressure. The temperature at the surface of the hot plate was measured with K-type thermocouples. AN was in powder form, and the size of binder and propellant was 6 mm  $\times$  6 mm  $\times$  3 mm. The sample was placed on a glass plate, which was placed on the hot plate.

### 2.3. Burning rate measurement

Each propellant strand was 10 mm in diameter and 40 mm in length. The burning behavior was investigated in a chimney-type strand burner (Kyouwa Kougyo KRS-RG-6085) that was pressurized with nitrogen. Each strand was ignited by applying 10 V to an electrically heated wire threaded at the top. Each propellant strand was combusted in the pressure range from 0.5 to 7 MPa at the initial temperature of 288 K. The burning phenomenon of the propellant was recorded by means of a high-speed video recorder (Photron FASTCAM-NET1000C). The burning rate was measured from the pictures from this high-speed video recorder. The combustion phenomenon was recorded with a shutter speed of 125 frames s<sup>-1</sup>. All measurements were checked in triplicate at each pressure, and average values were used in data analysis. The dispersion of data was less than 10%.

## 3. Results and discussion

### 3.1. Theoretical propellant performance

Theoretical calculations are useful as a means of evaluating and comparing the performance of various propellant formulations

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