



# Decomposition characteristics of an elemental sulfur doped polysulfide based ramjet fuel

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## ABSTRACT

The decomposition characteristics of five candidate polysulfide (LP-33) based ramjet solid fuel formulations are evaluated using thermogravimetric analysis and differential scanning calorimetry. The formulations are variants on the polymer crosslink mechanism and the doping mass fraction of elemental sulfur. Four fuel candidates are oxidatively cured and one formulation is cured with isocyanates. Post-cure, the Shore-A hardness of each sample is measured followed by TGA and DSC analysis. The objective of this research is to develop a solid fuel ramjet formulation with thermal decomposition onset temperatures lower than conventional ramjet fuels while maintaining a Shore-A hardness of at least 30. The results show that polysulfide based formulations decompose at temperatures approximately 120 °C lower than hydroxyl-terminated polybutadiene based fuels. Additionally, the results demonstrate that the decomposition onset temperature, and the heat of decomposition can be further reduced with the addition of elemental sulfur.

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## 1. Introduction

A primary consideration for solid fuel ramjet design is the decomposition temperature, the heat of decomposition, and the ignition characteristics of the fuel. As the decomposition and autoignition temperature of the fuel increases, the ramjet inlet recovery temperature at ramjet takeover must also increase to assure ignition of the solid fuel grain, if no secondary ignition source is to be used. To increase the inlet recovery temperature, the booster burnout velocity must also increase which translates into an increase in the required booster propellant mass and total impulse. Thus, the booster mass is linked to the ramjet's inlet thermal and pressure conditions at ramjet takeover. In addition to ignition at takeover, ramjet flame-out during high angle of attack maneuvers is another area of design consideration. Re-ignition of the extinguished fuel grain by free stream enthalpy alone, once the maneuver is complete, is a desirable feature. As with ignition at takeover, there is much advantage to ramjet fuel formulations that minimize the decomposition and autoignition temperature of the fuel grain in air.

The most prominent ramjet fuel formulations are based on the polymer hydroxyl-terminated polybutadiene (HTPB). This polymer provides excellent mechanical properties and aging characteris-

tics across temperature ranges that are typical for tactical military weapon systems. Thermochemical analysis and extensive test data shows that the polymer also produces high specific impulse values. Considerable research has been conducted in the development of high energy HTPB ramjet fuels to include formulations that contain aluminum and boron particulate up to mass fractions of 30–40% [1–4]. The decomposition temperature of HTPB is approximately 425 °C with a specific gravity of 0.913. Without metal additives, the specific gravity of HTPB produces a fuel with a low impulse density even though the specific impulse is high among typical fuel polymers. Maximizing impulse density is a performance characteristic that is of prime importance in ramjet design. Metal additives can potentially greatly increase the energy density of the fuel but to achieve good combustion efficiencies, high chamber temperatures, high chamber residence times or secondary air dumps are required. This problem is exacerbated at low equivalence ratios where the chamber temperature is well below that which is required for efficient aluminum or boron combustion. Additionally, thermochemical analysis shows carbon as the primary species of decomposition of HTPB which is an excellent fuel, but has an autoignition temperature in air near 700 °C.

The design of a high performance low cost and low weight ramjet necessitates that the fuel have a high impulse density and operate efficiently in a simple ramjet cycle over a range of equivalence ratios. In addition, a highly desirable feature is that the fuel should autoignite by free stream enthalpy at minimal inlet recovery temperatures allowing for the elimination of an igniter, and

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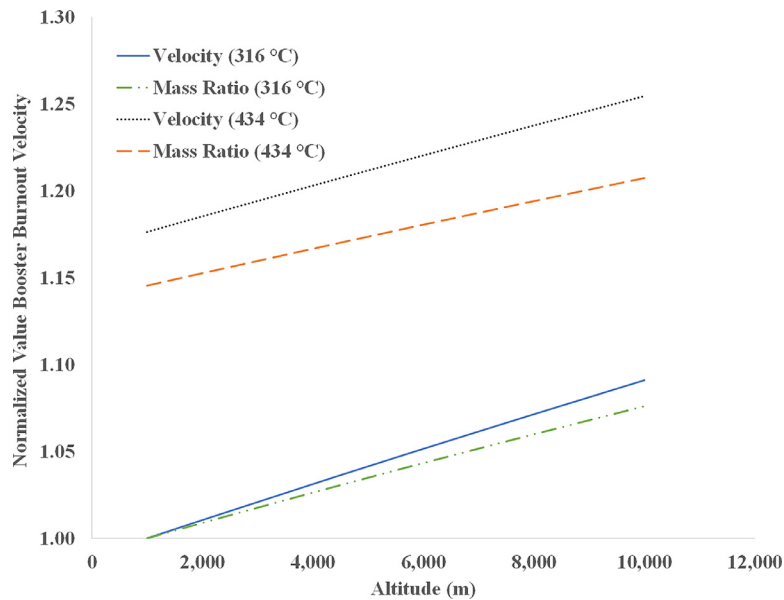


Fig. 1. Effect of fuel decomposition temperature on propellant mass fraction.

re-ignite should extinguishment occur during maneuver. Naphthalene has been investigated and used successfully as a ramjet fuel having a low decomposition temperature and high density [5,6]. As with HTPB, naphthalene decomposes into approximately 75% solid carbon with an autoignition temperature similar to HTPB and has a propensity to produce significant amounts of soot [7]. Figure 1 demonstrates the performance advantage of minimizing the decomposition temperature of the ramjet fuel, assuming that the autoignition temperature is near the decomposition temperature. In the figure, two fuel formulations are considered with the first having a primary decomposition temperature of 316 °C and the second having a primary decomposition of 434 °C representing polysulfide and HTPB based fuels respectively. The plot shows the normalized booster burnout velocity, as a function of altitude, that must be obtained to provide the inlet recovery temperature needed to decompose the respective propellant. Using the ideal velocity equation, the normalized propellant mass fraction is calculated for each corresponding velocity point. These calculations assume a booster  $I_{sp} = 250$  s and are independent of the boost propellant burn rate. The normalization is relative to the 316 °C lowest altitude data point. The graph shows that when the decomposition temperature is reduced from 434 °C down to 316 °C the propellant mass fraction is reduced by 14–20% across the considered altitude range. The objective of the figure is to show the additional energy that must be carried in the booster via propellant mass such that the minimum ramjet take-over velocity for autoignition is achieved at booster burnout. Ramjet  $I_{sp}$ 's have the potential to be 3–4 times that of a solid rocket propellants and thus quick conversion to ramjet operation can be a significant system performance advantage.

An initial screening of candidate fuel polymers was conducted using TGA to determine the decomposition peak mass loss temperature [8]. The results of this screening are shown in Fig. 2 and in Table 1. The survey considers several polymer materials that are commonly used as solid propellant binders to include HTPB, Poly(ethylene glycol), and polysulfide. Also considered are several materials that have been considered to be potential ramjet fuels by other researchers to include Poly(methyl methacrylate), naphthalene, and wood. The results show that naphthalene has the lowest decomposition temperature of materials tested, but its low melting point is undesirable for a tactical munition system. Wood also decomposes at low temperatures as compared to the standard HTPB

Table 1

Decomposition peak mass loss temperatures for potential ramjet fuel polymers.

Polymer	Peak mass loss temperature (°C)
HTPB	477.1
Polysulfide	316.0
Naphthalene	158.2
Polyethylene glycol (PEG)	408.7
Polyethylene terephthalate (PETE)	477.1
Polymethyl methacrylate (PMMA)	382.8
Polypropylene	462.5
Polystyrene	441.6
Wood	297.3

and has been considered by some as a low cost ramjet fuel, however it has a low stoichiometric air to fuel ratio which minimizes ramjet performance at higher air to fuel ratios where the benefits of an air breathing engine are realized. Polysulfide (bis-[ethylene oxy] methane) shows a sharp peak mass loss temperature at approximately 316 °C.

Presented in this paper are thermogravimetric (TGA) and differential scanning calorimetry (DSC) analyses of five polysulfide fuel formulations. Four formulations are doped with elemental sulfur up to a mass fraction of 6.8%. For comparison, one formulation is cured with isocyanate and does not contain any additional elemental sulfur. The results show that decomposition characteristics of the polymer can be altered as a function of the sulfur content to included broadening the temperature range of the decomposition process, lowering the heat of decomposition, and decreasing the peak mass loss temperature. The DSC results show the presence of an exotherm developing at temperatures prior to the peak mass loss temperature of the material with an amplitude that correlates with the sulfur doping fraction.

## 2. Experimental results

### 2.1. Polymer material selection and curing

The liquid polysulfide polymer LP-33 was purchased from Toray Fine Chemicals Company of Tokyo, Japan. LP-33 is a polymer of bis-(ethylene oxy) methane with mercaptan terminal end groups [9].

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