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ABSTRACT

While shedding light on green space propulsion, the development of non-toxic hypergolic bipropellants has provided alternatives to the conventional, highly toxic propellants. Non-toxic energetic hydrocarbon fuels promoted by sodium borohydride were produced, and their hypergolicity with rocket grade hydrogen peroxide was confirmed. Ignition delays ranged from 4 ms to 17 ms, depending on the oxidizer concentration. A 500 N scale thruster utilizing a non-toxic hypergolic bipropellant combination was also developed. With an oxidizer-to-fuel ratio of 5.48, the injector was designed with pentad unlike-impinging jets, one fuel jet orifice and four oxidizer jet orifices for enhancing the mix and atomization of the hypergolic propellants. The operation test was performed, and burning time was established to be 3.5 s. For the demonstrator, the rising time was approximately 158 ms, and the pressure oscillation inside the combustion chamber was approximately $\pm 7.69\%$. These results indicate the feasibility of this novel concept engine for future space missions.

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1. Introduction

Liquid-fueled propulsion systems have been developed over the past 100 years, and monopropellant or bipropellant chemical propulsion thruster systems have undergone several specific changes. The latter is the most commonly used in a variety of civilian and military applications, and in an attempt to increase its performance, various propellant combinations have been tested since Robert Goddard's first rocket. Above all, the hypergolic ignition system has been widely adapted for space propulsion applications, where reliable and repetitive ignition is essential.

A hypergolic bipropellant is a particular combination of fuel and oxidizer that ignites spontaneously upon contact. However, in the existing hypergolic propulsion systems, the use of hydrazine or its derivatives as a fuel, along with nitrogen tetroxide or nitric acids as an oxidizer, is indispensable, even if they are extremely toxic, corrosive, potently carcinogenic to humans and detonable. These utmost dangers that stem from the toxicity of the propellants significantly increase the cost and time required for development and launch of a rocket. The cost penalties are made up of diverse parts: the propellant, its handling, health surveillance, decontamination and disposal of residuals [1,2]. Furthermore, a curtailment in the

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budget of national space agencies for space missions, along with stricter environmental and safety regulations, emphasizes the need for non-toxic hypergolic propellants. Thus, extensive research has been conducted on non-toxic hypergolic bipropellants to remedy the drawbacks reported from the existing hypergolic propulsion systems, although follow-up implementation continues to remain relatively scarce [3–5].

The principal goal of this research is to demonstrate a novel concept of 500 N scale bipropellant thruster using a non-toxic hypergolic bipropellant combination. This paper then presents experimental demonstrations with respect to hypergolic interactions and thruster operation. The focus is placed on the use of high test hydrogen peroxide (HTP) as a green oxidizer, considering its environmentally benign nature, and the search for non-toxic energetic hypergolic fuels comparable to conventional toxic fuels.

In general, there are two different types of additives for granting hypergolicity in a rocket fuel: catalyst or strong reducer. A catalyst dispersed in fuel can vigorously decompose an oxidizer when they are brought into contact. This process represents a rapid exothermic reaction that generates enough energy to ignite the reactants. However, this catalyst additive is not directly involved in the combustion process, which could result in increasing ignition delay and in degrading performance. Unlike the catalytic additive, once a strong reducer, such as metal hydrides, comes into contact with an oxidizer, ignition occurs immediately because the strong reducer itself can easily combust with a strong oxidizer. Due to its



Fig. 1. Molecular structures of hydrocarbon solvents used for non-toxic reactive fuels.

active participation in the ignition process, the fuel promoted by a strong reducer, commonly referred to as a reactive fuel, provides faster and more reliable ignition. Therefore, although hypergolic interactions can be caused by the two different types of additives, reactive fuels promoted by strong reducers are typically preferred.

Three reactive fuels were produced by blending sodium borohydride (NaBH₄) powder, a strong reducing agent, into energetic hydrocarbon mixtures. Sodium borohydride was previously used as an ignition promoter for 90 wt.% hydrogen peroxide by R. Mahakali et al. [6]. In the previous work, although most of the fuels demonstrated hypergolic properties with the oxidizer, they were potentially not suitable for practical applications because the solvents used for sodium borohydride had low chemical potential energy. In addition, there was no attempt to evaluate the non-toxic hypergolic bipropellant in actual conditions. This work is dedicated to developing enhanced reactive fuels by using more energetic solvents compatible with sodium borohydride and applying them into a 500 N scale hypergolic bipropellant thruster with 90 wt.% hydrogen peroxide as an oxidizer. Through ground hot-fire tests, the feasibility and performance of the non-toxic hypergolic bipropellant thruster were evaluated.

2. Materials and experimental methods

2.1. Materials

Metal hydrides are typically introduced as reducing agents in chemical synthesis and in materials for hydrogen storage applications. Light metal hydrides, such as lithium or beryllium compounds, are preferred as additives in rocket fuels due to their low molecular weight, enhancing the rocket performance. They are, however, extremely expensive and difficult to handle; lithium compounds react explosively with moisture and water. Practically, sodium borohydride is a promising alternative when considering cost and handling, even if it has lower chemical potential energy than lithium or beryllium compounds. Moreover, the presence of boron and borane compounds has been reported to act as energetic additives or active promoters in the process of hypergolic combustion [7,8]. The borohydride is also part of the electron-rich groups that prefer hypergolic reactions with a strong oxidizer such as hydrogen peroxide. Other characteristics of sodium borohydride, which encourage its use, are described below [9]:

- The least expensive metal hydride commercially available (on a hydride equivalent basis).
- Safe with regards to storage and use & handling.
- Industrial implementation requires no or limited equipment investment.
- Ease of work-up (water soluble boron salts).

Table 1 shows the three different types of non-toxic hypergolic fuels that were considered in this work. All the candidates are reactive fuels promoted by sodium borohydride. Stock 0 was prepared by blending sodium borohydride powder into a tetraglyme solvent. A small Teflon stir bar was used for vigorous mixing for a minimum of one hour; the product was a colorless liquid. The composition of Stock 0 was the same fuel previously tested by R. Mahakali et al. [6]. Tetraglyme is a polar aprotic solvent that is miscible with sodium borohydride and has excellent chemical and thermal stability, along with high density (specific gravity: 1.013 at 20° C), low vapor pressure (< 0.01 mmHg at 20° C) and low toxicity [10]. However, its low chemical potential energy leaves room for improvement.

Stock 1 and 2 are reactive fuels prepared and improved by adding more energetic solvents, such as tetrahydrofuran and toluene to Stock 0. Comparing the newly introduced solvents, tetraglyme has lower chemical potential energy because the heat of combustion decreases with the rise of the oxygen percentage in the molecule; their molecular structures are pictured in Fig. 1. The production methods of Stock 1 and 2 basically followed the aforementioned method for Stock 0. The only difference between Stock 0 and the enhanced reactive fuels was the composition of solvents. The more energetic solvents did not retain the same compatibility with sodium borohydride, and thus precipitation was formed at the end of the process. After leaving the solvents under standard atmospheric conditions for a minimum of one day, we could finally obtain colorless liquids by collecting the limpid portion of the solution. Sodium borohydride powder with >98% purity, 99.8% pure tetrahydrofuran (THF) and 99.8% pure toluene were purchased from Samchun Chemical Inc. located at Daejeon, and >99% pure tetraglyme was purchased from Sigma-Aldrich Inc.

Theoretical performance data and physical properties of the reactive fuels are tabulated in Table 2. Density was measured by I.S.O 649 Standard Hydrometers, and viscosity was measured with an SV-10 VIBRO Viscometer. Vapor pressure of the mixtures was calculated using Raoult's law. The net energy content from combustion was measured by a Parr 1261 bomb calorimeter. All candidates have higher density than MMH, and Stock 2 is estimated to be the most energetic material among the candidates, even more than MMH. The observed unstable chemical behavior of Stock 1 during its storage is associated with its high volatility. Theoretical vacuum specific impulse (I_{sp}) and O/F ratio were estimated using NASA CEA code, where the chamber pressure was set to 30 bar [11]. Based on the data, the non-toxic bipropellant combination of Stock 2 fuel and 98 wt.% H₂O₂ oxidizer is expected to have a performance comparable to the conventional toxic hypergolic bipropellants, with a volumetric specific impulse $(\rho \cdot I_{sp})$ greater than 99% of MMH/RFNA and 97.5% of MMH/NTO. This slightly lower performance can be compensated by the cost savings and risk reduction in terms of handling, storage and environmental impact.

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