



Autoignition and combustion characteristics of sodium borohydride-based non-toxic hypergolic fuel droplet at elevated temperatures



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ABSTRACT

Non-toxic hypergolic propellants have considerably generated recent research interest in the field of green propulsion technology because they can replace highly toxic hypergolic combinations currently used. In this experimental research, sodium borohydride-based non-toxic hypergolic fuel was prepared by blending sodium borohydride in the mixture of energetic hydrocarbon solvents. In a drop test, sodium borohydride as an ignition source enabled the hydrocarbon mixture to initiate hypergolic interactions with H_2O_2 oxidizer. Two different heating methods were utilized to analyze the characteristics of autoignition and combustion of the hypergolic fuel. As a reference fuel, a non-hypergolic fuel which has the identical chemical compositions to the hypergolic fuel except for sodium borohydride was tested and compared. As a one of the heating methods, thermogravimetric analysis was not suitable for evaluating the inherent thermophysical properties of the hypergolic fuel. In a droplet combustion chamber test, the autoignition and combustion of the hypergolic fuel droplets occurred exposed to elevated temperatures (in a range of 400–800 °C) at atmospheric pressure (1 bar), whereas the non-hypergolic fuel droplet was automatically ignited only at 800 °C. The ignition delay and total combustion time of the hypergolic fuel droplet were lower than those of the non-hypergolic fuel droplet. According to the temporal histories of the droplet size, sodium borohydride-based hypergolic fuel droplets did not obey the d^2 -law of diffusion-controlled combustion, which indicates the droplet evaporation rate was not a controlling factor in the combustion process. Consequently, the addition of sodium borohydride into the hydrocarbon mixture expedited the autoignition and combustion process of the fuel at elevated temperatures.

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1. Introduction

A chemical propulsion system is indispensable in space missions. In modern rocket technology space-faring nations make efforts to develop advanced chemical propulsion systems utilizing storable propellants, with the purpose of not only improving propulsive performance but also reducing development cost, lead time and risk [1]. The systems are normally classified by the selection of the propellants. For a monopropellant propulsion system, hydrazine has been widely used as a conventional propellant. Since hydrazine causes serious technical problems due to its toxicity and

explosiveness, an alternative should be developed in line with the modern rocketry. In repose to the technical needs, the green energetic monopropellants, HAN-based or ADN-based liquid propellants, are developed as one of the most promising alternatives. In case of a bipropellant propulsion system, the conventional combinations are using hydrazine derivative fuels (MMH and UDMH) with NTO or WFNA as an oxidizer. For improving the system performance, much research has been conducted to raise the specific impulse by increasing chamber pressure and temperature [2]. From the point of view of a propellant feed system, active mixture ratio control devices and ultra-light propellant tank technology can also contribute to enhance the system performance. However, these research activities are not enough to ultimately resolve the technical problems caused by the toxicity of the bipropellants, which is inconsistent with the requirements of the modern rocket technology in terms of cost, time, and risk.

The advent of non-toxic hypergolic bipropellants draws attention from space-faring nations because this concept is expected to provide a fundamental solution to the insurmountable

Abbreviations: ADN, Ammonium dinitramide; HAN, Hydroxylammonium nitrate; HTP, High-test peroxide; MMH, Monomethylhydrazine; NTO, Dinitrogen tetroxide; TMEDA, N, N, N', N'-Tetramethylethane-1, 2-diamine; UDMH, Unsymmetrical dimethylhydrazine; WFNA, White fuming nitric acid.

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technical difficulties. Since early 2000s, various combinations of non-toxic hypergolic bipropellants have been suggested [3]. In many studies, high-test peroxide (HTP) was selected as a green oxidizer. Developing a non-toxic energetic fuel having hypergolic properties with HTP has been regarded as a matter of a technically challenging task. Up to date, two different types of non-toxic hypergolic fuels were mainly reported and demonstrated. Firstly, a catalytic fuel was produced by dissolving a catalyst agent in a fuel; the catalyst exists in the colloidal state in the fuel. When the fuel contact to hydrogen peroxide, the catalyst starts to decompose the oxidizer. The catalytic decomposition is a highly exothermic reaction, thereby initiating hypergolic interactions between the fuel and oxidizer. A variety of metallic oxide such as platinum oxide, manganese oxide and lead oxide can be used for the catalytic ignition source. Secondly, a reactive fuel was produced by blending a light metal hydride in a fuel. For the fuel, a reduction-oxidation reaction between the reactive fuel and hydrogen peroxide triggers a hypergolic initiation. Many kinds of strong reducing agents are regarded as an ignition source for hypergolic ignition, for example light metal hydrides such as lithium hydrides, lithium aluminum hydrides, and sodium borohydride. Unlike the catalytic fuel, the reactive fuel could provide relatively short ignition delay because the fuel can directly combust with the oxidizer by the reduction-oxidation reaction. Furthermore, it is expected that the reactive fuel provides higher specific impulse because the molecular weight of light metal hydrides is mostly smaller than those of catalyst agents. Overall, because of these advantages, the reactive fuel is likely to become more attractive as a promising alternative for an advanced non-toxic hypergolic propulsive system in future.

For a reactive fuel, the key technology is the selection of the chemical ingredients, especially for a light metal hydride as an ignition source. Lithium-based hydrides can provide superior performance and high reactivity because lithium belonging to the alkali metal group of chemical elements is the lightest one among the metal elements. However, it is somehow difficult to find a comparable solvent with lithium hydrides. Additionally, the storage and handling of lithium hydrides is demanding because it is extremely sensitive to humidity due to the nature of a water reactive chemical. Sodium hydrides are regarded as an attractive ignition source for a reactive fuel. In particular, the use of sodium borohydride (NaBH_4) provides many benefits [4]. Sodium borohydride has a relatively low molecular weight, and it is the least expensive one among commercial metal hydrides. Moreover, various kinds of comparable hydrocarbon solvents are also commercially available. Additionally, it is safe with respect to storage and handling because it does not violently react with water but is soluble.

It is important to explore the ignition and combustion characteristics of non-toxic hypergolic propellants in order to develop advanced chemical propulsion systems. With hydrogen peroxide as a green oxidizer, the ignition source dissolved in the non-toxic hypergolic fuel has great influence on the phenomena related to the ignition and combustion. Thus, much research in recent years has been performed to investigate the characteristics of combustion between the fuel and oxidizer, depending on a type of ignition sources. As the content of ignition source increases, both parameters, the delay of hypergolic ignition and the performance of specific impulse, are normally decreased. However, little work has been done to examine the combustion characteristics of non-toxic hypergolic fuel itself. Due to the existence of an ignition source in a non-toxic hypergolic fuel, pyrolysis and burning processes of the fuel could be dramatically changed, thereby affecting its combustion characteristics in terms of combustion efficiency and instability. In the technology of internal combustion engines, it is already well known that the burning process of a fuel droplet significantly affects overall system performance [5–7].

The purpose of this research is to scrutinize the autoignition and combustion characteristics of a non-toxic hypergolic fuel depending on the existence of an ignition source in the fuel. A sodium borohydride-based non-toxic hypergolic fuel (named as 'Stock 2') was prepared and utilized. The hypergolicity of the reactive fuel with HTP was demonstrated by a drop test. A non-hypergolic fuel (named as 'pure Stock 2'), which indicates the ignition source was not mixed together with the same mixture, was also tested as a reference fuel. Two different heating methods were introduced. Firstly, thermogravimetric analysis was implemented to compare the thermophysical properties of the fuels from the standpoint of a general approach. Secondly, the droplet burning rate of the fuels was measured and analyzed in a specially-designed combustion chamber capable of rapidly increasing the ambient temperature around the droplet.

2. Experimental approach

2.1. Sodium borohydride-based non-toxic hypergolic fuel

A sodium borohydride-based non-toxic hypergolic fuel was classified as a reactive fuel, and the hypergolic fuel used in this work was named as 'Stock 2'. Sodium borohydride was selected as an ignition source for hypergolic initiation with HTP oxidizer. The energetic fuel for the ignition source was produced by mixing three energetic hydrocarbon solvents (tetraethylene glycol dimethyl ether, tetrahydrofuran, and toluene), and then sodium borohydride was blended in the mixture. Tetraethylene glycol dimethyl ether is compatible with sodium borohydride but it has relatively lower chemical potential energy. Toluene as a more energetic material was used to increase the net energy content of the fuel, considering a decrease of oxygen percentage in the molecule. However, toluene potentially causes the storability issue of the fuel mixture because it is incompatible with the ignition source. Thus, tetrahydrofuran was additionally added to improve the storability because it is not only slightly comparable with sodium borohydride, but also can dissolve a wide range of non-polar and polar chemical compounds. The preparation of sodium borohydride-based non-toxic hypergolic fuel was described in detail within the precedent study [8]. Table 1 shows the differences in the physical properties of both the hypergolic and the non-hypergolic fuel. Density and viscosity were measured using I.S.O 649 Standard Hydrometers and SV-10 VIBRO Viscometer, respectively. A Parr 1261 bomb calorimeter was used to measure the net energy content of the fuels. By blending sodium borohydride in the mixture, its density and viscosity were increased but the net energy content was decreased.

The hypergolicity of the reactive fuel with HTP was demonstrated through a drop test. In the test, a small droplet of 90% HTP oxidizer fell down to the pool of the fuel so that the propellants were physically in contact. The diameter of the oxidizer droplet was approximately 3 mm and the drop height from the surface of the pool was about 12 cm. The hypergolic interactions were captured by a high-speed charge-coupled device (CCD) camera, X-Stream™ XS-3 model with 1000 frames per second. The hypergolic interactions occurred on all occasion, and the representative result was presented in Fig. 1. After a time lapse of 11 ms from the initial physical contact, hypergolic ignition occurred for the first time. Because of a rapid expansion of reactive vapor layers generated from the flame kernel, fuel droplets were splatted into the atmosphere. The green-colored flame, which was mainly observed during the early stage of the hypergolic reactions, indicates that boron atoms from the ignition source actively participated in the combustion process. The observation of light red-colored flames subsequently appeared after the green-colored flame seemed to imply that the hydrocarbons started to dominantly participate in the

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