The Developing Present and Prospect of High Energy Density Metallic Combustion

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Abstract. In this paper, we reviewed the developing present and future of high energy density metallic combustion. The review is mainly focused on the aluminium based alloys, born based alloys, hydrogen storage alloys three aspects. We discussed the present and developing trend of kinds of materials.

Keywords: Metallic combustion, Combustion energy, Energy containing materials, Aluminium based materials, Boron based materials.

1. Introduction

Metal incendiaries are widely used in various energetic materials such as propellants and explosives. With the continuous increase in the demand for high-efficiency lethality and high combustion heat indicators, the demand for high energy density metal incendiaries has received increasing attention from countries all over the world. At present, metal incendiaries mainly include several major types such as aluminum-based and magnesium-based alloys, boron-based alloys, and hydrogen storage alloys. Various metal incendiaries have certain mature application fields. However, as the demand for materials with higher energy density becomes increasingly urgent, metal incendiaries with higher energy density, better ignition conditions, more suitable sensitivity, and good comprehensive performance have always been the goal of the development of metal incendiaries.

2. Aluminum-based Alloys and Magnesium-based Alloys

The combustion heats of aluminum and magnesium are 30 MJ/kg and 25 MJ/kg respectively. During the metal combustion process of aluminum and magnesium, the oxides produced, especially the combustion products of aluminum, are easy to adhere to the surface of metal particles, which further inhibits the complete combustion of the metals, resulting in the incomplete release of their combustion heat and the inability to fully utilize their performance, and finally reduces the performance of gun propellants and warheads. Liu Xin et al. studied the combustion behavior of aluminum-based alloys in the AP/Al/HTPB composite propellant and proved that a viscous layer of Al2O3 was formed on the surface of aluminum particles during the combustion process [1]. In order to fully utilize the performance of aluminum-based alloys and magnesium-based alloys, on the basis of aluminum-based and magnesium-based incendiaries, people have developed aluminum-based alloys and magnesium-based alloys.

2.1. Aluminum-Magnesium Alloys

Compared with pure aluminum incendiaries, ball-milled magnesium/aluminum alloys have higher reactivity with water, and this kind of metal fuel propellant has a higher burning rate and explosion heat value. Meanwhile, ball milling improves the primary combustion effect of the metal fuel propellant reacting with water, and the secondary combustion products react more thoroughly. The aluminum content in the remaining secondary combustion products is lower, which improves the combustion efficiency of aluminum [2]. Researcher Ren Xiaoxue from the North Science and Technology Information Institute investigated the analysis of the development of foreign nano-thermite technology [3]. According to the current development status of metal incendiaries, it is pointed out that in-depth research on the nanostructure, properties, and preparation mechanism of aluminum-based alloys and seeking efficient, inexpensive, and environmentally friendly preparation methods suitable for industrial production are the key points for the future development of thermites.

2.2. Research on Nanostructured Thermites

The development of nanotechnology has promoted the development of the preparation technology of nanoscale metal incendiaries, making the research and application of nanostructured metal-based energetic materials possible. In 2010, the University of Missouri in the United States and the U.S. Army Armament Research, Development and Engineering Center jointly developed a nano-energetic composite with adjustable combustion performance [4]. In 2014, TDA Company in the United States also developed a new nano-particle modification technology for the field of energetic materials and used this technology to adjust the energetic and physical properties of nano-thermite composites [5]. The research results verified that the performance of nano-thermite can be adjusted by chemical modification. In China, E Xiutianfeng et al. also studied the improvement of combustion performance by nano-aluminum modification [6]. The results showed that using organic reagents to modify the surface of aluminum particles can improve the dispersibility of aluminum in organic gun propellants and thus improve its combustion performance. This may be because after the aluminum metal particles are coated, the interface structure between them and the organic gun propellants has changed, from the original metal-organic direct contact structure to the metal-transition interface-organic gun propellant structure, which changes the original interface compatibility and thus improves the dispersion and combustion performance of aluminum alloys. Li Meng et al. also studied the influence of nano-aluminum powder and micron-aluminum powder on the combustion performance of composite propellants [7]. The results showed that with the increase in the content of nano-aluminum, the standard theoretical specific impulse and characteristic velocity of the composite propellant reached their maximum values, the combustion temperature increased, and the average relative molecular mass of the combustion gas decreased. This should be due to the improvement of the combustion performance of the propellant caused by the presence of nano-aluminum.

2.3. Modification of Aluminum Alloys

In order to further improve the combustion performance of aluminum alloys, people have also studied the composite alloying of aluminum alloys. By alloying aluminum alloys, the structure of aluminum alloys is changed at the atomic structure level, and then the combustion performance of aluminum alloys is changed, inhibiting the adhesion of oxides generated during the combustion process of aluminum alloys on the particle surface, so as to achieve the purpose of improving the combustion performance of aluminum alloys. Song Guixian et al. doped aluminum with nickel for modification [8]. The results showed that the nickel-aluminum composite alloy had a nickel-coated aluminum structure, and the aluminum powder was completely wrapped by the nickel powder. The addition of low-content nickel-aluminum composite powder reduced the combustion pressure index of the propellant. After adding nickel-aluminum composite powder to the propellant samples, the flame structure changed significantly. Liu Jingru et al.'s research on the preparation and performance of aluminum-based composites for solid propellants showed [9] that the combustion heat of the composites of Al and MgH2 prepared by mechanical alloying was greater than 30 MJ/kg, and the combustion efficiency was greater than 94%. The combination of aluminum and hydrogen storage alloys can also improve its combustion performance and achieve good results. Many studies have shown that alloying aluminum with other metal elements, especially transition metals, is an effective method to improve the combustion performance of aluminum alloys. However, since the combustion heat of other metals, especially transition metals, is much lower than that of aluminum, although alloying can improve the combustion performance of aluminum alloys within a certain range, the final combustion heat is affected by the content of alloying elements. The author's research results show that alloying aluminum with elements such as rare earths can improve the combustion performance of aluminum alloys and adjust the combustion heat of aluminum alloys. However, when excessive rare earths are added, it is easy to lead to a decrease in the combustion heat.

Compared with alloyed aluminum-based alloys, metal aluminum-based nano-composite energetic materials, such as metastable intermolecular complexes, are in a nano-sized structure, and the metal incendiary aluminum particles and metal/non-metal oxides are in a metastable state, which greatly improves the activity of the materials and improves the combustion performance of the materials. Yin Qiushi et al. studied the preparation and combustion performance of flake aluminum powder [10]. The spherical aluminum powder was processed into flake shape by ball milling. The flake alloy after ball milling had the same crystal structure as the spherical alloy, but the surface area of the flake aluminum powder after ball milling was greatly increased, the combustion activity was enhanced, and finally the combustion performance of the alloy was greatly improved. The aluminum content in gun propellants also affects the performance of engines [11]. Under the condition that other components remain unchanged, with the increase in the content of aluminum powder in the unit mass of composite propellants, the flow velocity of combustion gas decreases, the pressure increases, and the thrust of the rocket engine first increases and then decreases.

Yang Weijuan et al. summarized the uses of aluminum alloys in the energy field [12]. They conducted a review from aspects such as propellants, hydrogen production, and aluminum batteries. Through improvements in technologies such as the improvement of aluminum-based fuel formulations, modification of aluminum particles, surface coating, and the perfection of system processes, the techno-economic and stability of aluminum energy utilization will continue to improve, and its position in energy utilization will become more and more important.

Aluminum-based alloys are currently high-energy metal incendiaries with the most mature application technology and the widest application range. Through the modification of nano-aluminum powder, magnesium-aluminum alloys, and various other modification methods, the comprehensive performance of aluminum alloys has been greatly improved, and good results have been achieved so far. In order to further increase the combustion heat of metal additives, on the basis of continuous improvement of aluminum alloys, people have also developed other metal additives. Among various additives, boron-based materials have attracted great attention due to their high combustion heat value.

3. Boron-based Alloys

Although the combustion heat of aluminum alloys can already meet the needs of some weapons, with the continuous extension of the range of long-range strikes and the continuous improvement of the requirements for high-efficiency damage performance, the demand for developing metal incendiaries with higher combustion heat and better combustion performance has received increasing attention from more countries. Compared with aluminum alloys, boron elements and boron-based alloys have the most practical potential in terms of combustion heat among current materials and have already obtained some applications. The atomic number of boron is 5, and its atomic weight is 10.8. It has relatively high reactivity, and its combustion heat when completely burned in oxygen can reach 58 MJ/kg, ranking third among currently known materials and 1.5 times that of aluminum. During the combustion process, boron generates B2O3. Since the generated B2O3 has the characteristics of a viscous flow state, it is extremely easy to adhere to the surface of boron particles, thereby preventing their full combustion. Therefore, how to improve the combustion performance of boron particles has become a current research difficulty.

In order to improve the combustion performance of boron, dispersing boron into other fuels is a relatively effective method. On the one hand, this improves the dispersibility of boron powder during the combustion process, and on the other hand, it is beneficial to the ignition of boron powder. Yang Dali et al. studied the combustion characteristics of directly adding boron powder into an alcohol solution [13]. The results showed that after adding boron to the alcohol solution, the incomplete combustion of boron powder was easily caused by micro-explosions during the combustion process. The occurrence of micro-explosions should be due to the large amount of heat released during the combustion process of boron, resulting in excessive heat concentration in some areas of the boron powder. Meanwhile, as the alcohol infiltrated in the boron powder rapidly vaporizes, volatilizes, and burns, finally, the pressure is overly concentrated in some areas and micro-explosions occur.

Since the density of boron powder is greater than that of liquid fuels, when boron powder is added to liquid fuels, the boron powder is prone to sedimentation. The sedimented boron powder is neither conducive to the stable combustion of the fuel nor beneficial to the operation of the engine. Therefore, how to improve the dispersibility and stability of boron powder in liquid combustion is an important issue faced by the application of boron powder. Pei Huixia et al. studied the feasibility of using solid particles to significantly increase the density and energy of liquid fuels [14]. The results showed that using tri-n-octylphosphine oxide to modify the surface of boron nanoparticles can reduce particle agglomeration and sedimentation, improve the dispersion stability of particles in liquid fuels, and the mass density and energy density of the fuel increase with the increase in the mass fraction of solid particles, while the viscosity of the fuel also gradually increases with the increase in particle content.

In order to further improve the combustion performance of boron, Chen Binghong et al. studied the influence of oxidant coating on the combustion performance of boron [15]. The results showed that after coating boron particles with four oxidants such as AP (ammonium perchlorate), AN (ammonium nitrate), NQ (nitroguanidine), and HMX by recrystallization, the thermal oxidation process of boron-based propellants was divided into two stages. Coating boron particles with AP was more conducive to improving the oxidation reaction intensity of boron-based propellants in the first stage, thereby promoting the participation of boron particles in the oxidation reaction in this stage. Compared with mechanically mixed samples, it was found that the recrystallization coating method was an important reason for the occurrence of rapid low-temperature combustion. The study also found that the ignition delay time of boron powder after being coated with AP was 330 ms, and the ignition speed was significantly faster than that of other samples, indicating that AP coating was beneficial to improving the ignition characteristics of boron-based propellants. Chen Binghong et al. also studied the coating of boron particles and its influence on combustion performance [16], and discussed the action mechanism, coating process, and characterization of coating effects of the influence of the surface coating of boron particles on their combustion performance. From the perspective of promoting the combustion effect of boron particles, five aspects, including removing the oxide film on the surface of boron particles, increasing the combustion temperature, reducing the ignition temperature of boron, improving surface compatibility, and catalyzing the oxidation reaction of boron particles, were proposed as the research directions for the surface coating materials of boron particles.

Pang Weiqiang et al. also studied the coating of boron powder and the composition of combustion residues of propellants containing coated boron powder [17]. The results showed that after being coated by a coating agent, a coating layer was formed on the surface of the boron powder. After coating, the pH value of the boron powder water suspension system increased significantly, which was beneficial to the pharmaceutical process of boron-rich fuel propellants. The combustion efficiency of the coated boron powder was significantly improved. Zhang Jiaoqiang et al. also studied the combustion performance of AP-coated ultrafine boron [18]. The boron powder pretreated with a silane coupling agent can make AP coat the surface of the boron powder more evenly, improving the coating effect. Using methanol or acetone as a solvent, the coated boron powder has relatively weak water absorption, is not easy to agglomerate after drying, and the combustion efficiency of the AP-coated boron powder is improved.

Hao Lifeng et al. comprehensively investigated the technical status and development trend of boron-rich fuel propellants [19]. They reviewed the research progress at home and abroad in aspects such as the ignition and combustion performance of boron particles, the coating and agglomeration granulation technology of boron powder, the influence of boron powder treatment on the performance of formulations, and the formulation and performance test and characterization means of boron-rich fuel propellants. They also summarized the current situation and existing gaps in the engineering application of boron-rich fuel propellants at home and abroad.

Currently, the modification of boron powder mainly focuses on the coating with oxides such as AP. To some extent, this can improve the combustion performance of boron powder and inhibit the agglomeration of boron powder. However, it does not fundamentally solve the shortcomings of combustion product accumulation, incomplete combustion, and the inability to fully utilize its performance during the boron combustion process. To fundamentally improve the combustion performance of boron, theoretically, modification should be carried out from the perspective of the characteristics of molecular structure. By improving the dispersion of boron atoms, the purpose of improving its combustion performance can be achieved. In future research, if boron can be alloyed with other elements, theoretically, the combustion performance of boron powder should be completely improved. However, due to the high melting point of boron, how to conduct alloying is the main difficulty faced by the research on boron-based alloys.

4. Hydrogen Storage Alloys

Hydrogen is currently the substance with the highest known combustion heat value. Its combustion heat can reach 140 MJ/kg, which is 2.4 times that of boron, 4.7 times that of aluminum, and 5.7 times that of magnesium. However, since there is currently no particularly effective way to store elemental hydrogen efficiently, how to effectively utilize hydrogen has always been the main concern. Among various hydrogen storage methods, hydrogen storage alloys are an effective way, and metals are also effective metal additives for gun propellants. Ma Yi summarized the application of hydrogen storage alloys in high-energy solid propellants [20]. Starting from metal hydrides, metal nitrogen-hydrogen compounds, and metal coordination hydrides, he analyzed the components of hydrogen storage material propellants, the improvement of combustion performance, and the action mechanism, and pointed out that hydrogen storage materials still need in-depth research in aspects such as stability, hydrogen storage capacity, and the mechanism of the reaction process.

Pei Jiangfeng et al. studied the energy characteristics of propellants containing metal hydrides [21] and pointed out that the energy contribution of metal hydrides such as AlH3 is higher than that of aluminum within a certain range, but the overall energy density of gun propellants is affected by the addition amount. In addition, the stability of metal hydrides is affected by their phase structures. During the use process, the stability of metal hydrides directly affects the storage, combustion, and other performances of gun propellants. In order to improve the storage stability of metal hydrides, organic substances or other substances can be used to coat metal hydrides. The research results show that paraffin coating can improve the storage stability of KBH4, NaBH4, and LiAlH4 [22]. Adding a certain amount of metal hydrides to gun propellants is beneficial to improving the propulsion performance of gun propellants, especially being able to significantly improve the specific impulse of hybrid propellants. The technology of metal hydride hybrid propellants will be an important research and development direction for propellants [23].

Besides metal hydrides, hydrogen storage alloys are also important hydrogen storage materials. Commonly used hydrogen storage materials include magnesium-based alloys, LaNi5-based alloys, zirconium-based alloys, and titanium-based alloys. Different hydrogen storage alloys have different hydrogen storage performances, and the hydrogen absorption and desorption plateau performances of alloys are also affected by the components of hydrogen storage alloys. Magnesium-based hydrogen storage materials can improve their energy levels by promoting the thermal decomposition process of energetic materials [24]. Meanwhile, their relatively high thermal stability is beneficial to improving the compatibility and stability of the components of energetic materials. Magnesium-based hydrogen storage alloy hydrides, magnesium hydride, and magnesium-based coordination hydrides can all significantly improve the performance of solid propellants and explosives. Therefore, magnesium-based hydrogen storage materials have broad application prospects in the field of energetic materials.

To further improve the combustion performance of hydrogen storage materials, people have also carried out nanometer processing on hydrogen storage materials. Yang Yanjing et al.

comprehensively studied the application of nano-hydrogen storage materials in propellants [25] and pointed out that nano-chemisorption hydrogen storage materials have lower hydrogen desorption temperatures and faster hydrogen desorption rates, which are beneficial to fully utilizing the hydrogen contained in hydrogen storage materials during the combustion process of propellants. Nano-chemistry-physical adsorption hydrogen storage materials can simultaneously serve as the hydrogen source and combustion catalyst of propellants. By adjusting the nanostructure of physical adsorption hydrogen storage materials, they can have the ability to rapidly absorb and desorb hydrogen at room temperature, improving their application prospects.

To improve the combustion performance of hydrogen storage alloys, people have also studied the process of coating hydrogen storage alloys with oxidants such as AP. The results show that after coating with AP and HTPB, the combustion performance of the hydrogen storage alloy/AP/HTPB composite propellant has been significantly improved [26, 27]. The research results also show that hydrogen storage alloys have a good catalytic effect on the condensed phase reaction of the AP/HTPB propellant, reducing the main decomposition temperature of the condensed phase of gun propellants and increasing the reaction heat. This indicates that hydrogen storage alloys can improve the combustion activity of gun propellants and increase their combustion heat.

5. Conclusions and Prospects

The application of metal materials in gun propellants has greatly increased the combustion heat of gun propellants and improved their combustion performance. Meanwhile, the catalytic performance of metal materials has also improved the combustion performance of organic gun propellants to some extent. Therefore, metal materials have broad application prospects in gun propellants.

Although metal materials play an important role in gun propellants, their application in gun propellants is still affected by many factors at present. Firstly, the preparation methods of metal materials need to be further improved. The preparation process of nano-metal materials, the surface modification of metal particles, and the interface structure performance between metal materials and organic gun propellants, oxidants, etc. also require further in-depth research. Secondly, the combustion mechanisms of some materials such as boron in primary and secondary combustions have not yet been reasonably explained. Thirdly, how to increase the content of high-combustion-heat materials such as hydrogen elements in gun propellants and maintain the structural stability and long-term storage stability of gun propellants is also a subject that gun propellant research needs to face.

All in all, metal materials have broad application prospects in gun propellants. The application of metal materials plays a crucial role in the development of high-performance gun propellants and warheads. A lot of work still needs to be done on the research of their preparation and combustion mechanisms.

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