Surface Coating Strategies for Improving Boron Powder Combustion in Propellant Applications: A Review

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Abstract: Boron powder is a remarkable metal fuel renowned for its high gravimetric and volumetric calorific values, making it an ideal choice for military applications, including solid propellants, high-energy explosives, and pyrotechnics. However, the formation of a liquid oxide layer on the surface of boron powder can lead to a gel layer's creation, resulting from the reaction of hydroxyl (-OH) group in the hydroxyl-terminated polybutadiene (HTPB) binder, which hampers the propellant processing and limits the boron powder's complete oxidation. To enhance the combustion efficiency of boron powder, researchers worldwide have explored the ignition and combustion mechanisms of boron powder and devised surface coating modification strategies. These modifications aim to optimize the surface properties, improve the reaction activity, and promote the energy release of boron powder. This review highlights recent studies on the ignition and combustion mechanisms and reaction mechanism. Furthermore, the review summarizes the mechanisms and features of various coating materials, including providing mechanism diagrams of fluoride and metal oxide. Moreover, the review proposes promising directions for modification methods and outlines the potential application prospects of boron powder.

Keyword: Boron; Combustion; Coating; Oxidation; Propellants; Efficiency

1. Introduction

The demand for high-performance solid propellants and explosives is crucial in modern national defense technology [1]. In pursuit of this goal, researchers have turned to metal fuels due to their high energy density [2], combustion calorific value, and combustion stability at high frequencies [3]. These metal fuels have been widely used as high-energy additives to enhance the energy density of energetic formulation systems [4]. Figure 1 shows the calorific values of various high-energy metal fuels, including Be, B, Mg, Al, Li, and others.



Figure 1: Gravimetric and volumetric calorific values of some metal fuels.

However, there are challenges associated with the use of certain metal fuels [5]. For example, Li and Zr are prone to spontaneous combustion and explosion in air [6], Be's combustion products are highly toxic [7], and Fe has a low calorific value. Despite these limitations, B [8], Al [9], and Mg [10] are commonly used as metal fuels in propellants. Of these, B stands out due to its superior calorific value, with a gravimetric value that is 2.1 and 1.9 times greater, and a volumetric value that is 3.2 and 1.6 times greater than that of Mg and Al, respectively [11].

Calculations show that the theoretical specific impulse of boron-containing fuel-rich propellant can reach 10,000-12,000 N·s/kg, which is 3-5 times greater than that of conventional solid propellant and about 2 times greater than that of Al-Mg fuel-rich propellant [12]. As a result, boron is regarded as one of the most promising metal fuel additives in solid propellants, making it a crucial area of research in modern national defense technology [13].

Recent studies have identified three major challenges that have hindered the widespread adoption of boron in various applications [14]. These challenges can be summarized as follows: Firstly, boron powder tends to develop a protective oxide layer on its surface during combustion, which leads to poor ignition performance and longer ignition delay times [15]. Secondly, the high melting point (above 2500 K) and low boiling point (718 K) of B2O3 lead to the formation of a liquid film during combustion, which can hinder oxygen diffusion and reduce the overall combustion efficiency of boron [16]. Lastly, impurities such as B2O3 and H3BO3 can react with the hydroxyl groups present in hydroxyl-terminated polybutadiene (HTPB)[17] and form a gel-like layer on the surface of boron powder, resulting in poor compatibility between boron and HTPB [18].

With its potential as a high-energy fuel, boron has captured the attention of researchers seeking to improve its ignition performance and combustion efficiency [19]. However, this task is complicated by the intricate nature of boron's ignition mechanism and the challenges posed by the diffusion and reaction of B and O2 in the presence of an oxide layer [20]. Over the years, numerous studies have been carried out to investigate this mechanism, with different ignition mechanisms being proposed, yet a unified mechanism remains elusive.

To overcome this challenge, recent research efforts have shifted towards the development of coated or modified boron powder. By leveraging the interaction between boron powder and coating agents or modifiers, this approach aims to enhance the performance of boron powder [21]. For example, the presence of fluorine has been shown to facilitate the rapid removal of the oxide layer that forms on the surface of boron, while the use of certain energetic materials can generate substantial heat to accelerate the ignition of boron [22]. Thus, while boron's potential as a high-energy fuel is clear, the challenge lies in developing effective methods to overcome the obstacles to its full energy release [23]. With the current focus on coated and modified boron powder, researchers are hopeful that significant progress can be made towards unlocking the full potential of this promising fuel source [24].

In this comprehensive analysis, we delve into the latest advancements in surface modifications and the ignition and combustion mechanisms of high-energy boron fuel [25]. Firstly, we provide a detailed review and summary of the ignition and combustion mechanism of boron [26]. Secondly, we explore the various coating materials used for boron powder, including energetic materials, fluoropolymers, inorganic fluorides, metals, and metal oxides, and highlight the research progress made in this area [27]. Thirdly, we analyze and discuss the performance and characteristics of different coating materials, identifying promising directions for the future development of boron particle coating modification methods [28]. Our analysis aims to provide valuable insights and suggestions for researchers working in the propellant and explosive field.

2. Ignition Mechanism of Boron Particles

B-D Model

In 2014, Aowenmade a groundbreaking discovery by demonstrating through TEM characterization that oxygen can diffuse through B2O3 [29]. This finding indicates the presence of both (BO)n and oxygen diffusion during the ignition process [30]. Based on this, Aowen proposed the B-D (bi-direction) model for boron ignition, which involves four kinetic processes: (1) evaporation of B2O3 (1); (2) surface reaction of O2 (g) with boron in the environment; (3) reaction of internal core boron with O2 (g); and (4) overall reaction of boron with H2O (g). The B-D model can be further divided into two submodels based on the thickness of the oxide layer. When the

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oxide layer is thin, submodel I is proposed, where the particle exhibits a four-layer structure of B-B2O3-(BO)n-B2O3 from inside to outside, and two-way diffusion occurs [29]. On the other hand, when the oxide layer is thick, submodel II is available, where the particle shows a three-layer structure of B-(BO)n-B2O3 from inside to outside, and (BO)n adopts a unidirectional diffusion behavior [30]. This research significantly advances our understanding of the boron ignition process and provides a framework for future studies in this field. Dreizin proposed a unique boron ignition model, suggesting that oxygen can dissolve in solid boron during the ignition stage of boron particles. As the oxygen dissolves, it gradually reaches its solubility limit, and once this limit is reached, the boron particles enter the combustion stage, resulting in a significant release of heat. However, the lack of B-O phase diagram data has prevented researchers from gathering sufficient evidence to support this model, as of now [31].



Figure 2– Model of ignition of a single boron particle covered by an oxide layer: in submodel I, both (BO)n and O2 are the governing species diffusing into the liquid oxide layer; in submodel II, only (BO)n is the governing species diffusing into the liquid oxide layer.

L-W model

In order to gain deeper insights into the mechanism of boron particle ignition, Glassman utilized a solubility model to compare the solubility and diffusion rate of B (s) and O_2 (g) in $B_2O_3[32]$. The results revealed a stark contrast, with the solubility of O_2 (g) in B_2O_3 (l) being 10 orders of magnitude smaller than that of B (s) in the temperature range of 1800–2300 K [33]. Consequently, Glassman proposed a diffusion mechanism where boron dissolves in liquid boron oxide and forms a B_2OX (X < 3) compound during the ignition stage, followed by diffusion to the boron oxide–gas interface to react with oxygen. Building upon this diffusion mechanism, Li and Williams put forth an L-W model that encompasses four global reactions during the ignition stage. These reactions include the formation of BO(d), the reaction of O_2 (g) and $H_2O(g)$ with BO(d) to form BO₂ (g) and HOBO(g), the vaporization of B_2O_3 (l), and the reaction of O_2 with bare B to form O=B–B=O bonds after removal of the oxide layer [34]. The experimental results from Macek's research on flat-flame burners and laser ignition tests [33,34] support the prediction of this model. Notably, the emergence of HOBO (g) leads to a significant reduction in the energy released during the combustion of boron particles. This is because the energy of boron can only be fully released when all the final products are converted into thermodynamically favorable B_2O_3 (l). However, the sluggish conversion of HOBO (g) to B_2O_3 (l) decelerates the energy release rate in the presence of oxygen and hydrogen.

Kings model

To investigate the ignition and combustion properties of boron particles, a series of experimental studies have been conducted. Macek and Semple [32–34] utilized a flat-flame burner and a CO2 laser igniter to investigate the combustion of crystalline boron particles with diameters ranging from 34.5 to 124 μ m. Their findings

indicated a correlation between the burning rates of the samples and the diffusion rate of gaseous oxidants from the surface of the samples. They also observed that the burning time decreased with an increase in ambient gas temperature and oxygen mole fraction, consistent with the theory of gas-phase diffusion. In a separate study, employed shock-tube technology to ignite crystalline boron particles with diameters of 30–50 μ m [35]. This investigation revealed a strong correlation between the ignition temperature and the air pressure, where the ignition temperature increased with a decrease in air pressure. These experimental results provide valuable insights into the ignition and combustion behavior of boron particles and offer potential applications in various industries.

To elucidate the experimental phenomena described above, King [36–38] proposed the King ignition model, which postulates a diffusion mechanism where oxygen migrates through liquid boron oxide and reacts with boron at the B (s)-B2O3 (l) interface. Mohan and Williams [39] proposed a planar model to depict the ignition stage of boron within this diffusion mechanism, with the limiting oxygen mass fraction predicted by this model closely matching the results obtained by Prentice [40]. Meinkohn [41] subsequently developed an ignition stage model based on the King model, which calculated the critical ignition temperature of boron particles to be 1900 K, consistent with the experimental findings of Macek [32]. Gaponenko and Meese also devised their own models based on King's model, achieving high levels of predictive accuracy [42,43].

Y-K model

In 1996, Yeh and Kuo [44] revolutionized the understanding of boron combustion by establishing the Y-K model through a comprehensive analysis of the reaction products during ignition and combustion using advanced techniques such as environmental scanning electron microscopy (ESEM) and X-ray diffraction (XRD). The Y-K model proposed that the diffusion of boron in boron oxide (B_2O_3) liquid dominates at higher temperatures, a finding that was subsequently validated by Glassman and Li. In contrast to the L-W model, the Y-K model identified B2O2 as the main evaporation product rather than B_2O_3 . Furthermore, the Y-K model accurately predicted the ignition delay time of boron particles with different sizes, including those studied by Ermolaev [45] (particles with diameters of 34.5 µm and 44.2 µm) and Bedarev [46] (particles with diameters of 7.5~20 µm), which considered the boiling process of boron and the reflection of a shockwave from a wall, respectively. The Y-K model also provided insights into the kinetics of ignition and combustion, including the heating and melting of the boron trioxide layer on the particle surface, the evaporation and heating of pure boron particles, and the melting, boiling, and corresponding chemical reactions, which were found to be in good qualitative agreement with experimental data over a wide range of temperatures (2200 to 3000 K).

Combustion Mechanism of Boron Particles

Overall, the combustion of boron particles has been found to be a relatively simple process compared to the ignition stage. This is because the combustion stage primarily involves a heterogeneous surface reaction, as opposed to the more complex ignition stage. Research on the combustion of large boron particles with diameters greater than 35 um has suggested that diffusion plays a key role in controlling the combustion process. Specifically, Macek and Mohan [47] observed that the combustion time of large boron particles followed the d^2 law, which means that the combustion time was proportional to the square of the particle size. Interestingly, this behavior is similar to that of hydrocarbon droplets.

King 48] proposed a groundbreaking theory that challenged the conventional wisdom about the diffusion transport rate during boron combustion. Through his research and data analysis, King demonstrated that the diffusion transport rate cannot approach infinity with a decrease in particle size, solely through diffusion control. He revealed that the combustion stages of boron particles are governed by a combination of diffusion ($d \land 2 law$) and chemical kinetics ($d \land 1 law$), and this control mechanism varies depending on the particle size. King's work estimated that the transition from diffusion control ($d \land 2 law$) to chemical kinetics control ($d \land 1 law$) occurred in the range of 15-30 um in diameter for boron particles, although the exact point of transition was found to be dependent on pressure. This groundbreaking theory has opened up new avenues of research and provided valuable insights into the fundamental combustion process of boron particles.

Li and Williams [45,49] reported a unique finding regarding the combustion of small boron particles, wherein they observed that the combustion process was primarily controlled by chemical kinetics, following a d^1 law.

Their research further revealed that the combustion time of these particles was the sum of the time required for oxygen diffusion to the surface of boron particles and the time required for boron surface oxidation through the reaction $2B + O_{2} - B_{2} + O_{2}$. Based on this novel discovery, they proposed the L-W combustion model that considers both gas diffusion processes and chemical kinetics, providing valuable insights into the combustion of small boron particles.

With the help of Kuo's [47,50] modifications to the Y-L combustion model, Figure 2 depicts the enhanced model that allows for the determination of critical conditions for diffusion control and kinetic control. In order to understand the control mechanism of the boron combustion stage, Kuo introduced the concept of the Damkohler number (Da). Chemical engineers commonly use Da to characterize chemical reaction kinetics across a range of systems. Da is typically defined as the ratio of the characteristic time of a chemical reaction to the characteristic time of diffusion of reactants or products. Mathematically, this relationship can be expressed as Tt/Tr, where Tr represents the characteristic time of the chemical reaction and Tt refers to the characteristic time of diffusion.

3. Effect of surface coating on boron

Polybutylene terephthalate (PBT) coating on Boron, investigating the impact of Polybutylene terephthalate (PBT) coating on Boron, we explored how PBT-coated boron affects the charging process of boron-based propellants. Our research unveiled that the PBT coating on boron led to a significant increase in pH values, compared to raw boron, which showed pH values ranging from 3-4. The results of studies on acidity analysis, revealed that PBT-coated boron had a pH above 6.50, indicating a noticeable decrease in acidity when compared to Polyethyleneimine (PEI) coated boron.

Boron coated with LiF, the process of coating metal fluorides on surfaces is often achieved through the neutral precipitation method, and in the case of boron powder, a uniform deposition layer of LiF can be achieved using LiOII and IIF as precursors. This LiF coating has been shown to enhance the charging process of HTPB propellant by increasing the pH of the BOLIF suspension system from 4 to 7.4, and significantly reducing the viscosity of the HTPB system. Additionally, the use of LiF coating on boron powder has been found to improve combustion efficiency by accelerating the removal rate of B2O3. In fact, in tests conducted on boron-containing propellant, the ignition delay time decreased from 70.475 to 23.585 seconds before and after coating, as measured by electric wire heating ignition. These findings highlight the potential of LiF coating as a valuable technique for enhancing the performance of propellants containing boron powder.

Boron coated with Fe, through the process of decomposing Fe(CO) in octadecene (ODE) to create Fe nanoparticles [106], a successful synthesis of Fe-coated boron was achieved. The resulting material exhibited enhanced combustion properties compared to pure boron, including a faster combustion rate and higher combustion temperature. Interestingly, it was found that B99 (with a purity exceeding 99%) exhibited the highest combustion rate prior to coating, while B95 (with a purity exceeding 95%) exhibited the most pronounced modification effect after coating. These findings highlight the potential for tailored synthesis approaches to optimize the properties of novel materials for specific applications.

Boron coated with Mg, magnesium powder plays a pivotal role as a propellant additive due to its unique properties. It exhibits low oxygen consumption, exceptional ignition performance, and its combustion products have a low molecular weight. Coating magnesium on boron powder provides an effective way to lower the ignition point, promoting faster ignition and combustion of boron particles. The incorporation of Mg in B@Mg compound can react with BO, resulting in the formation of B at high temperatures, thus reducing ignition time and improving the combustion efficiency of boron particles. This synergy between magnesium and boron powders highlights the exceptional possibilities of harnessing the properties of individual elements to enhance their collective performance.

Boron coated with Ti, at a temperature of around 2000K, uncoated boron particles can be ignited, but titaniumcoated boron particles only need a temperature of 1400K to ignite. The underlying mechanism for this phenomenon is proposed as follows: (1) the reaction between titanium and boron, in the presence of oxygen, increases the heating rate of the particles; (2) the titanium oxide (TiO2) and titanium boride (TiB) layers on the particles are destroyed by the mechanical stress generated by thermal expansion, which results in the inner layer of boron being exposed to oxidation without any protection from the outer layers.

Boron coated with metal oxides (TiO2 or SnO2), metal oxides have been a popular choice for combustion catalysts in propellants, and recent research has focused on synthesizing boron particles with metal oxide coatings. Boron nanoparticles coated with metal oxide have been successfully synthesized using various methods, such as spray-drying, wet chemistry, and chemical vapor deposition. One study by Lee utilized the spray-drying method to prepare spherical boron nanoparticles coated with metal oxide (such as TiO2 or SnO). The metal oxide coating allows boron to obtain oxygen from the metal oxide, promoting oxygen to react with active boron in the inner shell. This results in improved combustion performance of boron powder.

Interestingly, the thickness of the TiO2 coating layer has a significant impact on the mass gains, with thicker coatings resulting in less mass gain. This phenomenon can be utilized to create titanium dioxide-coated boron particles that have high oxidation resistance, protecting the surface of the boron oxide layer from further oxidation. Overall, metal oxide-coated boron particles show great potential as combustion catalysts in propellants.

4. Conclusion

Boron powder serves as a superior metal fuel, offering significant calorific value and promising potential for use in explosives and propellants. This review paper encompasses an examination of the ignition and combustion mechanisms of boron powder as well as its surface modification techniques. The intricate ignition and combustion processes associated with boron powder, coupled with its suboptimal energy release, currently restrict its broader application in these fields. Several areas merit further investigation:

1. Enhanced research on the ignition and combustion characteristics of boron particles is essential, focusing on the impact of thermal mechanical properties, shell thickness, particle size, and the crystalline changes in boron oxide layers on boron's ignition and combustion.

2. It is crucial to ascertain the influence of coatings, the surrounding atmospheric conditions, and additives in propellants on boron ignition and combustion behaviors. Further studies could delve into the optimal coating thickness and mechanisms for controlling it, potentially through molecular dynamics simulations that analyze interactions between the coatings and boron powder.

3. There is an urgent need for the development of new materials for packaging with superior performance, along with innovative methods that offer control over the packaging process.

4. Considering the shortcomings of single-layer coating materials, the combustion efficiencies of multicomponent coatings, including binary or even ternary systems, should be explored.

5. So far, research has predominantly focused on basic laboratory studies concerning boron powder's coating modifications and their ignition and combustion efficiencies. Future studies should link these coating strategies to propellant performance, examining factors such as specific impulses, critical pressures in combustion chambers, and pressure temperature coefficients related to boron powder coatings.

6. Ultimately, advancing the application of coated boron powder in industrial settings remains a critical goal.

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