Strategies to Enhance Ignition and Combustion Characteristics of Boron-Based Fuel-Rich Propellants for Ducted Rocket Applications

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Abstract— **Boron-based fuel-rich propellants have attracted substantial research interest in recent years, especially for applications in airbreathing propulsion systems due to their exceptional potential combustion energy. However, boron exhibits a distinctive two-step reaction mechanism, in which an oxide layer develops on the particle surface, inhibiting the effective diffusion of B and O_2 to the flame zone. In this context, the present article explores some strategies that have been investigated to improve the ignition and combustion efficiency of boron. These strategies include the use of nanoscale boron particles, incorporation of metal fuels with lower ignition temperatures, integration of fluorine-containing species into the propellant formulation, and application of surface coating techniques.**

Keywords— **Fuel-rich propellant, Boron, combustion.**

I. INTRODUCTION

The use of boron-based fuel-rich solid propellants for airbreathing applications has attracted recent research interests due to its exceptional theoretical volumetric/gravimetric heat of combustion. In comparison with a typical hydrocarbon fuel, e.g. kerosene or polymeric materials such as HTPB, amorphous boron exhibits a gravimetric energy density about 40 % higher [1], surpassed only by beryllium. However, Be and its compounds, such as BeO, are highly toxic, so their use in propulsion systems is no longer widespread. Boron also has the highest volumetric energy of all elements and a high stoichiometric air/fuel ratio (approx. 10 [2]), which is particularly attractive for ram-rocket engines.

Unfortunately, the theoretical energy potential of boron cannot be fully extracted during the burning process. Ulas *et al.* [3] suggest there are two major reasons for the ignition and combustion performance deficiencies of boron: (1) the presence of a naturally formed oxide layer on the surface of boron particles which causes a significant ignition delay and (2) the kinetically favorable formation of metaboric acid (HBO_2) in hydrogen-containing environments, which significantly reduces the energy release during the combustion process of boron particles.

Furthermore, boron-based fuel-rich propellants pose some additional challenges in comparison to typical composite propellants due to their high metallic fuel content and low oxidizer content [4]. These highly metalized propellants exhibit a reduced pressure dependence in the burning rate, which causes problems in thrust modulation. While a regular propellant's burning rate pressure exponent is typically above

0.5, boron-based fuel-rich propellants often exhibit a burning rate pressure exponent lower than 0.3 at pressures below 1.0 MPa [4].

The low oxidizer content in boron-based propellants also poses difficulties in achieving sustained combustion. Boron particles' combustion requires the surrounding gas temperature to be of the order of 1900 K [5]. At this temperature, the vaporization rate of the oxide layer is significantly increased, allowing for the fast oxidation of boron particles.

The occurrence of these phenomena highlights the need for further in-depth investigations into the combustion of boron. In this context, the present study undertakes a comprehensive literature review of recent methods utilized to improve the oxidation process of boron particles. To this end, the fundamental workings of TDR engines will be presented, followed by a brief overview of metal particle combustion.

II. THROTABLLE DUCTED ROCKET TECHNOLOGY

Throtablle ducted rockets, also known as ramrockets, constitute a prominent alternative for air-to-air missile propulsion systems [6]. They integrate the advantages of air-breathing systems, such as superior range, higher flight speeds, and thrust control capabilities, with lower costs, simplicity, and readiness of conventional solid rocket engines. TDR engines consist of two combustion chambers, as shown in Fig 1. The primary combustor works as a gas generator, where an end-burning grain produces fuel-rich gas which is injected into the secondary combustor. Subsequently, this fuel-rich gas burns with the compressed air admitted from the atmosphere through the air intakes [1].
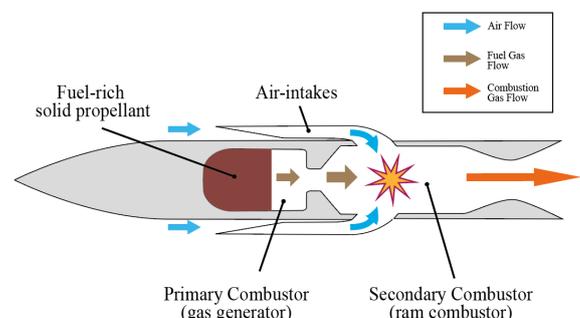

Fig. 1. Schematic of a throttleable ducted rocket

Integration with boosters is essential to accelerate the missile from its launch speed to the minimum operational speed

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required for the ramjet engine [7]. During the boost phase, the air intakes and air inlet ports leading to the ramcombustor must be closed and they need to be rapidly opened during the transition phase to minimize deceleration between the boost and sustain phases. Unlike mechanical moving parts found in turbine compressors, the air is compressed with shock waves induced by the geometric design of the air intakes [8].

Sustain propellants used in gas generators are central components in the design of ducted rocket engines. Distinct from conventional solid rockets, propellants used in air-augmented propulsion systems replace a considerable portion of the oxidizer in their formulations with fuel. However, the amount of fuel incorporated into the grain is limited by minimum combustion and processibility requirements. For example, the propellant formulation must be adjusted to achieve a high pressure exponent (n) of the burn rate (r), enabling active thrust modulation through a control valve [7]. Table I offers a comparative analysis of typical propellant formulations for solid and ducted rockets applications.

TABLE I

TYPICAL PROPELLANT FORMULATIONS [2].

		Solid Rocket	Ducted Rocket
Oxidizer	%	$60-90$	$20-55$
Metalic Fuel	%	$0-22$	$0-55$
Organic Fuel	%	$10-15$	$30-80$

Maximizing properties like density and energy release is crucial when choosing materials for Ducted Rocket fuel-rich propellants. Metal particles are the compounds that best fulfill these criteria and are widely employed in solid propellant formulations. In addition to the performance advantage among other metallic fuels, boron-based propellant also offers autoignition in the afterburner and isn't sensitive to combustion and flameout instabilities [7].

III. METAL PARTICLES BURNING

There are two types of metal combustion that can occur in oxidizing environments: homogenous combustion and heterogeneous combustion. To differentiate between these combustion processes, Glassman's criterion [9] was established. According to this criterion, if the oxide boiling/volatilization temperature is higher than the boiling temperature of the metal ($T_{vol} > T_b$), the combustion takes place in the vapor phase, and it is classified as homogeneous combustion. The temperatures of interest for metal fuels used in solid propellants are presented in Table II.

TABLE II

METAL FUEL AND OXIDE PRODUCT TEMPERATURES [9].

Metal	T_b [K]	Oxide	T_{vol} [K]
Al	2791	Al_2O_3	4000
Be	2741	BeO	4200
B	4139	B_2O_3	2340
Li	1620	Li_2O	2710
Mg	1366	MgO	3430
Ti	3631	Ti_3O_5	4000
Zr	4703	ZrO_2	4280

Note: T_{vol}, volatilization temperature under ambient conditions $T = 298$ K, $P = 1$ atm; T_b, metal boiling point at 1 atm.

It is important to note that the boiling temperature of a metal and the volatilization temperature of its oxide are dependent on pressure conditions [9]. Therefore, it's necessary to properly select the temperature values for the application of Glassman's criterion. For instance, the combustion of aluminum at atmospheric pressure (1 atm) occurs homogeneously, while at pressures above 200 atm, it transitions to heterogeneous combustion [9].

A. Homogenous combustion

When the volatilization temperature of the metal oxide exceeds the boiling temperature of the metal, the fuel and oxidizer undergo homogeneous combustion in the vapor phase, leading to the formation of a diffusion flame sheet above the surface of the particle [10]. This mechanism is similar to the burning of liquid hydrocarbon fuel droplets in air/oxygen. Specifically, in the case of aluminum combustion, gas-phase aluminum oxide is produced and rapidly nucleates to form a liquid vapor dispersion. This condensed phase disperses both inward and outward from the envelope, diffusing towards the particle and through the surrounding $O_2(g)$.

The aluminum oxide product accumulates on the surface of the liquid aluminum droplet, forming a growing oxide lobule [10], as illustrated in Fig 2. This phenomenon is attributed to the immiscibility of $Al_2O_3(l)$ in aluminum, combined with the high surface tensions of these species. Consequently, the system's temperature becomes limited by the volatilization temperature of $Al_2O_3(l)$ as the lobule absorbs a significant portion of the available thermal energy without volatilizing [9].

Homogeneous combustion of metal fuels presents significant advantages due to its capability to promote the combustion of reactants above the metal particle surface, enabling high diffusion rates through the flame envelope. This process contributes to improved combustion completeness and results in a more efficient energy release.
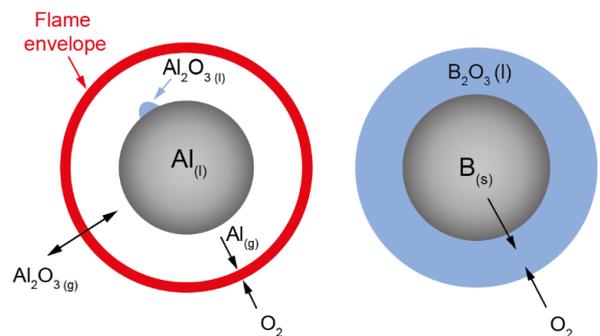

Fig. 2. Metal particles combustion mechanism in air. Left: Al homogeneous combustion. Right: B heterogeneous combustion. Adapted from [10].

B. Heterogeneous combustion

Heterogeneous combustion, on the other hand, occurs when the thermochemical characteristics of the fuel and the formed oxide do not meet Glassman's criterion. In such conditions, not all reactants interact in vapor phase, posing difficulties in the combustion mechanism. Under atmospheric pressure, according to Table II, boron ($T_b = 4139$ K) burns in oxygen heterogeneously within a flame zone at the surface of the boron particle to form boron oxide ($T_{vol} = 2340$ K). The

combustion mechanism of boron in oxidizing environments has been extensively studied and consists of two stages [11]–[13].

In the first stage of boron oxidation, a condensed phase boron oxide layer forms at the particle surface, since the flame temperature is lower than the oxide volatilization temperature. The particle tends to be uniformly coated by the generated product due to the low surface tension of B_2O_3, as shown in Fig. 2, unlike the localized agglomeration observed in aluminum combustion. As the reaction progresses, the oxide layer thickens, inhibiting the diffusion of O_2 and B species through the combustion product layer and gradually reducing the reaction rate [13].

When the system's temperature surpasses boron oxide volatilization temperature, the second stage takes place. During this phase, the liquid oxide layer starts to progressively be consumed, vaporizing into $B_2O_3(g)$. If this condition persists until the remaining oxide layer becomes thin enough, an abrupt temperature increase will occur [13]. As a result, the oxide layer will be completely removed, enabling the direct reaction between the neat boron particles and the surrounding oxygen on the particle surface.

The practical difficulties of ignition and combustion of boron particles are primarily attributed to the initial stage of the oxidation process. The oxide coating envelope leads to a diffusion-limited oxidation mechanism that is strongly dependent on the particle's size. Thus, removing the boron oxide layer plays a critical role in improving the ignition and combustion characteristics of boron-based propellants [10].

IV. STRATEGIES

Several methods have been explored to enhance the combustion performance of boron fuels. These strategies include (A) reducing the particle size of boron, (B) igniting in a fluor-containing environment, (C) incorporating metals with lower ignition temperature and higher volatility, and (D) implementing thin coatings. The subsequent sections briefly explain each method and an overview of the research progress.

A. Nanoscale boron

According to Yetter et al. [14], the reduction of metal particle size from micrometers to nanometers has been shown to strongly influence thermophysical properties and combustion characteristics. This observation is attributed to the significant increase in the ratio of surface atoms to bulk atoms in particles with diameters less than 10 nm [15], [16]. Due to the distinct thermal and physical properties of surface atoms compared to bulk atoms, the bulk material can start to exhibit properties similar to those of the surface atoms in small particles. These differences are evident in various properties such as melting point, heat of fusion, particle burning time, and ignition temperature [14].

Fig. 3 shows the burning times of nanosized and micron-sized aluminum particles [17]. In the case of large particles (on the order of 100 nm), the burning rate of metals was observed to be directly proportional to the square of the particle diameter, commonly referred to as the "d^2 law". However, for particle sizes smaller than 10 μm, the experimental data indicates a linear dependence on the particle diameter. A similar behavior can also be observed in other metallic fuels

[14]. Nanoscale particles also have higher specific surface area and better contact with oxidizers, which promote shorter ignition delays and combustion times.
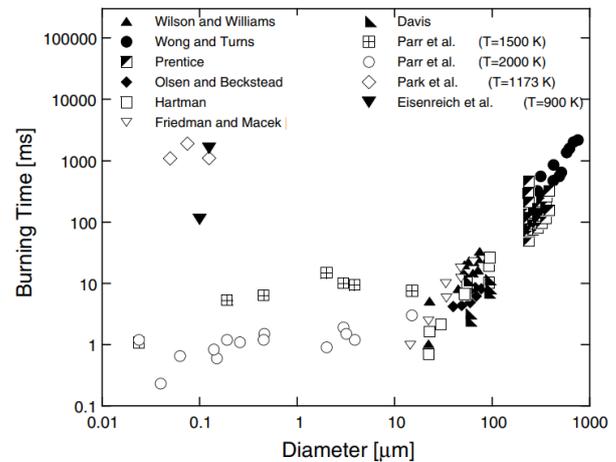

Fig. 3. Aluminum particles burning times of varying particle size [14]. References for the data can be found in [17].

In boron oxidation systems, it is expected that nanoscale boron would exhibit a thinner oxide layer compared with micron-sized boron. The surface area of nanoscale boron is approximately ten times greater than that of commercially available amorphous boron with a diameter of 0.4-0.9 μm [18]. Li and Williams [19] observations revealed a transition from diffusion-limited to kinetically-controlled combustion as the boron particle size is reduced to the range of 1-10 μm under the same pressure conditions. Furthermore, investigations conducted by Krier et al. [20] revealed that the ignition delay times for boron combustion experienced a significant reduction from $400 - 800$ μs to $30 - 50$ μs as the boron particle size was decreased from 15 μm to 1 μm, within high-pressure and high-temperature environments (approximately 2250 K and 8.5 atm) generated in a shock tube.

The utilization of nanoscale boron in propellant formulations also has been proven as an effective approach to enhance combustion performance. It was been demonstrated that the incorporation of nano boron in paraffin fuels for hybrid rocket combustor configurations leads to about 80 % boron combustion efficiency [21]. Recent research on the combustion of nano boron and sub-micron boron with poly-tetrafluoroethylene (PTFE) samples was conducted by Feng [10]. The study utilized a simultaneous differential scanning calorimeter/thermogravimetric analyzer (DSC/TGA) with on-line evolved gas analysis under both argon and air atmospheres. The results demonstrated that nano boron particles facilitated the reaction between boron and PTFE, resulting in a reduction of ignition temperature by 130 K at 3.2 MPa.

B. Fluorine-containing species

The addition of fluorine as an oxidizer into the combustion environment has been proven highly effective in reducing boron ignition delay and enhancing boron combustion [3], [22]–[24]. The reaction between fluorine gas and boron results in the formation of boron fluoride instead of boron oxide. The gas phase behavior of BF_3 is highly favorable for combustion

physics, as its boiling temperature at atmospheric pressure (about 173 K) is much lower than B_2O_3 boiling temperature. Fluorine species also can react with the formed diffusion-limiting boron oxide layer, as presented in (1), increasing the oxide removal rate and thereby accelerating the first-stage combustion over a wide range of combustion regimes [22].

$$B_2O_3(l) + 3\,F_2 \longrightarrow 1.5\,O_2 + 2\,BF_3(g) \qquad (1)$$
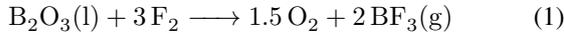
Incorporating fluorine species can also mitigate the formation of HBO_2 by promoting the favorable generation of gaseous OBF and BF_3 in hydrogen-containing environments [25]. This is particularly relevant for propellant applications, which typically contain hydrocarbons in their formulation. As a result, the first-stage burning time is significantly reduced, leading to a substantial increase in the overall energy release rate, as the slow conversion from $HBO_{2(g)}$ to $B_2O_{3(l)}$ is effectively reduced.

There are various ways to introduce fluorinated species into the combustion process. Notably, the use of fluorocarbon oxidizers is distinguished as one of the most easily obtainable sources of reactive fluorine in the context of metal-based energetic materials. For instance, a well-known example is the use of Magnesium/Teflon/Viton (MTV) compositions, which have been employed in flare applications [26]. In Brazil, through various experiments conducted by Barros *et al.* [27], it has been demonstrated that the MTV composition developed by the *Instituto de Aeronáutica e Espaço* (IAE) is qualified as safe for fabrication, handling, and application in the ignition systems of solid propellant rocket motors and military flares.

Polytetrafluoroethylene (PTFE), commonly known as Teflon, presents a prominent alternative for boron-based fuel-rich propellants, due to its hydrogen-free composition. The PTFE decomposition process starts slowly at temperatures around 530 K and significantly accelerates in temperatures above 670 K [28]. During the decomposition of PTFE, the major products consist of vapor phase C_2F_4, C_3F_6, and C_4F_8 [28]–[30], with C_2F_4 being the dominant component, comprising approximately 75% by weight of the fragments.

C. Metalic additives

An alternative approach to improve the efficiency of boron combustion is to focus on enhancing the available energy within the system during the initial stage of boron oxidation. The thermal diffusion barrier is removed by elevating the temperature of the oxide layer beyond its volatilization temperature, enabling self-sustained burning. This can be achieved by incorporating metal fuels with lower ignition temperatures and rapid volatilization properties [10].

Magnesium powder has emerged as a popular metal additive to enhance boron-based fuel-rich propellants' ignition and combustion performance. The inclusion of Mg fuels has notably resulted in improved boron combustion efficiency. Sandall *et al.* [28] results demonstrated that the addition of 10 wt.% magnesium to B/HTPB fuel grains led to a substantial increase in combustion efficiency, ranging from 84% to 90-94%, in a subscale model ramjet.

D. Coating boron particles

Surface coating of boron powder has attracted considerable attention across various research areas as a promising approach to harness the benefits of other strategies directly on the boron surface. Several studies highlight the potential of surface coating technology in enhancing the ignition and combustion performance of metal fuels [31]–[34]. Boron particles have been coated with a variety of materials, including metals [35], [36], oxidant [37] or energetic binders [38], and fluorine-containing compounds [39], [40].

At first, the surface coating of boron particles with metals such as Al, Mg, Ti, and Fe is promising due to their capability to react with boron and form metal borides with low ignition points. This reaction results in more energy release at high temperatures, effectively promoting the ignition and combustion of boron [41]. Previous experiments investigating the ignition temperature of Mg-coated boron, Mg, and aluminum alloy [42] demonstrate that the Mg-coated boron particles have a significantly lower ignition temperature of 469.07 K in oxygen, in contrast to Mg and Al alloy, which exhibited higher ignition temperatures of 599.73 K and 513.64 K, respectively. Moreover, according to Rosenband's theoretical studies on the ignition and combustion characteristics of titanium-coated boron particles [33], surface coating led to a significant reduction in the ignition temperature from 2000 K to 1400 K.

Glycidyl azide polymer (GAP) coating provides, besides high heat release, some processability advantages, including a reduction in slurry viscosity during propellant blending and casting, which is attributed to the acidity and porous nature of boron [38]. In this study, it has been found that GAP-coated boron particles exhibit lower ignition delay times compared to raw boron powder, with values of 7.6 ms and 16.2 ms, respectively.

Research conducted by Liu *et al.* [40] explored the effects of coating boron particles with lithium fluoride (LiF) and Viton A (fluorinated rubber) in boron-based fuel-rich propellants. The propellant formulation employed amorphous boron with a purity of 90%-92% combined with magnalium, ammonium perchlorate (AP), and HTPB.

The use of a LiF coating on boron particles was based on the concept that it directly enhances oxide removal through the reaction mechanism depicted in Fig. 4.
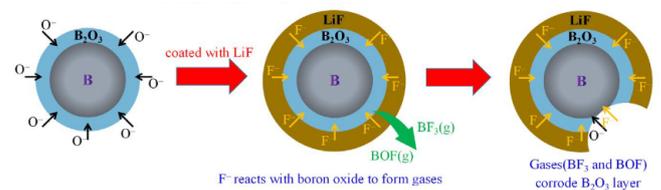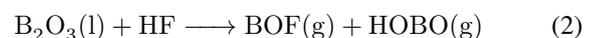

Fig. 4. Reaction mechanism of LiF-coated boron particle. Adapted from [41].

On the other hand, the addition of Viton A aimed to produce HF, which subsequently undergoes reactions with $B_2O_3(l)$ leading to the formation of BOF and BF_3 gases (3).

$$B_2O_3(l) + HF \longrightarrow BOF(g) + HOBO(g) \qquad (2)$$
$$B_2O_3(l) + 6\,HF \longrightarrow 2\,BF_3(g) + 3\,H_2O \qquad (3)$$

The study revealed that propellants with LiF-coated boron particles exhibited reduced burning rates at low pressure but showed no significant change at high pressure. In contrast,

propellants with Viton A-coated boron particles displayed a substantial reduction in burning rate [40]. Laser ignition tests confirmed that the propellant with LiF-coated boron particles demonstrated the shortest ignition delay, while the propellant with Viton A-coated boron particles exhibited the longest ignition delay [40].

V. CONCLUSION

This paper presents a comprehensive literature review concentrating on improving the combustion and ignition of boron-based propellants in TDR engines. The review provided some insights into the working principles of these engines, along with explanations of metal particle burning. It became evident that primary challenges in realizing the full potential energy release of boron reside in the formation of a boron oxide layer on the particle's surface and the creation of metaboric acid in hydrogen-containing environments. As a result, the strategies discussed primarily focus on removing the oxide layer or elevating the particle surface temperature until it reaches the boron oxide vaporization point.

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