Dual-stage Ignition of Boron Particle Agglomerate

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1   Introduction

Elemental boron is an attractive fuel for air-breathing propulsion applications since it has the highest volumetric energy density among all elements in reaction with oxygen, and the third highest energy density on a mass basis. Due to the low melting point of solid boron oxide and the high surface tension and viscosity of liquid boron oxide [1], agglomeration of the initially micron size (crystalline boron) or submicron size (amorphous boron) particles is an inescapable phenomenon occurring in all propulsion applications. Since the particle agglomerate has a large internal reactive surface area and non-uniform accessibility to oxygen, its ignition behavior considerably differs from that of a single micron-size boron particle or a low density boron dust cloud.

In order to further develop the applications of boron-based high-energy fuel, it is important to acquire a fundamental understanding of the key physical mechanisms responsible for the ignition characteristics of boron particle agglomerates. However, only two prior studies have attempted to experimentally investigate the ignition process of boron agglomerates. The first study by Shevchuck et al. [2] examined ignition of boron particle agglomerates suspended on a tungsten wire in a flow of a dry air heated by an electrical furnace up to 1100 K. They found that agglomerates with sizes from 2 to 5 mm comprised of powders of amorphous and crystalline boron can be ignited at temperatures as low as 800 K, in contrast to much higher temperatures, around 1900 K, that are characteristic for ignition of individual boron particles [3]. This dramatic reduction in ignition temperature did not, however, result in transition to full-fledged boron combustion as expected after a short period of rapid temperature rise; the agglomerates quenched and the degree of boron oxidation was low. The second study by Faeth et al. [4] investigated ignition and combustion of boron slurry agglomerates ($d = 0.175$-0.8 mm) supported by a quartz wire in the post-flame gases of a flat-flame burner in the higher temperature range between 1690 and 1950 K. By studying agglomerate images from high speed camera recordings and the surface morphology of the agglomerates quenched at different stages of ignition and combustion, the authors qualitatively identified a multistage ignition and combustion process consisting of the following sequence: 1) heat-up of a porous agglomerate with particles covered by a solid oxide, 2) melting of the oxide to form a liquid coating around each particle with a decrease in the overall agglomerate porosity, 3) gasification of the oxide layer that starts around 1900 K leaving an open, porous structure of reacting boron particles, and 4) melting of the boron (m.p. 2450 K) and transformation of the agglomerate to a molten drop consumed by combustion in the external diffusion regime. The authors emphasized observation of the two-stage ignition phenomenon that occurs consecutively. The first stage (the same as observed by Shevchuk et

al.) occurs at relatively low temperatures under the progressively increasing thickness of the oxide layer enveloping individual particles. The second stage follows the first after the agglomerate attain higher temperature that leads to evaporation and removal of the oxide from at least part of the agglomerate surface, thus creating conditions for self-sustained combustion. While the proposed scheme appears plausible, it can be considered only as a hypothesis since those authors did not study or explain critical parameters leading to the second stage ignition or quenching, nor did they performed any in-situ measurements of the agglomerate temperature during the ignition process.

The main focus of the current work is to numerically model the ignition process of boron particle agglomerates considering the oxygen diffusion and concentration gradient inside the agglomerate. The simulated results accurately capture the behavior of quenching after one-stage ignition and dual-stage ignition processes in response to the ambient flow conditions (i.e., flow temperature and oxygen concentration). Experimental investigation of the ignition characteristics of boron agglomerates were performed by introducing 2-3 mm agglomerates into the high temperature oxidizing flow generated by the combination of hydrogen combustion and electrical heating. Experimental results show a qualitatively good agreement with the simulated results in terms of the time history of the ignition process and its dependence on the flow parameters.

2   Model Description and Results

In order to model the ignition process of the boron agglomerate, the agglomerate is divided into a series of N equidistant concentric layers. At each time step, as shown in Figure 1(a), the oxygen concentration profile inside the agglomerate is approximated as a step function by solving the one-dimensional diffusion equation through a reacting media along the radial direction across each layer. As a fixed oxygen concentration is assigned to each layer, the rate of heat generation and boron oxidation is thus uniform across each layer at a given time step. In order to take the effect of the accumulated boron oxide reducing the porous diffusion channels width into account, the concept of a bottle-neck diffusion area, as schematically illustrated in Figure 1(b), has been built into this model. When in the process of the calculations, the thickness of the boron oxide film exceeds the chosen critical width of the bottle-neck area in the ith layer, the oxygen concentration and the rate of boron oxide evaporation are determined to be zero in the layers underneath. The calculations of the rate of heat generation, boron oxidation, and boron oxide evaporation for each individual, fixed-diameter boron particle are adapted from King's model [5]. A detailed mathematical formulation of this model will be shown in the full paper.
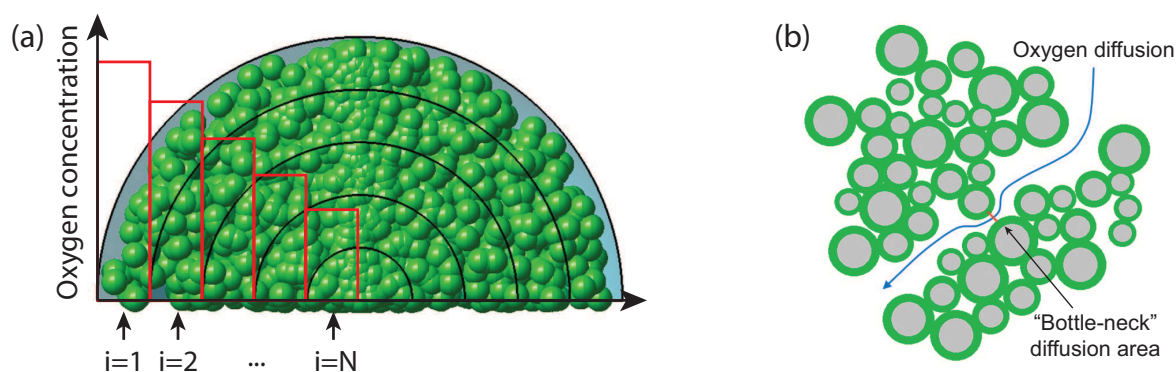

Figure 1: Three dimensional cross-sectional view of layered agglomerate model (a) and illustration of "bottle-neck" diffusion area concept (b)

The numerical simulation of the agglomerate reacting in dry oxidizing flow has been performed with

the model dividing the agglomerate into $N = 100$ equidistant layers. The calculated time-temperature history of the agglomerate that is rapidly inserted into the oxidizing flows having different temperatures but the same 20% oxygen content is shown in Figure 2(a). As shown in Figure 2(a), the model predicts a dual-stage ignition process of the agglomerate. Below 1150 K the agglomerate undergoes only an inert heating to the flow temperature. At about 1150 K, the thermal explosion within agglomerate rapidly elevates its temperature to the peak value of about 1500 K, after which the reaction practically stops and agglomerate starts to cool down to the flow temperature as an inert body. At flow temperatures above 1190 K, after the agglomerate temperature briefly plateaus at about 1600 K, the second ignition stage occurs and is manifested by a very sharp rise in the agglomerate temperature and the intensive evaporation of the boron oxide within the agglomerate. Figure 2(b) shows results of the model simulation of the agglomerate ignition process at different oxygen contents at a flow temperature of about 1050 K. The model predicts transition from the first to the second stage ignition at about 79% of O_2.
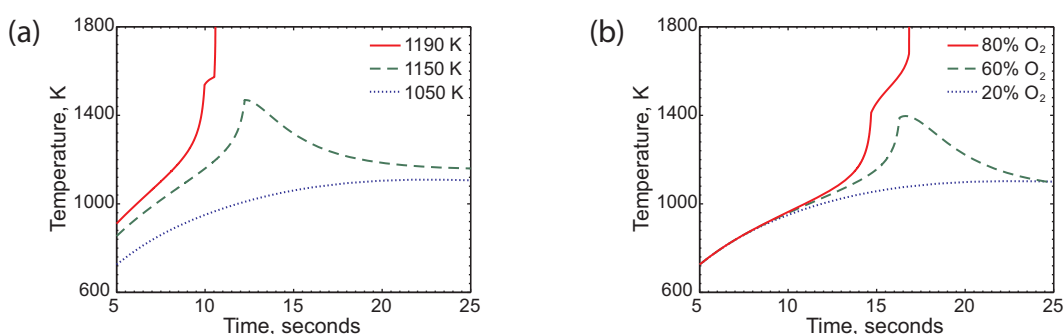
Figure 2: Model results showing time history of agglomerate temperature at various flow temperatures (a) and ambient oxygen concentrations (b)
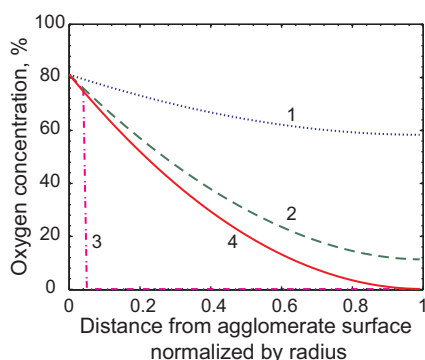
Figure 3: Model results showing oxygen concentration profiles inside the agglomerate at different stages of the ignition process

The model result shown in Figure 3 predicts the dynamical changing process of oxygen concentration profile inside the agglomerate during a dual-stage ignition process. The four curves represent the oxygen concentration with respect to the distance from the agglomerate surface at different ignition stages enumerated in a chronological order. This model result shows that the accessibility to oxygen is limited to the vicinity of the agglomerate surface at some point (curve 3) due to the blockage of the porous channels by the accumulated boron oxide. In this simulation, since the ambient oxygen concentration is above the critical condition, the agglomerate reaches a sufficiently high temperature to remove the boron oxide blocking the pores. Thus, the re-entry of oxygen into the deeper layers of the agglomerate after the reaction zone reducing to the minimum (curve 4) allows the agglomerate to undergo the second

stage of ignition. In the opposite case, when the flow parameters are below the critical conditions, the pore blockage progresses towards the surface of the agglomerate shrinking the reaction surface area to just external agglomerate surface as it quenches.

3 Experimental Set-up and Results

Crystalline and amorphous boron powders used in the experiments were obtained from Alpha Aesar. Average particle size and other parameters of both powders are summarized in Table 1. The boron agglomerates with diameter of 2-3 mm were prepared by drying boron/water slurry droplets at 90 °C for 24 hours. The densities calculated from the agglomerate weights and diameters were in the range of 0.75-0.82 g/cm^3 with standard deviation below 10%. The calculated average agglomerate porosity was about 70%.

Boron powder	Purity, % (metal basis)	Particle shape	Specific surface area, m^2/g	Diameter, μm
Crystalline	98.0	irregular	0.54	7.10
Amorphous	99.0	spheroid	26.0	0.21

Table 1: Characteristics of boron powders

The agglomerate to be tested was inserted into the heating flow using a sliding post on an optical bench. An end switch closed by the post at the moment of insertion triggered the computer data acquisition for the optical diagnostics. The heating flow was provided by thermoline tubular electrical furnaces using a combination of hydrogen combustion and electrical heating. The composition of the heating gas was adjusted by controlling the flow rate of each gas component (i.e., N_2, O_2, and H_2). The flow temperature at the exit of the ceramic nozzle was maintained and varied over a range of 700-1100 K, which was monitored by a thermocouple with 5 K accuracy.

The two-stage ignition process observed in the boron agglomerates was evident from light intensity traces registered by the photodiode, the dynamic spectral measurements of the agglomerate surface temperature, and the video recording of the ignition process. As can be seen from the photodiode traces shown in Figure 4, the sharp increase in agglomerate luminosity is observed after introduction of the agglomerate into an oxidizing flow having a temperature above some critical value. At low oxygen concentrations, however, the agglomerate rapidly quenches after the first ignition stage.
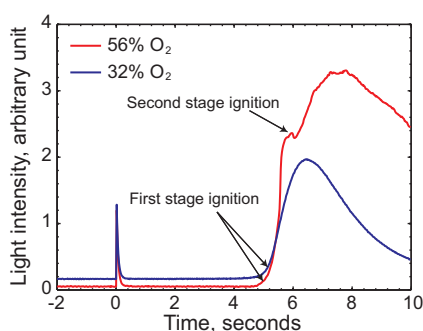

Figure 4: Experimental agglomerate luminosity history recorded by the photodiode during the first and the second stage ignition

Figure 5(a) illustrates the time history of agglomerate temperature during the first ignition stage followed by quenching. Figure 5(b) illustrates the case when the first ignition stage is immediately followed by

the second ignition stage, resulting in a self-sustained agglomerate combustion. The second ignition stage was observed only when the oxygen concentration in the flow was at or above some critical value that, as is shown below, was different for crystalline and amorphous boron agglomerates. As can be seen from Figure 5)(b), the trend in agglomerate temperature after the first stage ignition matches the luminosity trace and reaches a plateau around 1750 K before rapidly starting to rise again.
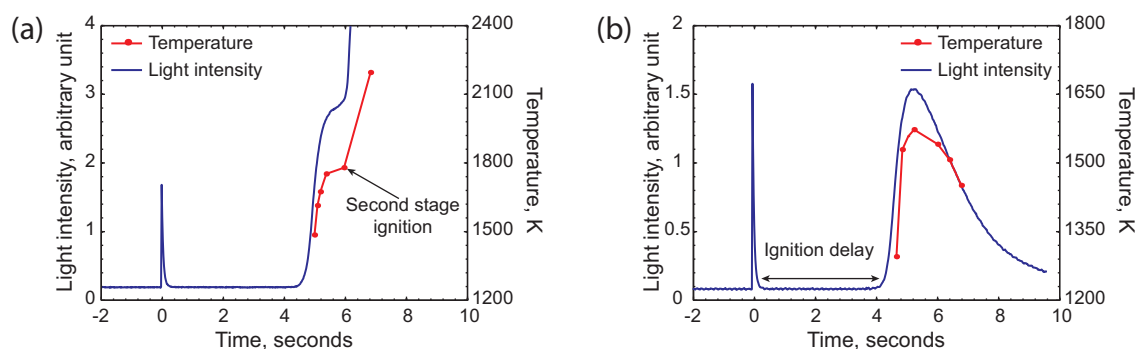

Figure 5: Time history of agglomerate temperature during the first stage ignition and quenching (a) and the dual-stage ignition (b) processes

The first stage ignition temperatures and the critical oxygen concentrations needed for the second stage ignition in both wet and dry flows were experimentally determined and are shown in Table 2.

Flow	Boron	1st ignition temperature, K	2nd ignition critical O_2 concentration, %
20 % H_2O	Crystalline	715	56
20 % H_2O	Amorphous	828	44
Dry	Crystalline	896	70
Dry	Amorphous	958	55

Table 2: Comparision of agglomerate ignition characteristcs in wet and dry flows

4 Conclusion

The model correctly predicts the qualitative behavior of the quenching and dual-stage ignition processes of boron agglomerate which were observed experimentally. The theoretical and experimental results also show a good agreement with the critical ambient oxygen concentration for the second stage of ignition in dry flow. Hence, this work further clarifies the fundamental understanding of the ignition characteristics of boron particle agglomerates. The consumption by boron oxidation and the reduction in the size of porous diffusion channels by the accumulation of boron oxide are found to be the key physical mechanisms which introduce an oxygen concentration gradient inside the agglomerate. Due to the non-uniform accessibility to oxygen, the agglomerate might undergo ignition and quenching or dual-stage ignition processes in response to the free flow conditions.

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