

Fabrication of TaB₂/mullite composites by combustion synthesis with excess silicon and B₂O₃ additions



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ARTICLE INFO

Article history:

Received 9 March 2016

Received in revised form

29 March 2016

Accepted 30 March 2016

Available online 1 April 2016

Keywords:

A. Powders: solid state reaction

B. Composites

D. Borides

D. Mullite

Self-propagating high-temperature synthesis (SHS)

ABSTRACT

Preparation of TaB₂/mullite composites from a cost- and energy-effective mixture of reactants was conducted by self-propagating high-temperature synthesis (SHS). The sample stoichiometry of 1.18Ta₂O₅ + (2.36x)B₂O₃ + 6Al + (2y)Si was formulated with x = 1.0–1.3 and y = 1.0–2.0 for the study of the effects of excess B₂O₃ (x > 1.0) and Si (y > 1.0) on combustion characteristics and product compositions. The synthesis reaction involved coreduction of Ta₂O₅ and B₂O₃ by Al and Si. The reaction exothermicity and combustion velocity increased slightly with increasing B₂O₃ but decreased considerably with Si. Formation of TaB₂ and mullite was improved by adopting excess B₂O₃ and Si to compensate for their evaporation loss at high combustion temperatures up to about 1600 °C. The sample with x = 1.3 and y = 1.5 was shown to yield the optimum formation of TaB₂ and mullite. Mullite grains with a tubing-like shape were produced from in situ formed SiO₂ and Al₂O₃ and had an atomic composition close to that of 3:2 mullite (3Al₂O₃ · 2SiO₂).

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1. Introduction

Mullite-based composites have attracted great attention due to the broadness of their industrial applications. Mullite with stoichiometries ranging from relatively silica-rich 3Al₂O₃ · 2SiO₂ (3:2 mullite) to alumina-rich 2Al₂O₃ · SiO₂ (2:1 mullite) is the only stable crystalline phase in the aluminosilicate system [1]. Owing to many favorable properties including high melting point, high creep resistance, good thermal stability in harsh oxidizing environments, good chemical resistance to alkalis and acids, high thermal shock resistance, low thermal expansion coefficient, low dielectric constant, and transmittance to infrared, mullite ceramics have been recognized as potential materials for use in the high-temperature structural, electronic packing, and optical applications [1–3]. For further improved fracture toughness, flexural strength, and thermal shock resistance, mullite-based composites with ZrO₂ and Al₂O₃ as reinforcements were extensively studied [4–8]. Lately, other ceramic and intermetallic additives, such as TiC, SiC, TiB₂, NbB₂, Al₂TiO₅, and MoSi₂, have gained increasing attention [9–17].

A variety of processing techniques with different starting materials have been employed to prepare mullite in either monolithic or composite forms, and they include the sol–gel method [18,19], reaction sintering [4,20], spark plasma sintering (SPS) [5,9,21],

solution combustion synthesis [22,23], self-propagating high-temperature synthesis (SHS) [11–13], and thermal explosion [16]. The SHS method takes advantage of highly exothermic reactions, and hence, has the merits of low energy requirement, short processing time, simplicity, high productivity, and structural and functional diversity of final products [24–26]. Moreover, combustion synthesis integrated with Al-based thermite reactions represents an in situ processing route of producing composite materials reinforced by Al₂O₃ [26,27].

In addition to the yield of Al₂O₃, high exothermicity of the thermite reaction is a great benefit for combustion synthesis. Therefore, the thermite-based SHS technique of different modifications has been utilized to prepare mullite combined with ceramic or intermetallic phases. For example, Zaki [11] fabricated the TiB₂/mullite composite by combustion synthesis from TiO₂–B₂O₃–Al–SiO₂ powder compacts preheated at 550 °C. With addition of ZrSiO₄ into thermite reagents of TiO₂–B₂O₃–Al, the TiB₂/ZrO₂/mullite composite was synthesized under a sample preheating temperature of 600 °C [12]. Because aluminothermic reduction of Nb₂O₅ was sufficiently exothermic, no prior heating was required for the Nb₂O₅–B–Al–SiO₂ sample to achieve self-sustaining combustion and produce NbB₂ and mullite in a composite form [13]. By means of high-gravity combustion synthesis, Wang et al. [14] obtained the Al₂TiO₅/mullite composite from a combustible mixture consisting of Al, NiO, TiO₂, and SiO₂ powders. In addition, Zaki et al. [16] prepared the MoSi₂/mullite composite by combustion synthesis in the thermal explosion mode with the MoO₃–SiO₂–Al powder compact.

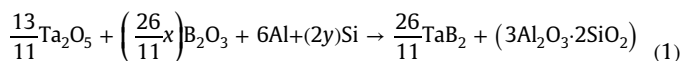
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The goal of this study is to adopt a low-cost and energy-effective mixture of reactants to prepare the TaB₂/mullite composite through combustion synthesis in the SHS mode. Tantalum diboride (TaB₂) is one of the promising ultra-high temperature ceramics (UHTCs) potential for the extreme environment applications requiring high thermal-shock resistance and durability in a rapid temperature change [28,29]. Mechanical properties of TaB₂ including bulk, shear, and Young's moduli, Poisson's ratio, and hardness are given in Ref. [30]. Starting materials adopted by this study included not only Ta₂O₅ and B₂O₃ to replace expensive Ta and boron, but also Al and Si powders as the reducing agents. Regarding the reaction exothermicity, the synthesis process involves coreduction of Ta₂O₅ and B₂O₃ by two reductants. Different from the previous studies with pre-added SiO₂ [11–14,16], the present study features the evolution of mullite from in situ formed SiO₂ and Al₂O₃. According to the literature [31,32], both B₂O₃ and Si have high vapor pressures and are volatile at elevated temperatures. Therefore, the specific objectives of this work are to investigate the need of excess B₂O₃ and Si additions for formation of TaB₂ and mullite, and to explore the influence of sample stoichiometry on combustion behavior.

2. Experimental methods of approach

The starting materials of this study include tantalum (V) oxide (Ta₂O₅) (Alfa Aesar, < 45 μm, 99.85%), boron oxide (B₂O₃) (Strem Chemicals, 99.9%), Al (Showa Chemical Co., < 45 μm, 99.9%), and Si (Strem Chemicals, < 45 μm, 99%). The stoichiometry of the reactant mixture is formulated based on the following equation.



where x and y are the stoichiometric coefficients for the measure of B₂O₃ and Si contents. The product composition shown in

Reaction (1) is balanced by $x=1$ and $y=1$, under which B₂O₃ and Si are in stoichiometric quantities. Excess amounts of B₂O₃ and Si are specified by $x=1.1$ – 1.3 and $y=1.25$ – 2.0 , respectively. Additional B₂O₃ and Si are to compensate for their volatilization loss during combustion. It should be noted that the loss of boron could be also caused by escape of gaseous boron oxide (e.g., BO), an intermediate phase generated from reduction of B₂O₃ [33,34].

Sample compacts were prepared in a cylindrical shape with 7 mm in diameter, 12 mm in length, and 50% in relative density by compressing the well-mixed reactant powders. Combustion experiments were performed under high-purity (99.99%) argon of 0.15 MPa. The combustion propagation rate and reaction temperature were precisely measured. Details of the experimental setup and technique were reported elsewhere [35].

3. Results and discussion

3.1. Combustion front velocity and combustion temperature

Typical combustion sequences illustrated in Fig. 1(a) and (b) are respectively associated with an exactly stoichiometric sample and a powder compact with excess B₂O₃ ($x=1.2$) and Si ($y=1.5$). It is evident that upon ignition the combustion front was readily formed and propagated progressively along the powder compact, confirming sufficiency of the reaction exothermicity. That is, the reactant mixture is energy-effective to sustain combustion without requiring additional heat. In contrast to those adopting SiO₂ powders that imposed a dilution effect on combustion [11–13], the synthesis process of this study gained thermal enthalpies from not only aluminothermic reduction of Ta₂O₅ and B₂O₃, but silicothermic reduction of these two oxides. When Si was utilized instead of SiO₂, mullite was produced from in situ formed Al₂O₃ and SiO₂. As clearly seen in Fig. 1(a) and (b), the self-sustaining combustion wave was composed of two or three localized reaction

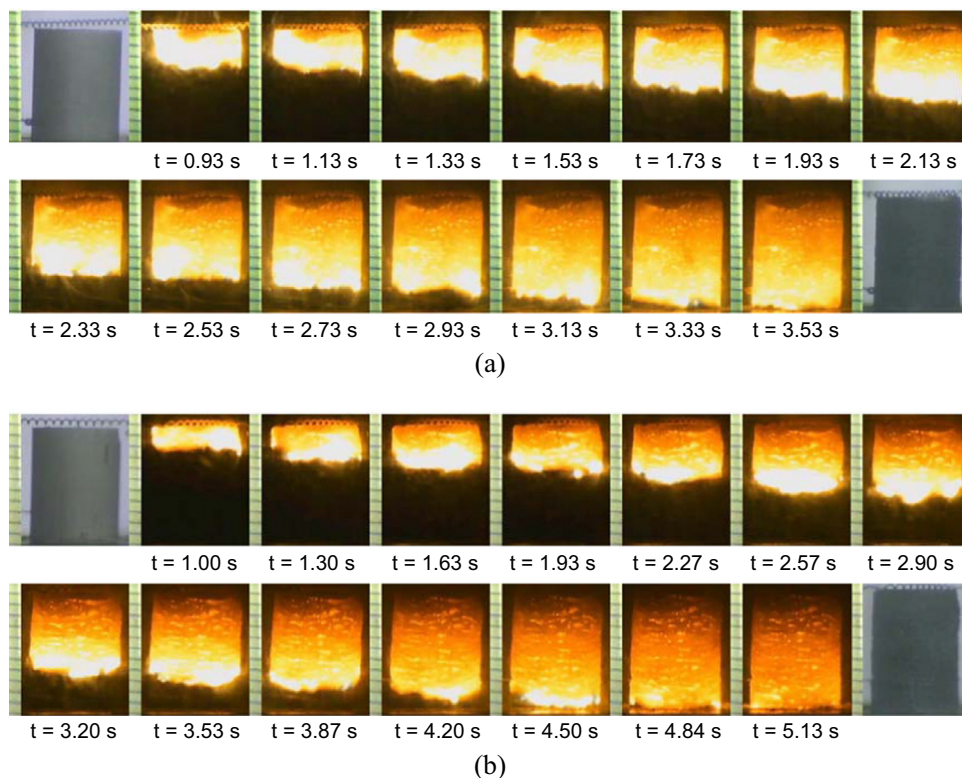


Fig. 1. Recorded sequences illustrating self-sustaining combustion wave traversing along samples of (a) $x=1.0$ and $y=1.0$ and (b) $x=1.2$ and $y=1.5$.

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