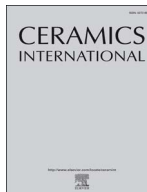




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# Preparation of TaB<sub>2</sub>-SiC oxidation protective coating for carbon materials by liquid phase sintering

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## ARTICLE INFO

### Keywords:

TaB<sub>2</sub>  
Powders  
Coating  
Liquid phase sintering  
Oxidation protection  
Borides

## ABSTRACT

To control the microstructure and amounts of TaB<sub>2</sub> phase in the TaB<sub>2</sub>-SiC coating, a novel liquid phase sintering method was developed on the basis of in-situ reaction method to prepare the TaB<sub>2</sub>-SiC coating, which includes synthesis of TaB<sub>2</sub> powders and further preparation of TaB<sub>2</sub>-SiC coating. With Ta<sub>2</sub>O<sub>5</sub>, B<sub>2</sub>O<sub>3</sub> and C employed as raw materials, hexagonal TaB<sub>2</sub> powders were prepared by carbothermal reduction method at 1500 °C, whose mean particle size is 491 nm. The TaB<sub>2</sub>, SiC, C powders, and the low melting point phases Si and silica sol were used to prepare the TaB<sub>2</sub>-SiC coating by liquid phase sintering at 2373 K. The thickness of the coating is about 350 μm. Compared with the SiC coating, the weight loss of the samples modified by TaB<sub>2</sub> decreased from 17.7% to 11.8%, and the average weight loss rate of the fastest weightloss zone reduced from  $-6 \times 10^{-3} \text{ mg cm}^{-2} \text{ s}^{-1}$  to  $-5 \times 10^{-3} \text{ mg cm}^{-2} \text{ s}^{-1}$ . During oxidation, the Ta-oxides would gradually dissolve in the silicate glass to form Ta-Si-O glass ceramics with dendritic structure, which significantly improved the toughness and stability of the glass layer. The Ta-Si-O glass ceramics possesses the ability of sealing and arresting the microcracks, which can enhance the oxidation protective ability of the coating.

## 1. Introduction

VB group transition metal diborides TaB<sub>2</sub> [1–5] belongs to the family of materials termed as ultra-high temperature ceramics (UHTCs) [6,7], which not only has high melting temperatures (> 3000 °C) and high hardness but also has favorable thermal shock resistance, excellent oxidation resistance and outstanding thermal conductivity [8–10]. However, despite the outstanding properties of TaB<sub>2</sub>, it has not been investigated as extensively as other transition metals diborides in the applications of ceramics or coatings, like ZrB<sub>2</sub> [11–13] or HfB<sub>2</sub> [14–16].

Due to the low density, low coefficient of thermal expansion and superior mechanical properties, carbon materials (graphite and carbon/carbon composites) are highly promising as a kind of engineering structural materials in extremely harsh environments, which can be used as rocket nozzles components, brake disk of airplanes and leading edges on reentry vehicles [17–19]. Unfortunately, the carbon materials will be gravely oxidized in oxidizing atmosphere above 723 K without protection [20–22], which severely restricts their potential applications. Hence, in order to enhance the oxidation resistance of carbon materials, Si-based anti-oxidation coating techniques have attracted extensive attentions these years, which can protect the carbon materials from oxidation through the formation of self-sealing silicate glass layer [23–28]. However, although the pure Si-based anti-oxidation coatings

present effective protective ability, they are not fully qualified for the application in extremely harsh environments, such as ultra-high temperature or dynamic oxidation environments.

Recently, because TaB<sub>2</sub> owns the promising properties that are suitable for applications as thermal protection structures in harsh ultra-high temperature environments, it was added into Si-based ceramic coatings to improve its oxidation protection ability for the carbon materials [29,30]. In our previous study, we have prepared the TaB<sub>2</sub>-SiC [31] coating by in-situ reaction method, and owing to the formation of oxides of Ta in silicate glass layer, the coating could effectively protect the C/C composites from oxidation at 1773 K in air for 300 h with only  $0.26 \times 10^{-2} \text{ g/cm}^2$  mass loss, thus demonstrating superior oxidation resistance potential to protect the carbon materials. Nevertheless, although the microstructure and distribution of TaB<sub>2</sub> phase in coating are essential for the oxidation protective ability of the TaB<sub>2</sub>-SiC coating, it is hard to control the microstructure and amounts of TaB<sub>2</sub> phase in the TaB<sub>2</sub>-SiC coating because the TaB<sub>2</sub> phase in coating is in-situ synthesized by the in-situ synthesis method.

Hence, to fully utilize the excellent properties of TaB<sub>2</sub> and broaden its applications as coating component, the microstructure and amounts of TaB<sub>2</sub> phase in the TaB<sub>2</sub>-SiC coating need to be controlled. Despite the shortcomings, the in-situ reaction method presents promising coating preparation methodology. Hence, in this paper, on the basis of in-situ

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<https://doi.org/10.1016/j.ceramint.2018.03.105>

Received 13 February 2018; Received in revised form 3 March 2018; Accepted 12 March 2018

Available online 13 March 2018

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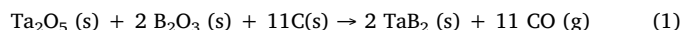
reaction method, a novel liquid phase sintering method which includes powder synthesis methodology and coating preparation technology was developed to prepare the TaB<sub>2</sub>-SiC coating. For the part of powders synthesis, the carbothermal reduction method, which has the advantages of short reaction time, low cost and simple facilities, was employed to synthesize the TaB<sub>2</sub> powders to control the microstructure of TaB<sub>2</sub> phase. For the part of coating preparation, TaB<sub>2</sub>, SiC, C, Si powders and silica sol, were used as raw materials to prepare the TaB<sub>2</sub>-SiC coating by liquid phase sintering method. The amounts of TaB<sub>2</sub> phase in coating can be controlled by adjusting the ratio of the raw materials. Furthermore, the existence of C, Si powders and silica sol in raw materials is to utilize the in-situ reaction mechanism, on the other side, the existence of low melting point phases Si powders and silica sol in raw materials is to create conditions for liquid phase sintering during heat-treatment to ensure the quality of the coating.

Therefore, in this paper, the TaB<sub>2</sub>-SiC coating was prepared by a novel liquid phase sintering method, which includes synthesis of TaB<sub>2</sub> powders and further preparation of TaB<sub>2</sub>-SiC coating. The microstructures of TaB<sub>2</sub> powders, and oxidation protective ability and mechanisms of the TaB<sub>2</sub>-SiC coating at dynamic TGA test from room temperature to 1773 K were investigated.

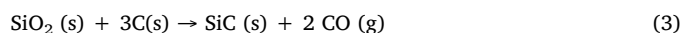
## 2. Experimental procedures

The preparation of the TaB<sub>2</sub> powders was shown in Fig. 1(a). The original powders were Ta<sub>2</sub>O<sub>5</sub> (purity > 99.9%, Guoyao Chemical Reagent Co., Ltd, Shanghai, China), B<sub>2</sub>O<sub>3</sub> (purity > 99.9%, Tianli Chemical Reagent Co., Ltd, Tianjin, China) and C (purity > 99.9%, Carbon Plant, Xi'an, China). Firstly, according to reaction (1), 0.006 mol Ta<sub>2</sub>O<sub>5</sub>, 0.015 mol B<sub>2</sub>O<sub>3</sub> and 0.066 mol C were weighted, which were employed as the precursor powders to synthesize the TaB<sub>2</sub> powders. Afterwards, the precursor powders were adequately mixed in a clean ball mill for 2 h. After ball-milling, the homogeneous powders mixture was loaded into graphite crucible and heated to 1773 K or 1973 K under flowing argon protective atmosphere for 2 h, respectively. The heating rate is

5 K/min. Finally, when the powders were cooled down to room temperature, the TaB<sub>2</sub> powders were obtained.



As an internal buffer layer, the inner SiC coating was processed as described previously [32]. Fig. 1(b) demonstrated the preparation of the outer 20 wt%TaB<sub>2</sub>-SiC coating. First of all, the prepared pure TaB<sub>2</sub> powders (20 wt%), Si powders (5–30 wt%) (Jiuling Smelting Co., Ltd, Shanghai, China), SiC powders (10–40 wt%) (China New Metal Materials Tec. Co., Beijing, China), graphite (5–20 wt%) (Carbon Plant, Xi'an, China) and silica sol (SiO<sub>2</sub>·nH<sub>2</sub>O) (the volume of silica sol to the weight of other raw materials was 0.5–1.5 ml/g) (City Fire Crystal Glass Co., Ltd, Dezhou, China) were used for the preparation of the original slurry. Then, the hybrid slurry was added onto the surface of the inner SiC coated graphite substrate by the method of brushing to obtain the pre-fabricated layer. Finally, the specimens were heated to 2373 K with a heating rate about 5–10 K/min and held for 2 h in normal argon protective atmosphere, where SiC can be made from in-situ reaction among Si, silica sol and C powders according to reaction (2) and (3). What is more, when the temperature is higher than 2000 K, the Si powders and silica sol, whose melting temperature are 1687 K and 1923 K, respectively, will become the liquid. Hence, during the heat-treatment process, the presence of Si powders and silica sol can create a floating condition for liquid phase sintering and largely enhance the densification of the coating.



Phase compositions of the TaB<sub>2</sub> powders and the TaB<sub>2</sub>-SiC coating were determined by X-ray diffractometry (XRD, Bruker D8 ADVANCE, BRUKER AXS, Germany). The Field Emission Scanning Electron Microscopy (FE-SEM, JSM-6700F, JEOL, Japan) and energy dispersive spectroscopy (EDS) were used to analyze the microstructure and elemental components of the samples. The particle size of the TaB<sub>2</sub>



Fig. 1. Illustration of the preparation of the TaB<sub>2</sub> powders (a) and the outer TaB<sub>2</sub>-SiC coating (b).

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