

Oxidation behavior of YAG-ZrB₂ ceramics prepared by spark plasma sintering

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ZrB₂ and YAG are widely applied because of some excellent properties, but ZrB₂ is easily oxidized in high-temperature air. To make ZrB₂ ceramics have better oxidation resistance, high-density YAG-ZrB₂ ceramics were prepared. The oxidation behavior of YAG-ZrB₂ ceramics is investigated at different temperatures. The results indicate that the enhanced weight change differs according to the temperature range, slightly in the temperature 900–1000°C, sharply from 1100–1300 and smoothly above 1300°C. The weight change rate of YAG-ZrB₂ ceramics is increased by prolonging the oxidation time, but the weight change rate of YAG-ZrB₂ ceramics is gradually reduced. The oxidation layer is porous as compared to the YAG-ZrB₂ ceramics layer.

Key words: ZrB₂, YAG, Ceramics, Oxidation, High temperature.

Introduction

Zirconium diboride (ZrB₂) has attracted substantial interest because of its extreme chemical and physical properties, such as, high melting point, superior hardness and low electrical resistance, ZrB₂ has several applications such as in Hall-Heroult cell cathodes for electrochemical processing of aluminum, evaporation boats, crucibles for handling molten metals, thermowell tubes for steel refining, thermocouple sleeves for high-temperature use, nozzles, plasma electrodes, or as a dispersoid in metal and ceramic-matrix composites for heaters and igniters [1–4]. However, ZrB₂ is easily oxidized in high-temperature air to impact on its high-temperature strength, which restricts its applied range [5–6]. Some excellent oxidation resistance materials are considered as an assistant phase of ZrB₂ materials to prepare ZrB₂ ceramics to improve the high-temperature performance of ZrB₂ materials, such as Al₂O₃-ZrB₂, SiC-ZrB₂, LaB₂-ZrB₂, and ZrO₂-ZrB₂ [7–9].

Yttrium aluminium garnet (YAG or Y₃Al₅O₁₂) adopts the cubic garnet structure, it is of great interest as a high-temperature engineering material, due to its high-temperature strength coupled with low creep rates [10–14], which indicates that YAG ought to be a suitable matrix and/or reinforcing material [15–16].

To make ZrB₂ ceramics obtain better oxidation resistance, high-density YAG-ZrB₂ ceramics were prepared using Al₂O₃-Y₂O₃/ZrB₂ shell-core composite particles by spark plasma sintering. In this paper, the oxidation behavior of YAG-ZrB₂ ceramics is investigated. Because the weight

of YAG-ZrB₂ ceramics is varied by the oxidation process, the degree of oxidation of YAG-ZrB₂ ceramics may be characterized through the weight change rate of YAG-ZrB₂ ceramics below a certain temperature, the weight change rate of YAG-ZrB₂ ceramics is discussed to investigate the oxidation behavior of YAG-ZrB₂ ceramics.

Materials and Experiment

Analytical grades of aluminum nitrate, yttrium nitrate, ammonia and commercially-available ZrB₂ powder (99.5% in purity) were used. ZrB₂ particles were coated with Al₂O₃-Y₂O₃ composite particles via a co-precipitation method, which has been reported before [17]. YAG-ZrB₂ ceramics were prepared using Al₂O₃-Y₂O₃/ZrB₂ shell-core composite particles by spark plasma sintering (SPS). YAG-ZrB₂ ceramics were oxidized by air in a furnace, the weight and microstructure of YAG-ZrB₂ ceramics were tested before and after oxidation. A process flow diagram is shown in Fig. 1.

YAG-ZrB₂ composite were prepared by SPS (Mode: SPS-1050, Japan) and oxidized in a furnace (Mode: Nabertherm LHT04, Germany). Phase analysis was performed by X-ray powder diffraction (XRD) (Model: D/Max-RB, Japan). Microstructural analysis was performed by scanning electron microscopy (SEM) (Model: JSM-5610LV, Japan). Elemental analysis was performed with electron probe microanalysis apparatus (EPMA) (Model: JXA-880R, Japan).

Results and Discussion

Effect of oxidation temperature on the oxidation of YAG-ZrB₂ composite

Fig. 2 shows the effect of oxidation temperature on the

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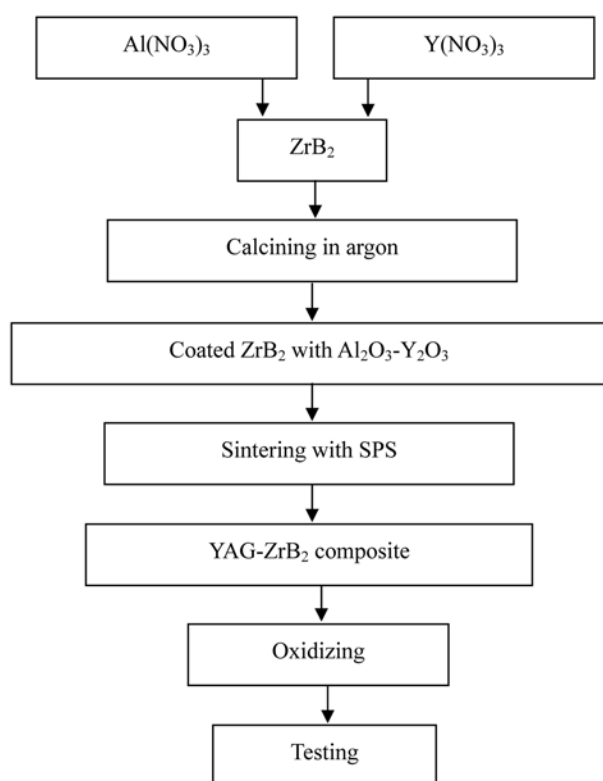


Fig. 1. The process flow diagram.

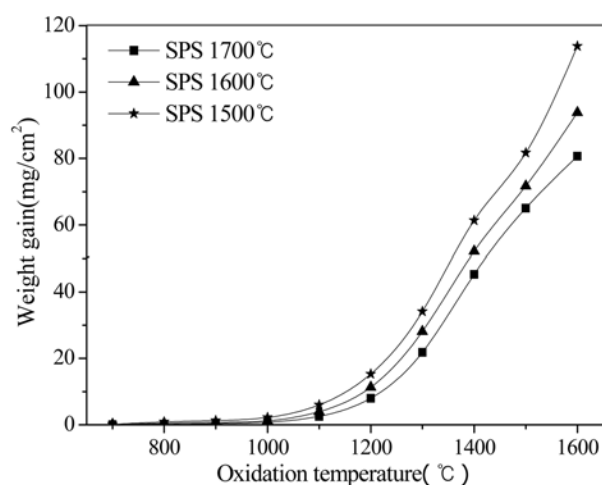


Fig. 2. Effect of oxidation temperature holding for 1 h on the weight change of YAG-ZrB₂ ceramics.

weight gain of fabricated YAG-ZrB₂ ceramics at different holding temperatures. The weight gain increased with an increase in the oxidation temperature, and the weight gain was decreased with an increase in the sintered temperature of the preparation of YAG-ZrB₂ ceramics. When YAG-ZrB₂ ceramics are exposed to air at temperatures lower than 900°C, the weight change rate approached zero, which indicates the ceramics are not oxidized. When YAG-ZrB₂ ceramics are exposed to air at temperatures from 900°C to 1100°C, the weight change rate of YAG-ZrB₂ ceramics is increased, which indicates ceramics are oxidized. When YAG-ZrB₂ ceramics are exposed to air at temperatures from

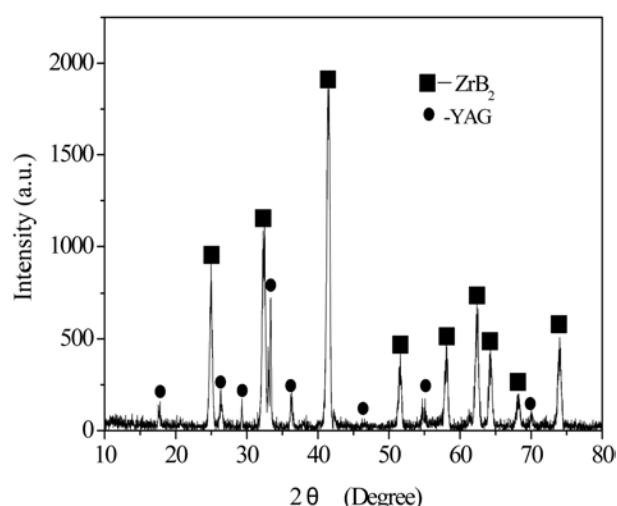


Fig. 3. XRD of ZrB₂-YAG ceramics.

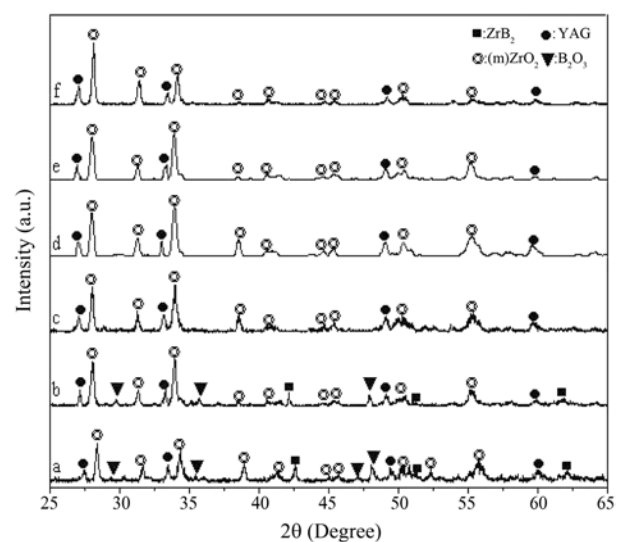


Fig. 4. Effect of oxidation temperature on the surface phases of ZrB₂-YAG ceramics (a-1100°C, b-1200°C, c-1300°C, d-1400°C, e-1500°C and f-1600°C).

1100°C to 1600°C, the weight change rate of YAG-ZrB₂ ceramics is greatly increased, which shows the oxidation rate of ceramics is quickened. Because the oxidation temperature higher, the reaction speed of ZrB₂ and O to form B₂O₃ and ZrO₂ is faster (Fig. 3 and Fig. 4) [18-20]. The density of YAG-ZrB₂ ceramics is increased with an increase in the sintering temperature of the preparation of YAG-ZrB₂ ceramics, the quantity oxygen diffusion through channels is reduced [21], which shows the oxidation resistance of YAG-ZrB₂ ceramics at high temperature is increased.

Fig. 5 shows surfaces of oxidized YAG-ZrB₂ ceramics with different oxidation conditions. The original surface is smooth. When the specimen is exposed to air at 1300°C for 1 h, small and symmetrical white spots are found on the surface, as shown in Fig. 5(b). As the oxidation temperature is increased to 1300°C, bigger white particles are

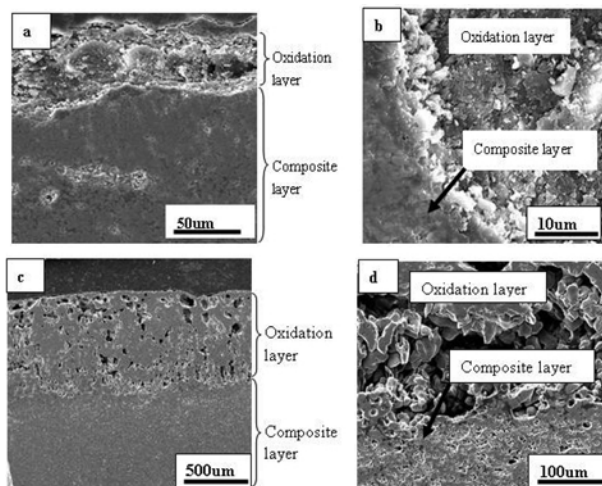


Fig. 5. SEM of YAG-ZrB₂ ceramics sintered at 1700°C after exposure to air at different temperatures for 1 h (a and b-1300°C, c and d-1600°C).

shown on the surface, because some B₂O₃ has melted and agglomerated. Increasing the oxidation temperature to 1500°C gains a quantity of white products with some pores which are seen in the oxidized surface. It is possible that a quantity of B₂O₃ has melted and agglomerated to form the white products layer, B₂O₃ is volatilized out of YAG-ZrB₂ ceramics through these pores.

Fig. 5 give the SEM results of YAG-ZrB₂ ceramics fabricated at 1700°C after exposure to air at different oxidation temperatures for 1 h, Fig. 5(b) and d are the high magnification from Fig. 5(a) and (c), respectively. The thickness of the oxidation layer is increased with an increase in the oxidation temperature by comparison of Fig. 5(a) and (c), because the diffusion rate of air in the YAG-ZrB₂ ceramics is increased, in addition, B₂O₃ is melted and volatilized, which offer more channels for diffusion of air [22-23]. The density of the oxidation layer is reduced with an increase in the oxidation temperature, as shown in Fig. 5(b) and Fig. 5(d), because B₂O₃ has melted and volatilized, which make more spaces in the oxidation layer.

Fig. 6 gives the EPMA results of oxidized YAG-ZrB₂ ceramics, the obvious oxidation layer and YAG-ZrB₂

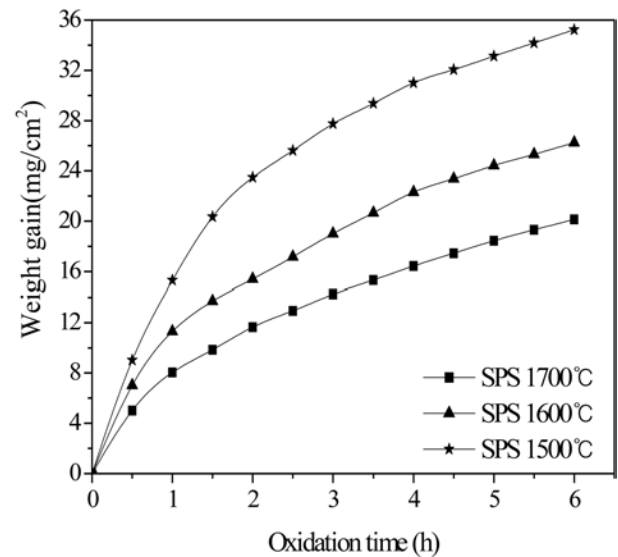


Fig. 7. Effect of oxidation time on weight gain of ceramics.

ceramics layer are shown. Some elements are analyzed through element flat scanning, the B analysis is obvious to know the oxidation process above 1300°C, which indicates the B content of the oxidation layer is less than that of YAG-ZrB₂ ceramics layer, because the B₂O₃ of the oxidation layer is volatilized out of YAG-ZrB₂ ceramics.

Effect of oxidation time on the oxidation of YAG-ZrB₂ composite

Fig. 7 is the relationship of the oxidation time and weight change rate of fabricated YAG-ZrB₂ ceramics formed at different sintering temperatures. The higher the sintering temperature of fabricated YAG-ZrB₂ ceramics is, the smaller the weight change rate of YAG-ZrB₂ ceramics, because the density of YAG-ZrB₂ ceramics is higher with a higher sintering temperature, which gives fewer channels for air diffusion in the YAG-ZrB₂ ceramics. The weight change rate of YAG-ZrB₂ ceramics is increased by prolonging the oxidation time, but the increasing speed of the weight change rate of YAG-ZrB₂ ceramics is gradually reduced, because the thickness of the oxidation layer is increased

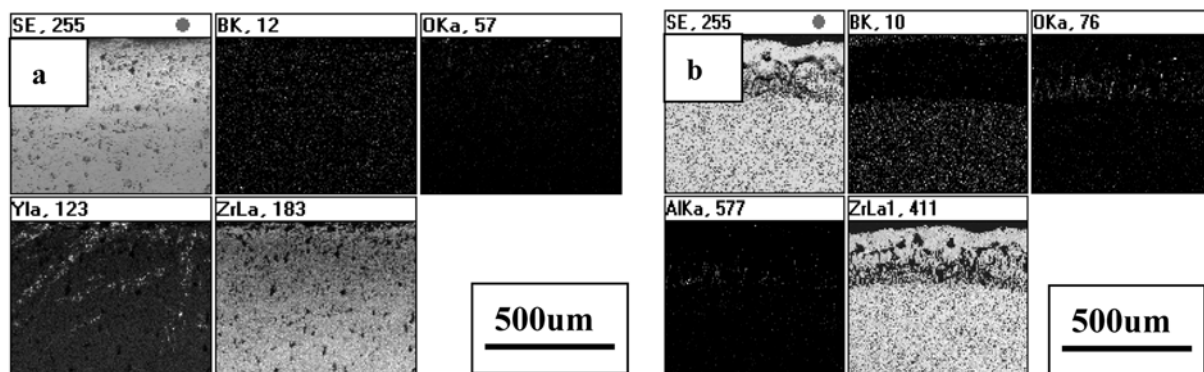


Fig. 6. EPMA of oxidized YAG-ZrB₂ ceramics (a-1300°C and b-1400°C).

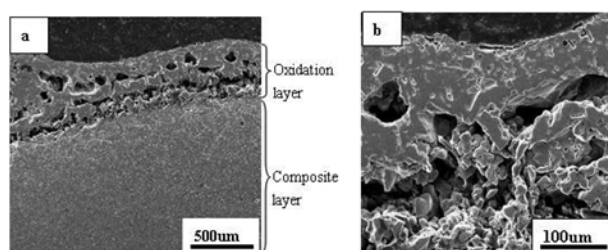


Fig. 8. SEM of sintered YAG-ZrB₂ ceramics at 1700°C after exposure to air at 1300°C for 6 h.

by prolonging the oxidation time, the length of air diffusion channels is increased [24], and oxidizing ZrB₂ particles is more difficult. The quantity of melted B₂O₃ is increased by prolonging the time, a continuous B₂O₃ layer is formed on the surface of YAG-ZrB₂ ceramics [25].

Fig. 8 gives the SEM results of oxidized YAG-ZrB₂ ceramics. Three layers, such as the inlay layer, oxidation layer and composite layer, can be found from top to bottom, as shown in Fig. 8(a). The oxidation layer is very porous as compared to the YAG-ZrB₂ ceramics layer (Fig. 8(a)). The top of the oxidation layer is more compact (Fig. 8(b)), because the quantity of melted B₂O₃ is increased and carried to the surface, which helps fill some spaces in enough time.

Conclusions

The weight gain is increased with an increase in the oxidation temperature, and the weight gain is decreased with an increase in the sintered temperature of prepared YAG-ZrB₂ ceramics. When YAG-ZrB₂ ceramics are exposed to air at temperatures from 1100°C to 1600°C, the weight change rate of YAG-ZrB₂ ceramics is much increased, which shows the oxidation rate is very rapid.

The density of YAG-ZrB₂ ceramics is increased with an increase in the sintering temperature of the prepared YAG-ZrB₂ ceramics, the quantity of channel for oxygen diffusion is reduced, which shows the oxidation resistance of YAG-ZrB₂ ceramics at high temperature is increased.

The weight change rate of YAG-ZrB₂ ceramics is increased by prolonging the oxidation time, but the speed of the weight change rate of YAG-ZrB₂ ceramics is gradually reduced.

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References

1. J.G. Song, L.M. Zhang, J.G. Li and J. R. Song, *J. Disper. Sci. Tech.* 28 (2007) 1072-1076.
2. I.B. Bankovskaya, M.P. Semov and A.E. Lapshin, *Glass Phys. Chem.* 31 (2005) 433-438.
3. J.G. Song, J.G. Li, J.R. Song and L.M. Zhang, *Mater. Manuf. Process.* 23 (2007) 475-478.
4. D. Sciti, S. Guicciardi and A. Bellosi, *J. Am. Ceram. Soc.* 89 (2006) 2320-2322.
5. J.G. Song, L.M. Zhang, J.G. Li and J.R. Song, *J. Reinf. Plast. Comp.* 26 (2007) 139-145.
6. A.L. Chamberlain, W.G. Fahrenholtz and G.E. Hilmas, *J. Am. Ceram. Soc.* 89 (2006) 450-456.
7. J.G. Song, L.M. Zhang, J.G. Li and J.R. Song, *Mater. Manuf. Process.* 23 (2008) 98-101.
8. A. Bellosi, F. Monteverde and D. Sciti, *Int. J. Appl. Ceram. Tech.* 3 (2006) 32-40.
9. J.G. Song, S.B. Li, Y.Y. Ju and D.M. Du, *Surf. Rev. Lett.* 16 (2009) 281-286.
10. A. Fernandez and J. Somers, *J. Mater. Sci.* 38 (2003) 2331-2335.
11. M.B. Kakade, S. Ramanathan and S.K. Roy, *J. Mater. Sci. Lett.* 21 (2002) 927-929.
12. F. Ivanauskas, A. Kareiva and B. Lapcun, *J. Math. Chem.* 37 (2005) 365-372.
13. X. Li, Q. Li, J. Y. Wang and S.L. Yang, *J. Alloy. Comp.* 421 (2006) 298-302.
14. M. Ono, Y. Waku and K. Habicht, *Appl. Phys. A.* 74 (2002) 73-75.
15. R.M. Laine, J. Marchal, H.P. Sun and X.Q. Pan, *Adv. Mater.* 17 (2005) 830-833.
16. S.J. Wang, Y.B. Xua, P.X. Lu, C.F. Xu and W. Cao, *Mater. Sci. Eng. B.* 127 (2006) 203-206.
17. J.G. Song, L.M. Zhang, J.G. Li and J.R. Song, *Surf. Rev. Lett.* 14 (2007) 117-122.
18. W.G. Fahrenholtz, *J. Am. Ceram. Soc.* 88 (2006) 3509-3512.
19. A. Rezaie, W.G. Fahrenholtz and G.E. Hilmas, *J. Am. Ceram. Soc.* 89 (2006) 3240-3245.
20. E.J. Opila and M.C. Halbig, *Ceram. Eng. Sci. Process.* 22 (2001) 221-228.
21. W.G. Fahrenholtz, *J. Am. Ceram. Soc.* 90 (2007) 143-148.
22. A. Rezaie, W.G. Fahrenholtz and G.E. Hilmas, *J. Am. Ceram. Soc.* 27 (2007) 2495-2501.
23. R.L. Gao, G.H. Min, H. S. Yu, and S. Q. Zheng, *Ceram. Int.* 31 (2005) 15-19.
24. D. Sciti, M. Brach and A. Bellosi, *Scripta Mater.* 53 (2005) 1297-1302.
25. C.M. Chen, L.T. Zhang, W.C. Zhou and Z.Z. Hao, *Comp. Sci. Tech.* 61 (2001) 971-975.