The effect of the oxidation conditions on the surface microdomains of ZrB_2-YAG ultra-high temperature ceramics

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Zirconium diboride is widely applied because of some excellent properties. The effect of the oxidation temperature on the surface microdomain of ZrB_2-YAG multi-phase ceramics was studied, it helps to improve the performance of ultra-high-temperature ceramics. The results show the oxidation layer thickness is increased with an increase in the oxidation temperature, the oxidation layer thickness is decreased by increasing the density of multi-phase ceramics at the same oxidation temperature. The ceramic surface shows the (m)ZrO_2, YAG, B_2O_3 and ZrB_2 phases below the the oxidation temperature at 1300 °C, but the ceramic surface do not show the ZrB_2 phase above the oxidation temperature at 1300 °C that is to say, the ZrB_2 phase of the ceramic suface is entirely oxidized. The oxidation layer thickness is increased with an increase in the oxidation time, however, the ratio of increasing thickness is less and less. The oxidation layer shows a loose structure during the initial stage of the oxidation, which lacks a barriers for the diffusion of the oxygen.

Key words: Ultra-high temperature ceramics, Zirconium diboride, Oxidation factor, Surface microdomain, Multi-phase ceramics.

Introduction

Zirconium diboride (ZrB_2) has attracted substantial interest because of its extreme chemical and physical properties, such as, a high melting point, superior hardness and low electrical resistance. ZrB_2 has several applications such as for Hall-Heroult cell cathodes for electrochemical processing of aluminum, evaporation boats, crucibles for handling molten metals, thermo well 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]. Yttrium aluminium garnet (YAG or $Y_3Al_5O_{12}$) adopts the cubic garnet structure and it is of great interest as a high-temperature engineering material, due to its high-temperature strength coupled with low creep rates, which indicates that YAG ought to be a suitable matrix and reinforcing material [5-6].

To make ZrB_2 ceramics have a better oxidation resistance, high-density ZrB_2-YAG ceramics were prepared using Al_2O_3-Y_2O_3/ZrB_2 composite powders with spark plasma sintering. In this paper, the effect of the oxidation conditions on the surface microdomains of ZrB_2-YAG ceramics is discussed.

Experimental

Analytical-grade aluminum nitrate, yttrium nitrate,

ammonia, and commercially available ZrB_2 powder (99.5% purity, $D_{50} = 12$ μm) were used. ZrB_2 particles were coated with Al_2O_3-Y_2O_3 composite particles via the coprecipitation method [7]. ZrB_2-YAG ceramics were prepared using Al_2O_3-Y_2O_3/ZrB_2 shell-core composite particles with spark plasma sintering (SPS) under different sintering temperatures, which a holding time of 4 minutes, and sintering pressure of 20 MPa. ZrB_2-YAG ceramics were oxidized by air in a furnace, and the weight and microstructure of ZrB_2-YAG ceramics were tested after oxidation. A process flow diagram is shown in Figure 1.

ZrB_2-YAG ceramics were prepared using SPS (SPS-1050, Japan). Oxidation treated in a furnace (Nabertherm LHT04, Germany). Phase analysis was made by X-ray powder diffraction (XRD, D/Max-RB, Japan). Microstructural analysis was performed by scanning electron
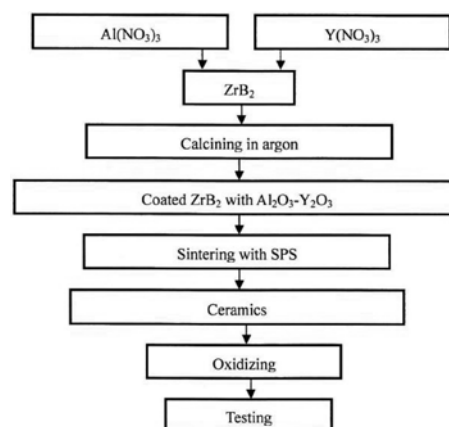

Fig. 1. The process flow diagram.

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microscopy (SEM, JSM-5610LV, Japan). Element analysis was performed with an electron probe microscope apparatus (EPMA, JXA-880R, Japan).

Results and Discussion

The effect of oxidation temperature on the surface microdomains of ZrB₂-YAG multi-phase ceramics

The effect of the oxidation temperature on the oxidation layer thickness of ceramics sintered at different temperatures via SPS is shown in Fig. 2. The oxidation layer microstuctures of ZrB_2-20 wt%YAG sintered at 1700 °C via SPS are shown in Fig. 3. The oxidation layer thicknesses are increased with an increased oxidation temperature and are decreased with an increased sintering temperature via the SPS. Because the oxidation temperature is higher, the reaction is activated between O_2 and ZrB_2, the diffusion coefficient of O_2 is higher in the ceramics [8-9], which make the oxidation layer thickness become thicker. When the sintering temperature is higher, the density of the ceramics is higher, the entry diffusion for O_2 becomes lesser [10], which make the amount of the reaction between O_2 and ZrB_2 become lesser. This shows that if the density of the ceramics is increased it may improve the oxidation resistance of ultra-high temperature ceramics.
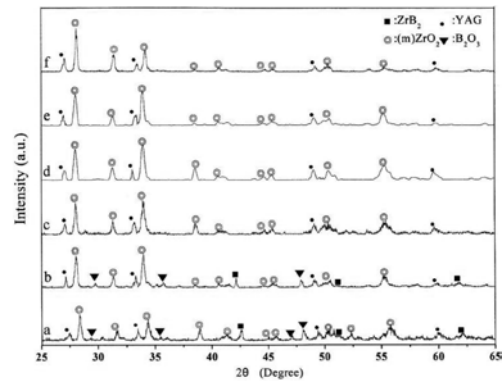
The surface XRD of oxidized ZrB_2-20 wt%YAG
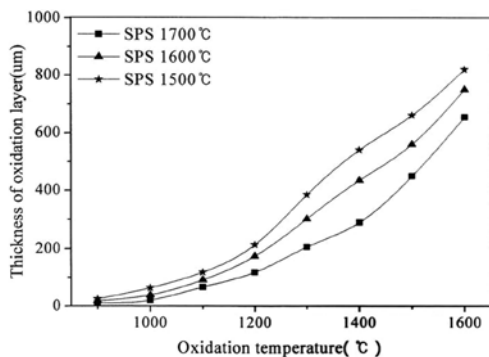
Fig. 2. The effect of the oxidation temperature on the thickness of the oxidation layer of ceramics.
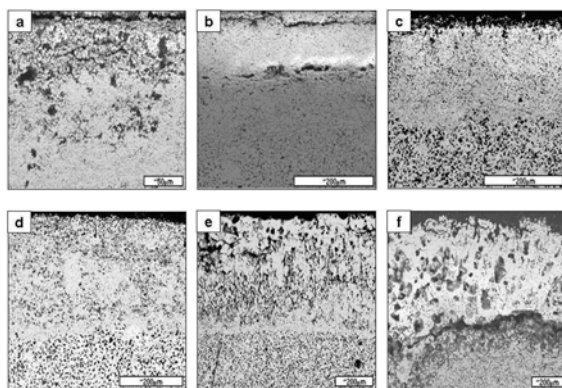
Fig. 3. The effect of the oxidation temperature on the oxidation layer of ceramics; (a)1100 °C, (b) 1200 °C, (c) 1300 °C, (d) 1400 °C, (e) 1500 °C, (f) 1600 °C.


Fig. 4. The effect of the oxidation temperature on the phases of ceramic surfaces; (a)1100 °C, (b) 1200 °C, (c) 1300 °C, (d) 1400 °C, (e) 1500 °C, (f) 1600 °C.
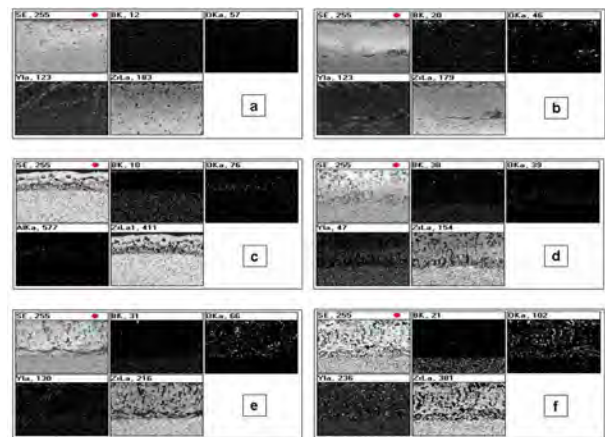

Fig. 5. EPMA of oxidized ceramics under different treating temperatures; (a)1100 °C, (b) 1200 °C, (c) 1300 °C, (d) 1400 °C, (e) 1500 °C, (f) 1600 °C.

ceramics sintered at 1700 °C via the SPS at different oxidation temperatures are shown in Fig. 4, which indicates the surface phases are varied with different oxidation temperatures. The surface phase contains the $(m)ZrO_2$, YAG, B_2O_3 and ZrB_2 when oxidizing below 1300 °C, the surface phases do not contain the ZrB_2 when oxidizing above 1300 °C, which shows the ZrB_2 of the ceramic surfaces are already oxidized entirely. The surface phases do not contain the $(m)ZrO_2$ and YAG when oxidizing above 1200 °C, which shows the ZrB_2 of the ceramic surfaces are already oxidized to form $(m)ZrO_2$ and B_2O_3, B_2O_3 is easily molted and volatilized. The B_2O_3 is molten and coats the surfaces of the ceramics at lower oxidation temperatures, the B_2O_3 is volatilized and removed from the surfaces of the ceramics at higher oxidation temperatures [10, 11], The boron electmental distribution from EPMA of oxidized ceramics are shown in Figure 5. The B element is homogeneously distributed in the ceramics oxidized at 1100 °C and 1200 °C, however, the B element has a non-uniform distribution in the ceramics oxidized above 1300 °C. The B element is homogeneously distributed in the unoxidized ceramic layer, it appears write. The B element analysis of the oxidation layer shows black, that
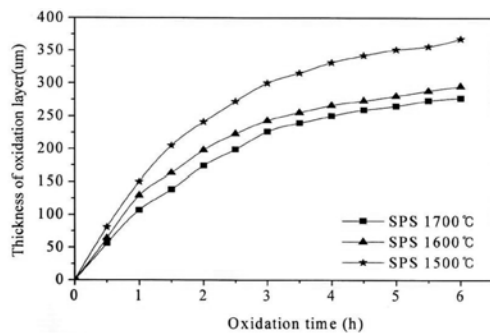
Fig. 6. The effect of the oxidation time on the thickness of the oxidation layer of ceramics.
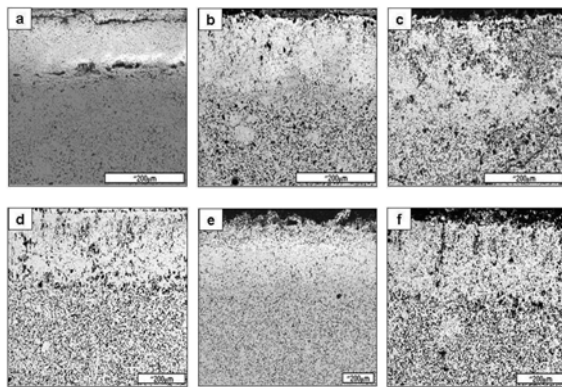
Fig. 7. The effect of the oxidation time on the oxidation layer of ceramics; (a)1100 $^{\circ}$C, (b) 1200 $^{\circ}$C, (c) 1300 $^{\circ}$C, (d) 1400 $^{\circ}$C, (e) 1500 $^{\circ}$C, (f) 1600 $^{\circ}$C.

is to say the B element exist. The effect of oxidation time on the surface microdomain of ZrB2-YAG multi-phase ceramics.

The effect of the oxidation time on the oxidation layer thickness of ZrB₂-20 wt%YAG ceramics

sintered at different temperatures via SPS and oxidized at 1200 $^{\circ}$C is shown in Fig. 6. The oxidation layer thickness is increased with an increase in the oxidation time, however, the amount of increase in the thickness is less and less. Because the oxidation layer thickness is thinner, the ZrB₂ is oxidized to form some pores and the molten B₂O₃ cannot fully fill the pores, it shows the oxidized layer is very loose, which promotes the diffusion behavior of O₂. The oxidation layer thickness is increased with an increase in the oxidation time, the molten B₂O₃ can fully fill the pores to decrease the diffusion of O₂. In addition to an increased oxidation layer thickness, the diffusion distance of O₂ is increased in the ceramics [11, 12], which slows down the oxidation behavior of ZrB₂, The oxidation layer microstucture of ZrB₂-20 wt%YAG sintered at 1700 $^{\circ}$C via SPS and oxidized at 1200 $^{\circ}$C is shown in Fig. 7.

Conclusions

The oxidation layer thickness is increased with an increase in the oxidation temperature and decreased with an increase in the density of multi-phase ceramics at the same oxidation temperature. The ceramic surfaces shows the (m)ZrO₂, YAG, B₂O₃ and ZrB₂ phases at oxidation temperatures under 1300 $^{\circ}$C, but the ceramic surfaces do not show the ZrB₂ phase above the oxidation temperature at 1300 $^{\circ}$C, that is to say, the ZrB₂ phase of the ceramic sufaces is entirely oxidized. The oxidation layer thickness is increased with an increase in the oxidation time, however, the amount of increase in the thickness is less and less. The oxidation layer shows a loose structure during the initial stage of the oxidation, which lacks a barrier to the diffusion of the oxygen.

Acknowledgments

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