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Preparation of High-density YAG/ZrB_2 Multi-phase Ceramics by Spark Plasma Sintering

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ZrB_2 and YAG are widely applied because of some excellent properties, but ZrB_2 is difficult to prepare as a high-density ceramic, and it is easily oxidized in high temperature air, which limits the application of ZrB_2 materials. To obtain high-density for better oxidation resistance ZrB_2 ceramic materials, in this paper, Al_2O_3 and Y_2O_3 are added into ZrB_2 raw material to prepare high-density YAG/ZrB_2 multi-phase ceramics by spark plasma sintering. Through the above analysis, it is proposed that 1600°C is regarded as a critical sintering temperature (Tc) to fabricate YAG/ZrB_2 multi-phase ceramics. Only if the sintering temperature is higher than Tc, can a nearly full density composite be achieved. The microstructure of sintered YAG/ZrB_2 multi-phase ceramics after 4 minutes at 1600°C is a better compact, the density and relative density are 5.5865 g/cm^3 and 99.40% respectively, which indicates that high-density YAG/ZrB_2 multi-phase ceramics have been fabricated successfully.

Key words: ZrB_2, YAG, Multi-phase ceramics, Densification behavior, Relative density, Spark plasma sintering.

Introduction

Due to some excellent properties, ZrB_2 is widely applied to many industries [1-5]. Although ZrB_2 has there excellent properties, it is difficult to prepare a high-density ZrB_2 ceramic, and it is easily oxidized in high temperature air, which limits the application of ZrB_2 materials [6-8]. Yttrium aluminium garnet (YAG or $Al_5Y_3O_{12}$) has attracted substantial interest, because of its extreme chemical and physical properties, especially high-temperature strength and stability, it is widely applied in some industries [9-11]. To overcame some problems with single phase ceramics, materials research has moved from single phase to multi-phase materials to prepare higher performance materials [12].

At the present time, there are a number of sintering methods for the preparation of ZrB_2 ceramics, such as, common sintering, high pressure sintering (HPS), self-propagating high-temperature synthesis (SHS), high isopressing sintering (HIP) and spark plasma sintering (SPS) [13-16]. The bulk is sintered using the stronger impulsive current through the electric mould or sintered materials in the SPS system. Accordingly SPS shows some merits, such as, fast sintering, short cycle time, high temperature, etc. [17, 18], which is used to prepare YAG/ZrB_2 multi-phase ceramics.

To obtain a high-density for better oxidation resistance ZrB_2 ceramics, in this paper, Al_2O_3 and Y_2O_3 are added into ZrB_2 raw material to prepare high-density YAG/ZrB_2 multi-phase ceramics by SPS.

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Materials and Experiment

Commercially available ZrB_2 powder (99.5% in purity) was used. Synthesized superfine Al_2O_3-Y_2O_3 composite powder with aluminum nitrate, yttrium nitrate and ammonia via the co-precipitation method was used. The Al_2O_3-Y_2O_3/ZrB_2 composite powder was prepared with the Al_2O_3-Y_2O_3 composite powder and ZrB_2 powder via a mechanical ball milling method. Then the Al_2O_3-Y_2O_3/ZrB_2 composite powder was loaded into in a graphite mould with an inside diameter of 20 mm. The temperature was automatically raised to 600°C, then monitored and regulated by an optical pyrometer aimed at the mould surface. A heating rate of 100 K·minute^{-1} between 600°C and various chosen temperatures was performed, the graphite mould was removed, and YAG/ZrB_2 multi-phase ceramics were obtained. A process flow diagram is shown in Fig. 1.

Materials were sintered using the SPS method (Mode: SPS-1050, Japan), and the density was established with a precise electronic balance via the Archimedes' principle (Model: Sartorius BS210S, Germany) and compared to the YAG/ZrB_2 theoretical density. Microstructural analysis was performed by scanning electron microscopy (SEM) (Model: JSM-5610LV, Japan).

Results and Discussion

Densification behavior during the SPS process

Figure 2 shows the shrinkage curves of samples

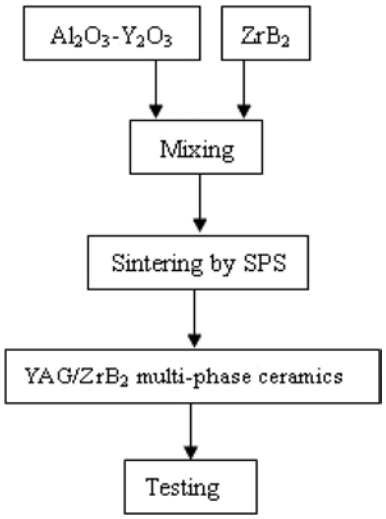

Fig. 1. A process flow diagram for preparing YAG/ZrB_2 multi-phase ceramic materials.

sintered at 1500, 1700, 1800°C. Owing to uniaxial pressure and high temperature sintering, the samples shrunk rapidly to accelerate densification. With further heating and a holding state, the shrinkage behavior retards and finally stops finally. From Fig. 2, it is discernible that the rate of shrinkage at different sintering temperatures is obviously different. The sample sintered at 1500°C continuously shrunk during heating and the holding state, and its shrinkage rate was more sluggish than those sintered at 1700 and 1800°C. It can be also seen from Fig. 2(b and c) that, when temperature is raised to 1600°C (dashed line), the samples stop shrinking, which indicates the samples are achieving nearly full density. The result of measured density supports the above explanation. The composites sintered at 1700 and 1800°C possess 99.36 and 99.40% of theoretical density, respectively, but the composites sintered at 1500°C only achieved 96.37% of theoretical density, as listed in Table 1.

During the SPS process, the action of the locally high temperature created by the plasma as well as the Joule heating from the pulse current results in producing YAG (Fig. 3) and melting YAG to promote shrinkage [19, 20]. When the sintering temperature was raised to 1600°C, the amount of molten YAG increased and

Table 1. Effect of temperature and holding time sintering parameters under 20 MPa pressure on the density of YAG/ZrB_2 multi-phase ceramics

Samples	T (°C)	Holding time (minute)	Experimental density (g/cm^3)	Relative density (%) (YAG/ZrB_2 theor.=5.62 g/cm^3)
1	1500	2	5.42	96.37
2	1600	2	5.57	99.16
3	1700	2	5.58	99.36
4	1800	2	5.59	99.40
5	1600	4	5.59	99.40
6	1600	6	5.59	99.40

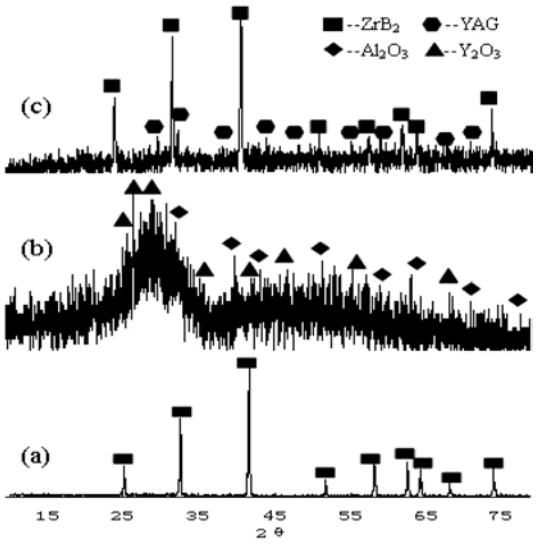

Fig. 3. XRD of materials (a) ZrB_2 raw materials, (b) Synthesized Al_2O_3-Y_2O_3 composite powder, (c) YAG/ZrB_2 multi-phase ceramic.

liquid phase sintering took place to lead the sample finally to stop shrinking and realize the sample densification. Thus, in the present study, it is proposed that 1600°C is regarded as a critical sintering temperature (Tc) to fabricate YAG/ZrB_2 multi-phase ceramics. Only if sintering temperature is higher than Tc, can a nearly full density composite be achieved.

Microstructure of YAG/ZrB_2 multi-phase ceramics

Microstructures sintered at different temperatures are shown in Fig. 4. It is clear that the amount of pores in

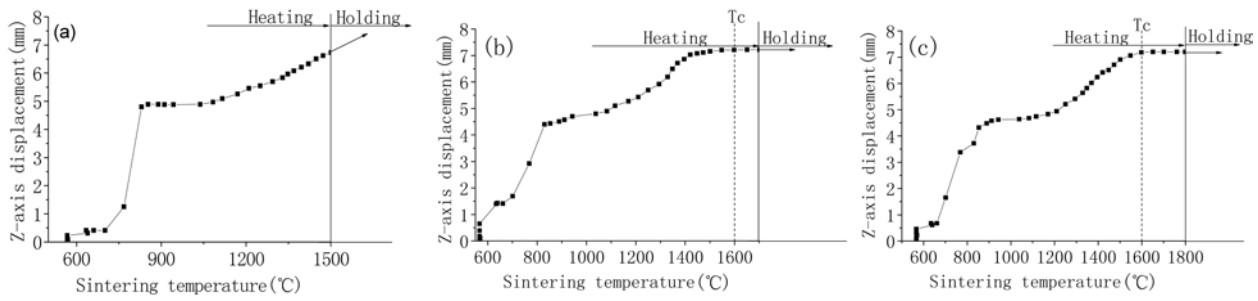

Fig. 2. Effect of sintering temperature on shrinkage characteristic of YAG/ZrB_2 multi-phase ceramics (a) 1500°C, (b) 1700°C, (c) 1800°C.

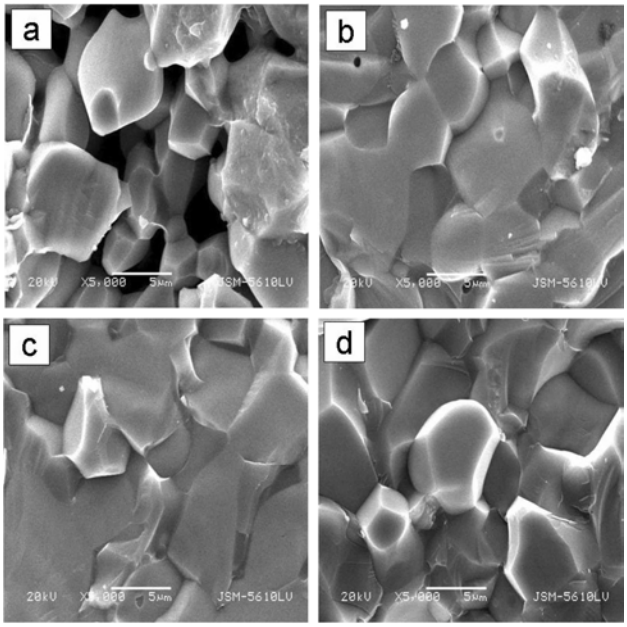

Fig. 4. Microstructure of YAG/ZrB_2 multi-phase ceramics (a) 1500°C, (b) 1700°C, (c) 1800°C, (d) 1600°C and 4 minutes.

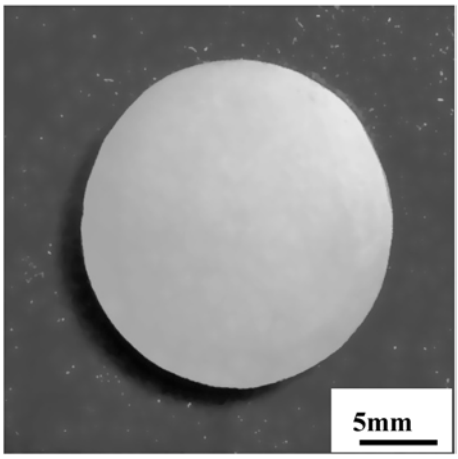

Fig. 5. Sample of YAG/ZrB_2 multi-phase ceramics.

the samples sintered at 1500°C is more than for those sintered at 1700°C and 1800°C, which is corresponding with the results of the measured density. With increasing sintering temperature, YAG molts to fill in the pores, which leads to the pores disappearing. With prolonging holding time, the pores are filled sufficiently by the molted YAG, which leads to increase the density, as listed in Table 1. High-density YAG/ZrB_2 multi-phase ceramics were successfully fabricated, the sample is shown in Fig. 5.

Conclusions

Through the above analysis, it is proposed that 1600°C is regarded as a critical sintering temperature (Tc) to fabricate YAG/ZrB_2 multi-phase ceramics. Only if the sintering temperature is higher than Tc, can a nearly full density composite be achieved. The microstructure of a YAG/ZrB_2 multi-phase ceramic sintered at 1600°C for 4 minutes is a better compact, the density and relative density is 5.59 g/cm^3 and 99.4% respectively, which indicates that high-density YAG/ZrB_2 multi-phase ceramics were successfully fabricated.

Acknowledgments

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