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Significance of hot pressing parameters and reinforcement size on sinterability and mechanical properties of ZrB_2–25 vol% SiC UHTCs

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Abstract

The influences of hot pressing parameters and SiC particle size on the bulk density, the average ZrB_2 grain size and Vickers hardness of ZrB_2–25 vol% SiC ultrahigh temperature ceramic composites were investigated. In this paper, the Taguchi methodology (An L9 orthogonal array) was used to specify the contributions of four parameters: the hot pressing temperature, holding time, applied pressure and SiC particle size. The experimental procedure included nine tests for four parameters with three levels which were employed to optimize the process parameters. The statistical analyses recognized the hot pressing pressure and temperature as the most consequential parameters affecting the density and hardness of ZrB_2–SiC composites. The SiC particle size and holding time were specified as the most effective parameters on the average ZrB_2 grain size. The bulk density, average ZrB_2 grain size, Vickers hardness and fracture toughness of the sample, hot pressed at optimal conditions (1850 °C, 90 min, 16 MPa and 200 nm), reached about 5.36 g/cm^3, 10.03 μm, ~17.1 GPa and 5.9 MPa m$^{1/2}$, respectively. The confirmation test, carried out under optimum conditions, showed that the experimental results were relatively equal to the predicted values from the Taguchi prediction model. Finally, the mechanisms of enhanced fracture toughness of the hot pressed ZrB_2–SiC ceramic composites were discussed.

Keywords: A. Hot pressing; A. Grain growth; C. Mechanical properties; ZrB_2–SiC composites; Taguchi method

1. Introduction

Zirconium diboride (ZrB_2) is a member of the family of materials known as ultrahigh temperature ceramics (UHTCs) [1]. Recently, remarkable studies have been accomplished in the field of ultrahigh temperature ceramics such as borides, and chiefly on ZrB_2-based ceramic composites [2,3]. ZrB_2-based UHTCs with particular compositions because of their unique combination of high melting point (3245 °C [1]), relatively low theoretical density (6.12 g/cm^3 [1]), high strength and hardness, high thermal and electrical conductivity, thermal shock resistance and chemical stability, have been significant for the high-temperature structural applications in recent years [1,4]. Moreover, ZrB_2 can be used for ultrahigh-temperature applications in aerospace [5,6]. Hence, preparing the fully-dense ZrB_2-based composites with improved mechanical properties (e.g. fracture toughness) is necessary. Recent studies [7–9] have shown that the presence of oxide impurities (ZrO_2 and B_2O_3) on the surface of ZrB_2 particles results in grain coarsening, which impedes the porosities to be completely removed. Therefore, enhanced densification of ZrB_2 is expected when grain coarsening is inhibited by removing the surface oxide impurities below the sintering temperature. Poor mechanical properties of ZrB_2, however, hinders the material from being beneficial in widespread range of applications. Its tendency to brittle fracture can reflect to sudden tragic failure; hence, its mechanical properties must be enhanced before considering any potential applications.

The addition of secondary phase has been recognized as an appropriate method for enhancing the mechanical properties of monolithic diboride ceramics. For purpose, the addition of SiC

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particles into ZrB_2 results in a ZrB_2–SiC ceramic composite which is stronger than monolithic ZrB_2. The mechanical properties of ceramics can be considerably enhanced by the addition of nano-sized particles into the matrix [10,11].

A ZrB_2 containing 10 vol% fine α-SiC ceramic composite, with mean particle size of ~3 μm and 0.8 μm, was fully densified by hot pressing at 1900 °C for 20 min under 40–50 MPa. The fracture toughness of this ceramic composite was 4.8 MPa $m^{1/2}$. It was suggested that, the addition of fine α-SiC particles was a key factor in enabling both the control of the ZrB_2 grain growth and the attainment of full density [11]. In another study, the ZrB_2–SiC ceramic composites, with SiC content of 5, 10, 15 and 20 vol%, were densified to near full density by hot pressing at 1900 °C under a uniaxed pressure of 45 MPa with mean particle size of 4–6 μm for ZrB_2 and 0.8 μm for SiC powders. The hardness and fracture toughness values of ZrB_2–15 vol% SiC were 21.7 GPa and 5.42 MPa $m^{1/2}$, respectively. It was expressed that the increase in SiC content, slightly increased the microhardness and improved the fracture toughness [12]. In another paper, three different ZrB_2–SiC ceramic composites, using various SiC sources, were produced by hot pressing at 1900 °C for 2 h under a 30 MPa, in order to indicate the effect of the SiC size on the microstructures and physical properties. It was clarified that the ZrB_2 or SiC grain size after hot pressing, highly depended on the initial particle size of SiC. The enhanced mechanical properties, using the fine SiC, are compatible with the Hall–Petch relation [13]. The ZrB_2–SiC composites with different amounts of SiC (0, 10, 20 and 30 vol %), whose mean particle size of ZrB_2 and SiC were ~1 μm and ~30 nm, respectively, were densified by hot pressing at 1900 °C for 60 min under a 30 MPa uniaxed pressure. It was found that the formation of the intragranular microstructure had improved the mechanical properties as the fracture toughness reached 6.8 MPa $m^{1/2}$ [14]. A ZrB_2 (2 μm) ceramic composite containing 20 vol% nano-sized SiC (~30 nm) was prepared by hot pressing at 1900 °C for 60 min under a 30 MPa uniaxed pressure. It was suggested that the formation of an unusual intragranular nanostructures was the main reason for the improvement in mechanical properties as the fracture toughness was 6.5 MPa $m^{1/2}$. It was illustrated that the intragranular nanostructures motivated the transgranular fracture mode [15]. In another paper, ZrB_2–SiC ceramic composites, which were fabricated by ZrB_2 powder (2 μm) containing 20 vol% nano-sized β-SiC particles (30 nm), were densified at 1900 °C for 30 min under a pressure of 30 MPa by hot pressing. It was indicated that the grain growth of ZrB_2 matrix was impressively graveled by adding submicron SiC particles, and the mechanical properties were strongly improved by incorporating the nano-sized SiC particles as a fracture toughness of 6.4 MPa $m^{1/2}$ was obtained [16].

In this paper, the influences of four parameters including the hot pressing temperature; the applied pressure; the holding time and the SiC particle size on the bulk density, average ZrB_2 grain size and Vickers hardness of hot pressed ZrB_2–SiC ceramic composites were studied using the Taguchi methodology.

2. Experimental procedure

2.1. Design of experiment

In this study, the Taguchi method was used to design the experiments. This method as a mathematical statistics determines the effect of different process parameters and specifies the relationship between input and output precisely [17]. The statistical design of the experiment is a beneficial technique in comparison with the classic methods, like the Factorial plan, to avoid applying a large number of experiments, save time and find the optimal conditions [18].

A property can be analyzed in three categories; "lower is better", "higher is better" or "nominal is better" conditions [19]. In this study, a high bulk density, high hardness and fine ZrB_2 grain size are desirable; hence the statistical analyses were performed with the "higher is better" choice for the bulk density and Vickers hardness, but "lower is better" option for the ZrB_2 grain size. The effects of each parameter on the bulk density, mean ZrB_2 grain size and hardness were indicated by the signal-to-noise (S/N) ratios [18]. The statistical analysis of variance (ANOVA) was performed to determine the significance and contribution of each process parameter over the output characteristics (bulk density, ZrB_2 grain size and hardness). Finally, the optimal experimental condition was predicted using S/N ratios and ANOVA analysis. In this part, all of the statistically analyses were performed by Qualitek-4 software (Automatic design and analysis of Taguchi experiments, Nutek Inc., USA).

In this research, the effects of four parameters with three levels for each one consisting of the hot pressing temperature (1700, 1775 and 1850 °C), holding time (30, 60 and 90 min), applied pressure (8, 12 and 16 MPa) and SiC particle size (5000, 200 and 20 nm) were studied. Unlike the factorial plan, in which the number of required tests is 81, the Taguchi offers only 9 experimental runs.

2.2. Materials and process

Commercially available ZrB_2 powder with size of ~2 μm (purity > 99%), supplied by Leung Hi-tech Co., China, α-SiC powder with size of ~5 μm (purity > 99%), supplied by Carborundum Universal Limited, India and two different β-SiC powders with mean size of ~200 nm and 20 nm (purity > 98%), supplied by PlasmaChem GmbH, Germany, were used as the raw starting materials. At first, the nano-sized SiC powders were dispersed in ethanol for 1 h using an ultrasonic stirrer (Mercury UC 4, Turkey). Mixtures of ZrB_2–25 vol% SiC then were prepared with different SiC particle size, and ball-milled using a polyethylene cup with zirconia balls at 90 rpm for 1 h. Afterwards, the mixed suspensions were dried by a rotating evaporator (80 °C) and then sieved. The prepared powder mixtures were loaded into a graphite die, with the surfaces of the die covered with boron nitride as a high separator lubricant and lined with a thin graphite foil, and then were hot pressed in a graphite resistance furnace (WEITAI High Temperature, China). Nine runs were