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Research paper

Fabrication and mechanical properties of $Al_2O_3/SiC/ZrO_2$ functionally graded material by electrophoretic deposition

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ABSTRACT

This study describes the synthesis of $Al_2O_3/SiC/ZrO_2$ functionally graded material (FGM) in bio-implants (artificial joints) by electrophoretic deposition (EPD). A suitable suspension that was based on 2-butanone was applied for the EPD of $Al_2O_3/SiC/ZrO_2$, and a pressureless sintering process was applied as a presintering. Hot isostatic pressing (HIP) was used to densify the deposit, with beneficial mechanical properties after 2 h at 1800 °C in Ar atmosphere. The maximum hardness in the outer layer (90 vol.% Al_2O_3 + 10 vol.% SiC) and maximum fracture toughness in the core layer (75 vol.% Al_2O_3 + 10 vol.% SiC + 15 vol.% ZrO_2) composite were 20.8 ± 0.3 GPa and 8 ± 0.1 MPa $m^{1/2}$, respectively.

The results, when compared with results from Al_2O_3/ZrO_2 FGM, showed that SiC increased the compressive stresses in the outer layers, while the inner layers were under a residual tensile stress.

1. Introduction

At present, functionally graded material (FGM) is often utilized as a means of remedying reliability and durability problems that arise when dissimilar materials that have different mechanical properties, such as variations in hardness, toughness, thermal and residual stresses, and strengths of the interfaces, are utilized within one application. FGMs are referred to as a class of advanced composites (Hvizdoš et al., 2007; Suresh and Mortensen, 1997) that operate by varying the microstructure of materials that have different mechanical properties in the layers, for instance, an object with a tough core and a hard surface (Mehrali et al., 2011; Novak et al., 2007; Put et al., 2003). A large range of processes for the production of FGMs are available, such as centrifugal casting, plasma spraying, spark plasma sintering, common powder metallurgy, physical vapor deposition (PVD), chemical vapor deposition (CVD), and colloidal processing (Kieback et al., 2003; Koizumi, 1997; Mortensen and Suresh, 1995). Among these techniques, electrophoretic deposition (EPD) is popular, as it constitutes a low-cost process that is capable of processing CGMs that have a complex geometry (Van der Biest et al., 2006; Zhitomirsky and Petric, 2001). As such, the production of FGMs using EPD processes is common, due to the high versatility in the use of the different materials and combinations that are produced (Besra and Liu, 2007; Boccaccini and Zhitomirsky, 2002). Electrophoresis is a technique that can be used for the purpose of forming functionally gradient and multi-layer composites using simple equipment (Besra and Liu, 2007; Sarkar et al., 1997). Silicon carbide (SiC) is a compound of silicon and carbon

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that can be bonded together via the process of sintering in order to form high-performance ceramics that can be used in applications that require high-endurance materials that can withstand high temperatures while retaining their strength, hardness, frequency, biocompatibility, corrosion resistance, and also maintaining a self crack-healing ability (Ando et al., 2004; Casady and Johnson, 1996; Kotzar et al., 2002; Rosenbloom et al., 2004; Thompson et al., 1995; Yakimova et al., 2007). These materials have a range of applications and are used in a number of tools; examples are gas turbines, heat engines, foam filters, cutting tools, and semiconductors (Greil et al., 2002; Ohnabe et al., 1999). Studies have shown that Al_2O_3–SiC and ZrO_2–SiC composite ceramics have significant crack-healing behavior (Ando et al., 2004; Houjou et al., 2009; Moffatt et al., 1996; Thompson et al., 1995). Nevertheless, SiC is a brittle material, and because of its strong covalent bonds, it is difficult to fully densify SiC using simple sintering techniques alone (Wang et al., 1996). In this study, a hot isostatic pressing (HIP) technique was used to sinter and densify the bulk material. The hot isostatic press utilized a high pressure that ranged between 100 and 300 MPa, which was applied to an encapsulated chamber that created and maintained a high dynamic force throughout the sintering process. Thus, a highly densified ceramic material could be formed that had a restriction in grain growth (Li et al., 1996). The ways in which the HIP influenced the physical properties of the materials in terms of shrinkage, porosity, and density were investigated. Using HIP technology, it was possible to simultaneously apply high temperature and high gas pressures to specimens, resulting in the ability to obtain full isotropic material properties (Bocanegra-Bernal, 2004).

Hip joints are commonly constructed using alumina. However, the restricted strength of this material presents an inappropriately high risk of fracture (Novak et al., 2007). The objective of this study was to vary the compositions of SiC and ZrO_2 (zirconia) in order to produce a material that had a tough core and hard outer layers and thus improved the properties of Al_2O_3/ZrO_2 FGM (Hvizdoš et al., 2007; Novak et al., 2007; Vleugels et al., 2003). A 2-butanone-based suspension was used for EPD and both homogeneous and functionally graded ceramic–ceramic, Al_2O_3, SiC, and ZrO_2, where the external layer was SiC and Al_2O_3 and the core layers were a homogeneous SiC, Al_2O_3, and ZrO_2 composite. The porosity and density were measured and the mechanical properties of the material were investigated using the Vickers indentation method. In addition to this, the microstructure of the sintered sample was examined.

2. Experimental procedure

The starting powders consisted of α-Al_2O_3 (99.95%, Alfa Aesar, Ward Hill, MA, USA), with an average crystal and particle size of 0.3 μm; β-SiC (99.98%, Alfa Aesar, Ward Hill, MA, USA), with an average particle size of 1 μm; and a partially stabilized ZrO_2 powder (5.3 wt% Y_2O_3, Sigma Aldrich Corp., St. Louis, MO, USA) that was of submicron size. The powders were ball milled in ethanol with zirconium oxide balls (Retsch GmbH, Haan, Germany) in a horizontal ball mill (9VS, Pascall Engineering Co. Ltd, Suffolk, UK) for 24 h in order to break up the hard agglomerates. After mixing, the ethanol was evaporated in an oven (Carbolite, Hope Valley, UK) at 100 °C over a period of 48 h. Following this, the powders were placed in a drying cabinet for one day. A 2-butanone, *n*-butylamine, and cellulose nitrate solution (2% in amyl acetate) was used for the suspension preparation. The slurries were prepared using a magnetic stirrer in order to suspend the powders in the solvent for 30 min. Dispersion of the particles and homogenization of the suspension were then carried out by placing it in an ultrasonic bath for 10 min. The EPD cell consisted of two stainless steel electrodes. The electrodes had a surface area of 10 cm^2 and a thickness of 0.5 cm. The distance between the electrodes was 3 cm, when they were positioned horizontally. The electrodes were weighed before and after EPD in order to determine the deposited mass.

A voltage of 300 V was applied for 30 min. The suspensions were pumped through the deposition cell by peristaltic pumps. Peristaltic tubing was used for the circulation system, and the diameter of the tubing was 6 mm. The maximum and minimum feed rates were 4 ml s^{-1} and 0.15 ml s^{-1}, respectively. The total deposition time for a functionally graded material was 30 min. A magnetic stirrer was used to mix the suspension during all the EPD steps.

The powder deposited on the anode, and the particles were negatively charged. After EPD, the deposit was removed from the electrode and the dried green body was placed in the drying cabinet for 48 h. Following this, the green body was sintered at 1550 °C for 2 h in a tube furnace. During this process, the sample was placed in an alumina crucible, covered by graphite, and subjected to an argon gas flow of 20 sccm to prevent oxidation. In this study, two sintering techniques were used to sinter the green body solid, namely pressureless sintering and hot isostatic pressing (HIP). After the pressureless sintering, the HIP was performed at 1800 °C and 150 MPa for 2 h in order to reduce the porosity and increase the density.

The sintered, cross-sectioned bulk was ground, polished, and thermally etched for one hour in a tube furnace at 1600 °C under argon atmosphere in order to create a non-oxidizing atmosphere for microstructural analysis. The bulk density of the green and sintered bodies before and after HIP was measured using the Archimedes method. The microstructure of the cross-section was observed by scanning electron microscopy (SEM; Philips XL30) and X-ray diffraction (XRD; Philips X'pert MPD PW3040), using Cu Kα radiation.

The properties for the FGM, i.e. the Young's modulus E and the Poisson's ratio [υ], were derived from the properties of component Al_2O_3, SiC and ZrO_2, levied by the respective volumetric fraction, as follows (Fuchiyama and Noda, 1995):

$$E = \frac{E_c\left\{E_c + \left(E_{Al_2O_3} - E_c\right)V_{Al_2O_3}{}^{2/3}\right\}}{E_c + \left(E_{Al_2O_3} - E_c\right)\left(V_{Al_2O_3}{}^{2/3} - V_{Al_2O_3}\right)} \quad (1)$$

$$\upsilon = \upsilon_{Al_2O_3}V_{Al_2O_3} + \upsilon_{ZrO_2}V_{Al_2O_3} + \upsilon_{SiC}V_{SiC}, \quad (2)$$

where subscript c stands for compositions of SiC/ZrO_2. The Vickers indentation technique is a simple way to estimate the fracture toughness of ceramics (Gong et al., 2002). By means of the indentation method, the mechanical properties of the materials were investigated. Vickers hardness loadings