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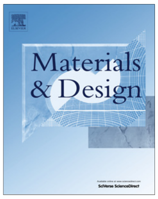

Design and fabrication of laminated–graded zirconia self-lubricating composites

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ABSTRACT

A new design method to achieve the integration of structure and lubricating function in ceramic was proposed by combining the functionally graded material (FGM) concept with the bionic design of ZrO_2(3Y) ceramic. ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/solid lubricant (SL) FGMs were prepared, and gradient exponent *p* was defined to determine the distribution of SL in the materials. The mechanical and tribological properties of the composites with different *p* were studied. ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/SL FGM shows excellent self-lubricating performance over a broad temperature from room temperature to 800 °C, and the bending strength of the sample with the *p* of 2 is four times higher than that of monolithic ZrO_2(3Y)/SL composites. The variation of *p* causes the change in the residual tensile stress generated from the thermal mismatch, and further influences the mechanical property of the materials.

1. Introduction

Tetragonal zirconia polycrystals stabilized by yttria are promising candidates for wear-resistance components owing to their excellent properties such as high strength and fracture toughness, resistance to corrosion, and oxidation stability at high temperature. Nevertheless, both of the coefficient of friction and wear rate of this material are high under dry sliding. To improve the tribological performance of ceramics, ceramic–matrix self-lubricating composites, which are with additives of solid lubricants, have attracted a lot of research attention [1,2]. These materials exhibit excellent self-lubricating properties in a wide range of temperatures since the lubricants can promote the formation of well-covered lubricating films on the surfaces of ceramics. Furthermore, due to the ceramic-skeleton, the good performance and corrosion resistance could also be maintained in high temperature. Actually, the ceramic self-lubricating composite is the only material that can be capable of the work condition above 1000 °C, while maintaining low density and excellent corrosion resistance.

A laminated structural design to ceramics, which could significantly improve the bending strength, fracture toughness and flaw tolerance of them compared with the monolithic ceramics, has been widely studied [3–5]. The principle of such laminated design comes from the imitation of structures found in nature such as biological hard tissues, shells and teeth, which are made of layered architectures combining materials with different properties that lead to laminates with mechanical behavior superior than that of the individual constituents [6]. Since the pioneering work performed by Clegg et al. [7], much effort have been devoted to the development of laminated composites. However, most of these studies focused on the enhancement in the mechanical properties of ceramics [8–10], and few literatures concerned on the tribological and lubricating characteristics of them [11–13].

Therefore, the combination of the bionic design of ceramic materials and self-lubricating ceramic–matrix composites with excellent lubricating property is a promising way to achieve the integration of mechanical and tribological properties. Unfortunately, the addition of soft solid lubricants would reduce the strength of the whole ceramic–matrix composites. In addition, studies have shown that the layered structure solid lubricants break up the continuity of substrate hard phase, which also reduce the mechanical property of the materials [1]. The aim of this work is to overcome the contradiction between mechanical and tribological properties by introducing the concept of a functionally graded material (FGM). The gradual distribution of layered structure component in a FGM realizes the transition from the tough ceramic core, which provides high strength for the whole material, to the wear-resistance surface layers containing solid lubricants that offer excellent lubricating function. Moreover, graded design of the materials is an effective method to eliminate the interface stress of dissimilar materials system [14]. This design is conductive to the combination of mechanical and tribological properties, while retaining all the advantages of these materials.

Based on the previous work [4,8,13], the goals of this work are as follows: (1) fabricate ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/solid lubrication (SL) ceramic composites through the design of bionic laminated–graded structure; (2) characterize the composition distribution,

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microstructures, tribological and mechanical properties of the composites; (3) propose the definition of gradient exponent-*p*, and investigate the effect of *p* on the strength of the laminated–graded composites.

2. Theoretical model

Fig. 1 illustrates the design concept of the ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/SL FGMs. The center area of the FGM is laminated ZrO_2(3-Y)-15% (mass fraction, the same below)Al_2O_3/ZrO_2(3Y) with 15 layers, which provides high strength for the whole material. The SL content is graded increased from center to two sides (10 layers on each side), and finally reaches 60% on the surface to ensure the excellent lubricating function of the materials. In the present study, each layer has the same thickness ($d_1 = d_2$). Each couple of ZrO_2(3Y)–Al_2O_3/SL and ZrO_2(3Y)/SL has the same SL content. The SL content of each couple *f*(*x*) is determined by the following equation:

$$f(x) = (x/m)^p \times 60\% \tag{1}$$

where *x* is the number of the couple; *m* is the total number of the couples in the gradient area (in the present study, $m = 5$); *p* is the gradient exponent.

3. Experimental details

Commercially available CaF_2, graphite and Al_2O_3 after being ball-milled with alumina ball and ethanol for 24 h were used in this study. ZrO_2(3Y) powders were synthesized using a coprecipitation method [15]. ZrO_2(3Y), Al_2O_3, and SL (CaF_2:graphite = 4:1, mass ratio) were carefully weighted and mixed according to different proportion. The powders were placed into a steel die layer by layer with designed composition ratio and then dry-pressed at 180 MPa for 5 min. After that, the green bodies of laminated composites were sintered by hot pressing at 1350 °C and 25 MPa in an Ar atmosphere. The monolithic ZrO_2(3Y)/SL ceramic composites with 60% SL content were also produced.

Macroscopic features of the ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/SL FGMs and microstructures of each layer with different SL content were observed under optical microscopy and JSM-5600LV scanning electron microscopy (SEM), respectively. The structures of the sintered composites were determined by X-ray diffraction (XRD) using Cu Kα radiation. The friction coefficients were measured using a standard SRV friction and wear tester (SRV-IV, Germany) with reciprocating motion against a $\varnothing$ 10 mm alumina ball with a Vickers hardness of 15 GPa. The tests were conducted at a frequency of 15 Hz for 1800 s, with a linear stroke of 1 mm and under a load of 20 N at the temperature range from 25 to 800 °C. Bending strength was determined by a three point bending test on a universal testing machine (DY35) with a span length of 20 mm and a crosshead speed of 0.5 mm/min at room temperature according to ISO 14704:2008(E) [16]. Sample dimensions were 25 mm × 4 mm × 3 mm and bending strength was achieved by the following formula:

$$\sigma = 3FL/2bd^2 \tag{2}$$

where *F*, *L*, *b*, *d* were load, span, width and height, respectively.

4. Results and discussion

4.1. Microstructure characterization

Fig. 2a is the optical photograph of the ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/SL FGM. The center bright area is laminated ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y) without SL. The enlarged SEM micrograph of this area is shown in Fig. 2b, in which the darker layers are ZrO_2(3Y)–Al_2O_3 and the lighter layers are ZrO_2(3Y). This area shows obvious layer structure without distinct pores or cracks. Each of the adjacent layers has the corresponding thickness, and the average thickness of each layer is about 100 μm. In addition, as can be seen from Fig. 2b, the layers in the composite materials prepared by the technology of layers stacking and hot pressing sintering in the present study are not very smooth. However, the tortuosity of interfaces increases contact area between the adjacent layers, which is beneficial to boundary strength of the materials [4].

The dark areas of the two sides of the ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/SL FGM are gradient layers with SL content increased from center to two sides. Fig. 3 shows the microstructures of fracture surfaces of gradient layers. As can be seen, the ceramic composites are mainly composed of ceramic phase and lubricating phase. The main composition of ceramic phase is well-crystallized ZrO_2(3Y) particles. Graphite distributed among them shows flaked structure, and CaF_2 presents with spherical form. With the increasing content of SL (d → a), large pores begin to appear and the amount of pores increases generally. This result is in agreement with the reports before [17,18]. The major reason for that is the addition of graphite which increases the densification temperature of the materials. Therefore, the density of the composite decreases with increasing SL content under the same sintering condition, and large holes begin to appear.

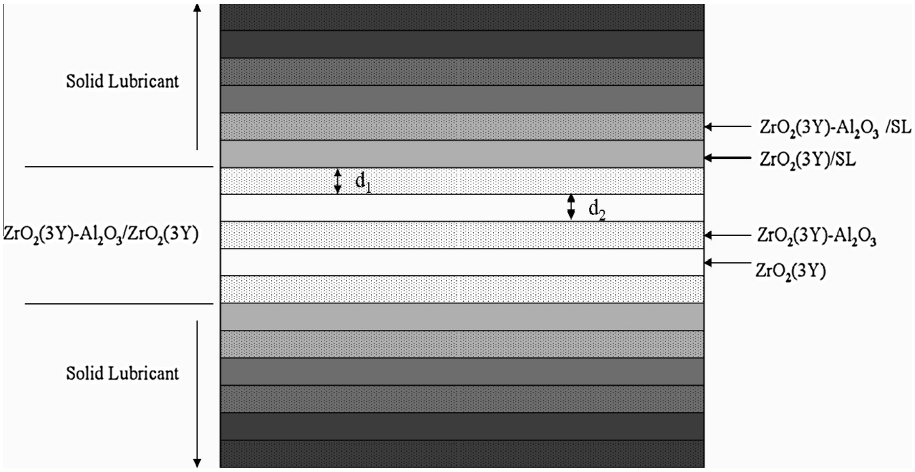

Fig. 1. Schematic of the ZrO_2(3Y)–Al_2O_3/ZrO_2(3Y)/SL FGMs.