Int. Journal of Refractory Metals and Hard Materials 64 (2017) 122–134

Contents lists available at ScienceDirect

Int. Journal of Refractory Metals and Hard Materials

journal homepage: www.elsevier.com/locate/IJRMHM

Residual stress and diffraction line-broadening analysis of Al_2O_3/Y-TZP ceramic composites by neutron diffraction measurement

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ARTICLE INFO

Article history:
Received 3 August 2016
Received in revised form 23 December 2016
Accepted 30 January 2017
Available online 01 February 2017

Keywords:
Ceramics
Ceramic composites
Tape casting
Neutron diffraction
Residual stress
Time of flight, Peak broadening
Microstrain

ABSTRACT

Time-of-flight neutron diffraction and Rietveld analysis have been used to investigate the residual stresses, crystallite structure and microstructural features in Al_2O_3/Y-TZP ceramic composites fabricated by two different green processing techniques (a novel tape casting and conventional slip casting) and with different zirconia content (5 and 40 vol.% Y-TZP). The change in lattice parameters, individual peak shifts and peak-broadening were analyzed, in order to calculate the uniform residual stress field (mean phase stresses and peak-specific stresses) and the non-uniform microstrains. Peak-specific residual stresses were calculated for different *hkl* reflections both for the Al_2O_3 matrix and the Y-TZP particulates. The sign and the magnitude of the peak-specific residual stress are highly dependent on the individual *hkl* reflection studied and on the volume fraction of zirconia. Peak broadening was observed in the Y-TZP reflections, due to non-uniform microstrains. Both the mean phase stress field and the non-uniform microstrains were mainly influenced by the Y-TZP content in the studied Al_2O_3/Y-TZP composites, irrespective of the measured direction and the fabrication process.

1. Introduction

Alumina-zirconia ceramics have received considerable attention in both engineering and academic fields due to the possibility to improve their mechanical properties with respect to pure alumina [1–4]. As one of the most popular alumina-zirconia systems, the Al_2O_3/Y-TZP (tetragonal polycrystalline zirconia stabilized with 3 mol% Y_2O_3) exhibits a noticeable performance, i.e. high strength and toughness combined with excellent biocompatibility, wear resistance and good resistance to corrosion and chemicals. This is the reason why it is considered as a candidate for structural and biomedical applications [5,6]. In order to improve the inherent brittleness associated to ceramics, new fabrication techniques have been employed to increase fracture toughness through different mechanisms. Besides the well-known stress-induced transformation toughening and microcracking toughening [7–11], it has been proposed that interphase residual stresses generated by the thermal and elastic deformation mismatches between alumina and zirconia could also contribute to toughening and enhance the structural performance of these ceramic composites [12–14]. Furthermore, for each phase, the thermal expansion coefficient is anisotropic, which may lead to the development of intergranular stresses. The generation of residual stresses, in addition to externally applied stresses, may induce a martensitic transformation (tetragonal to monoclinic) in zirconia [12]. This could produce a wide variety of possible toughening mechanisms: crack deflection, crack branching, crack bridging and microcracking [11, 15–17]. As a consequence, an accurate knowledge of residual stresses would provide valuable data to optimize the performance and reliability of ceramic composites [18].

In order to obtain defect-free laminated ceramic structures with operative reinforcing layers, residual stresses should be known. Besides the effect of laminate designs (layer stacking structure and thickness) on the macroscopic stress field between adjacent layers [18–21], detailed micro-residual stress information (i.e. residual stresses between different phases and grains) is needed. This would allow us to better understand the reinforcing mechanisms as well as to optimize the resulting residual stress field. Micromechanical analyses predicted that residual stresses of ceramics could be changed by an appropriate selection of the fabrication process and the materials design [6,22]. The effects associated to material composition and fabrication should be separated from those related to the laminate stacking design, in order to have a better control over the manufacturing process.

Several research works have been published on the residual stress (RS) analysis of alumina-zirconia composites [14,19,23–26]. In those works, different measurement techniques were used, namely Raman spectroscopy, X-ray and neutron diffraction. A range of RS results were reported depending on the fabrication route, composition design and measurement technique. Most of the works were focused on the uniform residual stresses between phases, without paying attention either to the elastic and plastic anisotropy effects, or crystal defects (i.e.

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http://dx.doi.org/10.1016/j.ijrmhm.2017.01.011
0263-4368/

dislocations and crystal vacancies). However, intergranular stresses are important because stress concentrations on the scale of individual grains may play a significant role on the initiation of cracks. Consequently, when different processing routes or materials are used, it is necessary to accurately quantify residual stresses at different scales by using the most suitable RS measurement technique in each case.

A novel tape casting route [27,28] was employed to fabricate the alumina-zirconia ceramic composite samples in the present work. In this case, green ceramics tapes were stacked by using low pressure at room temperature. Such processing route has important benefits over traditional methods both from the economic and environmental point of view, partly because lower content of organic additions is needed. In addition, the resulting laminates have very good properties because interface defects, anisotropy, porosity and even cracks are considerably reduced [27,28]. However, a detailed assessment of the residual stresses produced by this novel route is lacking. The conventional slip casting technique [29], which was accepted as a simple, reliable, flexible and economical process technology for ceramic preparation for many years, should be taken as a reference for comparison. The addition of zirconia to alumina has a significant influence on the microstructure and mechanical properties of the resulting ceramic composites [30]. In addition, co-sintering of layers with different compositions opens the door to new laminated materials with improved properties. The crystal structures in sintered ceramics, especially the ones of zirconia, should be accurately examined, due to the complex polymorph of ZrO_2 and the possible tetragonal to monoclinic phase transformation during the fabrication process.

Neutron diffraction (ND) allows the measurement of residual strains in bulk materials due to the high penetration of neutrons. When the time of flight technique (TOF) is used, the whole diffraction pattern can be collected simultaneously [31], which is key for residual stress determination in complex materials, such as ceramic composites. By applying Rietveld refinement [32] (whole profile structure refinement method), information on residual stresses can be extracted from the whole diffraction spectrum. Macrostresses at the continuum scale (type I stresses) and average phase stress (type II stresses) can be calculated from the change in the lattice parameter with respect to the strain-free value. In addition, intergranular stresses can be calculated from the peak shifts associated to individual reflections. Finally, peak broadening analysis [33–35], allows us to estimate the non-uniform microstrains at the subgrain scale (type III stresses), as well as the coherently scattering domain size.

In this paper, TOF neutron diffraction and Rietveld analysis were combined to precisely determine the uniform and non-uniform residual stresses in a series of Al_2O_3/Y-TZP composites with different Y-TZP content and prepared using different green processing techniques (a novel tape casting and conventional slip casting). Multi-phase qualitative and quantitative analyses, as well as crystal structure determination, were performed by Rietveld refinement. Line broadening analysis was carried out, using the “Double-Voigt” profile modelling, to obtain the microstructural information (domain size and crystal microstrain). The effects of the addition of second phase zirconia particulates and the green processing technique employed were discussed in terms of the obtained residual stress fields, microstrains and domain sizes.

2. Materials and methods

2.1. Materials

The studied Al_2O_3/Y-TZP (3 mol% Y_2O_3 stabilized zirconia) ceramic composites were prepared by using two different green processing methods: a novel tape casting route [27,28], and conventional slip casting. Two different zirconia contents were employed as reinforcement: 5 and 40 vol.%. The specimens were coded as A-5YTZP (slip), A-5YTZP (tape), A-40YTZP (slip), and A-40YTZP (tape), to describe their composition and fabrication technique.

The starting powders used to fabricate the composites were high-purity α-Al_2O_3 (Condea HPA 0.5, USA) and polycrystalline tetragonal zirconia stabilized with 3 mol% Y_2O_3 (TOSOH, Japan). A polyelectrolyte (Dolapix 64 CE, Zschimmer and Schwarz, Germany) was used for powder dispersion. Stable slurries were prepared by mixing the starting powders in deionized water. After mixing, the slurries were ball milled during 4 h in alumina jars using alumina balls. For each composition, two series of pieces were fabricated from the stable slurries. One series was fabricated by slip casting of the deflocculated suspensions in plaster mold and leaving them to dry for 24 h. The other series was fabricated by tape casting on a polypropylene film using a moving tape-casting device with two doctor blades using a 10 mm/s casting velocity and a 500 μm gap height between the blades and the carrier film. After drying, the final thickness of the green tapes varied between 480 and 520 μm. Monolithic pieces were prepared by stacking tapes of the same compositions together and applying a gluing agent (5 wt.% dilution in distilled water of the binder) under uniaxial pressure (18 MPa) at room temperature. For both the slip-cast and tape-cast green pieces, binder burnout and sintering were performed in a single thermal treatment. The binder burnout was carried out with a heating rate of 1 °C/min up to 600 °C, followed by a dwell time of 30 min. Sintering was performed at maximum temperature of 1500 °C, with a dwell of 2 h (heating and cooling rates of 5 °C/min). High-density materials (relative density > 98% the theoretical density) were obtained after sintering for all the studied Al_2O_3/Y-TZP composites, both of the slip-cast and tape-cast samples [36].

The specimens were ground and mirror-polished with diamond paste until 1 μm, and then chemically etched in 85% H_3PO_4 for 7 min at 200 °C for microstructural characterization. The analyses were performed by scanning electron microscopy (SEM, Zeiss DSM 950, Germany) and the average grain sizes of Al_2O_3 and Y-TZP particles were determined by the linear intercept method considering at least 200 grains for each phase.

2.2. Residual stress measurement

For neutron diffraction strain scanning, samples in the shape of parallelepipeds with dimensions $20 \times 20 \times 5$ mm^3 were employed. In the pieces fabricated by tape casting, the larger surfaces (20×20 mm^2) were parallel to the surface of the stacked tapes.

Residual strain measurements were performed by neutron diffraction on ENGIN-X time-of-flight instrument, a third-generation neutron strain scanner, operating at the ISIS spallation neutron source in the Rutherford Laboratory, UK. Details of this instrument are given in [37]. The experimental setup consists of two detector banks which are centered on Bragg angles of $2\theta_B = \pm 90°$, as shown in Fig. 1. This setup allows the simultaneous measurement of strain in two directions: the in-plane direction, parallel to the larger plane of the samples (20×20 mm^2), and the normal direction, perpendicular to it, which are assumed to be the principal stress component directions. The fabrication process is expected to produce a transversely isotropic stress state, where the stresses in the larger plane of the sample are equal. Consequently, only the in-plane and the normal directions would be needed for residual stress determination.

The measurement gauge volume for the ceramic composites was set to $15 \times 1 \times 1$ mm^3 by means of horizontal and vertical cadmium slits (incident beam) and a radial collimator slit in front of each detector bank. The centroid of the gauge volume defines the measurement location. Strain scanning was carried out along the sample thickness (approximately 5 mm), in 0.4 mm steps.

The stress-free reference lattice parameters of α-Al_2O_3 and Y-TZP were obtained by measuring both the α-Al_2O_3 and Y-TZP original powders. In this case, the gauge volume was enlarged to $16 \times 3 \times 1$ mm^3, in order to increase the diffracted intensity and thus to improve the measurement accuracy. CeO_2 standard powder was also measured to