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Thermal conductivity of ceramic/metal composites from preforms produced by freeze casting

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ABSTRACT

Porous alumina and zirconia preforms, processed by ice templating, have been used to manufacture ceramic/metal composites by aluminium alloy infiltration. The aim of the present work is to study the influence of the ceramic material nature and of the initial porous structure on the thermal conductivity anisotropy of the composite in order to assess potential applications in the field of thermal management. The materials are characterised in terms of pore volume fraction and pore size before and after metal infiltration. The freeze casted preforms exhibit anisotropic lamellar structures with ellipsoidal pores ranging from 35 μm to 40 μm and porosity fractions from 64 to 67%. After metal infiltration, composite parts present the same anisotropic morphology, which correspond to alternating ceramic and metal layers. Thermal conductivities have been determined, with an average of 80 $W m^{-1} K^{-1}$ and 13 $W m^{-1} K^{-1}$ parallel and perpendicular to the freezing direction respectively, for zirconia/metal composites. Theoretical values of thermal conductivity can be calculated using the Maxwell-Eucken relation, to handle the residual porosity, in combination with series and parallel resistance models to describe the overall anisotropic character. These give good agreement to experiment.

1. Introduction

Porous ceramics are used for diverse applications according to characteristics such as pore volume fraction and distribution, permeability and so on. As examples, porous ceramic structures are used as filters for molten metal [1], water [2] or gas [3] and microfiltration by membranes, but also to process fuel cell electrodes [4], for thermal energy storage devices [5], and for sound [6] or thermal insulation [7]. All these applications are typically developed with isotropic porous materials. In the case of materials exhibiting anisotropic porosity, similar applications are possible, with significant improvements in the properties as it has been demonstrated for the preparation of membranes [8], of piezoelectric substrates [9,10], electrodes for fuel cell (SOFC) [11], biomaterials [12] and ceramic matrix composites [13,14].

Freeze casting by ice templating of ceramic materials, developed in the last 15 years [15], consists of freezing stable ceramic slurry. The porous structure results in the growth of ice crystals and their interaction with the ceramic particles. A green body is then obtained by sublimation of the ice, without shrinkage or damaging the ceramic framework [16]. An anisotropic structure can be produced in specific conditions when the solidification of the slurry is oriented by a thermal gradient [17]. Consequently, the structure presents elongated and continuous pores oriented along the freezing direction. Some authors have shown that these porous materials exhibit anisotropic thermal conductivity [18–20].

An easy way to obtain a ceramic matrix metal composite consists of filling the porosity of a ceramic preform with a metal, by squeeze or die casting [14,21]. In most cases, such composites are made, using an isotropic [22] preforms, but some papers have reported composites prepared from freeze casted ones [23,24]. For example, Roy et al. [14] have filled porous alumina preforms with an aluminium alloy and they studied the elastic behaviour of such composites.

Recently, it has been shown that the orientation of carbon nanofibers in a copper matrix leads to anisotropic thermal conductivity [25]. In such perspective, the present work aims at

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investigating the anisotropic thermal conductivity behaviour of ceramic metal composites processed from freeze casted ceramic performs. The structures are characterised in terms of pore volume fraction and pore size before and after metal infiltration. An analytical model is used to calculate values, based on resistances in series and parallel to describe the anisotropic conduction, and the Maxwell-Eucken relation to handle the effect of residual porosity.

2. Experimental procedure

2.1. Materials and methods

Slurries are prepared by mixing 20 vol% of ceramic in deionised water with a small amount of ammonium polymethacrylate anionic dispersant (Dolapix CE64, from Zschimmer and Schwarz, Germany). The ceramic is an alumina powder (d50 ~0.4 μm, specific surface area = 8 $m^2 g^{-1}$, P172SB, Rio Tinto Alcan, Brazil) or a 3 mol% yttria stabilised zirconia powder (d50 ~0.3 μm, specific surface area = 7 $m^2 g^{-1}$, TZ-3YS, Tosoh, Japan). The slurries are then mixed for 24 h in Turbula® with alumina or zirconia beads (diameter of 2 mm). After removing the balls, 3 wt% of polyethylene glycol (Mw = 1000 mol g^{-1}, from Merk, Germany) is added as a binder.

Each slurry is poured into a cylindrical TEFLON mould (30 mm inner diameter and 50 mm high) and put on the freezing device. Fig. 1 illustrates the freeze casting device. The sample holder is cooled slowly down to −40 °C (1 °C/min), and then the temperature is kept constant until the full freezing of the slurry is achieved. The frozen samples are then placed in a freeze dryer (HETO CD8, Thermo Fisher Scientific, USA) for 48 h to remove the solvent at a pressure close to 10 Pa. All green bodies are fired at a constant heating rate of 5 °C/min and maintained during 2 h at 1600 °C (alumina) or 1450 °C (zirconia) and finally cooled down at 5 °C/min to room temperature.

After sintering, ceramic/metal composites are obtained by filling the ceramic preforms with an aluminium alloy, $AlSi_{10}Mg$ (Altara, France) with a purity of 99%, using a vacuum pressure metal casting device (VC-650 V, INDUTHERM, Germany) (Fig. 2). The ceramic part is placed inside a refractory plaster mould which was prepared by investment casting. Aluminium alloy is melted at 740 °C and poured into the mould heated at the same temperature. The mould is then cooled down to ambient temperature naturally. Composites exhibit cylindrical shape with a diameter and a height of about 20 mm. Slices of the composites are cut both perpendicularly and parallel to the freezing direction, to allow the determination of their thermal conductivity at room temperature and examined by SEM and X-ray tomography to evaluate the

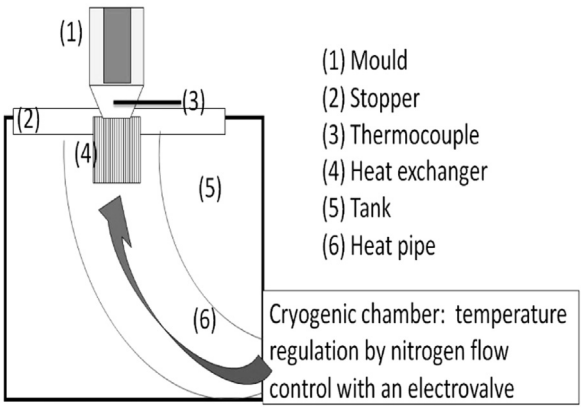

Fig. 1. Schematic representation of the freeze casting device.

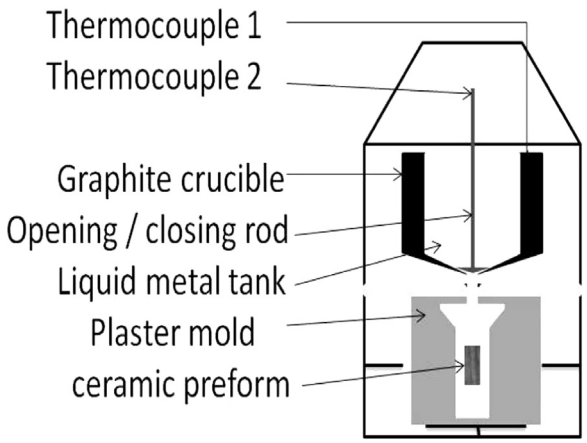

Fig. 2. Schematic representation of the metal infiltration device.

infiltration quality. Typical dimensions of the disk samples are 2 mm in thickness and 14 mm in diameter with parallel and unpolished faces.

2.2. Characterisation

2.2.1. Microstructure

The amount of porosity and bulk density are determined by using the method based on Archimedes' principle for ceramic preforms using deionised water as the liquid medium. Pore size is measured using image analysis of the SEM micrographs (HITACHI S-3500N and JEOL JSM 5900 LV, Japan). For each sample, 10 micrographs with more than 150 pores in total are investigated.

The sintering conditions lead to incomplete densification of the ceramic scaffold, which correspond to the ceramic wall porosity. To determine this micro porosity in the ceramic wall of the porous preforms, an ultrasonic method is used. The measurements are made on cylindrical samples (diameter and height of 20 mm) using two transducers (Ultrasonic tester, Brutsaert, Belgium) with a nominal frequency of 250 kHz, assuming that anisotropic lamellae will act as a waveguide. The transducers are placed on opposite sides of the sample using soft paraffin as a coupling agent.

Calibration curves of wave velocity as a function of porosity are recorded on isotropic alumina and zirconia samples with known porosity amount. These samples are prepared by uniaxial pressing of powder pellets followed by sintering at different temperatures. Three samples were processed for each reference material and the wave velocity is measured six times for each sample. The calibration curves are plotted in Fig. 3. The experimental data are in

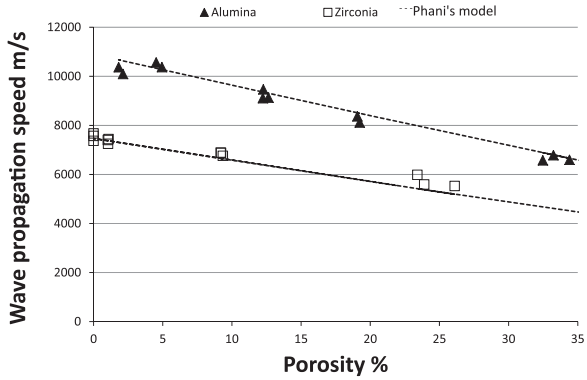

Fig. 3. Standard curves of ultrasound wave propagation speed as a function of porosity for isotropic porous ceramics. Black line correspond to calculated curves from Phani's relation.