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New approach to low thermal conductivity of thermal barrier protection with improved mechanical integrity

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Received 6 January 2016; accepted 9 January 2016

Abstract

Layered ceramic systems with designed stacks of dense and porous layers were investigated as alternative for thermal barrier protection system (TBPs). This approach gives the possibility to obtain low thermal conductivity with the impact protection of dense external layers whilst maintaining the relatively high mechanical properties. Different stacking configurations have been proposed utilizing in total a combination of up to 30 dense/porous layers. Porous layers were produced with two different nominal porosities 20 vol% and 40 vol%. For comparison uni-axial pressed samples with the same porosity level have been prepared. Thermal and mechanical characterization was performed on samples of tape cast (with different stacking designs) and uni-axial pressed fully stabilized zirconia TBPs. The layered fully stabilized zirconia (8YSZ) has 15–30 % lower thermal conductivity in comparison with the uni-axial pressed samples, nevertheless by the same Young`s modulus value. The results of the thermal and mechanical observation shows, that such an approach can be beneficial as an alternative for future thermal barrier protection systems.

Keywords: Thermal barrier protection; Zirconia; Tape casting; Thermal conductivity; Young`s modulus

1. Introduction

Structured layered materials attract attention of researchers in many fields including metallurgy [1], the polymer industry [2] and ceramics [3–5]. The reason of preparing structured materials in ceramics is to obtain a desired combination of properties. Namely to improve the fracture toughness, stiffness or strength of the final material laminar composite in comparison to conventional monolithic materials. There are two main methods to enhance the mechanical behaviour of multi-layered ceramic composites. The designing of compressive residual stresses due to the thermal mismatch of the combined layers can lead to a favourable increase in the fracture toughness. This approach was successfully described for oxide ceramics e.g. by Bermejo et al. [6] or Rincón et al. [7] and non-oxide ceramics Si_3N_4/TiN ceramics by work of Blugan et al. [8]. On the other hand, the integration of weak (porous) interfaces is the most common used technique to improve the strength and fracture toughness of multi-layered systems. The incorporation of porous layers was first described by Atkins in 1974 [9]. Henceforth, the stacking and effect of different-layered materials has been studied for several systems e.g. SiC composites [4,10], ZrO_2/(ZrO_2 + Ni) [11] or Al_2O_3 system in the recent work of Sellappan [12], mainly with regards to their mechanical behaviour. The most common technique to prepare such structured ceramics is tape casting followed by lamination of the tapes, burn-out and sintering [4,5,8]. Thus the chemical compatibility of the dense and porous layers is essential. Other

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http://dx.doi.org/10.1016/j.ceramint.2016.01.059
0272-8842/

approaches include electrophoretic deposition [13], extrusion [14] or slip casting [15].

Zirconia is a material with good mechanical and chemical properties [16]. It is widely used in medical applications; for fuel cells, as a dopant in other ceramics and as thermal barrier coatings [17]. Zirconia fully stabilized with yttria (8YSZ) is widely utilized in solid oxide fuel cells (SOFC) [18] due to the high ionic conductivity and low thermal conductivity. The above mentioned properties in combination with a high melting point makes it an excellent and widely used candidate for thermal barriers protection in high temperature insulation applications [19].

In a previous paper [20] we presented the results on aqueous and non-aqueous tape casting of 8YSZ. The present work is a continuation focused on obtaining multi-layered systems with low thermal conductivity whilst maintaining a relatively high Young`s modulus. Multi-layered materials consisting of 30 dense/porous layers of fully stabilized zirconia with and without pore former have been prepared. The properties of different laminate designs are compared to each other and also to bulk materials prepared via conventional uni-axial pressing. The experimentally obtained data from laser flash analysis of thermal conductivity and Young`s modulus are compared with the theoretical models.

2. Experimental

2.1. Materials and processing

The study was performed on materials produced from yttria fully stabilised zirconia powder - TZ-8YS (Tosoh Corporation, Advanced Ceramics Division, Tokyo, Japan). The primary particle size diameter, measured with a Beckman-Coulter LS 230 (Beckman Coultier, 250 South Kraemer Boulevard Brea, CA 92821, USA) particle size analyser gave a D_{50} of ~400 nm.

Spherical polymethyl methacrylate (PM_6) - Spheromers CA6 (Microbeads AS, Norway) with a mean diameter of 6 μm and coefficient of variation < 5 %, were used as pore formers.

Non-aqueous tape casting was used for preparation of dense and porous tapes. The preparation procedure followed the description previously presented in detail [20]. The recipes of the different compositions are summarized in Table 1.

The suspensions were cast using a 20 cm wide double doctor blade with a gap of 400 μm on a laboratory tape caster CAM-T0 (Heiku Keko Equipment, Žužumberk, Slovenia). Casting speed was 0.9 mm/s and the drying time at least 5 h at ambient conditions before releasing the tapes from the Mylar substrate.

Table 1
Recipes of suspensions for tape casting.

Suspension [vol%]						
Name	**ZrO_2**	**PM_6**	**Solvent**	**Surfactant**	**Binder**	**Plasticizer**
YSZ	25	–	57.2	2.3	7.7	7.8
YSZ+20PM$_6$	20	5	57.2	2.3	7.7	7.8
YSZ+40PM$_6$	15	10	57.2	2.3	7.7	7.8

The thickness of the dried tapes was between 130 μm at the casting start to 110 μm at the end of casting. The missing pressurized master feed tank in the tape casting machine caused this variation in the beginning and end of the casted tape. A simple doctor blade reservoir was used. Single layers were punched using a 47 mm diameter punch. Green laminated samples were stacked from 30 layers and heated up to 100 °C for 25 min before uni-axial pressing into laminates. A maximum pressure of 115 MPa was used to press the samples.

Binder burn-out was performed using a low temperature furnace Nabertherm LT40/12 (Nabertherm GmbH, Lilienthal, Germany). A moderate heating rate of 0.5 °C/min ensured slow burn-out of organic components along with the pore former. The maximum temperature of this step was set to 950 °C which causes some pre-sintering of the samples. After this the specimens could be handled and sintered.

Sintering of the final samples was carried out in a high temperature furnace HT 18/5 (Carbolite Gero GmbH & Co. KG, Neuhausen, Germany). The heating rate was 4 °C/min up to 1600 °C with a dwell time of 1 h.

2.2. Instrumentation

The relative density of sintered samples was measured by Archimedes method in water, from at least 5 samples for the dense materials and compared to the density calculated from dimensions and weight. The uncertainty of the measurements was ≤2%. The geometrical densities of samples containing porous layers have been calculated from the dimensions and weight. The total porosity of the samples (Φ), used in the Eq. (2), was calculated subtracting the theoretical density with the real density of the samples.

Microstructure characterisation was performed with scanning electron microscopy analysis (VEGA Plus 5136 MM, Tescan instruments, Czech Republic) on fractured surfaces of sintered samples.

The linear intercept method (software LINCE, TU Darmstadt, Germany) was employed to determine the mean grain size of zirconia and mean pore diameter. At least 250 intercepts were taken for each material with the correction factor of 1.56 [21].

Young`s modulus measurements were carried out using the impulse excitation technique with a GrindoSonic MK5 (J.W. LEMMENS N.V., Leuven, Belgium) in torsional and flexural vibration. The sample was impulse excited (gently tapped) and vibrations are analysed in accordance with the EN 843-2 standard. The obtained resonance frequencies are related to its stiffness, mass and geometry of the tested samples. The obtained samples had not been cut or polished before testing, diameter was 38 mm and thickness varied between 2.6–2.9 mm. The obtained value is an average at least from 4 samples measured 5 times.

Thermal diffusivity was measured using Laser Flash Analysis LFA 457 (NETZSCH-Gerätebau GmbH, Selb, Germany) in the temperature range from 25–1000 °C in Ar atmosphere at 10 temperature points. Samples were cut into squares of 10x10

Please cite this article as: M. Michálek, et al., New approach to low thermal conductivity of thermal barrier protection with improved mechanical integrity, Ceramics International (2016), http://dx.doi.org/10.1016/j.ceramint.2016.01.059