Journal of the European Ceramic Society xxx (2015) xxx–xxx

Contents lists available at www.sciencedirect.com

Journal of the European Ceramic Society

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

The effect of porosity and microcracking on the thermomechanical properties of cordierite

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ARTICLE INFO

Article history:
Received 27 April 2015
Received in revised form 3 August 2015
Accepted 12 August 2015
Available online xxx

Keywords:
Diesel particulate filter
Cordierite
Porosity
Microcracking
Micromechanical differential scheme

ABSTRACT

The effect of porosity and microcracking on the mechanical properties (strength, fracture toughness, Young's modulus, and fracture energy) and thermal expansion of diesel particulate filter (DPF) grade cordierite materials has been investigated. A method to deconvolute the effect of porosity and microcracking on Young's modulus is proposed. In addition, the microcrack density and the pore morphology factor are calculated by applying a micromechanical differential scheme. The values of the investigated mechanical properties are shown to decrease with an increase in porosity, but the thermal expansion values are insensitive to porosity. The variation in mechanical properties as a function of porosity leads to distinct porosity dependence of thermal shock resistance for crack initiation and crack propagation for DPF grade synthetic cordierite.

1. Introduction

The functional characteristics of diesel particulate filters (DPFs) are such that the porosity of the filter is a key determinant for filtration as well as mechanical properties. Cordierite (chemical formula—$Mg_2Al_4Si_5O_{18}$) is a low thermal expansion ceramic that is commonly used as a material in exhaust aftertreatment systems. The porosity needs to be optimized based on competing requirements for exhaust filtration efficiency and thermo-mechanical performance [1]. This is especially relevant since worldwide government regulations associated with particulate matter and NO_x emissions continue to become increasingly stringent [2]. The advantages of increasing the porosity [3] of the DPF include: (i) increased loading of washcoats and possibly catalysts, such that the filter can serve in a multifunctional manner including catalytic reduction, NO_x trapping, hydrocarbon oxidation in addition to trapping particulate matter, (ii) reduced engine back pressure; this leads to increased engine efficiency and (iii) increased mechanical compliance and, therefore, potential to increase resistance to thermal shock. Increasing the porosity, however, also leads to a decrease in the mechanical load-bearing ability and therefore, the disadvantages of increasing the porosity of the substrate can be summarized as follows: (i) decreased strength (and fracture toughness), (ii) decreased thermal conductivity of the substrate; both properties influence the thermal shock resistance [4–6]. A DPF with an optimal level of porosity will balance these competing effects.

DPF materials, and consequently the filters themselves, do not generally possess isotropic properties (thermal expansion, Young's modulus, etc.). Kachanov (see [7]) has shown that in the simplest case of *isotropic* microcracked (with penny shaped cracks) and porous materials, at least three parameters (microcrack density, porosity, and pore shape) are necessary to describe their effective properties. In general, one porosity and one microcrack density *tensors* are necessary to fully describe the effect of 'defects' in solids. For a porous and non-microcracked material, the typical empirical approach to tackle porosity effects on properties is to apply the pore shape factor, m [8,9]. This approach has its origin in the application of a micromechanical modified differential scheme [10,11]. For porous and microcracked materials, such as ceramics for DPF applications, Bruno and Kachanov [12] have shown that it is possible to apply a two-step homogenization approach, taking into account porosity and microcracks separately, since the two homogenization operations are commutative.

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http://dx.doi.org/10.1016/j.jeurceramsoc.2015.08.014
0955-2219/

In this work, we investigated DPF type cordierite materials of different origin (manufacturers) and conditions (bare and coated, after service and soot loaded) with porosity in the range of 50–70%, and compared their properties to those of lower porosity cordierite materials. The effects of porosity on some of the properties that determine the thermal shock resistance (for crack initiation and crack propagation [5,13]) were investigated. The properties that were measured include fracture toughness, characteristic strength, Young's modulus and coefficient of thermal expansion (CTE). Some of the available models to rationalize the above properties and the microstructural features (in particular the microcracks) linked to their variation with porosity are reviewed. It is shown that porosity and microcracking [14,15] can together have a strong influence on the thermal shock resistance of cordierite DPFs. These results can aid in the identification of a DPF substrate material with optimal thermal shock resistance that is balanced with functional requirements.

2. Materials and experimental procedure

A number of materials were acquired for this investigation and their description (including porosity), and the type of characterization performed on them are summarized in Table 1. A total of 11 cordierite-based materials are included in Table 1. All materials except the bulk cordierite material (BulkCO) and low porosity cordierite (LP) were acquired in honeycomb forms from as-fabricated DPF devices (Fig. 1(a)). The honeycomb structures had square channels (~1.5–2.5 mm size) and specimens for evaluation of properties were fabricated by following a series of dry cutting and grinding procedures reported elsewhere [16]. The porosity of specimens was measured by evaluating the specimen density and comparing it to the expected bulk density (2.46 g/cm^3) of the material. It was shown that this technique led to porosity values that were similar to those calculated with two dimensional image analysis of the Duratrap CO material [16]. Table 1 suggests that the porosity of the evaluated DPF materials varied in the range 50–70%. Since the low porosity cordierite material had some closed porosity, its porosity was determined by image analysis.

The specimen preparation and experimental procedure for mechanical and thermal expansion characterization has been described in detail in previous publications [15–17]. To summarize, fracture toughness testing was performed under ambient conditions on precracked double-torsion specimens with dimensions 20 mm × 40 mm × ground specimen thickness. The ground specimen thickness was ~0.25–0.40 mm with a flatness of 0.005 mm. The maximum load in the fast fracture test was used to calculate the fracture toughness value at room temperature [18]. The reported values are the average of five fracture toughness tests, and the error bars indicate the standard deviation. The flexural strength testing and analysis was performed on honeycomb shaped specimens (7 × 14 cells in cross-section) as per the ASTM standard C1674 – 11. A total of 30 specimens were analysed to compute the characteristic strength and Weibull modulus according to two parameter Weibull analysis.

The Young's modulus at room and elevated temperature was determined using resonant ultrasound spectroscopy (RUS) [15,19,20]. Thermal expansion measurements were performed in a thermomechanical analyser (TMA – Q400 TA Instruments). For the DPF specimens, measurements were performed both parallel and perpendicular to the extrusion direction whereas only one measurement was performed for the low porosity and bulk cordierite specimens (which are assumed to be thermally isotropic). The reported average linear CTE values are from the second heating cycle between 20 and 1000 °C (the thermal expansion curves are more stable in the second cycle where effects such as soot burning can influence the first cycle). Further details of the high temperature RUS and thermal expansion measurement procedure are reported elsewhere [15].

The microstructure of the porous cordierite samples was examined by field emission gun scanning electron microscopy (FEG SEM—Hitachi S4800) and by electron back scattered diffraction (EBSD). Specimens for EBSD characterization were prepared by embedding in cold-setting epoxy, polishing to a mirror finish, and then cutting the polished and embedded specimens into ~8 × 8 × 2 mm sections. These sections were then polished using argon ions in a Gatan Ilion + cross-section sample preparation tool. Final polishing used 3 keV argon ions. The specimen surface was lightly coated with carbon via evaporation, and conductive paint was applied to cover all other faces. EBSD was then performed in a JEOL6500F SEM, using a 15 keV, ~6 nA beam. The near-match of {0 1 0} and {0 0 1} lattice parameters resulted in "checkerboard" pseudosymmetry of cordierite crystals oriented along those axes.

3. Microstructure of porous cordierite for diesel particulate filters

The macro- and microstructure of porous DuratrapCO® cordierite for DPF applications is included in Fig. 1. The other DPF cordierite materials have qualitative features similar to the ones described in this section. Fig. 1(a) is an image of a 2 × 2 cell section of this material that shows the square channels with 2.25 mm sides. A low magnification SEM image showing the 50% porous microstructure is shown in Fig. 1(b). The higher magnification in Fig. 1(c) reveals the grain structure and the presence of microcracks. It is noted that some microcracks begin and terminate on the pores. The grains of cordierite are elongated along the *c*-axis of the crystal as described previously (and indicated in Fig. 1(c)) [14,15]. The microcracks may be preferentially aligned parallel to the long axis of grains within collections of neighboring grains with similar orientation termed 'domains' [14,15,21]. The presence of domains is shown in the EBSD image in Fig. 1(d) where the colors denote that several grains (separated by dark grain boundaries) have similar orientation. The microcracks in porous cordierite form at the domain level since the thermal mismatch stresses are generated at this microstructural scale. Finally, it is noted that the extrusion direction is within the image plane of Fig. 1(b) and (c), whereas, it is perpendicular to the image plane in Fig. 1(a) and (d). In Fig. 1(d), the grains, therefore, appear equiaxed but they appear elongated in Fig. 1(c). Since the crystallographic *c*-axis is the lowest thermal expansion direction [15], the overall thermal expansion is lower in the extrusion direction.

4. Mechanical and thermal property variations with porosity

4.1. Fracture toughness and strength

Fig. 2 indicates that the fracture toughness of porous cordierite specimens can change by a factor of over 20 in the investigated porosity range of 0–70%. For the cordierite DPF specimens (with porosity 50–70%), Fig. 2 shows that the double-torsion fracture toughness (K_{IC}) decreased by a factor of four (0.4–0.1 MPa$\sqrt{}$m) as porosity of the cordierite specimens increased. The following observations are made on the results in Fig. 2:

- The fracture toughness values of the cordierite specimens are most sensitive to the porosity of the specimen. Other factors, such as having a catalytic washcoating, service life or additional aging (Table 1), do not lead to a noticeable trend in fracture toughness

Please cite this article in press as: A. Shyam, et al., The effect of porosity and microcracking on the thermomechanical properties of cordierite, *J Eur Ceram Soc* (2015), http://dx.doi.org/10.1016/j.jeurceramsoc.2015.08.014