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ScienceDirect

Ceramics International 42 (2016) 3548–3555

CERAMICS INTERNATIONAL

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Mechanical and thermal properties of highly porous Al_2TiO_5–Mullite ceramics

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Received 16 September 2015; received in revised form 15 October 2015; accepted 1 November 2015
Available online 6 November 2015

Abstract

Al_2TiO_5–mullite porous ceramics were synthesized by direct foaming method using α-Al_2O_3, TiO_2 and SiO_2 as starting materials. The initial suspension for Al_2TiO_5 was prepared by adding TiO_2 suspension to the equimolar amount of partially hydrophobized colloidal Al_2O_3 suspension. A secondary suspension was prepared using following molar composition 3:2 Al_2O_3/SiO_2 and it was blended to the initial suspension in (0, 10, 20, 30 and 50) vol% to obtain the mullite phase in the sintered sample. Thermal expansion coefficient and thermal hysteresis curves of the porous ceramic samples with pure Al_2TiO_5 and of the Al_2TiO_5 ceramic with mullite addition were measured. Hertzian indentations are used to evaluate the damage behavior under constrained loading conditions. Mechanical behavior from indentation load–displacement curves is investigated.

Keywords: C. Mechanical properties; D. Al_2TiO_5; D. Mullite; Porous ceramics; Direct foaming; Thermal expansion co-efficient

1. Introduction

Al_2TiO_5 ceramics (AT) is known to be an excellent candidate material for refractory and engineering ceramics for high temperature applications because of its low thermal expansion coefficient ($\alpha_{20-1000}$ °C–1.5×10^{-6} K^{-1}), low thermal conductivity (0.9–1.5 Wm^{-1} K^{-1}), high melting point (1860 ± 10 °C) and low Young's modulus (10–20 GPa) [1]. Furthermore, its high thermal shock resistance, high refractoriness and good corrosion resistance are potentially advantageous for diesel particulate filters (DPF) and molten metal filtrations [2].

The material is characterized by prominent thermal expansion anisotropy along the crystallographic axes, resulting in distinct hysteresis loop and very low thermal expansion co-efficient [3]. This also induces the formation of microcracks which results in poor mechanical properties. Another disadvantage of AT ceramics is associated with its thermal instability, tending to decompose into α-Al_2O_3 and TiO_2-rutile within the temperature interval 800–1300 °C [4–6].

Earlier studies have shown that the thermodynamical stability can be improved by the addition of dopants such as MgO, Fe_2O_3 or TiO_2 which are isomorphous with the mineral pseudobrookite, thus form solid solutions with Al_2TiO_5 [7,8]. Another source of stabilization is the limitation of microcracking phenomenon by adding suitable second-phase materials like Al_2O_3, ZrO_2, mullite and kaolinite [9–11]. Mullite is a stable high-temperature phase and exhibits high deformation

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http://dx.doi.org/10.1016/j.ceramint.2015.11.002
0272-8842/

Table 1
Physical properties of Al_2TiO_5–Mullite porous ceramics in different stages of direct foaming process[20].

Sample name	Suspension characterization			Wet foam characterization					Sintered foam characterization	
	AT–Mullite volume ratio	Contact angle [θ]	Surface tension [Mn/m]	Air content [%]	Wet foam stability [%]	Adsorption free energy [J]	Laplace pressure [mPa]	Average bubble size [μm]	Pore size [μm]	Density [g/cm^3]
AT	100 : 0	45.99	56.13	76.42	80	2.7×10^{-13}	2.2333	50.13	51.78	1.71
ATM1	90 : 10	48.77	43.61	72.40	88	2.6×10^{-13}	1.6951	46.19	84.62	1.18
ATM2	80 : 20	49.92	37.12	73.61	90	2.4×10^{-13}	1.5683	45.55	97.05	1.11
ATM3	70 : 30	51.17	30.38	78.92	90	2.3×10^{-13}	1.5023	40.18	182.3	1.48
ATM5	50 : 50	55.23	23.56	82.99	92	2.2×10^{-13}	1.3038	36.18	410.71	1.34

resistance even at elevated temperatures. It also has properties like moderate thermal expansion coefficient (4.5×10^{-6} K^{-1}), low thermal conductivity (0.06 W cm^{-1} K^{-1}), low dielectric constant ($\epsilon \sim 7$), low fracture toughness ($\sim$2 MPa m$^{1/2}$) and reasonable mechanical strength (150–170 MPa) [12]. Mullite incorporation in the structure not only strengthens the material by reducing the grain growth but also improves the thermal stability at 1100 °C [13].

The mechanical behavior of porous ceramics is greatly influenced by their pore structure [14]. The introduced porosity affects and alters the mechanical properties, making it different from that of dense ceramics [15]. Therefore, mechanical measurement techniques commonly applied for dense ceramics might not be equally suitable for porous ceramics. Such porous ceramics ordinarily appear completely brittle in traditional strength tests. However, Hertzian indentation mechanics has been extensively used for the analysis and characterization of fracture and deformation properties of brittle ceramics including porous ceramics [16,17]. Hertzian fracture is commonly associated with energy dissipation by internal friction at sliding grains, platelet or whiskers, or other microstructural elements that bridge the crack wake [16,18,19].

In the present study, we report the evolution of mechanical and thermal properties of Al_2TiO_5–Mullite (ATM) porous ceramics with varying volume percentages of mullite, derived by direct foaming process. In direct foaming, air is incorporated to a concentrated colloidal suspension by mechanical frothing and open or closed cell structures are obtained after drying and sintering [20]. We found that the presence of mullite controls the exaggerated grain growth of Al_2TiO_5 phase, thereby improving the strength and minimizing thermal expansion.

2. Experimental

2.1. Suspension preparation

α-Al_2O_3 powder ($d_{50} \sim 4$ μm; KC, South Korea), TiO_2 powder ($d_{50} \sim 2$ μm, Junsei Chemicals Co. Ltd., Japan), and SiO_2 powder ($d_{50} \sim 3.5$ μm, Junsei Chemicals Co. Ltd., Japan) were added to de-ionized water, and the aqueous suspension was prepared separately. Homogenization and de-agglomeration of the suspension was carried out on a ball mill for at least 48 h at a rotation speed of 60 rpm, using polyethylene bottles, and zirconia balls (10 mm in diameter), with ball/powder ratio 2:1. After ball milling, propyl gallate (Fluka Analytical, Germany) as a surface modifier at 0.2 wt% amount was added to the Al_2O_3 suspension under mechanical stirring, to hydrophobize the surface of Al_2O_3 particles. The pH of the suspension was adjusted to 4.75, by adding 4 M NaOH and/or 10 N HCl (Yakuri Pure Chemicals, Japan) drop-wise. The solid content of the final aqueous suspension was set to 30 vol%, by adding the required amount of water. Then, the TiO_2 suspension, which was also homogenized and ball-milled, was added to the Al_2O_3 suspension in equimolar concentration [20].

For the mullite phase, Al_2O_3 suspension and aqueous suspension of SiO_2 powder, which was also homogenized and ball-milled in same condition, were mixed together in 3:2 Al_2O_3/SiO_2 mole ratio. This suspension of 10, 20, 30, and 50 vol% was added to the initial suspension containing equimolar concentration of Al_2O_3 and TiO_2, to form the mullite phase after sintering. Table 1 shows the volume ratio of Al_2TiO_5 and suspension added for the mullite phase in the final suspension.

2.2. Colloidal suspension and foam characterization

Surface tension and contact angle of final suspension were analyzed by the pendant drop method (KSV Instruments Ltd., Helsinki, Finland). The drop volume was fixed to a constant value within the range of 5–10 μl, for amphiphile containing suspension.

The energy of attachment or free energy gained (G) by the adsorption of a particle of radius (r) at the interface can thus be calculated using the following equation:

$$\Delta G = \pi r^2 \gamma_{\alpha\beta} \quad (1 - \cos\ \theta)^2 \qquad \text{where}, \theta < 90^\circ \tag{1}$$

where, γ is the surface tension of the suspension, and θ is the contact angle.

Foaming of the final suspension was carried out in room temperature, using a household hand mixer (150 W, Super Mix, France) at highest power, for 15 min. The mechanical