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Al_2TiO_5–mullite porous ceramics from particle stabilized wet foam

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Abstract

Aluminium titanate (Al_2TiO_5)–mullite porous ceramics were synthesized by a 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 an equimolar amount of partially hydrophobized colloidal Al_2O_3 suspension. A secondary suspension was prepared using molar composition 3:2 Al_2O_3/SiO_2, and blended to the initial suspension in (0, 10, 20, 30 and 50) vol%, to obtain the mullite phase in the sintered sample. The wet foam exhibits an air content of 80–92% and Laplace pressure from 1.30 to 2.23 mPa, which results in 68–83% foam stability. It also exhibits a much higher adsorption free energy of about 2.2×10^{-13} J to 2.7×10^{-13} J at the interface, which results in irreversible adsorption of particles at the air–water interface, leading to outstanding foam stability. The final suspension was foamed, and the wet foam was sintered at 1500 °C for 1 h. Phase identification was accomplished using X-ray diffraction, and microstructural analysis was performed by field emission scanning electron microscopy.

Keywords: D. Al_2TiO_5; D. Mullite; Porous ceramics; Direct foaming; Laplace pressure

1. Introduction

Al_2TiO_5 is one of the best ceramics, with excellent properties of low thermal expansion coefficient ($1.5 \times 10^{-6}\ K^{-1}$), low thermal conductivity (0.9–1.5 $W\ m^{-1}\ K^{-1}$), high temperature resistance (melting point 1860 ± 10 °C) [1], low Young's modulus, and good corrosion resistance [2,3]. These properties allow it to find application as a thermal barrier coating, offering corrosion resistance coating and anti-oxidation coating under high temperature [4], and as a refractory, structural, and insulating material, in the metallurgical industries and automotive industries [5–7]. However, practical applications of this material have been severely restricted, because of two major limitations: low mechanical strength, due to the microcracks induced by high anisotropy of the thermal expansion coefficients, which are -3.0, $+11.8$ and $+21.8 \times 10^{-6}$/K for its three crystallographic axes [8]; and thermal instability, in the temperature range between 750 and 1280 °C [8–11]. This decomposition occurs when the adjacent Al^{3+} (0.54 Å) and Ti^{4+} (0.67 Å) octahedra collapse, because the lattice site occupied by the Al^{3+} ion is too large. The thermal energy available from this collapse allows Al^{3+} to migrate from its position, and causes structural dissolution to rutile (TiO_2) and corundum (Al_2O_3). Following decomposition, the material exhibits neither a low thermal expansion coefficient, nor favourable thermal shock behaviour, rendering it useless for many practical applications [8].

Mullite, a second phase, has been used to reduce microcracking, as well as grain growth of the Al_2TiO_5 phase, and eventually to improve the mechanical behaviour in the temperature range 1300 and 1450 °C [8,11–15]. Huang et al., Yano et al., Morishima et al., and Kim et al. worked on the development of Al_2TiO_5–mullite composite to improve its mechanical strength.

Based on the excellent physicochemical properties of Al_2TiO_5, porous Al_2TiO_5 ceramic can be a promising candidate for use as a high temperature flue gas filtration supporter and substrate in catalytic converters for motor vehicles [16].

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Recently, porous Al_2TiO_5 has also been applied to third-generation diesel particulate filters (DPF) [17]. Authors, such as Hongzhi et al. [16] and Nishijima et al. [17], reported the synthesis of porous Al_2TiO_5 ceramic by a reactive sintering process, using corn starch and potato starch as pore modifying agents, respectively.

In this paper, porous Al_2TiO_5 was synthesized by a direct foaming method, using Al_2O_3 and TiO_2 as raw materials, Al_2O_3/SiO_2 in 3:2 mol ratio as modifying agents to impart mullite phase after sintering, and propyl gallate as a surface modifier. The direct foaming method is a popular technique for the manufacture of porous ceramics, because of its simplicity, low cost, and versatility. In this process, high volume stable wet foams are prepared by incorporating air into the colloidal suspension. In this paper, we characterized the wet foams, and investigated the bubble size, air content, Laplace pressure, adsorption free energy, and wet foam stability. Later, we dried and sintered the wet foam, to produce open or closed cell porous ceramics with excellent porosity and high mechanical strength.

2. Experimental

2.1. Materials

(i) α-Al_2O_3 powder (KC, South Korea), with average particle diameter, d_{50}, of 4 μm and density of 3.95 g/cm^3; (ii) TiO_2 powder (Junsei Chemicals Co. Ltd, Japan), with average particle diameter, d_{50}, of 2 μm and density of 4.23 g/cm^3; and (iii) SiO_2 powder (Junsei Chemicals Co. Ltd, Japan), with average particle diameter, d_{50}, of 3.5 μm and density of 2.65 g/cm^3 were used to prepare the suspension. The short chain carboxylic acid used for surface modification was Propyl gallate (Fluka Analytical, Germany). Further chemicals used for this study were 10 (M) HCl (Yakuri Pure Chemicals, Osaka, Japan) and 4 (M) NaOH solutions (Yakuri Pure Chemicals, Kyoto, Japan), for pH adjustments; and Double deionized water, for suspension preparation and volume adjustment.

2.2. Preparation of suspensions

2.2.1. Preparation of Al_2O_3–TiO_2 suspension

α-Al_2O_3 powder, TiO_2 powder, and SiO_2 powder 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 as a surface modifier at 0.2 wt% amount was added drop-wise to the Al_2O_3 suspension under mechanical stirring, to hydrophobize the surface of Al_2O_3 particles. The mixing speed was kept constant at 500 rpm, and the amphiphile was added as concentrate, no prior dissolution having been carried out. The pH of the suspension was adjusted to 4.75, by adding (4) M NaOH and/or (10) N HCl 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. Fig. 1 shows a schematic illustration of the experimental procedure.

2.2.2. Preparation of suspension for the mullite phase

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 vol% of Al_2TiO_5 and suspension added for the mullite phase in the final suspension.

2.3. Contact angle and surface tension

Surface tension and contact angle 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.

2.4. Foaming and foam characterization

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 frothing facilitates air incorporation throughout the whole volume of suspension. The air content was measured, by calculating the percentage of volume increase of the suspension after foaming.

$$\text{Air content} = \frac{\left(V_{\text{wet foam}} - V_{\text{suspension}}\right) \times 100}{V_{\text{wet foam}}} \quad (1)$$

where $V_{\text{wet foam}}$ indicates the wet foam volume after foaming, and $V_{\text{suspension}}$ indicates the volume of suspension before foaming.

The average wet bubble size was measured by analyzing optical microscope images using the software linear intercept (TU Darmstadt, Germany). The optical microscope (Somtech Vision, South Korea) in transmission mode was connected to a digital camera. For each sample, a minimum of 100 bubbles was evaluated.

The most critical issue in direct foaming methods is the approach used to stabilize the air bubbles incorporated into the suspension. In this experiment, propyl gallate (2 wt%) was used as a surface modifying agent, which imparts hydrophobicity to the particle surface, and thus improves foam stability. To investigate the wet foam stability, the wet foam samples were filled into cylindrical molds of constant volume, and left for 48 h. Foam stability was evaluated upon observing the percentage of volume loss of the foam.

$$\text{Wet foam stability} = \frac{V_{\text{Final}}}{V_{\text{Initial}}} \times 100 \quad (2)$$