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INFILTRATION KINETICS STUDY AND PROCESSING OF LAYERED GRADED Al_2O_3-ZrO_2 COMPOSITE

Dwi Asmi
Department of Physics, FMIPA - Lampung University
Jl. Sumantri Brojonegoro No. 1, Bandar Lampung 35145

ABSTRACT

INFILTRATION KINETICS STUDY AND PROCESSING OF LAYERED GRADED Al_2O_3-ZrO_2 COMPOSITE. Layered graded Al_2O_3-ZrO_2 composite was successfully synthesised through infiltration of porous Al_2O_3 preform with a solution containing $ZrOCl_2.8H_2O$. The infiltration rate of liquid into porous Al_2O_3 preform had also been investigated. It was found that the infiltration rate equation proposed by Washburn is most suitable for describing the effects of preform sintering temperature, viscosity and multiple infiltrations on the infiltration behaviour, whereas the influence of applied pressure is consistent with the model proposed by Darcy, where the applied pressure enhances the infiltration rate behaviour. Depth profilling of layered graded Al_2O_3-ZrO_2 composite was characterised by x-ray diffraction, the result shows that the concentration of ZrO_2 decreased with depth, while that of Al_2O_3 increased with depth.

Key words : Layered graded materials, Al_2O_3-ZrO_2 composite, infiltration technique

ABSTRAK

KAJIAN KINETIKA INFILTRASI DAN PEMROSESAN BAHAN KOMPOSIT LAPIS GRADUAL Al_2O_3-ZrO_2. Telah dibuat bahan komposit lapis gradual Al_2O_3-ZrO_2 dengan cara menginfiltrasikan bahan prabentuk Al_2O_3 berpori dalam larutan yang mengandung $ZrOCl_2.8H_2O$. Laju infiltrasi larutan ke dalam bahan prabentuk Al_2O_3 berpori juga diteliti. Hasil yang diperoleh menunjukkan bahwa persamaan laju infiltrasi yang diusulkan oleh Wasburn adalah yang paling sesuai untuk menjelaskan pengaruh suhu *sintering*, viskositas, dan infiltrasi berjenjang terhadap perilaku infiltrasi, sedangkan pengaruh tekanan yang diberikan lebih sesuai dengan model yang diusulkan Darcy, dimana tekanan yang diberikan meningkatkan laju infiltrasi. Profil kedalaman dari bahan komposit lapis gradual Al_2O_3-ZrO_2 dikarakterisasi dengan menggunakan difraksi sinar-x, hasilnya menunjukkan bahwa konsentrasi ZrO_2 turun terhadap kedalaman sedangkan konsentrasi Al_2O_3 naik terhadap kedalaman.

Kata kunci : Bahan lapis gradual, komposit Al_2O_3-ZrO_2. teknik infiltrasi

INTRODUCTION

The alumina-zirconia system has attracted attention due to its specific mechanical character, especially the presence of toughening behaviour [1]. Alumina is commonly used as a matrix in dispersed zirconia. The role of the dispersion of zirconia in the microstructure and the properties of alumina based ceramic materials is very important. The addition of ZrO_2 was effective in inhibiting grain growth of alumina despite its inability to produce a ceramic with satisfying density [2]. Addition of up to 5 vol % ZrO_2 controlled the grain growth of alumina from 6.9 μm in pure alumina to 2.82 μm [3]. Some research related to the processing of alumina-zirconia system has been conducted however there is not much information on the use of liquid infiltration method [4]. The use of liquid infiltration technique to produce layered graded Al_2O_3-ZrO_2 composite has not been well documented. The infiltration process to synthesize layered graded material is relative simple [5,6]. The method is to infiltrate porous preforms of the base material with appropriate bonding phase to specified depths in conjunction with pressureless sintering to fabricate dense layered graded materials. This technique has an advantage of producing cost-effective specimens with concentration gradients, contributing to new technologies for fabricating advanced materials.

In this paper, we presents results on the infiltration rate behaviour of liquid into porous Al_2O_3 preform, depth profiling and physical characteristics of infiltration-processed layered-graded composites based on the Al_2O_3-ZrO_2 system. The characteristics of liquid infiltration into Al_2O_3 preform and the physical properties of Al_2O_3-ZrO_2 composites are discussed in the light of phase relations.

EXPERIMENTAL METHOD

Al_2O_3 powder for fabrication of preforms was obtained by wet ball milling alumina (A-11, Japan) with 1 wt % polyvinil alcohol as antifloc culant in methanol for 10 h. The slurry was then dried in oven at 60 °C for 36 h then grounded and sieved. The porous alumina preforms were fabricated by uniaxial pressing of processed alumina powder at 60 MPa to yield a bar sample of dimensions (5 x 12 x 60) mm^3 and cylindrical pellet of dimensions 19 mm in diameter and 3 mm in height. Infiltration of the porous alumina preform was conducted at room temperature. The sample was immersed in a solution of zirconyl chloride octahydrate ($ZrOCl_2.8H_2O$) for 8 h. The infiltrated preform was then dried at room temperature for 24 h prior to heat-tretment in a high temperature furnace (Carbolite Model 1600) at 900 °C for 2 h followed by 1500 °C for 6 h, and then furnace cooled. Figure 1 shows the schematic flowchart of the *liquid-infiltration* method for fabrication of layered graded Al_2O_3-ZrO_2 composites.

Analysis of phases formed and their abundance was performed with a SHIMADZU XD 600 diffractometer with operating conditions used were CuKα radiation (λ= 1,54 Angstrom) produced at 30 kV and 30 mA over 2 theta range of 10 ° - 120 °, step size 0.05, scan mode continue and counting time 2 deg s/step. The apparent porosity and bulk density of the sample were measured by using Archimedes technique.

RESULTS AND DISCUSSION

Infiltration Rate Behaviour

Alumina preforms, with dimensions (5 x 12 x 60) mm^3, were pre-sintered at temperatures 1000, 1100 and 1200 °C for 3 h. The apparent porosity and bulk density of the preforms were measured by the Archimedes method. A summary of the apparent porosity and bulk density data is given in Table 1. As expected the apparent porosity decreased with an increase in temperature, whereas the bulk density increased with an increase in temperature. As can be seen from Table 1, the high volume fraction of porosity obtained for the alumina prefom pre-sintered at 1000 °C, resulted in a low density of 2.15 g/cm^3. However, attempts to conduct such an infiltration were not successful because the alumina preform cracked easily. Pre-sintering at 1100 and 1200 °C for 3h yielded suitable alumina preforms were strong enough to withstand subsequent processing steps.

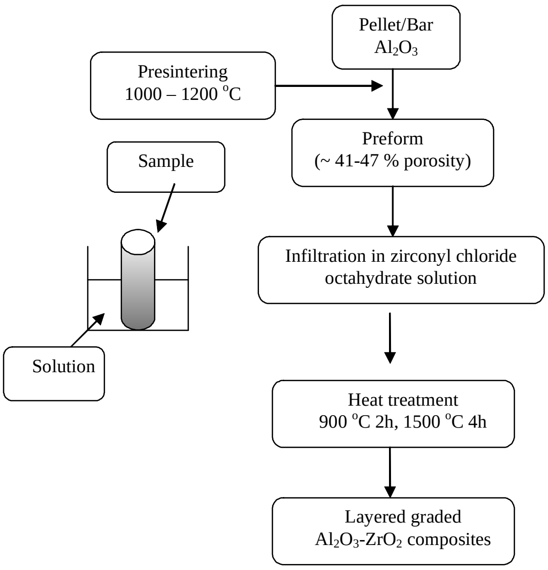

Figure 1. Schematic flowchart of the *liquid-infiltration* method.

Table 1. Apparent porosity and bulk density of the porous alumina preform pre-sintered at 1000, 1100, and 1200 °C.

Pre-sintering temperature (°C)	Apparent porosity (%)	Bulk density (g/cm^3)
1000	46.10 ± 1.50	2.15 ± 0.02
1100	45.11 ± 1.40	2.19 ± 0.03
1200	41.20 ± 1.40	2.35 ± 0.03

The alumina preforms sintered at 1100 °C and 1200 °C with dimensions of (5 x 12 x 60) mm^3 were used to study the infiltration rate behaviour of liquid into alumina preform because of their relatively high strength, which resulted in no cracks during infiltration in the solution of zirconyl chloride octahydrate. The height of liquid rise into alumina preform was recorded every 60 seconds. The influences of preform sintering temperature, liquid type, vacuum and non-vacuum and multiple infiltrations on the infiltration behaviour are described below.

Figure 2 shows the effect of Al_2O_3 preform pre-sintering temperature on the liquid infiltration rate behavior at room temperature. The result shows that the

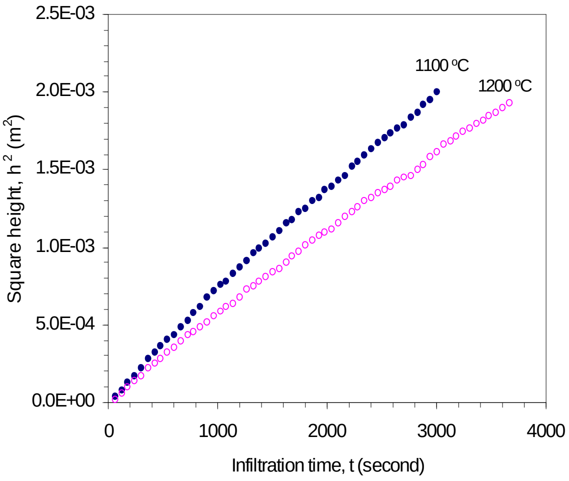

Figure 2. The effects of the preform pre-sintering temperature on the infiltration rate behaviour of liquid into Al_2O_3 preform.

rate of infiltration decreased with an increase in pre-sintering temperature. The decrease in infiltration rate can be attributed principally to the reduction in porosity with the increased Al_2O_3 preform pre-sintering temperature (see Table 1).

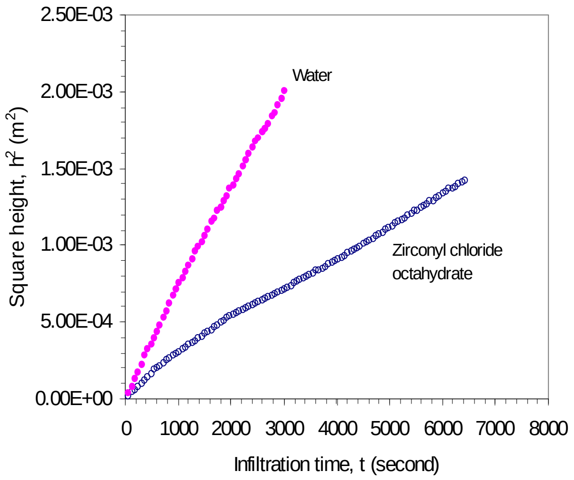

Figure 3. The effects of viscosity on the infiltration rate behaviour of liquid into Al_2O_3 preform.

The effect of viscosity solution on infiltration behaviour was studied using water and zirconyl chloride octahydrate solution. The Al_2O_3 preform sintered at 1100 ^{o}C was infiltrated with water under atmospheric pressure. The height square, h^2 as a function of infiltration time t is shown in Figure 3. The result shows that the infiltration rate of the zirconyl chloride octahydrate was lower than that of water into Al_2O_3 preform. This behaviour can be attributed to the viscosity of the zirconyl chloride octahydrate solution which is higher than water [7]. This result is also consistent with the Washburn model as would be expected because the rate of infiltration is inversely proportional to the viscosity of liquid. Washburn modeled [8] the rate of infiltration by assuming porous media to be a constant cross section capillary. In the case of good wetting, the liquid-vapor surface within a single pore channel will develop a concave curvature (meniscus) depending on the contact angle and the pore size. Across the meniscus a pressure difference called capillary pressure builds up, driving the liquid into the porous preform. Under this assumption, the flow in a capillary follows the Poiseuille's law [9], which states that

$$\frac{dV}{dt} = \frac{\pi\, r^4 dp}{8\mu\, h} \quad \text{......................................} \quad (1)$$

where dV is the volume of liquid which in time dt flows through a length, h, of a capillary having a radius, r. μ is the viscosity of liquid and dp is the total effective pressure acting to force the liquid along the capillary. So, by considering

$$dV = \pi\, r^2 dh \quad \text{......................................} \quad (2)$$

and the capillary pressure, dp by the following Young-Kelvin equation:

$$dp = \frac{2\gamma \cos\theta}{r} \quad \text{......................................} \quad (3)$$

where γ is the surface tension of liquid and θ is the contact angle between the liquid and the preform, equation (1) may be written in terms of a velocity:

$$\frac{dh}{dt} = \frac{r\gamma\ \cos\theta}{4\mu\, h} \quad \text{......................................} \quad (4)$$

Integration of equation (4) will give the expression for the relationship between the length of infiltration as a function of time:

$$h^2 = \left(\frac{r\gamma \cos\theta}{2\mu}\right)t \quad \text{......................................} \quad (5)$$

Dullien, El-Sayed and Batra [10] applied equation (5) (Washburn model) to a real porous media, in which the effective channel radius was defined by taking into consideration the varying segments of a real porous media. Einset [11] used the effective radius to replace the average pore radius in the Washburn equation, giving a successful explanation of the parabolic behaviour of the infiltrations of several organic solvents into compact formed from a mixture of carbon and silicon carbide powders.

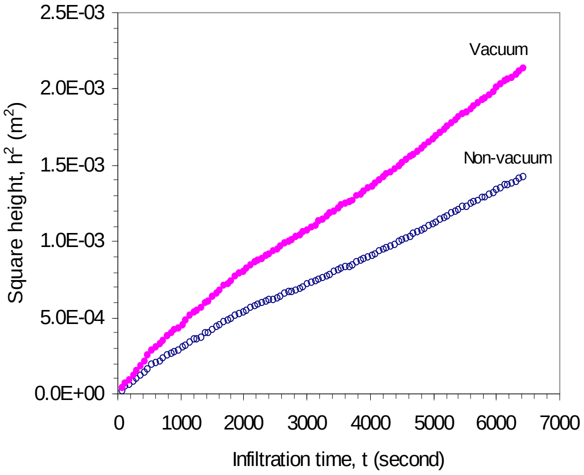

Figure 4. The effects of atmosphere condition on the infiltration rate behavior of liquids into Al_2O_3 preform.

A very important factor is the atmospheric condition under which the infiltration of the preform takes place. The experimental set-up considered is vacuum and non-vacuum. The effect of vacuum and non-vacuum on infiltration behaviour of water into alumina preform is shown in Figure 4. The alumina preform pre-sintered at 1100 ^{o}C was used in this experiment. The result shows that the rate of infiltration increased when experiment was conducted under vacuum. This result can be explained by the Darcy's law [12] in equation (6), where external pressure P_a play an important role in the process of the infiltration behaviour. In general increasing the applied pressure will enhance the infiltration rate.

$$h^2 = \left(\frac{2kP}{\eta}\right)t \quad \text{..} \quad (6)$$

where $P = P_c + P_a - P_i$ is the sum of the capillary pressure, the applied pressure, and the pressure of displaced gas, respectively. When the powder compact is modeled using identical spheres of diameter d with a relative density ρ and the porosity modeled with an equivalent capillary of radius r, the capillary pressure is given by [12]

$$P_c = \frac{2\gamma\cos\theta}{r} = \frac{6\gamma\cos\theta}{d(1-\rho)} \quad \text{..........................} \quad (7)$$

In equation 6, k is the permeability of porous media and is commonly expressed by the Kozeny-Carman equation [13]:

$$k = \frac{d^2(1-\rho)^3}{36T\rho^2} \quad \text{..} \quad (8)$$

where T is a constant that defines the shape and tortuosity of the pore channel.

Pre-sintered alumina preform at 1100 °C and zirconyl chloride octahydrate solution were used to study the effect of multiple infiltrations on infiltration behaviour. Experiment was conducted under vacuum. After the first infiltration, the infiltrated sample was dried at room temperature for 24 h followed by drying in oven at 70 °C for 12 h then fired in furnace at 900 °C for 30 minutes to decompose the infiltrant prior to continuing the next cycle. The results are shown in Figure 5. The rate of infiltration for the first cycle was higher than the second cycle. This indicates that after first infiltration the effective pore radius within the alumina preform decreased, therefore reduced the infiltration rate for the subsequent cycle.

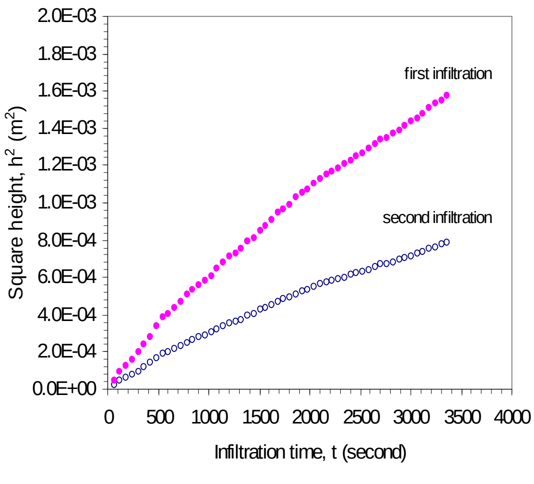

Figure 5. The effects of multiple infiltrations on the infiltration rate behavior of liquids into Al_2O_3 preform.

Depth Profiling of Layered Graded Al_2O_3-ZrO_2 Composites

Room temperature x-ray diffraction (XRD) was used to study the graded composition in layered graded Al_2O_3-ZrO_2 composites. Figure 6 shows the room-temperature XRD patterns for the alumina control sample and for the functionally-graded Al_2O_3-ZrO_2 composites obtained by the *gradual polishing* method. The sintered sample was polished with emery paper to depths of 0.0, 0.1, 0.15, 0.2 mm. α-Al_2O_3 was the only phase observed for the alumina control sample. The phases observed for the layered graded Al_2O_3-ZrO_2 sample were α-Al_2O_3 and ZrO_2 (monoclinic and tetragonal) phase. The peak intensities for alumina increased with depth, whereas the peaks for ZrO_2 decreased with depth. These changes in intensity with depth indicate that the layered graded Al_2O_3-ZrO_2 composite has a graded composition. The graded nature of these materials is clearly revealed and the concentration profile of ZrO_2 is in good agreement with other graded mullite/ZTA [14], AT/Al_2O_3 [15], Al_2O_3/CA_6 [16,17,18], and AT/ZTA [19] system.

CONCLUSIONS

A layered graded Al_2O_3-ZrO_2 composite has been successfully synthesized through infiltration of porous Al_2O_3 preform with a solution containing $ZrOCl_2.8H_2O$. The kinetics infiltration of liquids into porous Al_2O_3 preform was investigated. The variables that control the infiltration process such as preform pre-sintering temperature, viscosity, pressure, and multiple infiltrations were studied. It was found that, the samples pre-sintered at 1100 and 1200 °C for 2h resulted in most suitable Al_2O_3 preforms, were strong enough to withstand the subsequent processing steps.

The influence of Al_2O_3 preform pre-sintering temperature, viscosity and multiple infiltrations on the rate of infiltration was observed to obey the Washburn model, whereas the effect of applied pressure (vacuum and non-vacuum) was more consistent with the model proposed by Darcy where the external pressure enhances the infiltration rate. The depth profiling of layered graded Al_2O_3-ZrO_2 composites was clearly showed by x-ray diffraction results, where peak intensities for Al_2O_3 increased with depth, whereas the peaks for ZrO_2 decreased with depth.

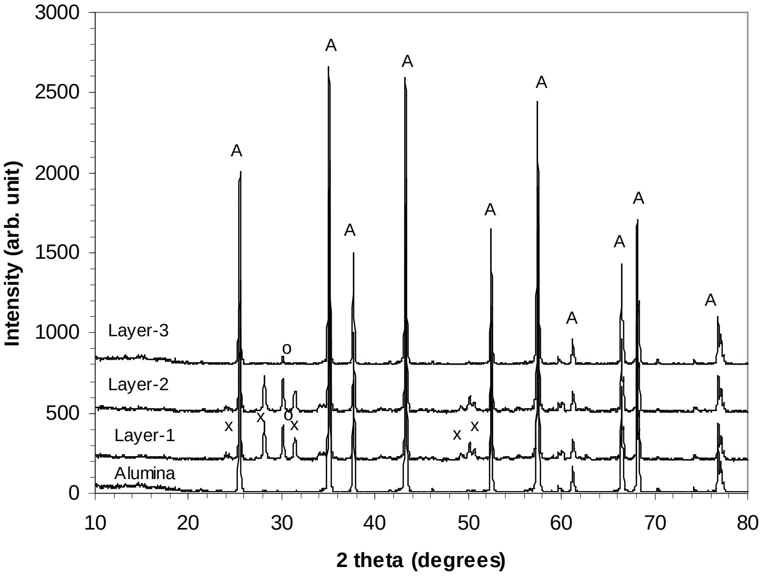

Figure 6. Room-temperature XRD patterns for layered graded Al_2O_3-ZrO_2 obtained by gradual polishing method at various depths (Layers 1 – 3) and alumina control. A: Al2O3, x = ZrO_2 (monoclinics), o = ZrO_2 (tetraghonal).

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