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Current-driven spontaneous infiltration of molten Al into a porous PSZ preform with a lamellar structure

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ABSTRACT

In this work we proposed a novel strategy for spontaneous infiltration of molten Al into a porous lamellar Y_2O_3 partially stabilized ZrO_2 (PSZ) architecture prepared by using an ice-templating method. The kinetics of liquid spreading and infiltration under the application of a 10 mA current was investigated. The electrochemically-induced improvement in wettability greatly facilitated the spontaneous infiltration.

1. Introduction

Ceramic–metal composites with a lamellar structure were demonstrated to possess some superior mechanical and physical performances to the counterparts with a uniform structure [1,2]. Hence, recent work has been focused on the development of lamellar ceramic–metal composites. Among various fabrication techniques, infiltration of a molten metal into a lamellar ceramic scaffold prepared by freeze casting has been proven to be a versatile processing approach [1,2].

As known, when melt-infiltration route is applied, the wettability of ceramic by molten metal plays a crucial role. Depending on their wettability, a molten metal can penetrate into the ceramic pore channels either spontaneously under a capillary force [3] or with the assistance of an external force [4]. For ZrO_2, because of its strong ionic bonding nature, it is not wetted by many metals such as Cu, Ag and Al [5]. For instance, the contact angle in the Al–PSZ system was approximately 145° at 1173 K [5]. In order to improve the wettability, in our previous work [6], we applied a minor direct current (DC > 1 mA) to the Al–PSZ couple during their wetting at 973 K and observed a noticeable effect. A transition from non-wetting ($\theta > 90°$) to good wetting ($\theta < 90°$) was readily obtained with increasing time. Based on this finding, in this work we attempted to take advantage of DC to achieve the spontaneous infiltration of molten Al into a lamellar ZrO_2 preform prepared by freeze casting so as to lay a foundation for the preparation of metal–ZrO_2 composites.

2. Experimental procedure

Commercial yttria partially stabilized zirconia (PSZ) powders ($D_{50} = 1.2$ μm) together with 3 wt% TiO_2 nanopowders (20–30 nm, ≥99.9%) were used as raw materials to prepare aqueous ceramic slurry. Lamellar PSZ scaffolds (ϕ 30 × 25 mm^2) with an initial solid loading of 30 vol% were prepared by ice-templating at −20 °C and then sintered at 1450 °C for 2 h [7]. The actual volume fraction of the ceramics in the lamellar scaffold after sintering reached 40 vol%.

The PSZ scaffold was then cut into ϕ 30 × 5 mm^2 and carefully polished. A pure Al (>99.99 wt%) ingot was cut into cubes with 4 × 4 × 4 mm^3 for the wetting test. Then they were ultrasonically cleaned in acetone and placed into a vacuum chamber. The chamber was evacuated to 3×10^{-4} Pa at room temperature and then heated at 20 °C/min to 900 °C. The DC-coupled wetting test was performed at this temperature and the details of the apparatus could be found in Ref. [6]. As the temperature was stabilized, the sharp-pointed upper graphite electrode was then lowered down to pierce into the molten Al drop. The DC power with a preset intensity (10 mA) and polarity (i.e., PSZ was connected to anode and Al to cathode) was switched on. Photographs were taken using a charge-coupled-device camera at a maximum rate of 2 frames

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per second. Contact angle and drop geometric parameters such as base diameter and height were measured from the captured drop profiles using a drop-shape analysis program. For comparison, the wetting of a dense PSZ substrate by molten Al was also performed under the same condition.

The microstructure in the sample after the wetting experiment was observed using a scanning electron microscope (SEM, VEGA 3 XMU, TESCAN, Czech). Phases were identified by X-ray microdiffraction (D8 Discover with GADDS, Bruker AXS, Germany) using a 300 μm beam diameter.

3. Results and discussion

Fig. 1 shows the backscattered electron images of ice-templated PSZ architecture. In the longitudinal section, the ceramic lamellae were predominantly oriented parallel to the freeze direction; whereas, in the transverse section, the lamellae orientation was basically random due to homogeneous nucleation of the ice crystals at the cold surface of the contact plane.

Fig. 2(a) shows the variation in contact angle (θ_d) and contact radius (R_d) for molten Al on the dense PSZ substrate with time under the application of a 10 mA DC. As indicated, θ_d decreased first rapidly and then gradually from 160° to 58°. At t = 700 s, θ_d reached 90°, implying a transition from non-wetting to wetting. During the isothermal dwelling, R_d always increased and the spreading rate, $U_d = \Delta R_d/\Delta t$ (calculated from the slopes of the fitting lines and represented by k_1, k_2 and k_3 in the figure), decreased from 5×10^{-3} mm/s in the initial stage ($0 < t < 250$ s) to 1.1×10^{-3} mm/s in the intermediate transition stage ($250 < t < 700$ s), and finally to 3.7×10^{-4} mm/s in the later long dwelling stage ($t > 700$ s). The spreading did not stop even after 4000 s, indicating the good wetting under the DC application. This significant effect was ascribed to the DC-induced formation of ZrO_{2-x} and metallic Zr [6]. As known, ZrO_2 doped with Y_2O_3 stabilizer is an oxygen ion conductor at elevated temperatures. The oxygen ions would move to the anode surface under the application of DC, leading to the formation of the substoichiometric ZrO_{2-x} phase and metallic Zr at the cathode surface [6]. The substoichiometric ZrO_{2-x} contains a large quantity of oxygen vacancies in its body, which favors the adsorption of a metal cluster at the solid–liquid interface [8]. Both the formation of the metal cluster and the adsorption of metallic Zr decreased the Al–PSZ interfacial free energy, thereby improving the wettability [6].

Fig. 2(b) shows the variation in apparent contact angle (θ_p) and contact radius (R_p) for molten Al on the porous PSZ substrate with time. At the beginning, θ_p and R_p remained almost constant, implying the pinning of the triple line due to either the presence of an oxide film on the Al surface or the large surface roughness or inferior ionic conductivity of the porous substrate. However, after an "incubation period" of 750 s, a noticeable decrease in θ_p was initiated, which changed from 163° to 20°, while R_p increased progressively until the melt separated from the upper graphite electrode. We noted that in this process the increase in the contact radius resulted only from liquid spreading while the decrease in the droplet height derived from both the liquid spreading and infiltration. Accordingly, the spreading rate, U_{spr}, can be still expressed as $U_{spr} = \Delta R_p/\Delta t$. At $750 < t < 1000$ s, the spreading rate was 5.5×10^{-3} mm/s, and thereafter it decreased almost linearly with the elapse of time with the values within 0.5×10^{-3}–2×10^{-3} mm/s (see Fig. 2(b_1)). In comparison with U_d, the slightly larger U_{spr} might be related to the lateral infiltration of the Al liquid to the front of the triple line as well as the improved conductivity of the porous substrate after the Al infiltration.

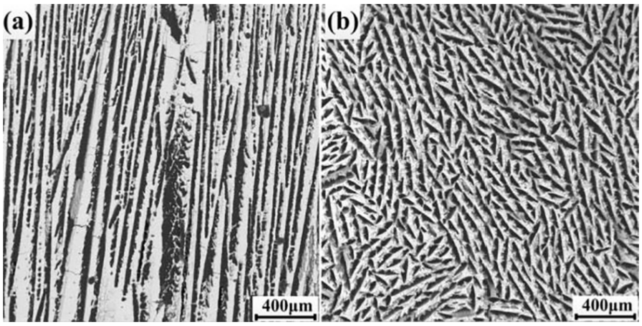

Fig. 1. Microstructures of the lamellar PSZ architecture in the (a) longitudinal and (b) transverse sections.

In order to investigate the melt infiltration kinetics, Fig. 2(c) shows the time variation in the infiltrated liquid volume, V_{inf}, calculated from the following equations:

$$V_{inf} = V_0 - V_p, \tag{1}$$

$$V_p = (\pi \times H_p \times (3R_p^2 + H_p^2))/6, \tag{2}$$

where V_0 is the initial drop volume, V_p is the dynamic drop volume, H_p is the dynamic drop height and R_p is the dynamic contact radius. The infiltration rate, $U_{inf} = \Delta V_{inf}/\Delta t$, was then calculated from the slope of the fitting line and broadly divided into three stages represented by k_4, k_5 and k_6, respectively, in Fig. 2(c). Note that the value of U_{inf} increased from 1.0×10^{-2} to 1.7×10^{-2} and further to 2.7×10^{-2} mm³/s during the wetting process, suggesting that a progressive improvement in the wettability facilitated the liquid infiltration. In order to further calculate the maximum infiltration depth, H_{inf}, of the infiltrated area, which was in the shape of a truncated cone (see Fig. 3(a_1)), we adopted a truncated cone model proposed by Voytovych et al. [9],

$$h(t) = 3V_{inf}/(\pi \times \alpha_{eff} \times (R_p^2 + R_0^2 + R_pR_0)), \tag{3}$$

where α_{eff} is the pore volume fraction in the PSZ architecture ($\alpha_{eff} = 0.6$), and R_0 and R_p are the initial and dynamic contact radii, respectively. The variation in H_{inf} with time was plotted together with V_{inf} in Fig. 2(c). The maximum H_{inf} was 2.6 mm after 3000 s, which is smaller than the measured value of 4 mm (see Fig. 3(a_1)). The deviation is presumed to result from the further penetration of the remnant Al into the PSZ architecture after the detachment of the liquid from the upper graphite electrode (in fact, all the liquid was found to infiltrate into the PSZ architecture after the sample was cooled down). Based on the above analysis, the entire spreading and infiltration process was schematically summarized in Fig. 2(d).

It might be worthwhile to further consider the effect of roughness for a porous substrate on the wetting behavior. To describe this effect, the Wenzel model [10] was adopted, in which an intimate contact is maintained for a liquid drop on a rough solid surface and the observed contact angle, θ_p, can be expressed as $\cos \theta_p = r \cos \theta_d$ ($r > 1$), where r is the ratio of the actual and projected surface areas of the substrate and θ_d is the equilibrium contact angle for the liquid on a smooth surface of the same chemical nature. According to this equation, the wettability should be deteriorated by the surface roughness when $\theta_d > 90°$ and it will be promoted when $\theta_d < 90°$. Therefore, we suggest that when molten Al began to spontaneously infiltrate into the channels between the PSZ lamellae under the capillary force, the local nano protrusions at the walls of the ceramic skeleton should promote the wettability and melt infiltration.

Fig. 3(a)–(d) show the longitudinal microstructure of the infiltrated sample after the wetting experiment. Clearly, molten Al spontaneously infiltrated into the porous PSZ architecture under DC, and the resultant composite inherited the lamellar structure. Due to limited drop volume, the PSZ sample consisted of a fully