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Thermal Conductivities of $LaPO_4/Al_2O_3$ Composites Fabricated by SPS

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Abstract. The thermal conductivities of the $LaPO_4/Al_2O_3$ composites that were fabricated by spark plasma sintering (SPS) were determined. The results revealed that their thermal conductivities displayed nearly a slow decrease with increasing temperature from 25°C to 800°C, having the classic 1/T dependence. In addition, the conductivities of the composites decrease monotonously with increasing the $LaPO_4$ content because of the lower thermal conductivity of $LaPO_4$. The calculated conductivities of the composites using Maxwell equation match well the experimental values at both the end members of $LaPO_4$ and Al_2O_3 being the continuous phase, but showing a little deviation at intermediate composition.

Introduction

Ceramics like Al_2O_3 have been attracting considerable attention and widely used in engineering components because of their high strength, high hardness, low density, and good chemical inertness. However, their brittleness has prevented their uses in structural applications to date. Lanthanum phosphate ($LaPO_4$), as a promising candidate for oxidation-resistant weak interfacial ceramics [1], has been extensively investigated due to its high temperature stability (melting point of 2075°C), relatively low hardness (5.5 GPa [1]), good chemical compatibility and weak bonding with Al_2O_3 [1-3]. It has been proved that $LaPO_4/Al_2O_3$ interface is weak enough to prevent crack growth by interfacial debonding and crack deflection [4]. Otherwise, monazite-type $LaPO_4$ has almost identical melting temperature and linear thermal expansion to those of Al_2O_3 [5-7]. Recently, it was found that $LaPO_4$ was stable with alumina at temperatures as high as 1750°C in air when the stoichiometric $LaPO_4$ is used [8], and $LaPO_4/Al_2O_3$ composites could be cut and drilled using conventional tungsten carbide metal-working tools instead of expensive diamond tools [4, 9, 10].

The potential application in high temperature field is one of the main objects for the $LaPO_4/Al_2O_3$ composites [8]. The thermal conductivities are undoubtedly important parameters for their practical applications. But as far as we know, there have been few reports on the thermal conductivities $LaPO_4/Al_2O_3$ composites [11]. In this paper, the thermal conductivities of the $LaPO_4/Al_2O_3$ composites that were densified by spark plasma sintering (SPS) were determined. The effects of $LaPO_4$ content on the thermal conductivities of the composites were also discussed.

Experimental

Monazite-type $LaPO_4$ powders were prepared according to Ref. [10]. Typically, La_2O_3 powders were slowly added to 85wt.% H_3PO_4, and a large amount of precipitates formed immediately at the reaction site. Subsequently, the synthesized solid was washed several times with de-ionized water until the PH value of the filtered water became close to 7. The La:P molar ratio was set to 1:1 in order to achieve $LaPO_4$ as the final product. After calcined at 800°C for 2h in air, the $LaPO_4$ powders with average particle size of 50 nm were used as starting materials. α-Al_2O_3 (Chongqing Tuoyuan Adv. Tech. Ltd., China) in this study was commercial powder with average particle size of 0.3 μm. The $LaPO_4$ powders were mixed with α-Al_2O_3 in the volume fraction of 0, 16.4, 34.4, 54.1, 75.6, 100vol.% by ball milling under ethyl alcohol with agate balls for 24 h and then dried using a rotary vaporizer, sieved through a 100-mesh screen. Sintering was carried out at 1350°C in vacuum (≤6Pa) with a holding time of 3 min, under a uniaxial pressure of 40 MPa in a cylindrical graphite mold of 20 mm diameter by SPS (Dr. Sinter 1020 SPS Sumitomo Coal Mining Co., Japan).

Thermal diffusivity was measured using the laser-flash method (NETZSCH LFA427, Germany). Heat capacity of the $LaPO_4/Al_2O_3$ composites were calculated from the literature values [12, 13] of $LaPO_4$ and Al_2O_3 by Neumann-Kopp rule. Thermal conductivity (k') of the sintered specimen was then determined using the formula:

$$k' = \alpha \bullet C_P \bullet \rho \quad (1)$$

where α is the thermal diffusivity, C_P is the specific heat, and ρ is the density. Since the sintered specimen still contained some micropores, thermal conductivity of the fully dense solid (k) was further normalized by the formula [14]:

$$\frac{k'}{k} = 1 - \frac{3}{2}\Phi \quad (2)$$

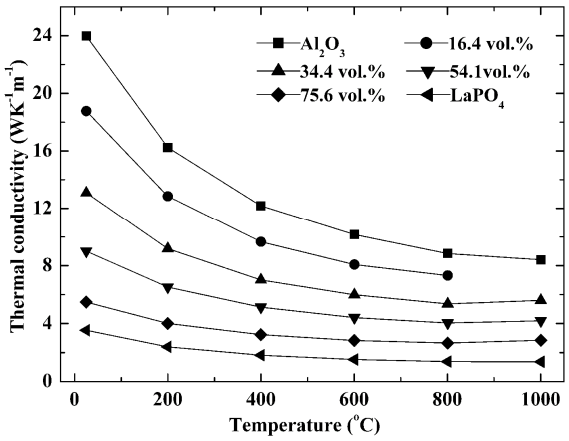

Fig.1 Thermal conductivities of the $LaPO_4/Al_2O_3$ composites versus tempersature

where Φ is the porosity, it was determined using the relation $\Phi = 1 - \rho/\rho_t$, where ρ_t is the theoretical density of the material, which were obtained from JCPDS. ρ is the density of the samples measured by the Archimedes method.

Results and Discussion

The relative densities of the sintered $LaPO_4/Al_2O_3$ samples are all higher than 97% [9], which may be attributed the characteristic of fully densifying at lower temperature and within very short time by SPS. There are no phases other than $LaPO_4$ and Al_2O_3 in all the samples, and the two phases are uniformly mixed [9]. The thermal conductivities of the composites are calculated according to Eq.(1) and shown in Fig.1. The deduced thermal conductivities are corrected for porosity according to Eq.(2), so they represent the thermal conductivity at zero porosity. It can be noticed from Fig.1 that the thermal conductivities of all the $LaPO_4/Al_2O_3$ composites decrease with increasing temperature from 25°C to 800°C, having the classic 1 / T dependence, with the exception of the thermal conductivities at 1000°C that show a slight increase. This is due to the contribution of radiation transport (internal radiation), which can be neglected at lower temperatures but becomes increasingly important at higher temperatures [15].

As we have known, the resulting thermal conductivities of the $LaPO_4/Al_2O_3$ composites depends on the amounts and arrangement of each phase presents as well as their individual conductivity [15]. Owing to the lower thermal conductivity of $LaPO_4$, the conductivities of the $LaPO_4/Al_2O_3$ composites decrease monotonously with increasing the $LaPO_4$ content (Fig. 2). In addition, the $LaPO_4/Al_2O_3$ composites can be thought to consist of a continuous major phase with a minor amount of a discontinuous second phase because of the uniform mixing between $LaPO_4$ and Al_2O_3 [9, 15]. Therefore, the thermal conductivities of the composites (k_m) can be calculated by Maxwell equation from the thermal conductivity of $LaPO_4$ and Al_2O_3 [15]:

$$k_m = k_1 \frac{1 + 2v_2(1 - k_1/k_2)/(2k_1/k_2 + 1)}{1 - v_2(1 - k_1/k_2)/(k_1/k_2 + 1)} \quad (3)$$

where v_1 and v_2 are the volume fraction of each component. k_1 is the thermal conductivity of the continuous phase, and k_2 the conductivity of the dispersed phase. when the $LaPO_4$ content is less than 50vol.%, Al_2O_3 is continuous with dispersed $LaPO_4$ grains within it. In samples containing smaller amounts of Al_2O_3, it is present as dispersed grains in a continuous $LaPO_4$ matrix. The calculated thermal conductivities of the $LaPO_4/Al_2O_3$ composites from Eq.(3) are shown in Fig.2. It can be observed that the calculated conductivities match well the experimental values at both the end members, but a little deviation at the intermediate composition. The reasons may be as follows: $LaPO_4$ and Al_2O_3 can be thought to be both continuous major phase at the end members. However, the continuous phase and the dispersed phase become indistinctness at the intermediate composition, so there present more deviations between the calculated conductivities and the experimental values.

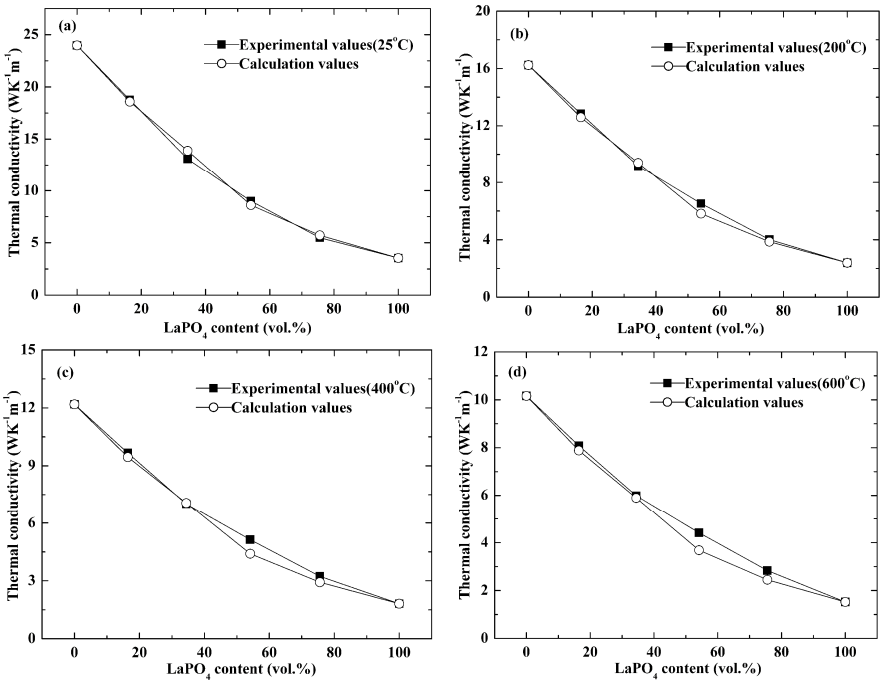

Fig. 2 Experimental thermal conductivities and calculated values of $LaPO_4/Al_2O_3$ composites at different temperature: (a) 25°C; (b) 200 °C; (c) 400°C; (d) 600°C

Summary

The thermal conductivities of the $LaPO_4/Al_2O_3$ composites with different $LaPO_4$ content, densified by SPS, have been determined. The results indicate that their thermal conductivities display nearly a slow decrease with increasing temperature from 25°C to 800°C, except the conductivities at 1000°C that present a slight increase. Otherwise, the conductivities of the $LaPO_4/Al_2O_3$ composites decrease monotonously with increasing the $LaPO_4$ content owing to the lower thermal conductivity of $LaPO_4$. The calculated conductivities of the composites using Maxwell equation match well the experimental values at both the end members of $LaPO_4$ and Al_2O_3, but showing a little deviation at intermediate composition.

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