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Influence of dopant concentration on the transparent and thermal properties of Nd_2O_3-doped alumina translucent ceramics

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Abstract: The transparent and thermal developments of high-purity Al_2O_3 doped with different levels of Nd_2O_3 were investigated. Dopant levels ranged from 500–1500 ppm (Nd/Al atomic ratio). The samples were characterized with X-ray diffraction (XRD), scanning electron microscopy (SEM), Raman spectroscopy, transmittance spectroscopy and specific heat measurement. Results revealed that with proper Nd doped, Nd^{3+} ions solid dissolved in Al_2O_3 lattice, resulting in small and uniform grain and high bonding vibration, which was beneficial to transparent and thermal properties. With 1000 ppm Nd doped, Al_2O_3 translucent ceramics showed a total transmittance of 89% and thermal conductivity of 41.7 W/m/K, indicating a potential application as substrate for effective heat dissipation and multi emitting surface in LEDs module.

Keywords: alumina translucent ceramics; neodymium dopant; thermal conductivity; substrate; LEDs; rare earths

For the last several decades, the effect of rare-earth additives on the sintering and microstructure development of alumina has received widespread attention[1–3]. It is reported that even at parts per-million (ppm) levels, rare-earth elements can greatly reduce the creep rate of alumina[4,5].

With respect to the effect of the rare-earth dopant on densification and grain growth, Fang et al.[6] have observed that for alumina doped with 1000 ppm yttrium the densification mechanism (at 1350 ºC) was consistent with control by grain-boundary diffusion, and densification kinetics were slowed by approximately one order of magnitude relative to undoped alumina. Wang et al.[7,8] studied the effect of Nd_2O_3 doping on the densification and abnormal grain behavior of high-purity alumina and reported that neodymium additives (100–1000 ppm) inhibited densification with a corresponding increase in the apparent activation energy. Furthermore, rare-earth doped transparent polycrystalline alumina ceramics have become a new focus recently as cheaper alternative to sapphire single crystals. Yang et al.[9] studied effect of La_2O_3 on the microstructure and transmittance of transparent alumina ceramics with total transmittance of 86% (1 mm thickness). Penilla et al.[10] prepared Tb^{3+} doped transparent alumina with fine-grained microstructure with 75% (1 mm thickness) of total transmission at a wavelength of 800 nm and with characteristic Tb^{3+} emission. Bodišová et al.[11] prepared transparent/translucent rare earth (Eu, Er, Nd, 2000 ppm) doped alumina with photo luminescent properties.

Although Bodišová et al. in their previous work[11] have suggested that 2000 ppm Nd_2O_3 doped Al_2O_3 ceramics have an in-line transmittance of about 10% (0.8 mm thickness) and with characteristic Nd^{3+} emission, the influence of dopant concentration on the transparent behavior has not been systematically studied. In addition, because of its high thermal conductivity, great strength and chemical durability[12], translucent poly crystalline alumina is a proper candidate of substrate for effective heat dissipation and multi emitting surface in LEDs module[13,14]. However, no research of dopant was processed on the thermal property of alumina transparent ceramics. Therefore, the object of the present study was to address these issues, with regard to neodymium doping.

1 Experimental

1.1 Preparation of Nd_2O_3-doped alumina translucent ceramics

High-purity powders of Al_2O_3 (99.95%, 0.25–0.45 μm, Alfa Aesar) and Nd_2O_3 (99.99%, 40 nm, Aladdin) were used as starting materials. The dopant concentrations were 0, 500, 1000, 1500 ppm (Nd/Al atomic ratio), respectively. And 500 ppm MgO (99.99%, 50 nm, Aladdin)

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was used as sintering aid. 40 g Al_2O_3 and corresponding contents of Nd_2O_3 and MgO were weighed and mixed by ball-milling in anhydrous alcohol for 12 h. The mixtures were dried in an oven at 70 ºC for 4 h, sieved under 200 meshes sieve, dry-pressed under 100 MPa into Φ20 mm discs and finally cold-isostatically pressed under 250 MPa. The Nd_2O_3-doped alumina translucent ceramics were prepared at 1770 ºC for 5 h under vacuum and then annealed at 1450 ºC for 20 h in air.

1.2 Characterization of Nd_2O_3-doped alumina translucent ceramics

The total transmission spectra of optically polished samples with the thickness of 2 mm were recorded between 300 to 800 nm using a spectrophotometer (UV 3600, SHIMADZU, Japan). The specific heat measurements of samples were obtained on a laser-flash apparatus (Flashline 3000 K2, Anter Corporation, American) at room temperature. Thermal conductivity (λ) was calculated from bulk density (ρ), heat capacity (C_p) and thermal diffusivity (α) according to the following equation: $\lambda=\rho\cdot C_p\cdot\alpha$. And the density of samples was obtained by draining water law of Archimedes.

The phase identification of samples was carried out by X-ray diffraction (XRD, D8 ADVANCE, Bruker, Germany). The diffraction data were collected within 10º–90º 2θ range and a scanning step of 5 (º)/min were used. The fracture surfaces of samples were investigated by scanning electron microscopy (SEM, SU 8010, HITICHI, Japan). The Raman spectra of samples were recorded from 200 to 1000 cm^{-1} in a micro-Raman spectrometer (inVia, Renishaw, UK).

2 Results and discussion

2.1 XRD analysis

Fig. 1 shows the XRD patterns of 0, 500, 1000 and 1500 ppm $Nd:Al_2O_3$ translucent ceramics. The results reveal that all the diffraction patterns of the samples match well with trigonal α-Al_2O_3 phase (JCPDS No. 10-0173), and no other impurity peaks are detected, independent of different Nd dopants.

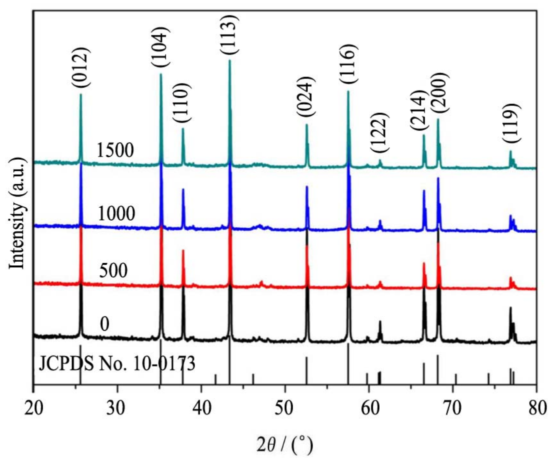

Fig. 1 XRD patterns of 0, 500, 1000 and 1500 ppm $Nd:Al_2O_3$ translucent ceramics

However, as shown in Fig. 2, the position of the diffraction peaks moves towards smaller angles with the Nd concentration increasing from 0 to 1000 ppm, indicating the expansion of the host lattice. Meanwhile, based on the least square fitting of the diffraction peaks[15], the lattice parameters of the samples are calculated and found to increase gradually with the increasing Nd concentration, as shown in Table 1, which results from the substitution of Al^{3+} ($R_{Al}{}^{3+}$=0.051 nm) ions with the larger Nd^{3+} ($R_{Nd}{}^{3+}$=0.098 nm) ions[7,8]. The expansion of the host lattice is expected to play an important role in the variation of the transparent and thermal properties.

2.2 Transparent properties

Fig. 3 shows the optical total transmittance of 0, 50, 100 and 150 ppm $Nd:Al_2O_3$ translucent ceramics (2 mm in thickness). It can be seen clearly that compared to undoped samples, samples with 500, 1000 and 1500 ppm Nd doped have a higher transmittance of 6%, 15% and 3%, respectively. The results indicate that the transmittance of Al_2O_3 ceramics can be improved by Nd doping, and the optimal dopant concentration is 1000 ppm. The differences in transmittance resulting in different Nd dopant concentrations are evident also from the macro photographs shown in Fig. 4.

2.3 Thermal properties

Fig. 5 shows the density and thermal conductivity of 0,

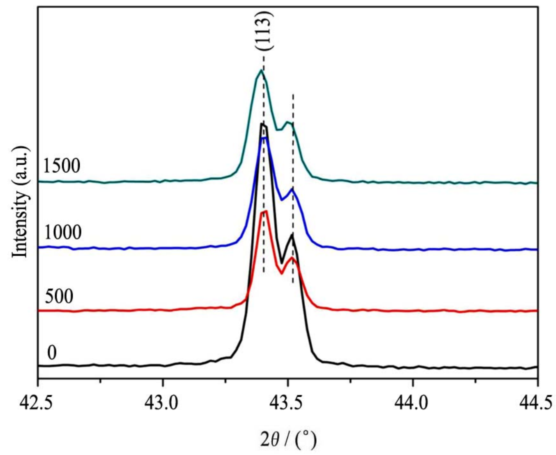

Fig. 2 Enlarged XRD patterns of (113) of 0, 500, 1000 and 1500 ppm $Nd:Al_2O_3$ translucent ceramics

Table 1 Lattice parameters of 0, 500, 1000 and 1500 ppm $Nd:Al_2O_3$ translucent ceramics

Concentration of Nd/ppm	Lattice constant		
	a/nm	*c*/nm	*V*/nm^3
0	0.476507	1.300054	0.25564
500	0.476424	1.300205	0.25558
1000	0.476444	1.300991	0.25576
1500	0.476137	1.301949	0.25562