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Feature article

Optical, mechanical and fractographic response of transparent alumina ceramics on erbium doping

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ABSTRACT

Alumina ceramics found their utilisation in many applications which can be further extend by attaining functional properties; in our case the transparency obtained through precise processing and photoluminescence due to erbium (Er) doping. In order to examine the optical, mechanical and fractographic response of transparent alumina on Er doping, slip casted samples containing 0–0.15 at.% of erbium nano-oxide were pre-sintered by two-step sintering regime and then hot isostatically pressed. Prepared samples exhibited fully dense submicron microstructure and corresponding high transparency (RIT up to 60%). Positive influence of doping on the Vickers hardness resulted in values up to 27 GPa (at 10 N load). Moreover, the comparison of the Vickers hardness determined at different loadings with literature data showed that the Er doped alumina is one of the hardest material in this category. The samples were characterised also in terms of fracture toughness and fractographic behaviour.

1. Introduction

Polycrystalline alumina is the most widely used structural ceramics in many common applications. When the alumina has extremely fine microstructure (low grain size) without residual porosity (final density ≥ 99.95%) then it becomes transparent [1]. Transparent polycrystalline alumina has a great technological potential for highly demanding applications which take advantage of its superior mechanical properties like high hardness, wear resistance, and high strength, in addition to its optical performance in the infrared and visible domain [2]. Moreover, in relation to the post-machining costs and inferior mechanical properties of sapphire (single crystal of alumina) the fine polycrystalline alumina exhibits economic and ecological benefits [3]. Krell et al. suggested application potential of this material in several directions: (i) lighting, (ii) optical components for different wavelengths, (iii) infrared emitters, (iv) orthodontic appliances and (v) ceramic armours [1,4].

To ensure transparency of the alumina few processing conditions must be fulfilled. The alumina powder should be extremely pure (99.99%) with nearly nanometre particle size and the grain growth during sintering has to be limited. The authors mainly utilise advantage of short sintering times using spark plasma sintering (SPS) [5–7]. However, to ensure other optical performances, i.e. photoluminescence, the doping with e.g. rare earth (RE) elements is needed [8–10]. By doping with the aim to obtain another functional property, it must be taken into account that the addition of sufficient amount of dopant can lead to the transparency deterioration. On the other hand, through the doping the alumina obtains interesting optical properties and pinning effect of dopant allows keeping low grain size. As doping elements for microstructure optimisation there are commonly used metals as chromium [11], titanium [12], magnesium [12–14], zirconium [15], or rare earth metals as yttrium [13], lanthanum [13]. Less common is doping with terbium [8], cerium [16], europium [9] or erbium [17,18].

Generally, mechanical properties and fractographic analysis of the transparent and/or photoluminiscent alumina is not well described in literature; only few publications on this topic can be found in the relevant literature. Krell et al. [19] doped commercial corundum powder with 0.03 wt.% MgO and 0.2 wt.% ZrO_2. In this transparent material no evidence of spinel phase and hardness 20–21 GPa (HV10) were obtained. The same combination of dopants used Braun et al. [20] to prepare transparent polycrystalline alumina with sub-micrometre microstructure. They studied the influence of different MgO and ZrO_2 dopant levels on the densification, microstructure development and in-line transmittance values. Nevertheless, authors provided information about hardness measurement valid only for undoped transparent alumina samples.

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The reported hardness ~11.6 GPa was relatively low probably due to higher value of grain size. In case of alumina doping with the rare earth elements Biswas et al. [3] reported hardness 21.4 GPa (HV5) of transparent sub-micrometre alumina doped with 0.1 wt.% lanthanum oxide.

Due to the lack of literature data this work is focused on investigation of optical, mechanical and fracture properties of transparent and/or photoluminescent alumina doped with various amount of erbia. Hardness measurement of alumina samples is extended to various load forces in order to enable better discussion with literature. Both hardness and fracture toughness are related to the observed microstructure. To the best of our knowledge, no similar study was carried out on this type of transparent and photoluminiscent material before.

2. Experimental

The commercial alumina powder (TM-DAR, Taimei Chemicals Co., Japan) with average particle size of 150 nm and commercial erbia powder (GNM, Getnanomaterials, USA) with average particle size of 20–30 nm were used for preparation of stable suspensions. Suspensions contain 45 vol.% of alumina, certain amount of erbia dopant (0, 0.10, 0.11, 0.125 and 0.15 at.%), electrostatic stabilizer (Darvan CN, Vanderbilt Minerals, USA) and deionized water. To remove aggregates the erbia powder was centrifugated at 800 rpm for 2 min before adding into suspension. Suspensions were homogenized for 24 h in a plastic bottles filled by alumina milling balls with diameter 4 mm. The balls to powder milling ratio of 2:1 was used.

The homogenous suspensions were casted into PVC dishes. Erbia doped alumina discs with diameter approximately 50 mm were first dried at room temperature for three days and then at 80 °C for 5 h. The discs were pressureless pre-sintered up to closed porosity stage (density range 95–96%) [18,21] using two-step sintering (TSS) in ambient atmosphere. The first step of TSS was realized at 1440 °C without dwell time followed by the cooling (20 °C/min) to the temperature 1280 °C with dwell time 10 h. The amount of open pores was checked by means of Archimedes method (EN 623-2). The two-step pre-sintered samples were consequently hot isostatically pressed under 200 MPa pressure of argon atmosphere at 1280 °C to completely eliminate residual closed porosity.

Sintered discs were grinded and polished up to 1 μm surface roughness, final thickness of the discs was approximately 0.8 mm. The real in-line transmittance (RIT) of the samples was measured with a non-polarized He-Ne laser (λ = 632.8 nm) at 860 mm sample to detector distance and 0.5° opening angle. The samples were thermally etched and investigated using a Lyra 3 microscope (Tescan, Czech Republic) and Titan Themis 60-300 (FEI, Czech Republic) equipped by Quantax EDX (Bruker Nano, Germany). During this investigation at least 5 micrographs of microstructure were obtained. The mean grain size (MGS) was determined on each micrograph using linear intercept method. The final average grain size value was multiplied by correction factor 1.56 [22].

Hardness was measured using an instrumented hardness tester (Z2.5, Zwick/Roel, Germany) at loading from 1 to 100 N. The number of measurements for each sample was set to 15. The fracture toughness was calculated from the length of the cracks originating from the corners of the indentation using equation [23]:

$$K_c = \propto \sqrt{\frac{E}{H}} \frac{P}{c^{3/2}}, \quad (1)$$

where α is an empirically determined constant (0.016), E is Young's modulus, H is the hardness, P is the applied load and c is the length of the crack grown from indent corner. Length of indent diagonals and cracks were measured using confocal microscope LEXT OLS 3100 (Olympus, Japan).

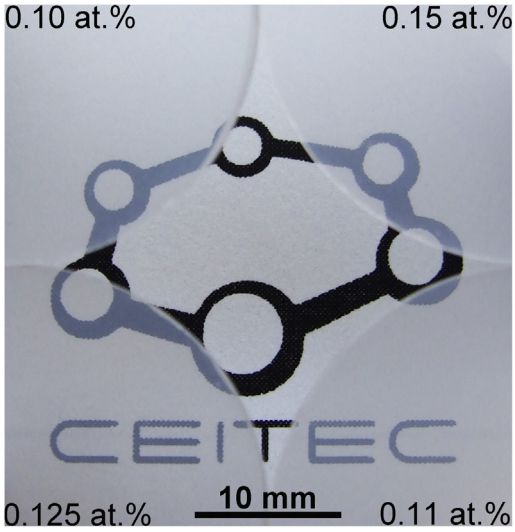

Fig. 1. Transparent alumina discs doped by different Er^{3+} concentration at 5 mm distance over the printed paper.

Table 1
Summary of microstructural and optical properties of undoped and Er^{3+}-doped alumina.

Concentration of Er^{3+} (at.%)	Density (%)	MGS (μm)	RIT (%)	RIT_r[a] (%)
undoped	≥99.95	0.50	60	64
0.10		0.35	56	59
0.11		0.34	–	–
0.125		0.33	53	55
0.15		0.35	53	55

[a] RIT_r = RIT recalculated to 1 mm thickness and related to the maximum theoretical transparency value for alumina (86% at 840 nm).

3. Results and discussion

Processing of the RE-doped transparent alumina was described in our previous work [18]. In the present work, one undoped and four Er^{3+}-doped transparent alumina discs were prepared. The photograph of transparent alumina discs doped by Er^{3+} in concentrations of 0.10, 0.11, 0.125 and 0.15 at.% is shown in Fig. 1. The samples were placed 5 mm above the printed paper. It is evident that transparency is similar for all samples. The real in-line transmittance of doped samples was in range 53–56% whereas the real in-line transmittance of undoped alumina was slightly higher (60%). Summary of RIT values is given in Table 1. The obtained RIT values were also recalculated to 1 mm thickness and related to the maximum theoretical transparency value for alumina (86% at 840 nm) for easier comparison with literature data [24]. The RIT of Er^{3+}-doped alumina in the range of 53–59% was already established in our previous work [18]. Here it was compared with RIT of undoped alumina prepared by the same way. The lower RIT value of doped alumina was expected because the relative high concentration of the dopant probably exceeded the amount being able to the segregate at the grain boundaries (e.g. lanthanum grain boundary solubility limit in alumina was observed at ~200 ppm [25]). Therefore, the higher content of the dopant then most likely results in decrease of optical transmittance.

Only few works reported optical transparency of photo- or thermoluminiscent alumina doped by Tb [8], Eu [9] and Ce [16]. In those cases the obtained transparencies were low, suffering from