

Fabrication of transparent $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics via vacuum sintering

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ABSTRACT

$\text{Y}_2\text{Hf}_2\text{O}_7$ transparent ceramics were successfully fabricated via vacuum sintering at 1900 °C for 6–12 h by the solid-state reaction using the corresponding nitrate and oxide powders without additives. The fabricated transparent ceramics showed a well-defined microstructure and no pores or other defects were observed. The linear optical transmittance reached to 60% in the visible wavelength region, which is about 76% of the corresponding single crystal. The refractive index of the $\text{Y}_2\text{Hf}_2\text{O}_7$ transparent ceramics was above 2.01 in the region between 400 nm and 800 nm.

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1. Introduction

Transparent ceramics have attracted lots of interests recently for their excellent properties in optical engineering application fields. Many transparent ceramics, such as Al_2O_3 [1], Y_2O_3 [2], (Y, Gd) $_2\text{O}_3$ [3], YAG ($\text{Y}_3\text{Al}_5\text{O}_{12}$) [4,5], Mg–Al spinel (MgAl_2O_4) [6], and ALON ($9\text{Al}_2\text{O}_3 - 5\text{AlN}$) [7], have been developed for many applications in face shields, windshields, special window, scintillators and solid state lasers. As a promising candidate for scintillators and high refractive index materials, the yttrium hafnate ($\text{Y}_2\text{Hf}_2\text{O}_7$) has attracted our interests a lot.

Ternary oxides with the formula of $\text{A}_2\text{B}_2\text{O}_7$, where A is a 3+ ion and B a 4+ ion, have been of great interest for many researchers over a number of years. These compounds often adopt a pyrochlore structure (space group $Fd3m$) and in some case a defective fluorite structure (space group $Fm3m$), which accommodates a wide range of solid solutions between BO_2 and A_2O_3 [8]. Because of the extensive compositional range, there are a variety of actual and potential applications for these materials, examples including catalysts [9,10], thermal barrier coatings [11], solid electrolytes [12,13], nuclear waste forms [14], and host materials for luminescence centers [15,16]. Additionally, it is also possible to fabricate transparent ceramics due to its cubic crystal structure without birefringence for $\text{A}_2\text{B}_2\text{O}_7$ materials. When A = Y and B = Hf, the $\text{Y}_2\text{Hf}_2\text{O}_7$ shows high density and high refractive index. If high transparency can be acquired using ceramic processing techniques, $\text{Y}_2\text{Hf}_2\text{O}_7$ transparent ceramics will present the potential applications in high-energy nuclear medical fields such as computer tomography (CT) and positron emission tomography (PET). However, to the best of our knowledge, few reports about the fabrication of $\text{Y}_2\text{Hf}_2\text{O}_7$ transpar-

ent ceramics have been found in the literature due to the high melting point. In 2009, $\text{Y}_2\text{Hf}_2\text{O}_7$ powders were synthesized by oxalate co-precipitation method and the morphologies were modified by surfactant PEG6000 [17], but the corresponding ceramics (in the follow-up work) were hardly transparent. Then in 2011, $\text{Y}_2\text{Hf}_2\text{O}_7$ transparent ceramics were obtained by using combustion synthesized powders and vacuum sintering, but the in-line transmittance was about 50% in the visible spectral region [18].

In this paper, polycrystalline, transparent $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics were successfully fabricated by a traditional solid-state reaction method via vacuum sintering. The phase composition, SEM morphologies, the linear optical transmittance and the refractive index of the as-prepared $\text{Y}_2\text{Hf}_2\text{O}_7$ transparent ceramics were investigated. The theoretic transmittance in the visible spectral region was calculated.

2. Experimental procedure

The $\text{Y}_2\text{Hf}_2\text{O}_7$ powders were prepared using yttrium nitrate ($\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, 99.9%), hafnium (IV) dioxide (HfO_2 , 99.0%) as the starting materials in the present work. Firstly, stoichiometric amounts of $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ and HfO_2 were weighed out and mixed by a planetary ball mill with ZrO_2 balls in anhydrous alcohol for 12 h. Subsequently, the powder mixtures were dried and calcined in alumina crucibles at 800 °C for 2 h with the heating rate 2.0 °C/min in muffle furnace to decompose $\text{Y}(\text{NO}_3)_3$ into Y_2O_3 . The calcined mixtures were ball milled again, then dried, sieved, and finally dry pressed and cold isostatic pressed at 200 MPa into $\Phi 20$ mm disks. The compacted green bodies were sintered at 1900 °C for 6–12 h with the vacuum of 2.0×10^{-3} Pa.

The microstructures of the green body and the sintered $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics were observed by scanning electron microscopy (SEM, JSM-6300, JEOL, Japan). The phase of product was identified by

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X-ray diffractometer (XRD, Model D/MAX-2550V, Rigaku Industrial Corporation, Japan) with $\text{CuK}\alpha$ radiation ($\lambda = 1.54178 \text{ nm}$). The mirror-polished specimens on both surfaces were used to measure the optical transmittance on a UV–VIS spectrometer (UV-2501PC, SHIMADZU, Kyoto, Japan). Then, they were thermally etched at 1600°C for about 4 h to observe the surface morphologies. The refractive index was measured by spectroscopic ellipsometry (UVI-SEL/460-VIS-AGAS, JOBIN YVON, France) in the wavelength region between 400 nm and 800 nm.

3. Results and discussion

Fig. 1 shows SEM image of fracture surface of the $\text{Y}_2\text{Hf}_2\text{O}_7$ green body. It can be seen that the starting powders are uniform in size. The average particle size is about $0.1\text{--}0.3 \mu\text{m}$, which is beneficial to fabrication of transparent ceramics. The green body is densified well and no big holes exist. Since the densification of the ceramics is directly affected by the corresponding green body, the uniform distribution of particle size and relatively small size of pores of the compacted powder are expected for the fabrication of high dense ceramics.

The XRD patterns of the as-prepared $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics vacuum sintered at 1900°C for 12 h are shown in Fig. 2. Because the JCPDS data files corresponding to $\text{Y}_2\text{Hf}_2\text{O}_7$ are still not available, the standard XRD pattern of $\text{Zr}_2\text{Gd}_2\text{O}_7$ (JCPDS 80-0471) which belongs to the same crystal structure with $\text{Y}_2\text{Hf}_2\text{O}_7$ (space group: $Fm\bar{3}m$ (225)) is chosen as a reference. It can be seen that all the diffraction lines can be indexed to the inserted reference patterns of $\text{Zr}_2\text{Gd}_2\text{O}_7$, and no impurity phase was found. Four main peaks (111), (200), (220) and (311) are characteristic of fluorite structure [19]. The SEM images of the fracture surface of $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics sintered at different temperatures for 12 h are shown in Fig. 3. The fracture mode of the ceramics sintered at 1800°C for 12 h is mainly intergranular fracture, and many micropores exist on the fracture surface and grain boundary (Fig. 3a). When the sintering temperature rises to 1900°C , the transgranular fracture occurs, the grain boundary is clean and nearly no residual pores can be observed. From the comparison, we can conclude that the sintering temper-

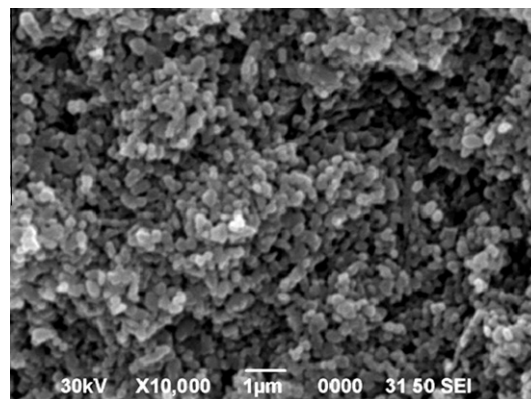


Fig. 1. SEM image of the $\text{Y}_2\text{Hf}_2\text{O}_7$ green body.

ature is important for the densification of the $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics, and only when the sintering temperature is high enough, the sintering driving force can be achieved to eliminate the residual micropores completely though the dwell time.

Fig. 4 shows the SEM images of the polished surface of the specimen sintered at 1900°C for 12 h after thermal etching at 1600°C for 4 h. It shows that the $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics consists of a well-defined microstructure and few micropores are observed. The grain size ranges from about 10 to $25 \mu\text{m}$. In addition, some relatively small grains of about $2\text{--}3 \mu\text{m}$ still exist (Fig. 4b), indicating that the sintering and densification of ceramics is actually the process that the big grains grow and the small grains dissolve.

Fig. 5 shows the photographs of the mirror-polished $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics sintered at 1900°C for 6 h (on the left) and 12 h (on the right) with the thickness of about 1 mm. Both ceramics exhibited high optical transmittance. However, the optical quality of the right sample is much better than that of the left due to the longer dwell time.

Fig. 6 presents the linear optical transmittance curves of $\text{Y}_2\text{Hf}_2\text{O}_7$ transparent ceramics sintered at 1900°C for different holding time.

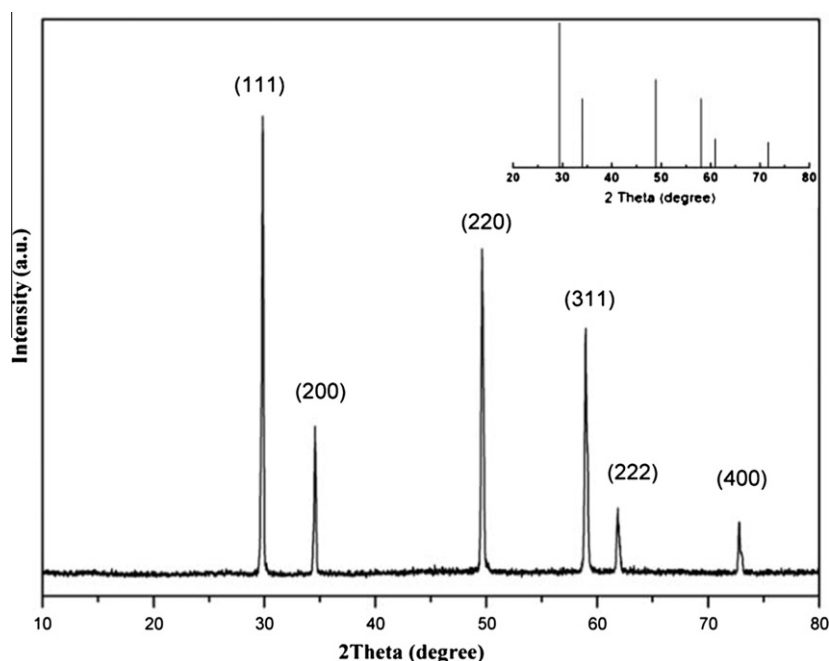


Fig. 2. The XRD patterns of the $\text{Y}_2\text{Hf}_2\text{O}_7$ ceramics fabricated via vacuum-sintering at 1900°C for 12 h, inserted is the standard diffraction patterns of $\text{Zr}_2\text{Gd}_2\text{O}_7$ (JCPDS No. 80-0471).

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