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Short communication

# Fabrication of $LaGdZr_2O_7$ transparent ceramic

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**Abstract**

A simple combustion method was used to synthesize $LaGdZr_2O_7$ powder and $LaGdZr_2O_7$ transparent ceramic was prepared by vacuum sintering at 1850 °C for 6 h. The final transparent ceramic, with a density of 6.46 g/cm$^3$, has an in-line transmittance of 70.7% at 1000 nm and a refractive index of 2.08 at 632.8 nm.


*Keywords:* Pyrochlore; $LaGdZr_2O_7$; Transparent ceramics; Combustion method; Vacuum sintering

## 1. Introduction

Pyrochlore rare-earth zirconate and hafnate ($RE_2Zr_2O_7$ and $RE_2Hf_2O_7$) have attracted much attention of many researchers due to their various interesting applications such as thermal barrier coating,[1] solid electrolytes for solid oxide fuel cells (SOFCs),[2] catalysts,[3,4] matrices for immobilization of highly active radionuclides from nuclear wastes[5] and host materials for scintillators.[6] Except for high density and high effective atomic number, the transparency is critical for scintillator materials.[7] However, with high melting point, $RE_2Zr_2O_7$ and $RE_2Hf_2O_7$ single crystals are difficult to grow. Hence, fabricating transparent ceramics becomes a good choice.

Recently, several attempts have been made to prepare $RE_2Zr_2O_7$ and $RE_2Hf_2O_7$ transparent ceramics. In 2004, transparent $La_2Hf_2O_7$ ceramics with an in-line transmittance above 70% (0.6 mm thick) were successfully fabricated from combustion synthesized powders.[8] Then in 2011, $La_2Zr_2O_7$ transparent ceramics were prepared by reactive spark plasma sintering,[9] but the transmittance was not high enough in the visible region. More recently, Zou et al. fabricated $Y_2Zr_2O_7$ transparent ceramics by using combustion synthesized powders and vacuum sintering, and the in-line transmittance was up to 68% (1.0 mm thick) in the visible spectral region.[10] However, the density and effective atomic number which are essential for host materials of scintillators of $La_2Zr_2O_7$ or $Y_2Zr_2O_7$ ceramics are not high enough. Compared with zirconate, hafnate has high density and high effective atomic number. Nevertheless, it is known that hafnate are much more expensive than zirconate, and the doping of Gd onto La site in $La_2Zr_2O_7$ with higher atomic number can improve the density and effective atomic number. In our present work, $LaGdZr_2O_7$ transparent ceramics were fabricated by vacuum sintering method using self-made powders from a rapid and simple combustion method. Morphology, phase composition of the $LaGdZr_2O_7$ powder, crystal structure, microstructure and the in-line transmittance of the resultant ceramics were investigated.

## 2. Experimental procedure

The starting materials were $Zr(NO_3)_4{\cdot}3H_2O$ (A.R.), $La(NO_3)_3{\cdot}6H_2O$ (99.99%) and glycine (A.R.). Gadolinium nitrate solution was prepared separately at first using $Gd_2O_3$ (99.99%) dissolved in excess nitric acid solution. Stoichiometric amounts of the reactants were mixed and stirred thoroughly till the solutions were clear. After that an appropriate amount of ammonia solution was added to adjust pH value of the solution

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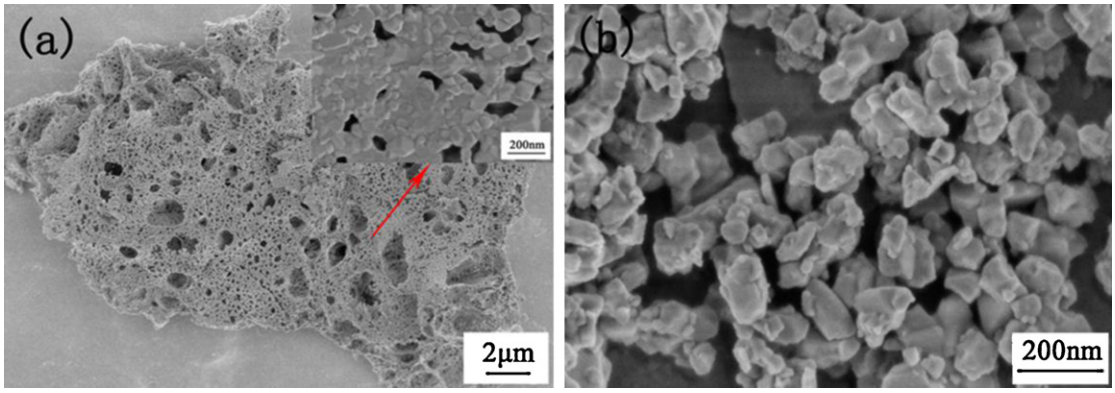


Fig. 1. SEM images of (a) the as-burnt powder calcined at 800 °C for 2 h, (inset) partial enlarged figure and (b) after ball milling for 20 h.

to 4. Then the solution was filtered and transferred into a quartz crucible. On a hot plate, the solution was heated. With the evaporation of water, the solution became sol and gel, finally the combustion reaction took place within a few minutes to form the primary fluffy powder. The as-synthesized powder was calcined at 800 °C for 2 h. Then the powder was ball-milled with a speed of 250 rpm using zirconia balls as grinding media on a planetary ball mill. After ball milling for 20 h, the powder was cold isostatically pressed into pellets (Ø20 mm × 2.5 mm) at 200 MPa for 2 min. The green pellets were sintered at 1850 °C for 6 h in vacuum (vacuum level: $10^{-3}$–$10^{-4}$ Pa) and then annealed at 1500 °C for 5 h in air. Finally, the resultant ceramics were polished to a thickness of 1 mm for test.

The phase compositions of the resultant ceramic were analyzed by X-ray diffraction (XRD) using a Japan Rigaku D/MAX 2200PC diffractometer (Tokyo, Japan) with Cu Kα radiation ($\lambda = 0.15418$ nm) in the range of $2\theta = 10$–80°. Morphology of the powder, thermal-etched surface and fracture surface of the ceramics were observed with scanning electron microscopy (SEM, JSM-6390, JEOL, Tokyo, Japan). The BET-specific surface area of the ball-milled powder was determined by nitrogen gas adsorption–desorption isotherms at 77 K on a porosimeter (Tristar 3000, Norcross, Georgia). The transmittance was measured using a UV-VIS spectrometer (UV-2501PC, SHIMADZU, Kyoto, Japan) in the range of wavelength between 200 nm and 1100 nm. The refractive index was measured on a prism coupler (Metricon Model 2010, Schott, Suzhou) at the wavelength of 532 nm and 632.8 nm. The density was measured by the Archimedes method in distilled water.

## 3. Results and discussion

Fig. 1a and b shows SEM images of the as-burnt powder before and after ball milling for 20 h. From Fig. 1a and the inserted picture, it can be seen that the powder calcined at 800 °C for 2 h is severely agglomerated with porous structure. The formation of the porous network structures may be originated from the release of gases, such as $CO_2$, $N_2$ and $H_2O$ during

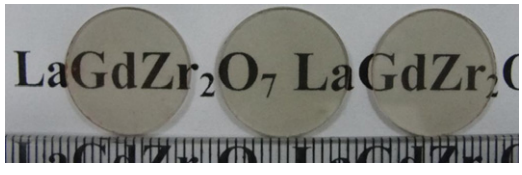


Fig. 2. Photos of $LaGdZr_2O_7$ ceramics vacuum sintered at 1850 °C for 6 h and annealed at 1500 °C for 5 h in air (1 mm thick).

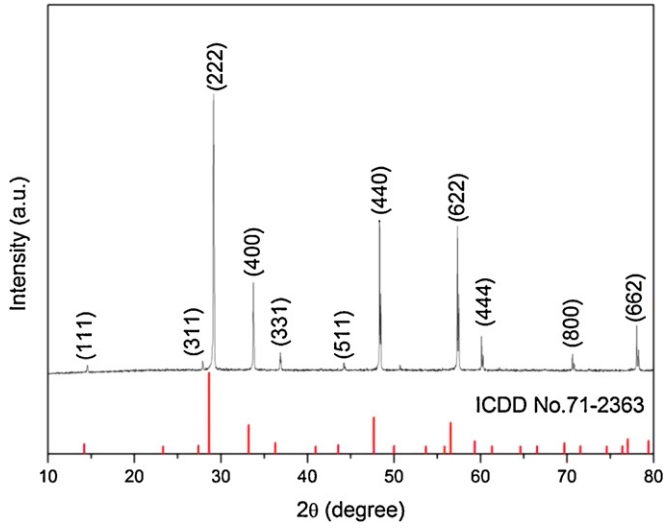


Fig. 3. XRD pattern of $LaGdZr_2O_7$ ceramic vacuum sintered at 1850 °C for 6 h and annealed at 1500 °C for 5 h in air.

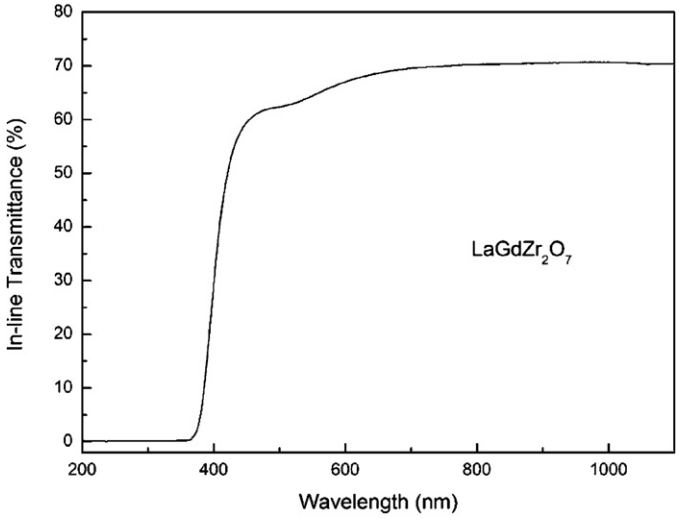


Fig. 4. In-line transmittance curve of the mirror-polished $LaGdZr_2O_7$ ceramic (1 mm thick).