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Fabrication and characterization of millimeter-scale translucent La_2O_3-doped Al_2O_3 ceramic hollow spheres

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ABSTRACT

Millimeter-scale translucent La_2O_3-doped Al_2O_3 ceramic hollow spheres have been successfully prepared using the oil-in-water (paraffin-in-alumina sol) droplets as precursors made by self-made T-shape micro-emulsion device. The main crystalline phase of the obtained hollow sphere is alpha alumina. The prepared translucent La_2O_3-containing Al_2O_3 ceramic hollow spheres have diameters of 500–1300 μm, wall thickness of about 23 μm and the degree of sphericity of above 98%. With the increase of the La_2O_3 content, grains and grain-boundaries of the alumina spherical shell for the prepared millimeter-scale hollow spheres become regular and clear gradually. When the La_2O_3 content is 0.1 wt.%, the crystal surface of the obtained Al_2O_3 spherical shell shows optimal grains and few pores, and its transmittance reaches 42% at 532 nm laser light. This method provides a promising technique of preparing millimeter-scale translucent ceramic hollow spheres for laser inertial confined fusion.

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1. Introduction

Many scientific researches are dedicated to the development and utilization of new energy sources, since the human society is suffering serious energy crisis with increasing consumption of traditional natural resources. The laser inertial confined fusion (ICF) as a new ideal environmental energy which is clean and lone-term use gets the attention of scientists [1]. Meanwhile, the approach to prepare appropriate millimeter-scale translucent hollow target pellet is still a great challenge, which is an importantly decisive part for ICF energy [2]. So far, millimeter-scale glass hollow spheres [3], metal alloy and metal-coated spheres [4] are the main candidate target pellets. However, it is hard to control the diameter, mechanical strength and uniformity of these targets. Comparing with the above-mentioned hollow spheres, translucent Al_2O_3 ceramic hollow sphere can retain structure stability for a long time in liquid helium temperature due to its oxidation resistance and physicochemical stability [5]. Thus, translucent Al_2O_3 hollow spheres become one of the best promising target pellets for ICF energy.

The typical characteristic of a target pellet is that the diameter of the sphere should be range from several hundred micrometers to several millimeters. Meanwhile, the sphere should show a single hollow core and translucent sphere shell [6]. Many methods have been used to synthesize ceramic hollow spheres [7–11]. Kang et al. prepared mesoporous TiO_2 hollow spheres with average diameters of 700 nm and wall thickness of 90 nm via a simple solvothermal method [12]. Tang et al. synthesized γ-AlOOH hollow spheres with diameter in the range of 500–900 nm via an ionic liquid-assisted hydrothermal synthesis method [13]. Generally speaking, most hollow spheres that the researchers had prepared were of diameters from several nanometers to several micrometers. Therefore, all the synthesized hollow spheres mentioned above can not be used suitably as ICF hollow target pellets. Millimeter-scale Al_2O_3 ceramic hollow spheres were synthesized in our previous work, but the transparency was not considered [14]. To fulfill the requirement of ICF usage, the improvement of the transparency of Al_2O_3 hollow spheres needs to be further studied.

The successful preparation of transparent Al_2O_3 ceramic inspires the fabrication of translucent Al_2O_3 ceramic hollow spheres. Many studies showed that the transparency of ceramics is mainly related to the microstructures including pores, grain sizes and grain boundaries and so on. The addition of dopants (such as MgO, Y_2O_3 and La_2O_3) has great influence on the microstructure and optical properties of Al_2O_3 ceramic [15]. Especially, lanthanum is often used as a dopant to improve the properties of Al_2O_3 ceramic. Fang et al. studied that the lanthanum doping can enhance the densification rate and make the grain-boundaries of Al_2O_3 ceramic become regular and clear [16].

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Rolf et al. investigated that 300 ppm MgO-doped alumina ceramic shows fine-grains and good light transmission properties [17]. Wang et al. studied that the alumina grains keep in a uniformly fine-equiaxed shape after the doping of appropriate content of La_2O_3 [18]. Rani et al. claimed that the high toughness of La-doped alumina is attributed to the microstructure modification and the effect of crack-bridging [19]. Moreover, the sintering atmosphere is also an influence factor. Gustavo et al. studied sintering kinetics and intragranular porosity of alumina ceramic by comparing with samples sintered in air and in high vacuum atmospheres [20].

In this work, on the basis of our previous work, the effect of La_2O_3 addition on the microstructure and transparency of the Al_2O_3 hollow spheres was further discussed in detail. Moreover, the preparation method was improved using paraffin liquid as inner oil phase which can be removed totally and the self-made T-shape channel was newly designed to make it easier for double droplets formation.

2. Experimental

2.1. Pure and La_2O_3-doped Al_2O_3 sol preparation

High-purity La_2O_3 (99.99%, 30 nm, Chengdu Kelong Chemical Co., Ltd.) was used as dopant with contents of 0 wt.%, 0.05 wt.%, 0.1 wt.%, 0.3 wt.%, 0.5 wt.% and 1 wt.% (Table 1). Firstly, 20.4 g La_2O_3-doped aluminum isopropoxide (AR, Chengdu Kelong Chemical Co., Ltd.) was added into 180 mL deionized water. A hydrolysis reaction was followed by increasing the system temperature to 85 °C and kept at this temperature for 0.5 h with vigorous continuous magnetic stirring. Afterwards, 12.5 mL dilute nitric acid (AR, Chengdu Kelong Chemical Co., Ltd.) ($2\ mol\ L^{-1}$) was added into the above solution in the mole ratio of $C_9H_{21}AlO_3$:H_2O:HNO_3 = 1:100:0.25 with continual stirring for 10 h at 85 °C. Finally, uniform and stable alumina sol was obtained after stewing the mixed solution in a drying oven (DHG-9070A, Keelrein Instrument Co., Ltd.) for 24 h at 90 °C.

2.2. Formation of oil-in-water emulsion droplets

The oil-in-water emulsion droplets were fabricated through the T-shape micro-emulsion channel (Fig. 1(a)) which mainly made of two different diameter capillaries. Two syringes were filled with La_2O_3-doped Al_2O_3 sol (water phase:continuous phase) and paraffin liquid (oil phase:dispersion phase) (AR, Chengdu Kelong Chemical Co., Ltd.) respectively. Controlling the two micro-injection pumps (BYZ-810, BEYOND Medical Devices Co., Ltd.) with different flow rates, the internal liquid could directly inject into the external fluids droplets at the capillary orifice (Fig. 1(b)). As time passed, the dispersion phase as soft template was gradually coated by continuous phase, and the volume dose of dispersion phase-in-continuous phase emulsion droplets became bigger. When Laplace force inside the droplets and shearing force of continuous phase became a pair of counterweight, the droplets would detach from constraints of dispersion phase, and instantly became a closed sphere under the action of surface tension. At last, the prepared core-shell type oil-in-water (paraffin liquid-in-alumina sol) droplets flowed out of the channel continuously and stably (Fig. 1(c)), and dropped into a round bottom flask filled with simethicone which provided a liquid environment and kept the core-shell structure of com-droplets stable [21]. The particle size and wall thickness were controlled by the flow rate of continuous phase and dispersion phase.

Table 1
Compositions of La_2O_3-doped aluminum isopropoxide sol.

Compositions	Sample.1 (wt.%)	Sample.2 (wt.%)	Sample.3 (wt.%)	Sample.4 (wt.%)	Sample.5 (wt.%)	Sample.6 (wt.%)
La_2O_3	0	0.05	0.1	0.3	0.5	1.0
$C_9H_{21}AlO_3$	100	99.95	99.9	99.7	99.5	99.0

2.3. Preparation of translucent hollow spheres

The procedure of preparing millimeter-scale translucent La_2O_3-doped Al_2O_3 ceramic hollow spheres was as follows four steps (Fig. 2): firstly, the fabrication of the double emulsion droplets had been narrated and discussed in Section 2.2. Secondly, the produced droplets were moved into a rotary evaporator and curdled gradually, semi-solidified La_2O_3-doped Al_2O_3 gel hollow spheres were obtained after rotating and evaporating for 12 h at 100 °C. The gel hollow spheres were washed three times by diethyl ether and deionized water respectively to remove the simethicone outer, then dried in an oven at 90 °C. Thirdly, these La_2O_3-doped Al_2O_3 gel hollow spheres were firstly sintered at 300 °C for 4 h at the rate of $1\ ^{\circ}C\ min^{-1}$ in air to remove the inside paraffin liquid phase, then heated to 1200 °C at rate of $3\ ^{\circ}C\ min^{-1}$ in air for 4 h, La_2O_3-doped Al_2O_3 spheres became dense gradually and formed hollow structure. Finally, the obtained ceramic hollow spheres were sintered at 1700 °C for 6 h in vacuum atmosphere. Millimeter-scale translucent La_2O_3-doped Al_2O_3 ceramic hollow spheres were obtained.

2.4. Characterization

A Ultra 55 Field-Emission Scanning Electron Microscope (FE-SEM) was used to characterize the morphology and size of the grain of hollow spheres. X-ray Diffraction (XRD) patterns of the prepared hollow spheres were recorded on an X'Pert PRO diffractometer (PANalytical Company (formerly Philips Analytical), Holland) with Cu Kα radiation (k = 0.15406 nm) over a 2θ scan range of 10–80° at rate of $8^{\circ}\ min^{-1}$. An Oxford IE450X-Max80 Energy Disperse Spectroscopy was used to analysis the elementary composition of the prepared hollow spheres. The light transmittance (L) of the samples was tested by measuring the laser energy E_1 before the pulse laser light (λ = 532 nm) passing through the sample at first, then measuring laser energy E_2 after the pulse laser light passing through the sample, the transmittance is calculated according to the formula $L = (E_2/E_1) \times 100\%$ [22]. The schematic of the samples' transparency testing is shown in the literature [5].

The degree of sphericity is calculated according to formula $S = \left(1 - R_0/\overline{D}\right) \times 100\%$. Where S is the degree of sphericity; R_0 is the differences between the maximum and minimum values; $\overline{D}$ is the average values of diameter in horizontal direction (D_0), vertical direction (D_{90}) and 45° direction (D_{45}) [23]. Ten randomly selected samples are measured and calculated by the formula.

3. Results and discussion

3.1. XRD analysis

Fig. 3 shows XRD patterns of the La_2O_3-doped Al_2O_3 ceramic hollow spheres calcined in vacuum 1700 °C for 6 h. It is obvious that the diffraction peaks are sharp and the main crystalline phase of the prepared Al_2O_3 ceramic hollow spheres is alpha alumina. When aluminum isopropoxide are mixed with water at 85 °C, the hydrolysis reaction will happen [24], the principle of the reaction can be expressed simply as following: $Al(OC_3H_7)_3 + 3H_2O \rightarrow Al(OH)_3 + 3(CH_3)_2CHOH$. Sintering in high temperature, aluminum hydroxide will decompose according to

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