

Yb:Sc₂O₃ Transparent Ceramics Fabricated from Co-precipitated Nano-powders: Microstructure and Optical Property

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Abstract: Sc₂O₃, as a host for solid-state laser gain materials, has advantage of high thermal conductivity and easy matching with activating ions, which is promising in high-power laser applications. Currently, Yb-doped Sc₂O₃ ceramics have been fabricated at very high sintering temperatures, but their optical quality and sintering process still need further improvement. In this work, 5%Yb:Sc₂O₃ (in mass) nano-powders were obtained by co-precipitation, and then transparent ceramics were fabricated by vacuum pre-sintering and hot isostatic pressing (HIP) post-treatment. The cubic Yb:Sc₂O₃ nano-powders with good dispersity and an average crystallite of 29 nm were obtained. Influence of pre-sintering temperatures (1500–1700 °C) on densification process, microstructure changes, and optical transmittance of Yb:Sc₂O₃ ceramics was detected. Experimental data revealed that all samples have a uniform microstructure, while the average grain sizes increase with the increase of the sintering temperatures. Impressively, the optimum in-line transmittance of Yb:Sc₂O₃ ceramics, pre-sintered at 1550 °C after HIP post-treatment, reaches 78.1% (theoretical value of 80%) at 1100 nm. Spectroscopic properties of the Yb:Sc₂O₃ ceramics reveal that the minimum population inversion parameter β_2 and the luminescence decay time of 5%Yb:Sc₂O₃ ceramics are 0.041 and 0.49 ms, respectively, which demonstrate that the optical quality of the Yb:Sc₂O₃ has been improved. Meanwhile, their best vacuum sintering temperature can be controlled down to a lower temperature (1550 °C). In conclusion, Yb:Sc₂O₃ nano-powders are successfully synthesized by co-precipitation method, and good optical quality transparent ceramics are fabricated by vacuum pre-sintering at 1550 °C and HIP post-treatment.

Key words: nano-powder; Yb:Sc₂O₃; transparent ceramic; hot isostatic pressing; optical property

Mainstream laser equipment includes gas lasers, solid-state lasers (SSLs), and fiber lasers^[1]. Compared to gas lasers, SSLs usually have smaller laser cavities, which make them more mobile and portable^[2]. Although fiber lasers have advantages in stability, typical SSLs can withstand higher laser peak energy, making them widely used in high-power laser facilities^[3]. Furthermore, for the

same reasons, advanced technological solutions based on SSLs have broad applications in industrial processing, military weapons, and medical surgery^[4-7].

Yb³⁺ doped laser media became well established in high-power SSLs since Yb³⁺ ions are well matched to the InGaAs laser diode (LD) pump wavelength (940–980 nm). Moreover, Yb³⁺ has a large absorption cross-section and

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a long luminescence lifetime, which contributes to the absorption and storage of pump energy. The laser emission mechanism of Yb^{3+} near 1 μm is a quasi-three level^[8]. Therefore, Yb^{3+} has high theoretical quantum efficiency (>90%) and is not prone to severe concentration quenching, up-conversion, and cross relaxation problems compared to other activated ions (Tm^{3+} , Er^{3+} , Ho^{3+} , etc.)^[9]. In conclusion, Yb^{3+} as an activating ion is suitable for high-power laser applications and has gained increasing popularity^[10-12].

Laser gain materials, mainly containing garnet (YAG), fluoride (CaF_2) and sesquioxide (Lu_2O_3 , Sc_2O_3 and Y_2O_3), are key components in the SSLs for light-to-light conversion and pump light amplification, determining the laser output efficiency and quality. To realize high-power SSLs, laser gain materials require good optical quality, good thermo-optical properties, and thermal shock resistance^[13]. Compared with the YAG laser gain host material, sesquioxide materials have better thermal shock resistance and thermal conductivity^[2]. In recent years, Sc_2O_3 material for laser gain host material has gained much attention due to the highest thermal conductivity (17 W/(m·K)) compared to Y_2O_3 (14 W/(m·K)) and Lu_2O_3 (13 W/(m·K)), large Stark splitting, and relatively low phonon energies^[14-18].

The high melting point of Sc_2O_3 reaching 2400 °C makes it difficult to produce large-sized and uniformly doped Sc_2O_3 single crystals (SCs). In addition, due to the slow crystal growth methods, the preparation of Yb^{3+} -doped Sc_2O_3 SCs is long-time and high-cost^[17-18]. However, preparing $\text{Yb}:\text{Sc}_2\text{O}_3$ transparent ceramics can effectively reduce the preparation temperature and time while also achieving larger size and uniform Yb^{3+} doping.

The fabrication of transparent ceramics with high optical quality requires powders with high purity, low agglomeration, and high sintering activity. The common methods to fabricate the $\text{Yb}:\text{Sc}_2\text{O}_3$ powders contain solid-state reaction method, precipitation method, Sol-Gel method, and self-propagating synthesis^[14-16, 19-27]. Bravo^[26], Dong^[28], and Li^[29] *et al.* studied the difference between solid-state reaction method and precipitation method. Through this research, the precipitation method was found to have the advantage of obtaining $\text{Yb}:\text{Sc}_2\text{O}_3$ powders with a uniform distribution, better dispersion, high sintering activity, and smaller particle sizes, which could significantly reduce the sintering temperature to promote the densification process of the ceramics. Dai *et al.*^[16, 21, 30] studied the influence of ammonium hydrogen carbonate on metal ions molar ratio and calcination temperature of co-precipitated nano-powders. $\text{Yb}:\text{Sc}_2\text{O}_3$ nano-powders with an average particle size of 32.4 nm and a good dispersion were successfully

fabricated through the co-precipitation method. Poirot *et al.*^[27] used the Sol-Gel method to synthesize well-dispersed Sc_2O_3 nano-powders. After spark plasma sintering at 1400 °C for 1 min, the ceramics are nearly fully dense and have an average grain size of around 100 nm. Permin *et al.*^[15, 25] studied the effects of fuel type, lithium fluoride aid, and calcination temperature on the powder made by self-propagating synthesis. $\text{Yb}:\text{Sc}_2\text{O}_3$ nano-powders with an average particle size of 31 nm and a good dispersion were successfully fabricated through this method. Although the Sol-Gel method, self-propagating high-temperature synthesis, and precipitation method can all synthesize nano-scandium oxide powder with high sintering activity, the precipitation method has the advantage of high yield, low cost, and controllable and stable process. Therefore, $\text{Yb}:\text{Sc}_2\text{O}_3$ nano-powders with an average particle size of 29 nm were obtained by the co-precipitation method in this work. The chemical reactions during the titration process and the powders calcination process were investigated by measuring pH and the thermogravimetric-differential thermogravimetric (TG-DTG) curve. The morphology, phase, particle size, purity, and agglomeration of the as-synthesized precursor and calcined powders were also evaluated.

The common methods of fabricating $\text{Yb}:\text{Sc}_2\text{O}_3$ transparent ceramics include vacuum sintering, hot pressing, and vacuum pre-sintering with HIP post-treatment^[15-16, 31-32]. Compared to hot pressing and vacuum sintering, the vacuum pre-sintering with HIP post-treatment can better eliminate the closed pores within the ceramics by controlling the microstructure of the pre-sintering samples^[33]. By eliminating the closed pores inside the ceramics, the optical quality of the ceramic can be improved. Therefore, the vacuum pre-sintering with HIP post-treatment method gains attention in synthesis of high optical quality transparent ceramics^[34-36]. $\text{Yb}:\text{Sc}_2\text{O}_3$ transparent ceramics, showing the best optical quality, were prepared by Permin *et al.*^[15], which reached an in-line transmittance of 78% at 1040 nm. Accordingly, this work aimed to explore an effective short-term two-step sintering method (vacuum pre-sintering with HIP post-treatment) to synthesize $\text{Yb}:\text{Sc}_2\text{O}_3$ transparent ceramics with high optical transparency. By adjusting the vacuum pre-sintering temperatures, the microstructure and the densification degree of the ceramics could be controlled, making the elimination of pores more complete in the HIP post-treatment. To this end, the effects of pre-sintering temperatures on the microstructure, optical transmittance, and densities of ceramics were investigated in this study. The optimum pre-sintering temperature were also determined, which yielded suitable spectroscopic properties for the use of

the resulting ceramics in laser medium applications.

1 Experimental

High-pure Sc(NO₃)₃ and Yb(NO₃)₃ were prepared by dissolving the commercial oxide powders of Sc₂O₃ (99.99%, Jingyun Material Technology Co., Ltd., China) and Yb₂O₃ (99.995%, Golden Dragon Rare Earth Co., Ltd., China) in an excess of nitric acid while heating and stirring. The concentrations of Sc(NO₃)₃ and Yb(NO₃)₃ were determined by wet chemical analysis using the UV-visible spectrophotometer (Specord 50 plus, Analytik Jena AG, Germany). Sc(NO₃)₃ and Yb(NO₃)₃ were mixed with the stoichiometric ratio and diluted to 0.2 mol/L to prepare Sc_{0.95}Yb_{0.05}(NO₃)₃ solution. A specific molar ratio of ammonium sulfate ((NH₄)₂SO₄, 99.0%, Sinopharm Chemical Reagent Co., Ltd., China) was added into the mixed solution as the dispersant. The precipitant solution was prepared by dissolving a certain amount of ammonium hydrogen carbonate (AHC, 99.99%, Sinopharm Chemical Reagent Co., Ltd., China) into the deionized water. The concentration of the precipitant was 1 mol/L.

During the co-precipitation process, the precipitant solution was dropped into the Sc_{0.95}Yb_{0.05}(NO₃)₃ mixed solution at a speed of 3 mL/min at room temperature (RT). When the process of co-precipitation finished, the resultant suspension was aged for 3 h, and then washed three times with deionized water and twice with ethanol (99.7%, Shanghai Lingfeng Chemical Reagent Co., Ltd., China). Then the precipitate was separated from the solvent by centrifugation. The rinsed precursor was dried at 70 °C for 48 h in the oven and sieved through a 200 mesh (74 μm) screen. 5%Yb:Sc₂O₃ (in mass) nano-powders were fabricated by calcining the precursor at 1100 °C for 4 h in air. The synthesized nano-powders were uniaxially dry-pressed into φ18 mm pellets under 40 MPa and then cold-pressed isostatically under 250 MPa. The green bodies were pre-sintered at different temperatures (1500–1700 °C) for 2 h with HIP post-treatment at 1700 °C for 3 h under 176 MPa in argon atmosphere. Finally, the ceramics were annealed at 1200 °C for 10 h in air and double-sided polished to the thickness of 1.3 mm, followed by thermal etching at 1250 °C for 3 h in air.

The TG-DTG analyses (Thermopiles EVO II, Rigaku, Japan) were measured at a heating rate of 10 °C/min from RT to 1300 °C. The specific surface area (*S*_{BET}) of the Yb:Sc₂O₃ nano-powders was measured by nitrogen chemisorption using an automatic surface area analyzer (BET, Quadrasorb SI, Micromeritics, USA). The X-ray

diffractometer (XRD, Model D/max2200 PC, Rigaku, Japan) was used to identify the powders' phase in the range of 2θ=10°–90° using Cu Kα1 radiation (λ=0.15418 nm) at a scan rate of 5 (°)/min. The densities of the samples were calculated using the Archimedes method. The morphologies of the resultant precursor and powders and the microstructures of the thermally etched pre-sintered and HIP post-treated ceramics were observed by field emission scanning electron microscopy (FESEM, SU8220, Hitachi, Japan). The average grain sizes (GS) were measured by the common linear intercept analysis (more than 200 grains were counted), according to the equation $GS=1.56L_1$, where L_1 is the mean intercept. The in-line transmittance and absorption spectra of the ceramic samples were measured using a UV-Vis-NIR spectrophotometer (Model Carry-5000, Varian, USA). The luminescence decays were excited by the 980 nm laser and recorded by a photoluminescence spectrometer (FLS980, Edinburgh Instrument, UK).

2 Results and discussion

Fig. 1 shows the TG-DTG curves of the precursor. According to the TG-DTG analysis, the mass loss of the precursor can be divided into 4 stages. The first stage is between 100 and 200 °C. The rate of mass loss in the DTG curve at 126 °C is very rapid. The corresponding TG curve shows the weight loss below 200 °C is about 12.2%, which is primarily caused by evaporation of molecular and surface absorbed water^[27, 37–38]. The second stage is 200–600 °C with a weight loss of 14.3%, which is generally considered to be the dehydroxylation^[31, 37, 39]. Another DTG peak of maximum weight loss at 643 °C indicates the formation of cubic Sc₂O₃^[31, 39–40], which means the weight loss of the precursor has come to the third stage. The corresponding weight loss at 600–800 °C is about 16.2%. The fourth stage is 800–1300 °C with a weight loss of 3.0% and a slow

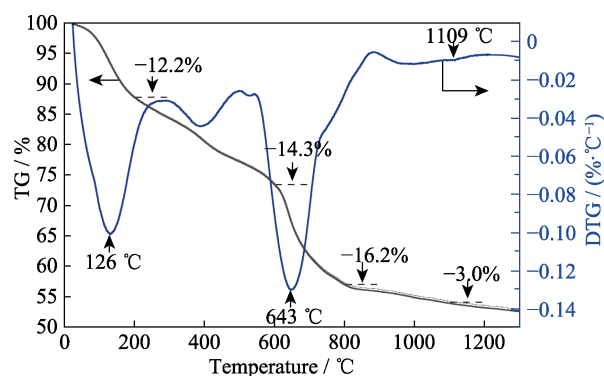


Fig. 1 TG-DTG curves of the precursor

weight-loss rate peak at 1109 °C. In this stage, the weight of the precursor changes very little, indicating that the precursor has completely decomposed and formed a stable cubic phase of Yb:Sc₂O₃. The result is consistent with the XRD pattern (Fig. 2) of the powders calcined at 1100 °C for 4 h.

Since XRD patterns possess no obvious peak in the range of $2\theta=65^\circ\text{--}90^\circ$, to provide a clearer and better representation of useful information, Fig. 2 presents the XRD patterns of the precursor, the calcined 5%Yb:Sc₂O₃ powders and the cubic Sc₂O₃ standard card (PDF# 43-1028) in the range of $2\theta=10^\circ\text{--}65^\circ$. The peaks in the ranges of $2\theta=15^\circ\text{--}20^\circ$ and $2\theta=25^\circ\text{--}30^\circ$ indicate that the precursor has a crystalline phase^[41]. Meanwhile, the XRD peaks of the calcined powders suit well with the cubic Sc₂O₃ standard card. Therefore, it illustrates that the precursor fully transformed into 5%Yb:Sc₂O₃ nano-powders after 1100 °C calcined for 4 h in air. According to Scherrer's formula and the full width at half maximum (FWHM), the average crystallite size (D_{XRD}) of the 5%Yb:Sc₂O₃ nano-powders is 29 nm. Scherrer's formula can be expressed as follow^[42]:

$$D_{\text{XRD}} = \frac{K\lambda}{\beta_1 \cdot \cos\theta} \quad (1)$$

where K is the dimensionless shape factor, typically value of K is 0.89; λ is the wavelength of Cu K α 1 radiation and β_1 is the FWHM of the diffraction peak at Bragg's angle (θ).

Fig. 3 shows that the precursor consists of dumbbell nanoparticles and has little agglomeration. After calcination at 1100 °C for 4 h, the particle sizes of the calcined powders clearly decrease due to the decomposition of the synthesized precursor. The shape of the powder has changed into globular compared to the synthesized precursor. Through primary particle size statistics, the average particle size obtained by SEM (D_{SEM}) of the calcined powders is 43 nm. The nano-powders exhibit a specific surface area (S_{BET}) of 25 m²/g with an average particle size (D_{BET}) of 58 nm which is calculated according to the formula (2):

$$D_{\text{BET}} = \frac{6}{\rho \cdot S_{\text{BET}}} \quad (2)$$

where ρ is the theoretical density of synthesized 5%Yb:Sc₂O₃ nano-powders determined from the XRD, 4.15 g/cm³. The agglomeration coefficient (N_1) is calculated by the equation $N_1=(D_{\text{BET}}/D_{\text{XRD}})^3$. From Table 1, N_1 of the 5%Yb:Sc₂O₃ nano-powders approaches 6, indicating the powders have a good dispersity.

In order to explore a suitable pre-sintering temperature that can produce dense ceramics sintered at a lower

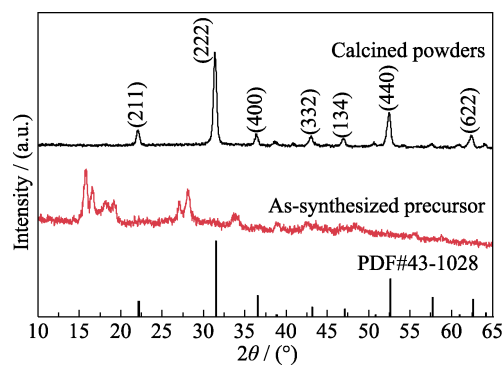


Fig. 2 XRD patterns of the precursor and the powders calcined at 1100 °C for 4 h

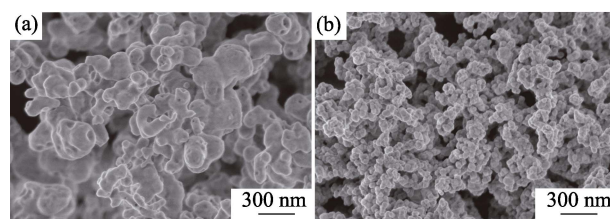


Fig. 3 FESEM micrographs of (a) the precursor and (b) the powders

Table 1 Analysis of particle size and agglomeration degree of 5%Yb:Sc₂O₃ nano-powders

Sample	$S_{\text{BET}}/(\text{m}^2 \cdot \text{g}^{-1})$	D_{BET}/nm	D_{SEM}/nm	D_{XRD}/nm	N_1
5%Yb:Sc ₂ O ₃ nano-powders	25	58	43	29	6

temperature, Fig. 4 shows the FESEM micrographs of the thermally etched surfaces of the 5%Yb:Sc₂O₃ samples pre-sintered at different temperatures in vacuum. It follows that the samples pre-sintered at different temperatures all have a uniform structure, and no abnormal grain growth is detected. However, the pre-sintering temperature can greatly influence the microstructure. As the pre-sintering temperature gradually increases from 1550 to 1700 °C, the number of residual pores along grain boundaries decreases because the high sintering temperature produces a high sintering driving force, which promotes the grain growth and the densification process^[43].

Fig. 5 shows the average grain sizes and relative densities of 5%Yb:Sc₂O₃ ceramics with different pre-sintering temperatures. With the increase of the pre-sintering temperature, the average grain sizes increase from 1.1 to 5.8 μm, while the relative densities rise from 89.2% to 99.0%. The sintering method of vacuum pre-sintering with HIP post-treatment requires the pre-sintering samples to have relative densities of 92%–97% because the HIP post-treatment provides a strong grain driving force to eliminate the residual pores in the ceramics^[44]. The samples pre-sintered at 1550

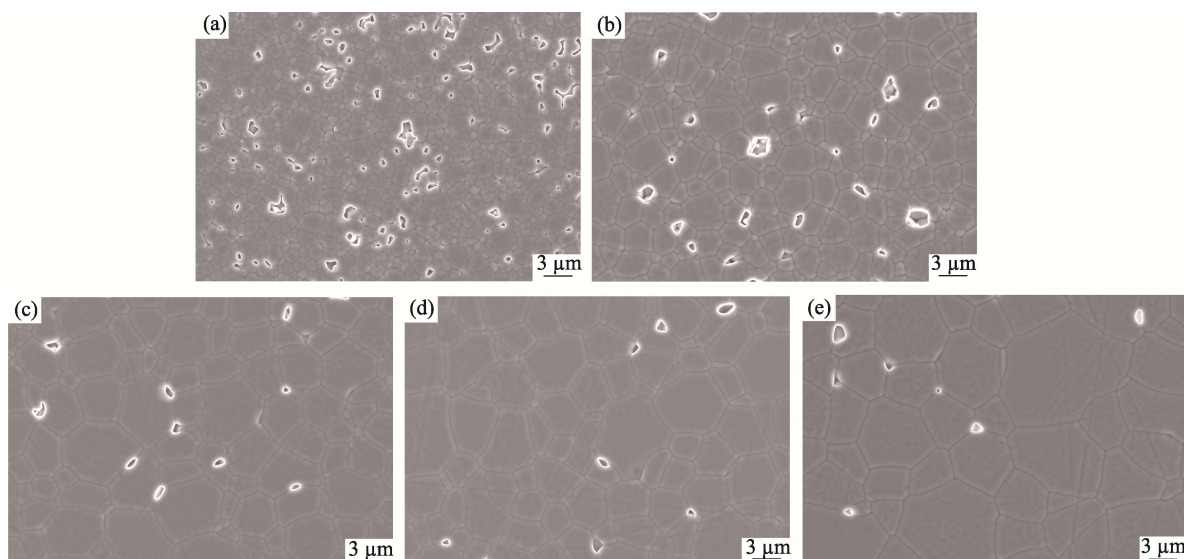


Fig. 4 FESEM micrographs of thermally etched surfaces of the 5%Yb:Sc₂O₃ ceramics pre-sintered at different temperatures

(a) 1500 °C; (b) 1550 °C; (c) 1600 °C; (d) 1650 °C; (e) 1700 °C

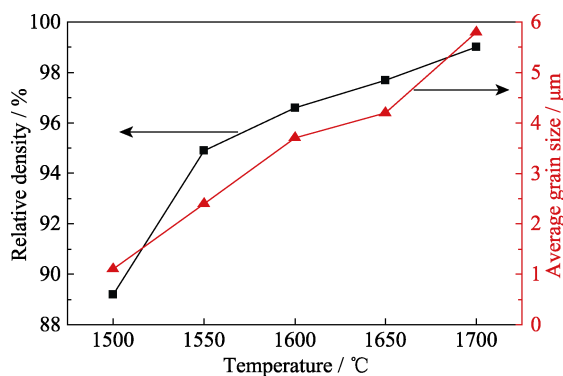


Fig. 5 Average grain sizes and relative densities of 5%Yb:Sc₂O₃ ceramics with different pre-sintering temperatures

and 1600 °C have appropriate relative densities (94.9% and 96.6%, respectively) and small grain sizes, which provides good conditions for the preparation of laser ceramics with high optical quality.

Fig. 6 presents FESEM micrographs of the thermally etched surfaces of the ceramics pre-sintered at different temperatures with HIP post-treatment. The pre-sintering ceramics have a uniform microstructure, and there is no abnormal grain growth. In particular, the ceramics pre-sintered at 1500 °C after HIP post-treatment still have many residual pores between the grain boundaries. The reason for this phenomenon is that the pre-sintering

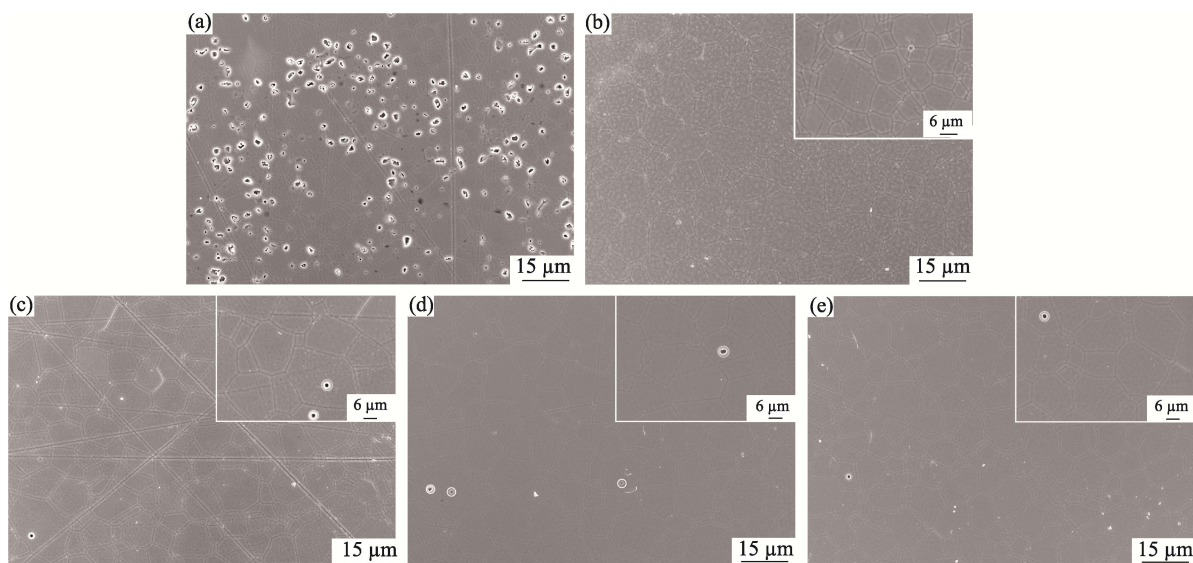


Fig. 6 FESEM micrographs of thermally etched surfaces of the synthesized 5%Yb:Sc₂O₃ ceramics
Ceramics pre-sintered at (a) 1500, (b) 1550, (c) 1600, (d) 1650, and (e) 1700 °C before HIP post-treatment at 1700 °C

sample has many interconnected pores, which cannot be eliminated by the HIP post-treatment. The ceramics pre-sintered at 1550 and 1600 °C with HIP post-treatment show merely dense microstructure, the residual pores are almost removed, leading to transparent ceramics. There are still a few submicron pores in the ceramics vacuum pre-sintered at 1550 °C (Fig. 6(b)), which makes it difficult for HIP post-treatment to completely remove them. In addition, some intragranular pores are found in the ceramics pre-sintered at 1600, 1650, and 1700 °C, which also cannot be eliminated by the HIP post-treatment. During the densification process, the mass diffusion rate is slower than the grain boundary diffusion rate at a high HIP post-treated temperature and pressure, resulting in the formation of intragranular pores^[45].

Fig. 7 shows the average grain sizes and relative densities of 5%Yb:Sc₂O₃ ceramics sintered by vacuum pre-sintering and HIP post-treatment. As pre-sintering temperature increases, the average grain sizes of the ceramics increase from 6.3 to 9.0 μm. The ceramics are almost densified at the pre-sintering temperature of 1550 °C.

Fig. 8 presents the photographs and in-line transmittance of the 5%Yb:Sc₂O₃ ceramics vacuum pre-sintered at different temperatures for 2 h and HIP post-treated at 1700 °C for 3 h. After HIP post-treatment, the ceramics pre-sintered at 1500 °C is opaque due to the light scattering caused by many residual pores, which matches the result of the thermally etched microstructure of the sample. With the increase in pre-sintering temperature from 1550 to 1700 °C, the in-line transmittance of the ceramics gradually decreases. This result can be explained by the increasing intragranular pores shown in Fig. 6, leading to severe light scattering. The ceramics pre-sintered at 1550 °C for 2 h with HIP post-treatment have the highest in-line transmittance of 78.1% at 1100 nm, which is close to the theoretical value of

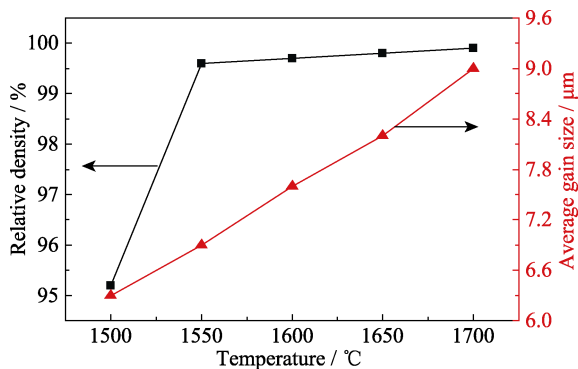


Fig. 7 Average grain sizes and relative densities of 5%Yb:Sc₂O₃ ceramics sintered by vacuum pre-sintering and HIP post-treatment

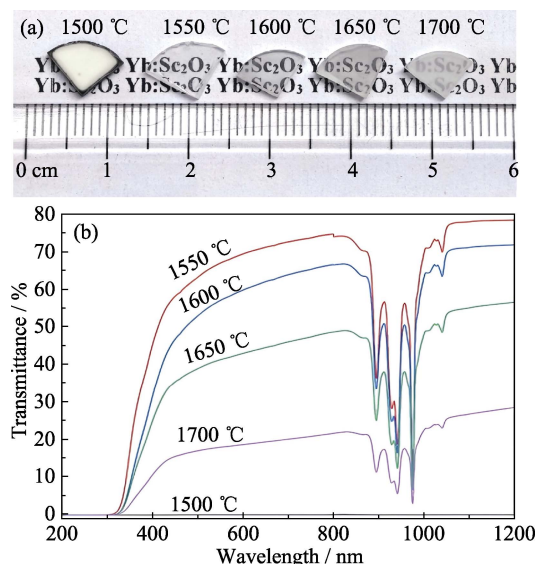


Fig. 8 Photograph (a) and in-line transmittance (b) of the 5%Yb:Sc₂O₃ ceramics

80%^[46]. The sharp decrease of in-line transmittance at wavelengths between 200 and 800 nm may be caused by light scattering from the residual submicron pores whose size is close to the incident wavelength (~600 nm), leading to Mie scattering^[47]. Generally, it indicates that the pre-sintering temperature has an effective regulation on the optical quality of transparent 5%Yb:Sc₂O₃ ceramics.

Other optical properties of the best optical quality transparent ceramic (in-line transmittance of 78.1% at 1100 nm) were further evaluated. Fig. 9 shows the absorption and emission cross-sections of the best optical quality 5%Yb:Sc₂O₃ transparent ceramic. The absorption cross-sections (σ_{abs}) are calculated by the following equation (3)^[21]:

$$\sigma_{\text{abs}} = \frac{-2.303 \cdot \lg \frac{I}{I_0}}{N_2 \cdot L_2} \quad (3)$$

where $\lg \frac{I}{I_0}$ is the absorbance of the ceramics, I_0 is the

incident light intensity and I is the transmitted light intensity; N_2 is the number of doping ions in the unit volume ($1.7 \times 10^{21} \text{ cm}^{-3}$ for 5%Yb:Sc₂O₃ ceramics) and L_2 is the thickness of the sample. The absorption cross-sections have five prominent peaks at 894, 929, 942, 975, and 1041 nm, which match the Yb³⁺ energy transitions from ²F_{7/2} to ²F_{5/2}. The absorption cross-sections at 975 and 942 nm are 1.18×10^{-20} and $0.68 \times 10^{-20} \text{ cm}^2$, respectively. The broad absorption band between 894 and 1041 nm makes 5%Yb:Sc₂O₃ transparent ceramics suitable for laser diode pumping. Since the second critical property for laser gain material is an appropriate emission property to achieve laser emission, the emission

and gain cross-sections need to be investigated. Since the second equally critical property is that the material has the appropriate emission properties to achieve laser action, a parameter must be determined to confirm whether the ceramics under investigation have the characteristics of good luminescent medium. The reciprocity method can be used to determine the emission cross-sections (σ_{em}) with the data from the absorption cross-sections^[48], which is one of the main objective parameters to evaluate these characteristics, using the following equation (4):

$$\sigma_{em} = \sigma_{abs} \frac{Z_l}{Z_u} \exp \left[\frac{hc}{kT} \left(\frac{1}{\lambda_{zl}} - \frac{1}{\lambda} \right) \right] \quad (4)$$

where Z_l and Z_u are the lower and upper manifold partition functions, the values of which are 1.175 and 1.179, respectively^[49]; h is the Planck constant, k is the Boltzmann constant, c is the velocity of light, T is the temperature, and λ_{zl} is the wavelength of the zero-phonon line, which locates at 975 nm. There are four main peaks (942, 975, 1041, and 1094 nm) of the emission cross-sections. The calculated emission cross-sections at 975 and 1041 nm are 1.13×10^{-20} and 1.14×10^{-20} cm², respectively. The FWHMs of the 5%Yb:Sc₂O₃ transparent ceramics at the wavelengths of 975, 1041, and 1094 nm are about 6, 10, and 26 nm, respectively, as shown in Fig. 9.

Fig. 10 presents the gain cross-sections of 5%Yb:Sc₂O₃ transparent ceramics pre-sintered at 1550 °C for 2 h and HIP post-treated at 1700 °C for 3 h. Finally, based on the data of absorption and emission cross-sections, the gain cross-sections (σ_g) can be estimated by the equation (5) below^[50]:

$$\sigma_g(\lambda) = \beta_2 \sigma_{em}(\lambda) - (1 - \beta_2) \sigma_{abs}(\lambda) \quad (5)$$

where β_2 is the population inversion parameter. The strongest emission peak is located at 1041 nm. The

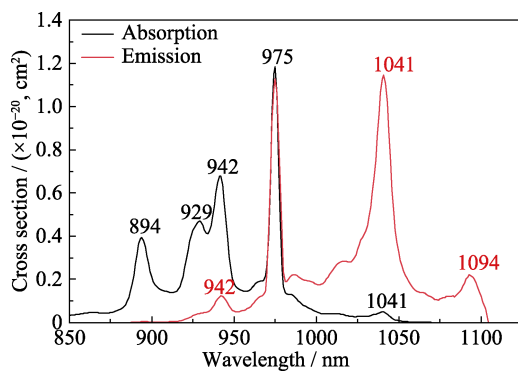


Fig. 9 Absorption and emission cross-sections of the 5%Yb:Sc₂O₃ transparent ceramics pre-sintered at 1550 °C with HIP post-treatment

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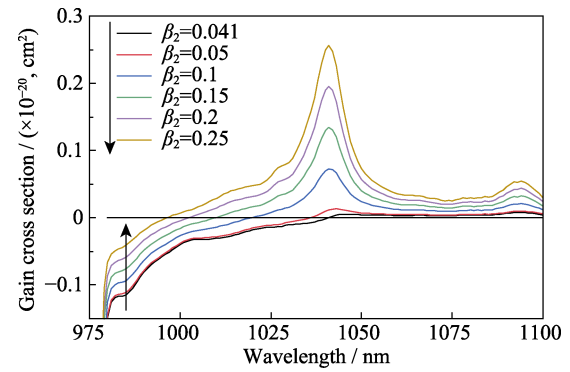


Fig. 10 Gain cross-section of the 5%Yb:Sc₂O₃ transparent ceramics pre-sintered at 1550 °C with HIP post-treatment

minimum β_2 is determined to be 0.041, which is smaller than that of Yb:YAG (0.055)^[49]. A smaller β_2 indicates that the laser diode pump source can more easily excite Yb³⁺ ions in Yb:Sc₂O₃ than in Yb:YAG, because a long luminescence lifetime of Yb³⁺ ions is important for laser gain material to output more laser energy.

In addition, to determine whether 5%Yb:Sc₂O₃ transparent ceramics still show further potential to improve their emission properties, the luminescence decay was investigated. Fig. 11 illustrates the ²F_{5/2} luminescence decay of 5%Yb:Sc₂O₃ transparent ceramics. The decay time conforms to a single exponential decay curve. The lifetime of the Yb³⁺ upper energy level is calculated as 0.49 ms. Noticeably, the result is smaller than the value of 0.8 ms reported in the Yb-doped Sc₂O₃ single crystal^[51]. The reasons can be explained by the energy transmission in Yb³⁺ as well as the energy transmission between Yb³⁺ and other impurity ions, which cause the self-quenching effect. When the concentration of Yb³⁺ rises, this phenomenon becomes more significant^[52].

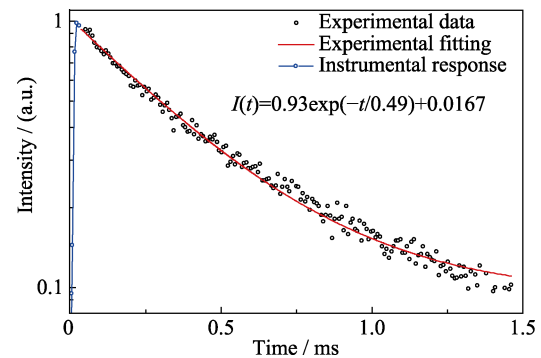


Fig. 11 Room temperature ²F_{5/2} luminescence decay of 5%Yb:Sc₂O₃ transparent ceramics excited by 980 nm laser and detected at 1041 nm wavelength

3 Conclusions

In order to find an appropriate synthesis process for high-quality optical Yb:Sc₂O₃ transparent ceramics, the co-precipitation method was chosen to fabricate Yb:Sc₂O₃ nano-powders with good dispersion and high sintering activity. The average particle size calculated by the specific surface area is 58 nm, while the average crystalline size calculated by the XRD pattern is 29 nm. According to the FESEM micrographs, the powders have a worm-like shape and low agglomeration.

The green bodies were pre-sintered at different temperatures, and the microstructure of the pre-sintering samples was studied. It is found that the average grain sizes of the pre-sintering samples increase from 1.1 to 5.8 μm , while the number of residual pores decreases as the temperature increases from 1500 to 1700 $^{\circ}\text{C}$. After HIP post-treatment at 1700 $^{\circ}\text{C}$ under 176 MPa argon pressure, the ceramics with the best optical transparency were fabricated at a pre-sintering temperature of 1550 $^{\circ}\text{C}$ with an in-line transmittance of 78.1% at 1100 nm, which was close to the theoretical value. The average grain size of the ceramics rises from 6.3 to 9.0 μm with the increase in pre-sintering temperature. When the temperature reaches 1550 $^{\circ}\text{C}$, the ceramics are almost densified.

The absorption cross-sections of 5%Yb:Sc₂O₃ ceramics at 975 and 942 nm are found to be 1.18×10^{-20} and $0.68 \times 10^{-20} \text{ cm}^2$, respectively. The emission cross-sections of 5%Yb:Sc₂O₃ ceramics at 975 and 1041 nm are 1.13×10^{-20} and $1.14 \times 10^{-20} \text{ cm}^2$, respectively. The FWHMs of 5%Yb:Sc₂O₃ ceramics at the wavelengths of 975, 1041, and 1094 nm are about 6, 10, and 26 nm, respectively. The minimum β_2 value is determined to be 0.041, proving that the 5%Yb:Sc₂O₃ transparent ceramics have potential in high-power solid-state lasers. The lifetime of the Yb³⁺ upper energy level is calculated as 0.49 ms, suggesting that the Yb³⁺ ion doping concentration may be too high, and structural defects may exist in the transparent ceramics.

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共沉淀纳米粉体制备 Yb:Sc₂O₃ 透明陶瓷的 微结构与光学性能

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摘要: Sc₂O₃ 作为固体激光增益材料的基质, 具有热导率高、与激活离子匹配性好等优点, 在高功率固体激光应用中有着广阔的应用前景。目前, Yb 掺杂的 Sc₂O₃ 陶瓷可以在很高的烧结温度下合成, 但陶瓷的光学质量和烧结温度还有待进一步优化。本研究采用碳酸氢铵共沉淀法制备了分散性好、平均晶粒尺寸 29 nm 的立方相 5%Yb:Sc₂O₃ (掺杂量为质量分数) 纳米粉体, 并采用真空预烧结和热等静压 (Hot Isostatic Pressing, HIP) 后处理制备了 5%Yb:Sc₂O₃ 透明陶瓷, 详细研究了真空预烧结温度 (1500~1700 °C) 对 Yb:Sc₂O₃ 陶瓷的致密化过程、微观结构变化及光学透过率的影响。结果表明, 所有样品均具有均匀的微观结构, 且随着烧结温度的升高, 样品的平均晶粒尺寸增大。值得注意的是, 1550 °C 预烧的 Yb:Sc₂O₃ 陶瓷经 HIP 后处理后, 在 1100 nm 处的最佳直线透过率达到 78.1% (理论值为 80%)。同时, 还评估了 Yb:Sc₂O₃ 陶瓷的光谱性能。5%Yb:Sc₂O₃ 陶瓷的最小粒子数反转参数 β_2 和发光衰减时间分别为 0.041 和 0.49 ms。综上所述, 采用共沉淀法成功制备了 Yb:Sc₂O₃ 纳米粉体, 通过 1550 °C 真空预烧结和 HIP 后处理得到光学质量良好的 Yb:Sc₂O₃ 透明陶瓷。

关键词: 纳米粉体; Yb:Sc₂O₃; 透明陶瓷; 热等静压烧结; 光学性能

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