

Synthesis and characterization of high concentration Nd³⁺ doped YAG nanopowders for laser applications

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Herein, neodymium-doped yttrium aluminum garnet (Nd:YAG) ceramic powders were fabricated via a wet chemical reaction method followed by heating in air. The structural studied confirmed the formation of single phase Nd:YAG nanocrystallites without any intermediate phase at 1000 °C, and the average particle size was calculated to be 260 nm. According to the reflectance results, the highest absorption of the synthesized powders was observed at 808 nm. Therefore, A laser diode (808 nm) with power output about 1000 mW was used as a pump source and the emission spectra was recorded. Upon IR excitation, the present sample showed intense emission at around 1064 nm and found to be potential for solid state laser devices.

Introduction

Recently, a potential research has been devoted on Nd³⁺ doped yttrium aluminum garnet (Y₃Al₅O₁₂; YAG) ceramics for laser applications owing to its excellent chemical stability, good optical and high temperature creep resistance [1, 2]. YAG possesses low vibration energy and the minimization of the quenching of the excited state of Nd³⁺ activator [3-5]. Nd:YAG single crystals are known to be important for fabricating solid-state laser [1-6]. However, the growth of Nd:YAG single crystals is a complicated process. It is very difficult for Nd³⁺ to replace Y³⁺ owing to the considerable difference between the radius of Nd³⁺ and Y³⁺. Because of the inhomogeneous distribution of Nd³⁺ ions, unwanted phenomenon such as thermal stresses, non-uniform optical properties, and fluorescence quenching could occur [7]. Owing to the above reasons, the Nd doping level in a YAG single crystal should be low. However, it is very difficult to produce high quality crystals for generating high power lasers with a low Nd doping content [8]. In comparison with the growth technique for Nd:YAG single crystals, the fabrication of polycrystalline Nd:YAG ceramics does not require any special techniques. Most importantly, large size and high concentration (>1mol%) Nd³⁺ doped samples can be fabricated via using polycrystalline Nd:YAG powders [9, 10]. Moreover, multilayer active elements and multi-functional powders can be easily fabricated together unlike the single crystal growth method. Thus a lot of effort has been made to try to synthesize polycrystalline YAG ceramics doped with high amount of Nd³⁺ ions. The Nd:YAG powders are usually synthesized by the various wet chemical synthesis methods, including co-precipitation [11], sol-gel combustion [12], sol spray [13], and the gel combustion methods [14]. These processes achieve symmetrical mixing on the molecular level, lowering the crystallization temperature. In this work, high concentration of Nd³⁺ doped YAG microparticles were synthesized via the wet chemical reaction method. The crystal structure and photoluminescence were studied in detail for the laser applications.

Experimental

Nanosized Nd:YAG powders were prepared via wet chemical reaction method taking Y(NO₃)₃, Al(NO₃)₃ and Nd(NO₃)₃ as starting raw materials. Nd³⁺ doping was fixed at 5 mol%. The flowchart of the synthesis process has been summarized in Fig. 1. The X-ray diffraction (XRD) patterns of the prepared samples were recorded using a Rigaku, Ultima IV X-Ray diffractometer with a Cu target ($\lambda = 0.154056$ nm).

The morphologies and particle sizes were investigated using a scanning electron microscope (SEM, Hitachi S-800). The emission spectra in the wavelength range from 850 to 1500 nm were recorded by exciting the samples using a 808 nm diode laser through a monochromator that was attached to a photomultiplier tube.

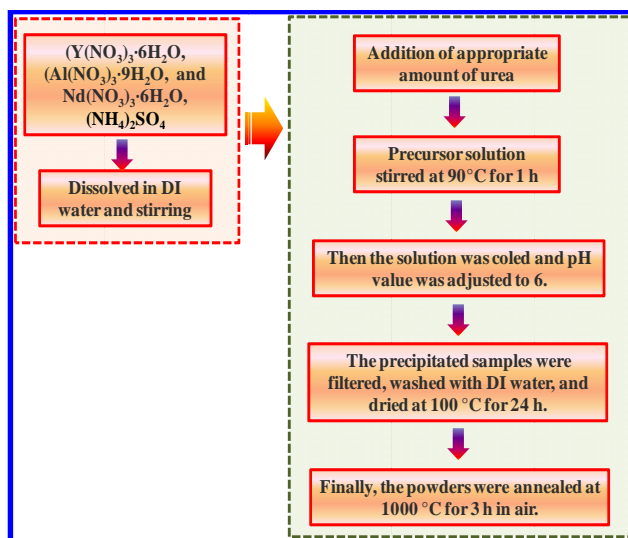


Fig. 1: Flow chart for the synthesis of Nd:YAG powders

Results and discussions

Figure 2(a) shows the XRD patterns of the obtained powders annealed at 1000 °C. According to Fig. 2(a), the XRD patterns of the powders are consistent with those of the standard YAG of JCPDS Card No. 88-2048. Because of the heat treatment in air, a complete transformation of the amorphous powders to the YAG phase was occurred.

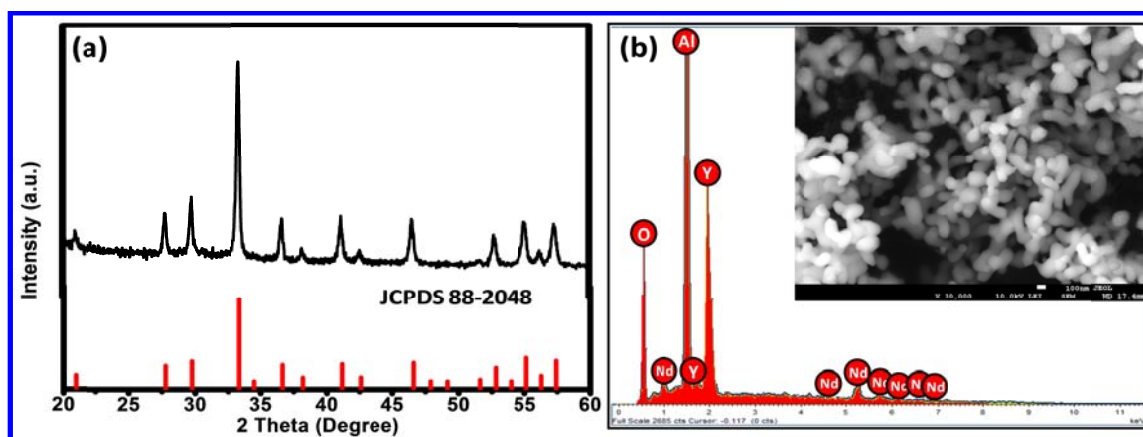


Fig. 2 (a) XRD and (b) EDX pattern of Nd:YAG powders. Inset: SEM of of Nd:YAG powders.

The elemental analysis of prepared sample was carried out via a energy dispersive X-ray spectroscopy. According to the EDX results shown in Fig. 2(b), all the elements Y, Al, O, and Nd are present in the sample stoichiometrically, and indicating the formation of required sample. The inset of Fig. 2(b) shows the SEM micrograph of the heat treated Nd:YAG powders. The micrographs clearly indicate that the crystallites are less agglomerated, distinct, moderately uniform, and have nearly spherical morphology. According to the SEM results, the synthesized powders have average particle size of 260 nm, and spherical in nature.

Figure 3(a) shows the reflectance spectra of Nd:YAG powders. The main absorption bands are indicated in the Fig. 3(a). The absorptions at around 585, 680, 740 and 808 nm are attributed for the absorption transitions from $^4I_{9/2}$ to the excited states, states $^4H_{11/2}$, $^4F_{9/2}$, ($^4F_{7/2}$, $^4S_{3/2}$) and $^4F_{5/2}$ of Nd^{3+} ions, respectively. However, the absorption corresponding to $^4I_{9/2} \rightarrow ^4F_{5/2}$ transition (808 nm)

was found to be stronger than the others. According to the reflectance spectra, the sample can be efficiently excited into $4f-4f$ Nd^{3+} absorption transitions and able to exhibit strong Nd^{3+} emission in near IR region. Figure 3(b) illustrates the emission spectra of Nd:YAG powders. As seen in Fig. 3(b), the spectra consist of groups of narrow lines in the range of 925–1450 nm, which correspond to the characteristic $f-f$ transitions between $^4\text{F}_{3/2}$ and $^4\text{I}_J$ ($J = 9/2-13/2$). The most intense emission was observed for $^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$ transition of Nd^{3+} ions at about 1064 nm. Such intense IR emission can be suitable for fabricating solid state laser.

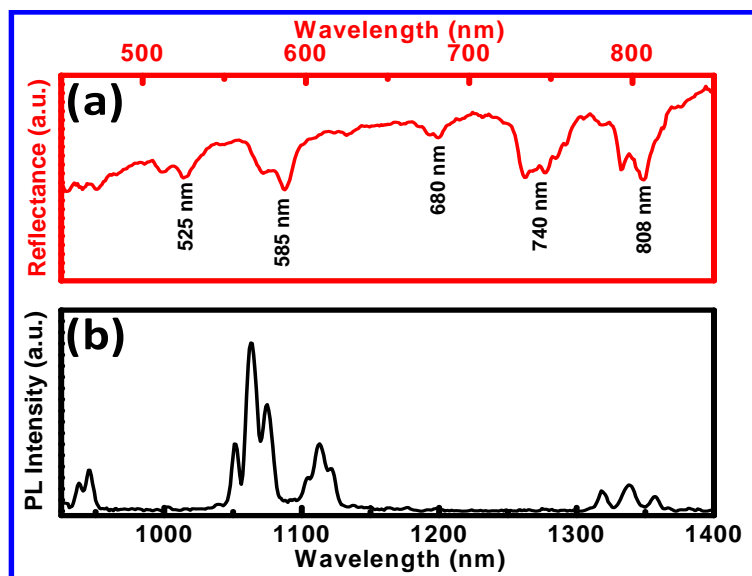


Fig. 3 (a) Reflectance and (b) PL spectra of Nd:YAG powders.

Conclusions

Herein, fine Nd:YAG powders were synthesized via a wet chemical reaction method followed by heating in air. The wet reaction promoted the homogeneity of Nd^{3+} ions in to the YAG host and yielded nanocrystalline powders. The present samples can absorb lights in the range of 925–1450 nm very efficiently and exhibited intense near IR emission at around 808 nm owing to the $^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$ transition of Nd^{3+} ions at about 1064 nm. The present powders can be suitable for laser applications.

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