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Optical and Near-Infrared Scintillation Properties of Nd^{3+}-Doped YVO_4 Crystals

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In this paper, we report on the optical and near-infrared (NIR) scintillation properties of YVO_4:Nd (0.5 and 3.0 mol%) crystals grown by the conventional floating zone technique. In optical transmission spectra, a number of absorption lines owing to Nd^{3+} appeared in the ultraviolet (UV)–NIR wavelength regions. Photoluminescence and X-ray-excited radioluminscence spectra showed characteristic Nd^{3+} emission peaks centered at approximately 1064 and 1319 nm, which correspond to the transitions from the $^4F_{3/2}$ excited state to the $^4I_{11/2}$ and $^4I_{13/2}$ states of Nd^{3+}. The NIR scintillation intensities showed good linearity to irradiation dose in the range from 1 to 1000 mGy X-ray. Thermal quenching of the NIR scintillation for the 3.0 mol% Nd-doped crystal was less than that of the 0.5 mol% Nd-doped crystal.

1. Introduction

Solid-state scintillation materials are widely used in ionizing radiation detection systems for security inspection to medical imaging, well logging, and radiation monitoring at nuclear power plants.[1–4] Common systems are based on the detection of visible (VIS) and ultraviolet (UV) light emitted by scintillators excited by high-energy photons or charged particles. Meanwhile, the scintillation light in the near-infrared (NIR) region is not exploited for detection systems. The utilization of NIR scintillation can expect to lead to a new scintillator that has high light output and good energy resolution because the energy of an NIR quantum (1.5–0.5 eV) is smaller than that of a visible photon. Some researchers reported the NIR scintillation characteristics of various materials, such as oxides, halides, and metals.[5–9] Our group has recently studied some oxide laser materials activated with rare-earth ions, and they were found to show intense NIR $4f^n$–$4f^n$ emission peaks and good irradiation dose response under exciton with X-rays.[10–13] Thus, the present study focuses on a Nd^{3+}-doped yttrium vanadate YVO_4 crystal because it is one of the most efficient laser host crystals that provide 1064 nm light owing to the Nd^{3+} ions.[14–16] The scintillation characteristics of the undoped YVO_4 crystal have been studied, and the crystal was found to exhibit the blue emission band owing to the transition from the triplet state of $(VO_4)^{3-}$.[17,18] To the best of our knowledge, the

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NIR scintillation characteristics of the Nd^{3+}-doped YVO_4 crystal have not been clearly understood up to now. In this work, we report and discuss the optical (transmittance, photoluminescence) and scintillation (radioluminescence, dose response, temperature dependence) properties of the Nd^{3+}-doped YVO_4 crystals with different Nd concentrations.

2. Experimental Procedure

Single-crystal samples of 0.5 and 3.0 mol% Nd-doped YVO_4 were produced by OXIDE Corporation by the conventional floating zone technique. The as-grown crystal was cut and polished for optical and radiation measurements, as shown in Fig. 1. The optical transmission spectra of the Nd-doped YVO_4 crystals were recorded on a V-670 spectrometer (JASCO) in the wavelength range from 190 to 2700 nm. The photoluminescence and radioluminescence spectra of the Nd-doped YVO_4 were recorded on a SR-163 spectrometer in combination with a DU492A-1.7 charge-coupled device (CCD) detector (Andor), which was cooled to 210–230 K by a Peltier module.[10–13] A xenon-lamp was used as the excitation source for photoluminescence measurements. In the radioluminescence ones, the X-ray source was our original X-ray generator (XRB80P Monoblock, Spellman) equipped with a tungsten target. The generator was supplied with a voltage of 60 kV and current of 2.5 mA. The NIR photons emitted from the crystals were sent to the spectrometer through an optical fiber to avoid the direct irradiation of the CCD by the excitation sources. The dose response of the NIR scintillation was also evaluated with similar instruments. X-rays at rates of 1, 10, 100, and 1000 mGy/min irradiated the crystals and the integrated Nd^{3+} NIR emission peak intensities were recorded. The temperature dependence measurement of the radioluminescence spectra was demonstrated using a constant-temperature bath (Espec SH661). The crystal sample and optical fiber were set in the temperature bath and the X-ray was irradiated to the sample through an irradiation port. The X-ray generator and CCD-based spectrometer connected to the optical fiber were outside the temperature bath.

3. Results and Discussion

Figure 2 illustrates the recorded transmission spectra of the Nd-doped YVO_4 crystals. In the spectra, the crystals showed more than 70% transmittance for wavelength from the UV to the NIR region. Additionally, a large number of absorption lines were observed in the wavelength range for the crystals and the absorption line intensities increased with increasing Nd concentration.

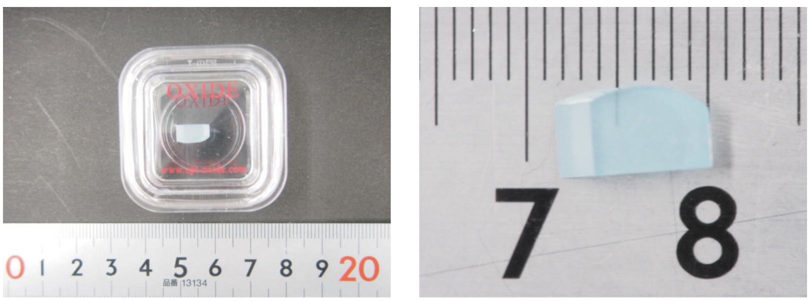

Fig. 1. (Color online) Polished crystal samples of Nd-doped YVO_4 grown by floating zone method.

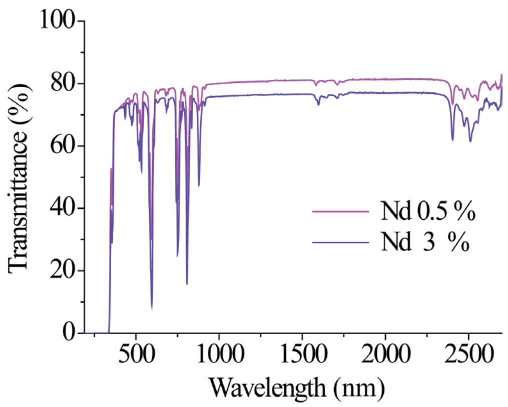

Fig. 2. (Color online) Optical transmission spectra of the Nd-doped YVO_4 crystals.

The absorption lines are due to $4f^n$–$4f^n$ forbidden transitions of Nd^{3+}.(12) The photoluminescence spectra of the Nd-doped YVO_4 crystals under excitation at 800 nm are presented in Fig. 3. The excitation wavelength of 800 nm is a typical pump wavelength for laser oscillation.(14–16) In the spectra, dominant NIR emission peaks at 1064 and 1319 nm were observed, which correspond to transitions from the $^4F_{3/2}$ excited state to the $^4I_{11/2}$ and $^4I_{13/2}$ states.(12–16) Figure 4 illustrates the radioluminescence spectra of the Nd-doped YVO_4 crystals. Like photoluminescence spectra, the emission peaks at 1064 and 1319 nm appeared and these peaks were ascribed to the $^4F_{3/2}\rightarrow{}^4I_{11/2}$ and $^4I_{13/2}$ transitions of Nd^{3+}. In particular, the 0.5 mol% Nd-doped crystal exhibited higher NIR emission peak intensities than the 3.0 mol% Nd-doped crystal. Scintillation is usually dependent sensitively on the energy transfer process from the host crystal to the emission center.(19,20) Energy migration under UV excitation between the $(VO_4)^{3-}$ molecular complex in the YVO_4 crystal and the emission centers with rare-earth ions has been reported in other papers.(21,22) Thus, it is suggested that the result is related to the energy migration from the YVO_4 host crystal to the Nd^{3+} emission center. In this case, the energy transfer efficiency for the 0.5 mol% Nd-doped crystal maybe higher than that for the 3.0 mol% one. The dose response of the NIR emission intensity is plotted in Fig. 5. The NIR emission intensities linearly increased with increasing irradiation dose for both Nd-doped YVO_4 crystals. Using our experimental setup, a good linear response of the NIR scintillation was observed in the 1–1000 mGy X-ray dose range. Figure 6 illustrates the variation of the characteristics with the temperature of the NIR scintillation. It is observed that for the 0.5 mol% Nd-doped crystal, the NIR peak intensities markedly changed with temperature. The peak intensities decreased with increasing temperature from −60 to 140 °C. The observed NIR scintillation intensity is proportional to the product of the energy migration efficiency from the host to Nd^{3+} ions and the emission efficiency of the excited state of the Nd^{3+} ions. The temperature dependence of the NIR scintillation intensity shown in Fig. 6 is discussed on the basis of the temperature dependence of the energy migration and emission efficiency. Because the emission is due to the transition in 4f orbitals, which is well shielded by the filled 5s and 5p subshells, it is reasonable to assume that the temperature dependence of the emission efficiency is weak. Thus, the observed temperature dependence of the NIR scintillation intensity is mainly attributed to that of the energy migration efficiency. For the 0.5 mol% Nd-doped crystal, the thermal quenching during the energy migration is more pronounced.

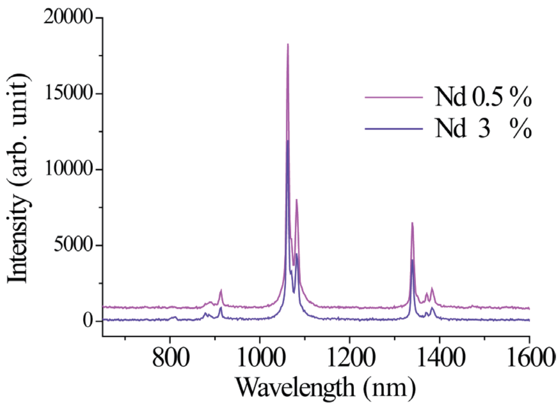

Fig. 3. (Color online) Photoluminescence spectra of the Nd-doped YVO_4 crystals under excitation at 800 nm.

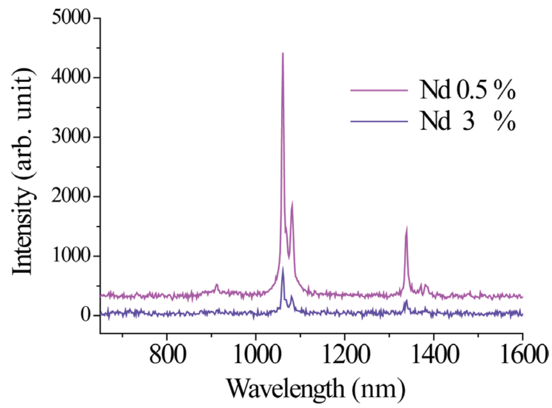

Fig. 4. (Color online) X-ray-excited radioluminescence spectra of the Nd-doped YVO_4 crystals.

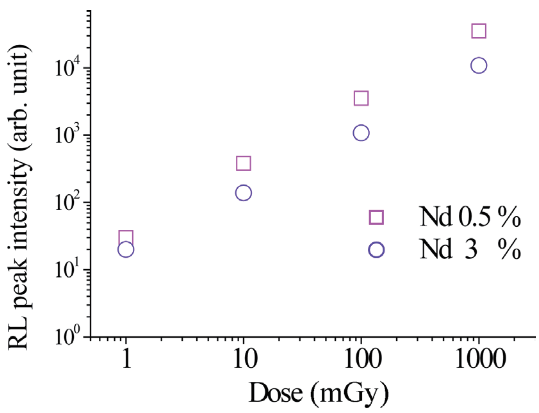

Fig. 5. (Color online) Irradiation dose response of NIR scintillation of the Nd-doped YVO_4 crystals.

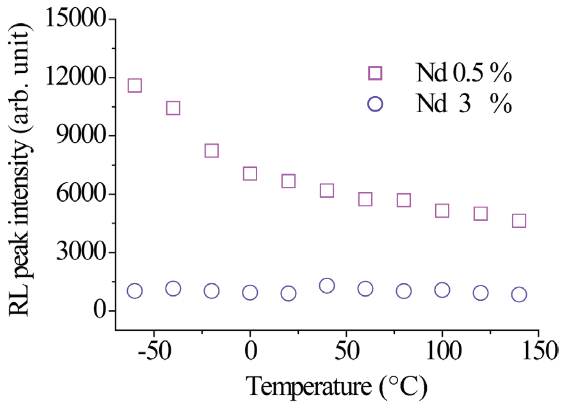

Fig. 6. (Color online) Temperature dependence of NIR scintillation of the Nd-doped YVO_4 crystals.

4. Summary

We presented the optical and NIR scintillation characteristics of 0.5 and 3.0 mol% Nd-doped YVO_4 crystals. The crystal samples were grown by the conventional floating zone technique. In the transmission spectra of the Nd-doped crystals, sharp absorption lines attributed to the Nd^{3+} $4f^n$–$4f^n$ transitions were observed at wavelengths from the UV to NIR regions. The photoluminescence spectra of the Nd-doped crystals showed Nd^{3+} 4f–4f forbidden transitions ($^4F_{3/2}\rightarrow{}^4I_{11/2}$, $^4I_{13/2}$) at 1064 and 1319 nm. When the X-ray excited the Nd-doped crystals, characteristic Nd^{3+} NIR emission peaks appeared as well as the photoluminescence. The Nd-doped crystals exhibited Nd^{3+} NIR emission proportional to irradiation dose in the dose range from 1 to 1000 mGy. From this study, the Nd-doped YVO_4 crystals were found to have potential for radiation monitoring in the charge integrating mode.

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