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Near-infrared Scintillation Properties of Neodymium-doped Cadmium-rich Borate Glasses

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Cadmium-rich borate glasses doped with different Nd concentrations were synthesized by the conventional melt-quenching technique, and their photoluminescence (PL) and scintillation properties were investigated. They exhibited PL and scintillation in the near-infrared region, originating from Nd^{3+}. The 3.0% Nd-doped sample exhibited the highest scintillation intensity among all the samples, with a lower detection limit of 7.5 mGy/h.

1. Introduction

Many radiation detectors use phosphors that convert the energy of radiation into low-energy photon emission, enabling indirect detection coupled with photodetectors.[1] These phosphors can be classified as storage phosphors or scintillators. The former ones store absorbed radiation energy in the form of trapped carriers, which afterward recombine under thermal or optical stimulation to emit photons, allowing the estimation of the irradiated dose within a certain period of time, from the emission intensity.[2] The latter ones emit photons instantly upon irradiation.[3] These two types of phosphor are essential in various radiation measurement fields, including high-energy physics,[4,5] security,[6–9] environmental monitoring,[10–12] medical diagnosis,[13–16] radiation therapy,[17,18] and individual dose monitoring.[19,20] As different applications require different properties of these phosphors, research and development on them is ongoing in several material forms, including crystals,[21–38] glasses,[39–52] and ceramics.[53–56]

Traditionally, scintillators emitting ultraviolet–visible (UV–vis) photons have been developed, because common photodetectors are sensitive in that range. Recently, photodetectors responsive to NIR photons have become available, enabling research on NIR-emitting scintillators for monitoring high-radiation fields,[57] such as nuclear power plants. In the proposed technique,[58] scintillation light is transmitted via an optical fiber to a remote photodetector, avoiding radiation damage to the photodetector and electrical circuits. In this application, enough distance (at least several hundred meters) from the radiation field is important to avoid the radiation exposure of operators. UV–vis photons are inefficiently transmitted through optical fibers hundreds of meters long,[59] especially when the fiber is

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radiation-damaged,[60,61] whereas NIR photons can propagate effectively, driving active research on NIR-emitting scintillators.

Neodymium-doped cadmium-rich borate glasses are promising materials for NIR laser applications.[62] They are also attractive candidates for high-performance NIR-emitting scintillators, because their cadmium-rich borate host likely exhibits high radiation stopping power; the host glass has a density of 3.97 g/cm^3[62] and an effective atomic number[3] (Z_{eff}) of 45, which are comparable to those of the NaI scintillator (density: 3.7 g/cm^3,[63] Z_{eff}: 51). Although their optical and basic physical properties have been studied with Nd-doped[62] and nondoped[64] cadmium-rich borate glasses, respectively, the scintillation characteristics of the Nd-doped glasses remain unexplored. In this study, we investigated the NIR scintillation properties of these glasses.

2. Materials and Methods

Neodymium-doped cadmium-rich borate glass samples were synthesized by the conventional melt-quenching technique. An x% Nd-doped 80CdO–20B_2O_3 glass sample (x = 0.3, 1.0, 3.0, and 10.0) was prepared from Nd_2O_3, CdO, and B_2O_3 powders mixed at a molar ratio of x/2: 80:20, melted at 1200 °C for 1 h and quenched to 300 °C. Disk-shaped glass plates were obtained and then cut and polished into 8.0 × 8.0 × 1.3 mm^3 plates. Following the synthesis, their photoluminescence (PL) and scintillation properties were investigated in the same sequence as in previously reported NIR-emitting glasses,[65–67] in line with tradition.

3. Results and Discussion

Figure 1 shows the appearance and XRD patterns of the synthesized samples. All the samples appeared to be homogeneous and transparent. All the XRD patterns exhibited an amorphous halo, suggesting successful glass synthesis. Figure 2 shows their diffuse transmittance spectra. All the samples exhibited a high transmittance (~80%) in the NIR region, demonstrating efficient emitted photon extraction. Sharp absorption peaks corresponding to 4f-4f transitions of Nd^{3+}[62,68–70] were observed, and their intensity increased with Nd concentration.

Figure 3(a) shows the PL excitation/emission map of the 1.0% Nd-doped sample as a representative, exhibiting three NIR emission lines at 890, 1060, and 1320 nm, corresponding to the $^4F_{3/2}\rightarrow{}^4I_{9/2}$, $^4F_{3/2}\rightarrow{}^4I_{11/2}$, and $^4F_{3/2}\rightarrow{}^4I_{13/2}$ transitions of Nd^{3+},[68,71–75] respectively. The other samples showed similar spectral shapes with different intensities. Figure 3(b) shows the PL decay curves and decay time constants of the samples. The decay time constants of the 0.3, 1.0, and 3.0% Nd-doped samples were 104, 93, and 57 μs, respectively. These decay time constants around 100 μs at low Nd concentrations of 0.3 and 1.0% agreed well with those of the previously reported Nd-doped cadmium-rich borate glass laser media.[73] The shorter decay time of the 3.0% Nd-doped sample was probably due to concentration quenching. The 10.0% Nd-doped sample exhibited a decay curve similar to the instrumental response function (IRF), and its decay time could not be estimated. This is probably due to the intense effect of concentration quenching. The PL QY values of the 0.3, 1.0, and 3.0% Nd-doped samples were 31, 21, and 8%,

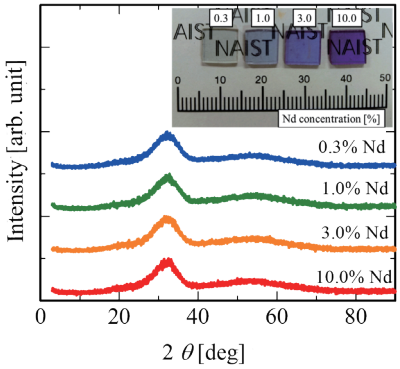

Fig. 1. (Color online) Appearance and XRD patterns of the samples.

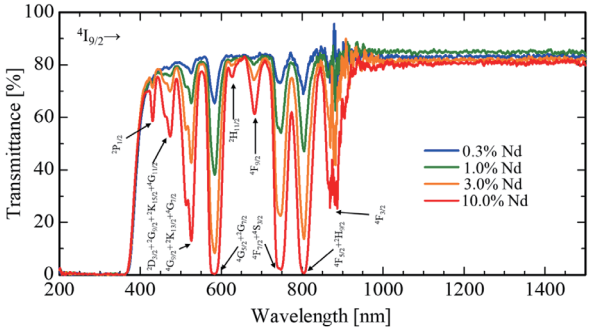

Fig. 2. (Color online) Diffuse transmission spectra of all the samples in the UV–NIR region.

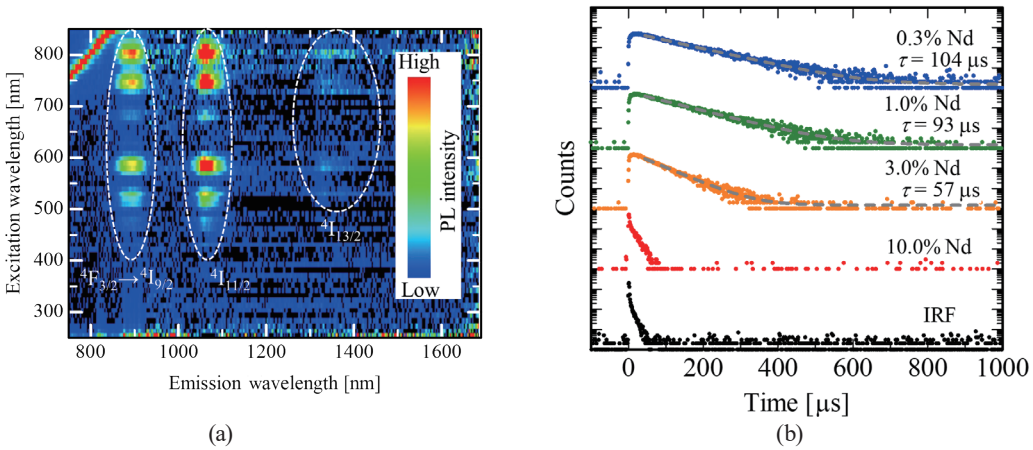

Fig. 3. (Color online) (a) PL excitation/emission map of the 1.0% Nd-doped sample and (b) PL decay curves of all the samples.

respectively, further confirming the quenching effect, and the PL QY of the 10.0% Nd-doped sample could not be measured, owing to its low PL intensity resulting from the intense effect of concentration quenching.

Figure 4(a) shows the scintillation spectra of all the samples under X-ray excitation, exhibiting peaks at 890, 1060, and 1320 nm, corresponding to the $^4F_{3/2}\rightarrow{}^4I_{9/2}$, $^4F_{3/2}\rightarrow{}^4I_{11/2}$, and $^4F_{3/2}\rightarrow{}^4I_{13/2}$ transitions of Nd^{3+}, respectively.[66,67,76] The $^4F_{3/2}\rightarrow{}^4I_{9/2}$ emission shifted to longer wavelengths with increased Nd concentration, likely due to the $^4I_{9/2}\rightarrow{}^4F_{3/2}$ self-absorption near 880 nm as shown in Fig. 2. Figure 4(b) shows the scintillation decay curves, each consisting of fast and slow components attributed to the IRF and scintillation decay, respectively. The decay constants for the 0.3, 1.0, 3.0, and 10.0% Nd-doped samples were 82, 76, 50, and 22 μs, respectively, consistent with the 4f–4f transitions of Nd^{3+}[66,67] and similar to their PL decay time.

Figure 5 shows the X-ray dose rate response functions of the samples. The 3.0% Nd-doped glass exhibited the highest scintillation intensity among the samples, with a linear response in the dose range of 15–45000 mGy/h. The lower detection limit of X-ray dose rates was estimated as 7.5 mGy/h by the 3σ method. This detection limit demonstrated that the 3.0% Nd-doped

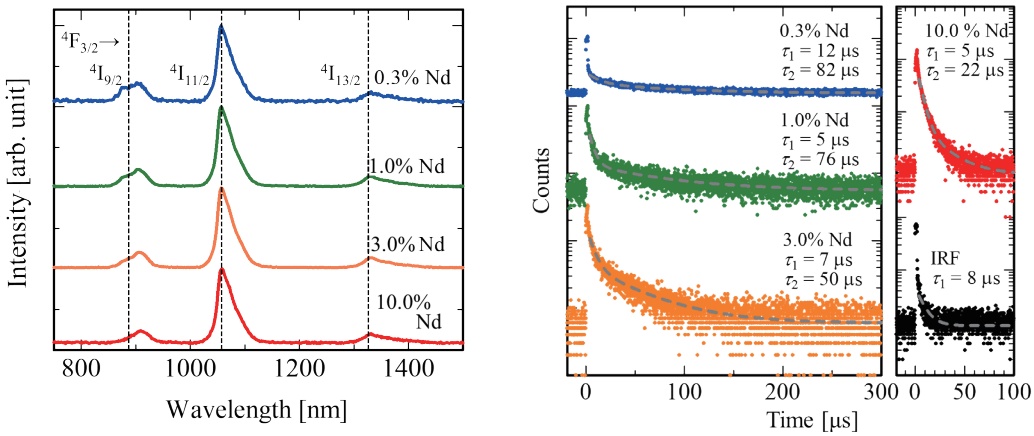

Fig. 4. (Color online) (a) Scintillation spectra and (b) scintillation decay curves.

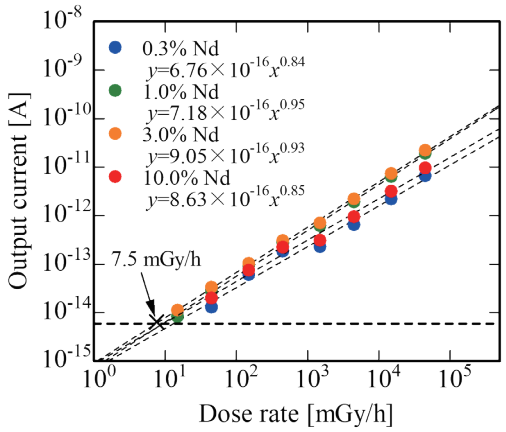

Fig. 5. (Color online) Dose rate response functions of the samples.

$80CdO\text{-}20B_2O_3$ glass can be used for monitoring the primary loop area of light water reactors during operation and the near reactor vessel of light water reactors during inspection.[58]

Although scintillation intensity theoretically correlates with PL QY,[77] the 3.0% Nd-doped sample outperformed the 0.3% Nd-doped sample, which exhibited the highest PL QY. This suggests enhanced energy transfer from the glass matrix to Nd^{3+} centers with increasing Nd concentration. It has already been shown that the energy transfer efficiency of a scintillator inversely correlates with the thermoluminescence intensity;[78] therefore, we will investigate their afterglow (i.e., thermoluminescence at room temperature) properties in the future to understand the energy transfer mechanism in depth.

4. Conclusions

$80CdO\text{-}20B_2O_3$ glasses with 0.3, 1.0, 3.0, and 10.0% Nd concentrations were successfully synthesized by the melt-quenching technique. The amorphousness and transparency of the samples were respectively confirmed from the XRD patterns and diffuse transmittance spectra. The samples exhibited PL and scintillation in the NIR region, and the origin of both types of luminescence was confirmed to be 4f–4f transitions of Nd^{3+} from the obtained PL and scintillation decay curves, respectively. The dose rate response measurement revealed that the 3.0% Nd-doped sample exhibited the highest scintillation intensity among all the samples. The lower detection limit with the 3.0% Nd-doped sample was 7.5 mGy/h, and this glass can be applied to the radiation monitoring of certain areas of nuclear power plants.

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