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15.5 A $SrBPO_5:Eu^{2+}$ phosphor for neutron imaging

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Abstract

A $SrBPO_5:Eu^{2+}$ phosphor material has been investigated for neutron imaging. This phosphor showed photostimulated luminescence (PSL) by illumination of 635 nm laser light after X-ray irradiation. The spectral characteristics of the phosphor were similar to those of $BaFBr:Eu^{2+}$, which is a commonly used phosphor of imaging plates. In addition, we found that this phosphor also showed PSL for neutron irradiation. It comes from the fact that it contains atomic boron in base matrix. Therefore, this phosphor can be used for neutron imaging without adding neutron sensitive materials such as Gd in commercially available neutron imaging plates. The PSL intensity and the neutron detection will be increased by using enriched boron instead of natural boron.

1. Introduction

The imaging plate (IP) made of storage phosphors is one of the most promising alternatives to conventional position sensitive detectors (PSDs) for nuclear radiation [1]. The IP has good properties as a PSD such as a wide-dynamic range, high-spatial resolution, large-detection area, and reusability. Therefore, the IP, originally developed for a two-dimensional PSD for medical diagnostics [2], is also used X-ray diffraction experiments [3]. The neutron imaging plate (NIP) has been also developed, where the plate contains neutron sensitive materials such as Gd [4]. Although the NIP has made a great success in the field of neutron scattering study, there is a disadvantage when the NIP is used for neutron detection in a field where both neutron and gamma ray exist. Since the NIP is also sensitive to gamma ray, it is difficult to discriminate neutron signal from gamma ray one when the NIP is read out. To overcome the problem, the authors have been searching new storage phosphors consisting of light materials.

Recently, a new $SrBPO_5:Eu^{2+}$ storage phosphor was reported [5], where the phosphor shows photoluminescence and photostimulated luminescence (PSL) due to Eu^{2+} transition of 390 nm. The phosphor has a low density compared to that of $BaFBr:Eu^{2+}$, which is usually used as a phosphor material of IP. It will be favorable for reducing gamma ray influence on the signal in the field where both neutron and gamma ray exit because phosphors with lower densities are less sensitive to gamma ray. The report motivated us to make the phosphor and to investigate the characteristics. In this paper, spectral characteristics of the $SrBPO_5:Eu^{2+}$ storage phosphor are described.

2. Experimental

Starting materials to prepare powder samples were $SrCO_3$, H_3BO_3, $(NH_4)_2HPO_4$, and $EuCl_3 \cdot 6H_2O$. Purity of these materials was more than 99.5%. After appropriate amounts of them were mixed in a mortar for 20 minutes, the resulting mixtures were transferred into an aluminum crucibles covered with lid for carrying out the solid-state reaction. Finally Eu^{2+}-doped boro phosphate ($SrBPO_5:Eu^{2+}$) samples were obtained by firing them in a muffle furnace in a nitrogen atmosphere with a computer-controlled heating schedule at 600℃ for 2 hours and at 800℃ for 2 hours. The flow rate of nitrogen gas is 2 liters/min.

Photoluminescence spectra of the samples were measured using a Hitachi F-2500 spectroflourometer at room temperature. PSL decay characteristics were measured using an experimental setup shown in Fig.1. For this purpose, the samples were continuously illuminated by a He-Ne laser (Audio Technica SU-31E, 6 mW) after irradiation by X-ray or neutrons. To eliminate the background light, a filter (made by MeresGriot) was set in front

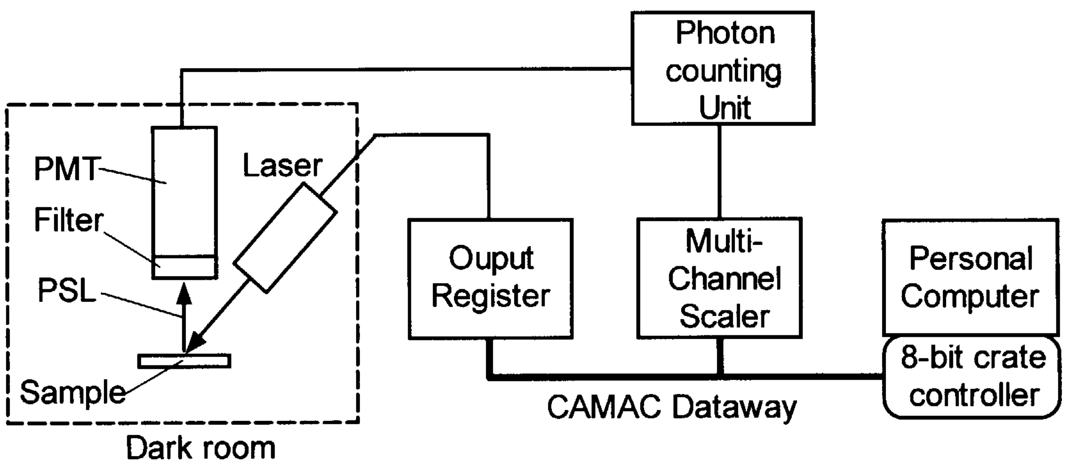

Fig.1 The experimental setup for PSL measurement.

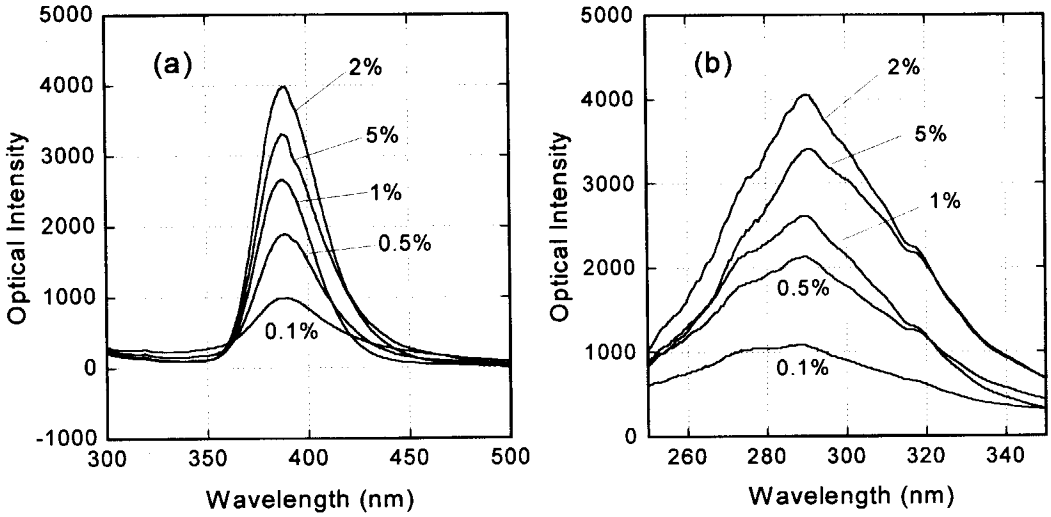

Fig.2 (a) Photoluminescence emission spectra and (b) excitation spectra of the samples.

of the photomultiplier tube (Hamamatsu R647P). The signal from the photomultiplier tube were amplified and discriminated from noise by a photon-counting unit (Hamamatsu C3866). The output pulses from the unit were counted by a multi-channel scaler (Hoshin C022). An output resister (Hoshin C004) was used to control the laser. The multi-channel scaler and the output resister were controlled by a personal computer through an 8-bit crate controller (Hoshin CCP-F).

3. Results and Discussions

Figure 2 (a) shows photoluminescence emission spectra of samples excited by 290 nm UV light. The concentrations (mol %) of Eu in the samples were 0.1%, 0.5%, 1%, 2%, and 5%. There is a broad peak at about 390 nm that is attributed to the transition of 5d-4f of Eu^{2+}. The photoluminescence spectrum was similar to that of $BaFBr:Eu^{2+}$. The lifetime of the 390 nm emission from the samples was estimated to be 0.67 μs [6] while that from a commercially available IP containing $BaFBr:Eu^{2+}$ (Fuji Film Co. Ltd.) was 0.80 μs, which corresponds to a value in the literature [7]. Although an optimal concentration of about 1% has been reported [5], it may exist between 2% and 5% in our experiment. Figure 2(b) shows photoluminescence excitation spectra of the samples while the emission at 390 nm was monitored. There was a broad peak at about 290 nm. The dependency of the peak intensity on the Eu concentration is the same as that of photoluminescence emission spectra.

Figure 3 shows PSL decay curves of luminescence at 390 nm of an X-ray irradiated sample, which was taken with the experimental apparatus shown Fig.1. The concentration of Eu in the sample was 1%. The X-ray irradiation was carried out for 2 minutes at KEK BL-27A irradiation facility. The X-ray energy and flux were estimated to be about 3 keV and 2.8 x 10^{10} $cm^{-2}s^{-1}$, respectively. Results of the same sample that was stored for 27 hours

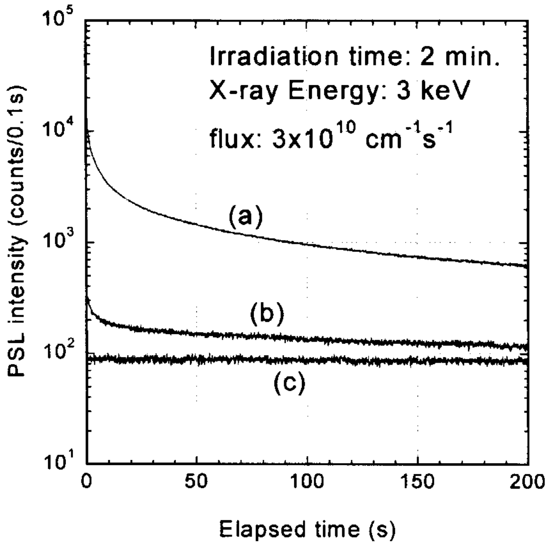

Fig. 3 PSL decay curves of luminescence of 390 nm of an X-ray irradiated sample. (a) just after the irradiation, (b) then stored in the dark 27 hours, and then (c) optically bleached by a white lamp for 5 min.

in the dark after the measurement and then optically bleached for 5 minutes, were also shown in the same figure. Since no photoluminescence could not be observed in unirradiated samples (not shown in the figure) and optically bleached samples, the emission at 390 nm is not due to up-conversion or multi-photon processes but to a PSL phenomenon.

As stated above, the NIP containing Gd_2O_3 as a neutron sensitive material is very effective in many fields such as neutron protein crystallography and neutron radiography. However, storage phosphors containing neutron sensitive materials other than Gd_2O_3, such as boron, have not been reported yet (Storage phosphors containing atomic lithium was trialy fabricated and reported in Refs. [4], [8]). One of the reasons may be that the ranges of secondary particles produced by nuclear reactions of boron with neutrons are very short in the phosphor. For example, the range of an alpha particle produced by a reaction of $^{10}B(n,\alpha)^7Li$ is estimated to be 5.3 μm in the BaFBr:Eu^{2+} phosphor. The range of ^7Li is much shorter than that of the alpha particle. Since the phosphor should contain an organic binder and the volume filling factor is less than 100%, it is almost impossible to store radiation image in the phosphor by adding ^{10}B materials. However, the atomic ^{10}B is attractive for neutron detection because of the large neutron cross section of 3840 barn and production of high energy secondary particles. If neutron image storage phosphors using $^{10}B(n,\alpha)^7Li$ reaction can be found, the phosphors will be a candidate for a new neutron imaging plate. Since the $SrBPO_5:Eu^{2+}$ phosphor investigated in this work has atomic boron in the base matrix and the natural abundance of neutron sensitive ^{10}B in natural boron is about 20 %, there is a possibility that the phosphor itself is sensitive to neutrons without adding any neutron sensitive materials.

The neutron irradiation on samples was carried out for 2 seconds at JAERI JRR-3M neutron radiography (NRG) irradiation facility. The neutron flux was 2 x 10^8 $cm^{-2}s^{-1}$. Although the NRG facility can provide a neutron field, there also exists gamma ray in the field. Therefore, to estimate the influence of gamma ray on the sample, two samples with the same Eu concentration of 2% were prepared and irradiated at the same time. One was set before 4 pieces of a Li block with a thickness of 1 cm (sample A) and another was set

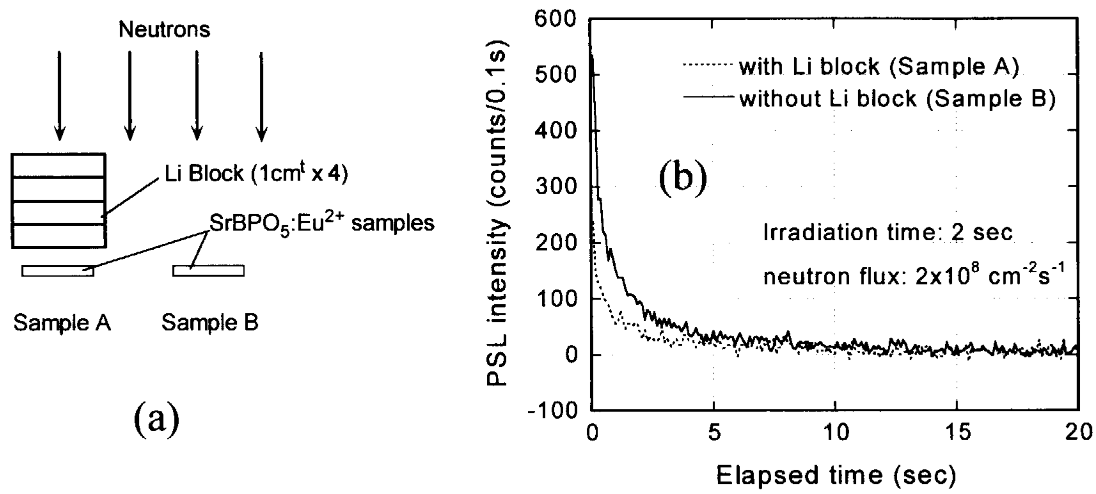

Fig. 4 (a) Experimental setup of the two samples. (b) The PSL decay curves of luminescence of 390 nm after neutron irradiation.

without any Li blocks (sample B), as seen in Fig. 4 (a). The influence of the Li blocks on neutron and gamma ray fluxes has been estimated in Ref. [8]. Incident neutron flux on the sample A was attenuated with the Li blocks and could be ignored while no affect on gamma ray flux was observed.

Figure 4 (b) shows PSL decay curves of luminescence at 390 nm of samples A and B. The output from the sample B shows PSL by both neutrons and gamma ray. The output from the sample A shows PSL by gamma ray. Therefore, the difference of the outputs between two samples corresponds to PSL by actual neutrons. The figure clearly shows that the sample itself was sensitive to neutrons. The neutron detection efficiency, E_f, of the phosphor can be calculated by a well-known formula

$$E_f = 1 - \exp(-\kappa N \sigma t) \qquad (1)$$

where κ is a filling factor, N is an atomic density of ^{10}B, σ is the total neutron cross section, and t is the thickness of the phosphor. Figure 5 shows the calculated detection efficiency of thermal neutrons (0.025eV) as a function of the effective thickness κt. The detection efficiency of the phosphor with a thickness of 200 μm and $\kappa = 0.6$ is estimated to be 7.7% at thermal neutrons. However, it will be 31 % by replacing natural boron in the phosphor by enriched boron (93%).

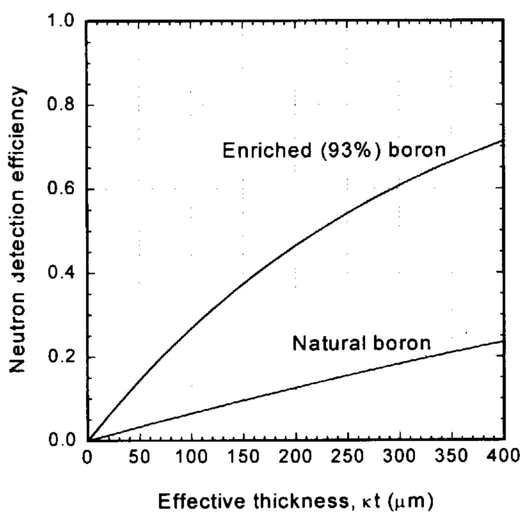

Fig. 5 The calculated detection efficiency as a function of the effective thickness.

4. Summary

Spectral characteristics of a SrBPO5:Eu^{2+} phosphor were investigated. The phosphor shows intense photoluminescence at 390 nm by UV ray excitation. The spectrum was similar to that of BaFBr:Eu^{2+} which is commonly used for an X-ray storage phosphor. The optimal Eu concentration for the luminescence existed between 2 and 5 (mol %). The phosphor shows PSL by illumination of 635 nm laser light after X-ray irradiation. It was also confirmed that the phosphor itself showed PSL after neutron irradiation without adding any neutron sensitive materials. The storage phosphor which is sensitive to neutrons through a nuclear reaction of $^{10}B(n,\alpha)^7Li$ will be a candidate for a new neutron imaging plate. Although the PSL intensity was relatively weak and the neutron detection efficiency was relatively low compared to those of commercially available NIPs, they will be increased by replacing natural boron in the phosphor by enriched boron.

5. References

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