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# Search for luminescent materials under 14 MeV neutron irradiation

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**Abstract**

A radiation detection system utilizing radiation-resistant optical fibers and luminescent materials will have several advantages, such as compactness and robustness, and can be a strong candidate for diagnostics of fusion neutrons. Luminescent materials – important components of an optical system – that exhibit luminescence under fast neutron irradiation were studied. Commercially available luminescent materials, such as ZnS:Ag, ZnS:Cu, $SrAl_2O_4$:$Eu^{2+}$, $Dy^{3+}$, and $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ exhibit luminescence under 14 MeV neutron irradiation. The materials excluding ZnS:Ag showed radiation resistance for a fast neutron fluence of up to $10^{14}$ n/cm$^2$. The luminescence characteristics of $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ that have three apparent peaks were also examined under proton irradiation. The compounds exhibited the same luminescence peaks under fast neutron irradiation, which can be attributed to their dopants, namely, $Dy^{3+}$ and $Eu^{2+}$, and the intensity ratio of $Eu^{2+}$ to $Dy^{3+}$ showed dependence on incident energy and radiation.


## 1. Introduction

Several features are desired for radiation-flux monitors in a fusion reactor such as the ITER. The monitors require features such as a good radiation resistance, wide dynamic range, wide and detailed detection area, and robustness against electrical noises; this is because the monitor is usually placed in a harsh environment including under heavy irradiation and in a large electromagnetic field. Furthermore, monitors require simplicity and compactness for easy maintenance and deployment. In view of these requirements, we propose an optical measurement system using luminescent materials with radiation-resistant optical fiber cable. In this system, the radiation-flux is measured by the intensity of luminescence, which is transmitted from a luminescent material to an optical detector by an optical fiber cable. They are free from the requirements of the application of high electrical voltage and insensitive to electromagnetic force and radiation-induced electrical phenomena such as radiation-induced conductivity (RIC) and radiation-induced electromotive force (RIEMF) [1]. Therefore, malfunctions caused by the degradation of electrical insulations in a harsh environment are not of concern.

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Recent advances in the development of radiation-resistant fused-silica-core optical fibers have greatly enhanced the prospects for optical diagnostics in fusion reactors such as the ITER [2–5]. A fusion-neutron diagnostic system should operate reliably in a neutron flux of up to $10^9$ n/cm$^2$s and an associated gamma-ray dose rate up to a few Gy/s in an ITER-like machine. To be compatible with a realistic and cost-effective maintenance scenario, a system must endure a neutron fluence of $10^{17}$ n/cm$^2$. Among the several optical diagnostics systems, the radiation-induced luminescence devices will be candidates for the measurement of nuclear-photon intensity (which is proportional to the nuclear fusion reaction rate) and the incoming particle flux at the first wall. In this study, luminescent materials – important components of an optical system that exhibit luminescence under fast neutron irradiation – were studied with a view to apply them in DT fusion reactors.

## 2. Experiment

Fast neutron irradiations were performed at the deuterium–tritium neutron irradiation facility of the fusion neutronics source (FNS) at the Tokai Research Establishment of the Japan Atomic Energy Agency (JAEA). The energy spectrum of the neutrons exhibits a sharp peak at 14 MeV and the flux was in the range of $10^6$–$10^9$ n/cm$^2$s. The irradiation was carried out for several days with an irradiation time of 7 h per day. A schematic diagram of the irradiation setup at the FNS and a magnified image of the irradiated sample are shown in Fig. 1. Several materials that were expected to have luminescence were attached at one end of radiation-resistant optical fibers with a core diameter of 0.2 mm. The luminescence generated by the fast neutron was detected by a photonic multichannel analyzer (Hamamatsu Photonics, PMA-10) via an optical fiber cable, which covered a distance of 40 m from the irradiation area to the detector. The optical detector can measure optical signals in the wavelength range of 300–800 nm. The optical fibers connecting the luminescent materials and the optical detector were generally placed in a low radiation area and shielded from direct exposure to the fast neutrons. Thus, radiation-induced phenomena in the optical fibers were minimal.

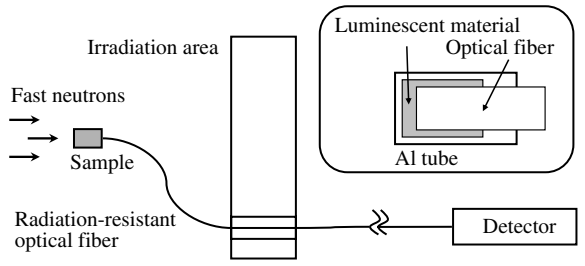


Fig. 1. Irradiation setup and magnified image of a luminescent material sample.

## 3. Results and discussion

Commercially available scintillators – silver-activated zinc sulfide (ZnS:Ag) and copper-activated zinc sulfide (ZnS:Cu) – and two types of long lasting phosphors (LLPs) – strontium aluminate doped with europium and dysprosium ($SrAl_2O_4$:$Eu^{2+}$, $Dy^{3+}$ and $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$) – exhibit fast neutron-induced luminescence. The luminescent spectra of the ZnS group (ZnS:Ag and ZnS:Cu) and the LLPs ($SrAl_2O_4$:$Eu^{2+}$, $Dy^{3+}$ and $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$) under the 14 MeV neutron irradiation are shown in Fig. 2(a) and (b), respectively. Here, we integrated the optical signals for 50 s in order to obtain clear spectra for the fast neutron-induced luminescence. All samples show intrinsic luminescent peaks, and $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ shows three apparent peaks at 400, 480, and 570 nm. The ZnS group and luminescence of LLPs at 570 nm showed an immediate end of the luminescence when the fast neutron production was stopped. Fig. 3 shows the time dependence of the luminescent intensity of ZnS:Ag at 450 nm, with time zero set when the fast neutron irradiation was stopped. Here, some scatter of the measured intensity can be observed; this was caused by a large fluctuation in the 14 MeV neutron flux, which resulted from the fluctuation of the deuteron beam current striking the tritiated target. The time resolution is less than a few seconds. The abrupt drop of the luminescence shows it was not caused by radioactivity induced by the product nuclide but was caused by the recoil nucleus and/or the prompt gamma-ray generated by the nuclear reaction between the host material and fast neutrons. ZnS:Cu and luminescence of LLPs at 570 nm which is caused by the $Dy^{3+}$ behaved in a similar manner. In the case of the LLPs except for the luminescence at 570 nm, the luminescence continued for tens of minutes after the 14 MeV neutron irradiation was stopped. The elapsed time dependence of $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ at 480 nm after the fast

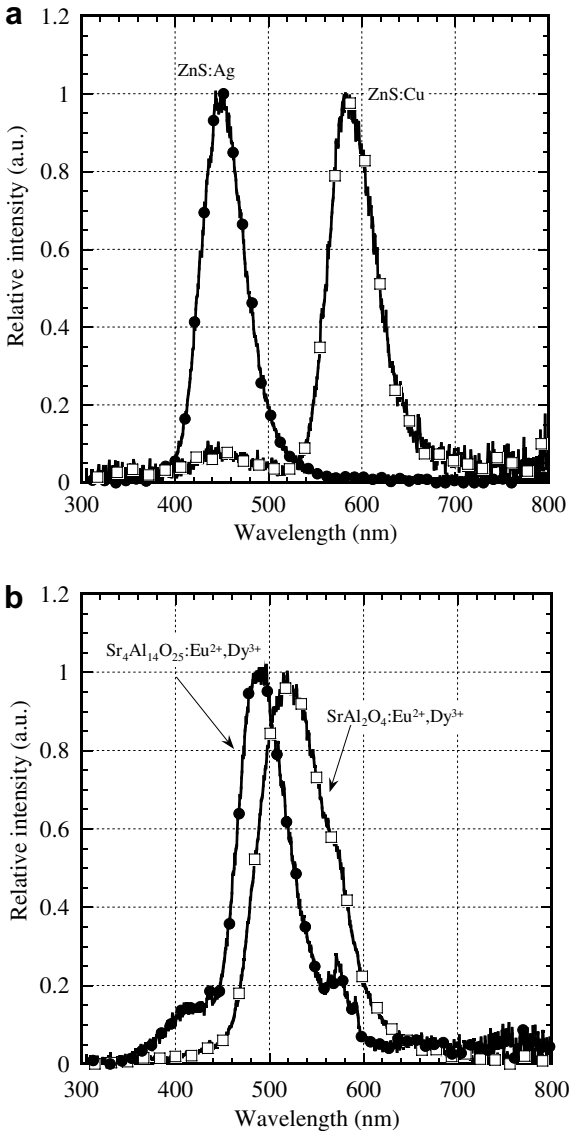


Fig. 2. Luminescent spectra of (a) ZnS:Ag and ZnS:Cu and (b) $SrAl_2O_4:Eu^{2+}$, $Dy^{3+}$ and $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$.

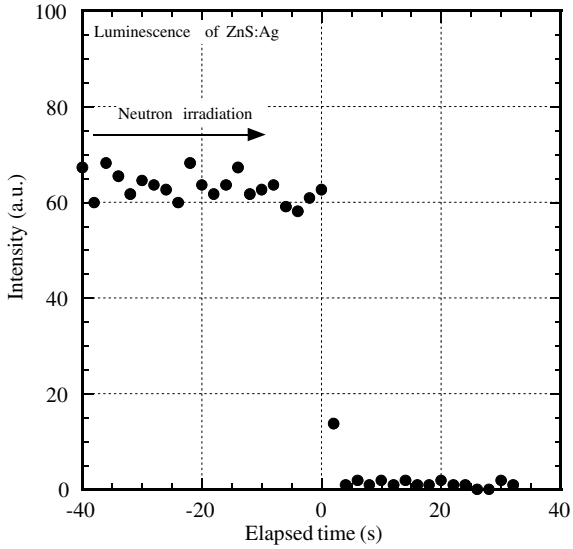


Fig. 3. Change in luminescent intensity of ZnS:Ag after neutron irradiation. Irradiation was measured tens of seconds before it was stopped.

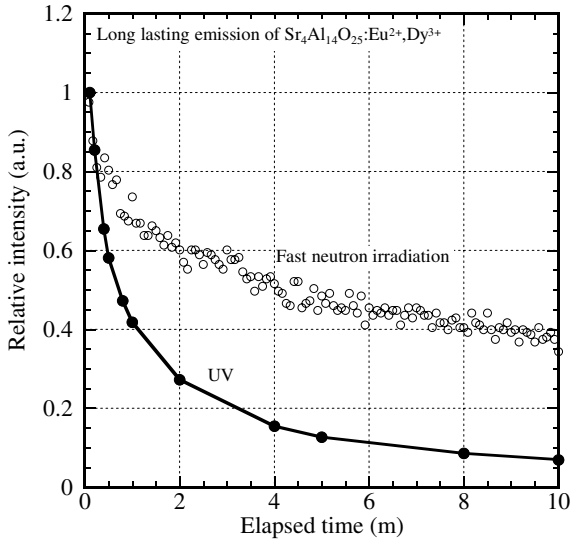


Fig. 4. Long lasting emission of $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$ after termination of 14 MeV neutron or UV irradiation.

neutron irradiation was stopped is shown in Fig. 4. Here, some scatter of the measured intensity is caused by the low optical intensity of the signal. This long-decay time luminescence after the cessation of irradiation is usually referred to as long lasting emission [6–8]. This study presents the first observation of long lasting emission for 14 MeV neutron irradiation, although this emission has previously been observed under the ultra violet (UV) and under the electronic excitation. The long lasting emission after the UV irradiation is also shown in Fig. 4. It can be seen that the decay time of the long lasting emission for 14 MeV neutron irradiation is longer than that for UV irradiation. The decay time of the LLPs is mainly attributed to the energy levels of the hole trap centers. The reason for the longer decay time after the fast neutron irradiation is not clear, but it is assumed that the material structure of $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$ is modified by the displacement effects of the fast neutron irradiation; this results in changes in the energy levels and/or

generates new hole trap levels and an increase in the decay time.

Changes in the luminescent peak intensities of the ZnS group and the LLPs are shown in Fig. 5(a) and (b), respectively, as a function of the elapsed time from the start of irradiation on the first day. The large scatter in the measured intensity is also mainly due to the fluctuation in the 14 MeV neutron intensity and a low optical intensity of the obtained luminescence. ZnS:Ag exhibited the strongest luminescence among the four materials; however, its intensity decreased as the irradiation fluence increased. The other materials exhibited a relatively weak luminescent intensity, but showed good radiation resistance, with little change on increasing irradiation time. ZnS:Cu and the LLPs maintained their respective luminescent intensities up to a fast neutron fluence of $10^{14}$ n/cm$^2$. Therefore, for high sensitivity, ZnS:Ag is the best among the four; however, ZnS:Cu and LLPs are preferable for long-term stability since they would not require frequent re-calibration or replacement.

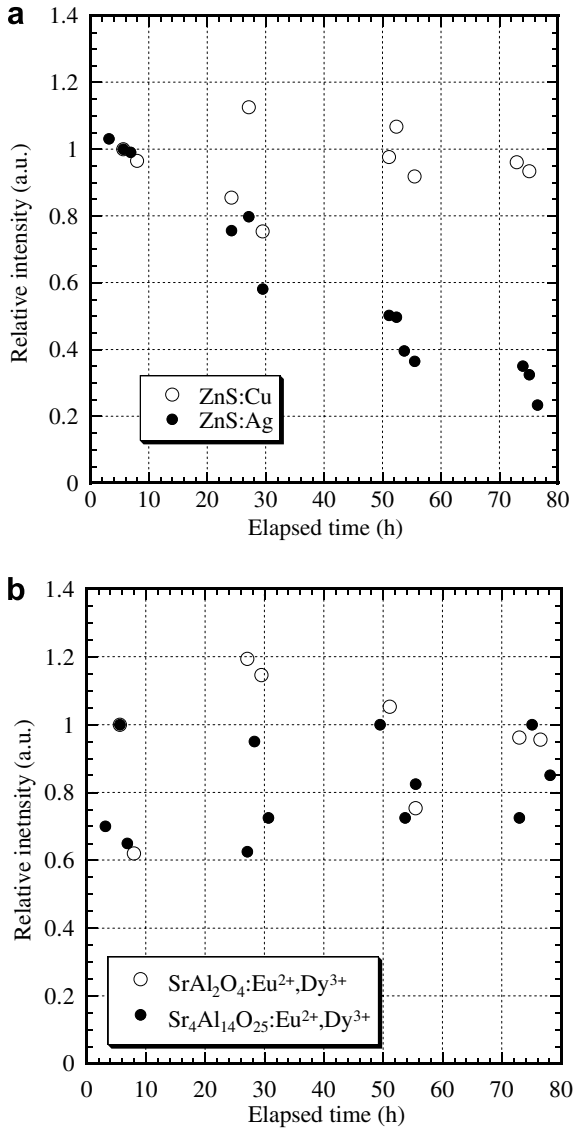


Fig. 5. Irradiation time dependence of the relative luminescence peak intensity of (a) ZnS:Ag and ZnS:Cu and (b) $SrAl_2O_4$:$Eu^{2+}$, $Dy^{3+}$ and $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$. Zero denotes the initial irradiation time on the first day.

$Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ shows three apparent characteristic peaks of different origins. For a more detailed study of its luminescent behavior, the luminescence characteristics of $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ were examined under proton irradiation by varying the incident energy. The irradiations were carried out using a 1.7-MV tandem accelerator at the Institute for Materials Research, Tohoku University. The incident proton energy was varied from 0.5 to 2.0 MeV; the flux was less than $5.0 \times 10^{13}$ p/cm$^2$ s. Luminescence generated under the proton irradiation was measured by means of a radiation-resistant optical fiber connected to an optical detector. $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ exhibited three luminescent peaks at 400, 480, and 570 nm that are similar to the spectra under the fast neutron irradiation. The luminescent peaks at 400 and 480 nm are attributed to $Eu^{2+}$, while the peak at 570 nm is attributed to $Dy^{3+}$. The luminescent intensity ratios of $Eu^{2+}$ to

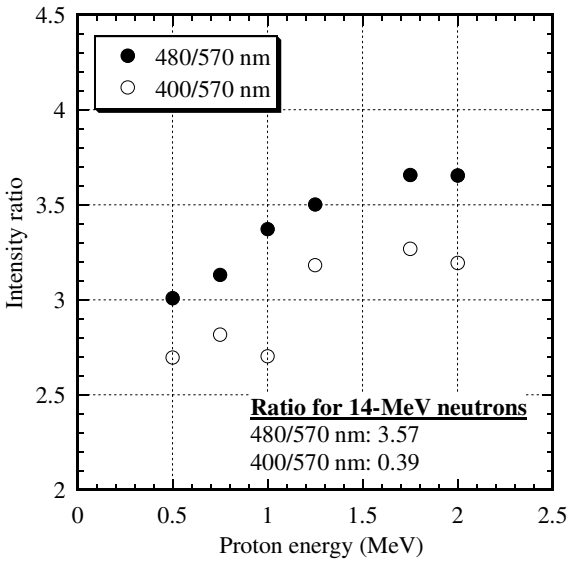


Fig. 6. Energy dependence of luminescent intensity ratios of 400 and 480 nm to 570 nm for $Sr_4Al_{14}O_{25}$:$Eu^{2+}$, $Dy^{3+}$ by protons.

$Dy^{3+}$ (400/570 and 480/570 nm) obtained by varying the proton energy are shown in Fig. 6. The ratios of 400/570 and 480/570 nm under the 14 MeV neutrons were 0.39 and 3.57, respectively. The peak intensity ratio is different for all proton energies, and their ratios are different from those under the fast neutron irradiation. $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$ can potentially be utilized in the identification of the incident particle type and energy.

## 4. Conclusions

We have proposed an optical radiation detection system using luminescent materials attached at one end of a radiation-resistant optical fiber. Luminescent materials – important components of the system – were studied under 14 MeV neutron irradiation. The commercially available scintillators – ZnS:Ag and ZnS:Cu – and long lasting phosphors (LLPs) – $SrAl_2O_4:Eu^{2+}$, $Dy^{3+}$ and $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$ – exhibit 14 MeV neutron-induced luminescence. Although the other materials possessed good radiation resistance for a fast neutron fluence of up to $10^{14}$ n/cm$^2$, the luminescence of ZnS:Ag decreased as the fluence increased. $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$ has apparent characteristic peaks that arise from the dopants – $Dy^{3+}$ and $Eu^{2+}$ – and the decay time of the long lasting emission increases due to the 14 MeV neutron irradiation. The luminescence characteristics of $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$ were also examined under proton irradiation with varying incident energy. It exhibited similar luminescence peaks to those under fast neutron irradiation, but the luminescent intensity ratio of $Eu^{2+}/Dy^{3+}$ exhibited proton energy dependence. The ratios are different from those under 14 MeV neutron irradiation. Therefore, the compound possesses the capability to distinguish between different types of incident radiations and particle energies. The present results support $Sr_4Al_{14}O_{25}:Eu^{2+}$, $Dy^{3+}$, which has a varying sensitivity to ions and 14 MeV neutrons, as a candidate for use as a luminescent material in an optical radiation detection system.

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