

Investigation of lanthanide-doped APLF scintillators for neutron detection

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Abstract. Investigating the fusion plasma is necessary to control the fusion process and to understand the plasma dynamics. Down-scattered (DS) neutrons detection is much suitable for measurement of the areal-density. We have demonstrated that lanthanide-doped $20\text{Al}(\text{PO}_3)_3\text{-}80\text{LiF}$ (APLF) has excellent characteristics as DS neutron scintillators. In this paper, the properties of lanthanide-doped APLF scintillators for neutron detection was investigated. Required temporal resolution of scintillators for DS neutron detection is less than 10 ns. The temporal resolution depends on fluorescence wavelength of luminescence center ion. Different lanthanide-doped APLF samples were prepared by melt-quench method for evaluation. Time-resolved spectroscopy using vacuum ultraviolet laser was used to assess the fluorescence decay times of each sample. Among all samples, the Nd-doped has the fastest time of 6.6 ns. When compared to the fluorescence decay time of a standard scintillator, GS2 with 38.1 ns, the lanthanide doped APLFs. The fluorescence decay time of a Pr-doped APLF was also evaluated using different radioactive sources. The Pr-doped APLF exhibits fast fluorescence lifetime compared to GS2 by one order of magnitude regardless of the excitation source. We have studied the potential of lanthanide-doped APLF as scintillators for neutron detection. Lanthanide-doped APLFs have faster fluorescence decay time compared to GS2. Nd-doped APLF exhibits the fastest decay time of 6.6 ns. The fluorescence of the Pr-doped and GS2 using three different radiation sources were also compared. Though the light output of the doped APLF is lower, the fluorescence intensity can still be detected. Lanthanide-doped APLFs can be better alternatives to conventional scintillators for neutron detection. Further developments on lanthanide-doped APLF based neutron scintillators are highly anticipated. In addition, this Pr-doped APLF scintillator was applied to development of a multichannel low-energy neutron spectrometer, and was actually used to detect DS neutrons in the recent fusion experiment using GEKKO XII. As a result, DS neutrons were successfully detected. We are confident that our scintillator will be a powerful tool of fusion experiment.

1. Introduction

We have demonstrated that lanthanide-doped $20\text{Al}(\text{PO}_3)_3\text{-}80\text{LiF}$ (APLF) has excellent characteristics as down-scattered (DS) neutron scintillators. In this paper, the properties of lanthanide-doped APLF scintillators for neutron detection was investigated. The lanthanide rare earth dopants include cerium (Ce^{3+}), praseodymium (Pr^{3+}), neodymium (Nd^{3+}). The Nd-

doped APLF has the fastest fluorescence time of 6.6 ns with ultraviolet (UV) excitation. The Pr-doped APLF has also a faster decay time compared to a conventional scintillator, GS2 with radioactive source excitation. Lanthanide-doped APLFs have the potential of being better alternative to conventional scintillators for neutron detection.

Investigating the fusion plasma is necessary to control the fusion process and to understand the plasma dynamics. DS neutrons detection is much suitable for measurement of the areal-density since DS neutrons are less susceptible to temperature of fusion plasma and applicable to present demonstration experiment of ignition through actual burning fusion plasma. Scattering cross-section is in proportion to the areal-density. In the case that this measurement of the areal-density by DS neutrons detection is realized, discussion about improvement of burning ratio becomes possible. In doing so, DS neutron diagnostics is implemented, but measuring DS neutrons is challenging because it requires highly selective scintillators. DS neutrons should be discriminated from primary neutrons, other scattered neutrons, and strong X-ray signals. A material, like APLF, containing lithium (${}^6\text{Li}$) can be used as a neutron scintillator, because resonant peak of cross section of ${}^6\text{Li}$ for scattered neutron is well-fit to the back-scattered neutron spectrum peak and ${}^6\text{Li}$ is an ideal receptor for initially scattered neutrons. In addition, a sufficiently fast response time is required because DS neutrons are interfered by the primary neutrons or X-rays. In typical arrangement, DS neutrons is detected in the time range from 25 to 50 ns after a fusion reaction. Thus required temporal resolution of scintillators is less than 10 ns. The temporal resolution depends on fluorescence wavelength of luminescence center ion. Generally, shorter wavelength fluorescence has shorter decay time. Ce, which has fluorescence from 300 to 400 nm, is used in general Li-silica scintillators, however Nd or Pr, which has fluorescence under 300 nm, cannot be used because of its UV self-absorption. In this paper, we introduced APLFs, which does not absorb deep UV light.

2. Experiment

2.1. Optical properties of lanthanide-doped APLFs

Lanthanide-doped APLF samples were prepared by melt-quench method. The preparation details were previously reported elsewhere [1]. Time-resolved spectroscopy was used to assess the fluorescence decay times of each sample. Taking into consideration the fluorescence peak of each lanthanide-doped APLF, two different excitation sources were used. A Ti:sapphire laser (217 nm, 1 kHz, 150 fs) was used for the Ce-doped (350 nm fluorescence peak) and Pr-doped samples (278 nm fluorescence peak). A F_2 laser (157 nm, 10 kHz, 5 ns) was used for the Nd-doped (185 nm fluorescence peak) and Er-doped samples (168 nm fluorescence peak). The spectral decay profiles of the lanthanide-doped APLFs are shown in Figure 1. The fluorescence decay times are determined by fitting exponential decay functions from 50% to 10% of the peak intensity. In general, the decay constant consists of two or more values, so we choose this appropriate limit of the finite region. Among all samples, the Nd-doped has the fastest time of 6.6 ns. The Nd-doped is then followed by the Pr-doped with 19.5 ns and by the Ce-doped with 23.3 ns. All these values come from well-fitted single exponential decay functions. The Er-doped has a slowest decay time of 31.0 ns from double exponential fitting. This observation is unexpected, but the Er-doped sample has low signal-to-noise ratio. When compared to the fluorescence decay time of a standard scintillator, GS2 with 38.1 ns, the lanthanide doped APLFs exhibit better decay times by more than 10 ns.

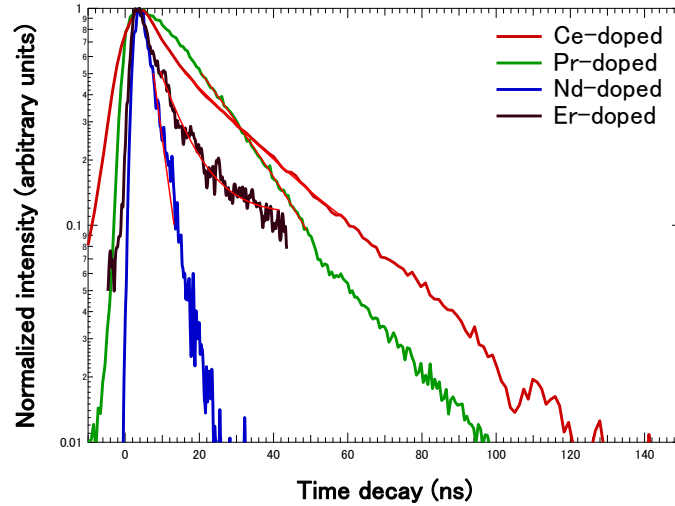


FIG. 1. Fluorescence decay profiles of the lanthanide-doped APLFs

From the dopants excluding erbium, Nd^{3+} has the smallest ionic radius, and the Nd-doped APLF has the shortest peak wavelength and decay time. In the contrary, Ce^{3+} has the largest ionic radius, and the Ce-doped APLF has the longest peak wavelength and decay time. These observations are attributed to their electronic configurations. The lanthanide ions have emission bands in the UV region due to their $5d-4f$ transitions. The $4f$ orbital electrons are shielded from the external electric field by the $5s$ and $5p$ outer orbital electrons. As compared to the $5d$ level, the $4f$ energy level is not affected much by the environment. The $5d$ electrons are heavily affected by the APLF matrix glass composition. For such reason, the Ce-doped APLF has a fluorescence peak at longer wavelength and a longer decay time.

The fluorescence decay time of a Pr-doped APLF was also evaluated using different radioactive sources. The radioactive sources used were americium (^{241}Am) with 5.5 MeV alpha (α) particles, cobalt (^{60}Co) with 1.33 MeV gamma (γ) particles, and californium (^{252}Cf) with broadband neutrons (n) of 1 MeV central energy. Figure 2 shows the fluorescence decay profiles of the Pr-doped sample and the GS2 scintillator. The Pr-doped APLF exhibits fast fluorescence lifetime compared to GS2 by one order of magnitude regardless of the excitation source. The drawback, however, is a lower light yield. Though this is the case, the fluorescence intensity is still sufficient for detection. Pr-doped APLF can be a good alternative to a standard neutron scintillator.

We have studied the potential of lanthanide-doped APLF as scintillators for neutron detection. Lanthanide-doped APLFs have faster fluorescence decay time compared to GS2. Nd-doped APLF exhibits the fastest decay time of 6.6 ns. The differences on the decay times of the samples were attributed to the dopants' electronic configuration, particularly to the $4f-5d$ transitions. The fluorescence of the Pr-doped and GS2 using three different radiation sources were also compared. Though the light output of the doped APLF is lower, the fluorescence intensity can still be detected. Lanthanide-doped APLFs can be better alternatives to conventional scintillators for neutron detection. Further developments on lanthanide-doped APLF based neutron scintillators are highly anticipated.

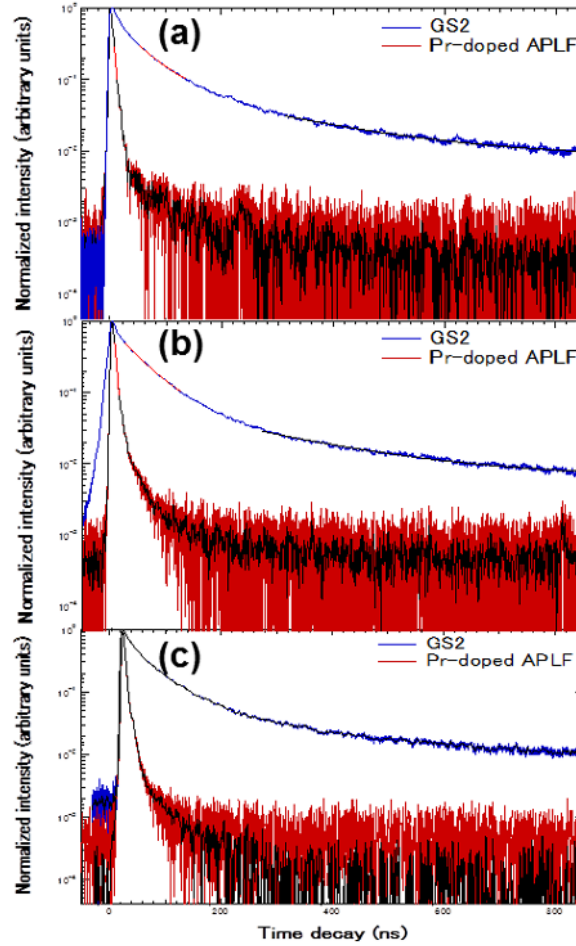


FIG. 2. Fluorescence decay profiles of Pr-doped APLF and GS2 with (a) ^{241}Am , (b) ^{60}Co , and (c) ^{252}Cf excitations

2.2. Detection of DS neutrons

In addition, this Pr-doped APLF scintillator was applied to development of a multichannel low-energy neutron spectrometer [2], and was actually used to detect DS neutrons in the recent fusion experiment using GEKKO XII. As a result, fuel areal density converted by count of detected DS neutrons is under analysis and discussion, however DS neutrons were successfully detected (Fig.3). We are confident that our scintillator will be a powerful tool of fusion experiment.

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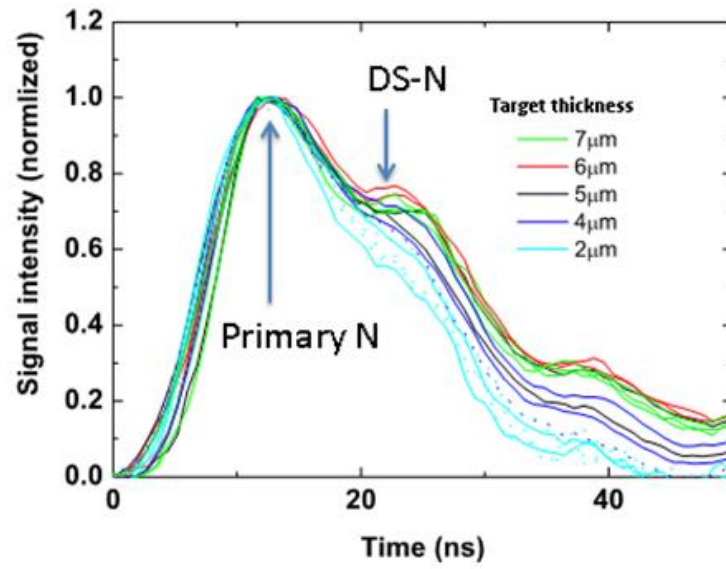


FIG. 3. Detected DS neutrons signal in GEKKO XII fusion experiment.

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- [2] Y. Arikawa et al., Rev. Sci. Instrum. 85, 11E125 (2014)