Nuclear Instruments and Methods in Physics Research B xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Analysis of excitation density effects on the scintillation properties of Ce: Gd_2SiO_5 (GSO) crystals

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ARTICLE INFO

Article history:
Received 21 November 2016
Received in revised form 27 March 2017
Accepted 1 April 2017
Available online xxxx

Keywords:
Scintillator
GSO
LET
Excitation density
Ce3+

ABSTRACT

We analyzed the effects of the excitation density or linear energy transfer (LET) on the scintillation temporal profiles of Gd_2SiO_5(GSO):Ce by using pulsed ion beams of 20 MeV H^+, 50 MeV He^{2+}, and 220 MeV C^{5+}. The rise was faster at higher LET, while the decay was similar at largely different LETs. The LET effects on the rise are explained in terms of the competition between the quenching owing to the interaction between the ^6P_J excited states at neighboring Gd^{3+} ions and the energy transfer to Ce^{3+} ions.

1. Introduction

Scintillation at high linear energy transfer (LET) has long been a technological concern. Low scintillation light yield at high LET has long been recognized. For example, Birks has proposed a simple formalism to account for the low light yield at high LET [1]. The low light yield at high LET inhibits the simultanous spectroscopy of low-LET radiations such as gamma-ray and high-LET radiations. Additionally, in the case of thermal neutron detection via nuclear reactions with ^{6}Li or ^{10}B, fission fragments (alpha-particles and high-energy ^{3}H or ^{7}Li) generate electron-hole pairts at much higher LET than those of gamma-ray. The significant difference in the LET leads to a difference in the scintillation temporal profiles, leading to a difference in the detection signal shape. Thus, pulse shape discrimination has been a powerful tool to discriminate the gamma-ray and thermal neutron detection events.

In contrast to the technological advances, the understanding of the basic processes of scintillation at high LET is still poor. The above-mentioned Birks model is the most famous one; however, the model treats the light yield in a phenomenological and time-independent manner. As for the experimental data, many reports have been publised on the scintillation light yield at high LET, which is also the time-integrated value. Actually, the scintillation at high LET should be analyzed from the viewpoint of dynamical aspects. The LET-dependent scintillation temporal profiles carry an information on the dynamics, however, previous researches have focused on the pulse shape discrimination capability rather than the temporal profiles themselves [2–4].

Recently, we started to analyze the scintillation temporal profiles at high LET. Many inorganic scintillators are comprised of insulator host crystals and dopant ions as luminescence centers. In the case of scintillation, most of electronic excitations occur in the host. Scintillation process can be described as the energy transfer from the host to the luminescence centers and subsequent radiative transition at the luminescence centers. We focused on the energy transfer process, which is manifested in the rise of the scintillation temporal profiles. We found LET-dependent rise in two kinds of scintillators for thermal neutron detection, a Li-glass scintillator [5] and Ce-doped $LiCaAlF_6$ [6]. The LET dependence of the rise is attributed to a competition between the energy transfer from the host to the luminescence centers and quenching owing to interaction between the excited states in the host. Based on these results, we hypothesize that such competition occurs for other scintillators with a slow rise in the scintillation temporal profiles.

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http://dx.doi.org/10.1016/j.nimb.2017.04.001
0168-583X/

In this study, we analyzed the LET dependence on the scintillation properties of Gd_2SiO_5:Ce (GSO:Ce) which is one of the common scintillator for γ-ray detectors [7,8], and known to have a slow rise in the scintillation temporal profile [9]. In addition, the rise depends on the Ce concentration in GSO [9]. We analyzed the scintillation temporal profiles of GSO:Ce with different Ce concentrations with main focus on the rise in the temporal profiles.

2. Experimental

The samples were GSO:Ce with Ce concentrations of 0.5, 1.0, and 1.5 mol%. The size of the crystals were 5 mm × 5 mm × 5 mm. The samples were irradiated with ion beams in air. The irradiation was performed using an azimuthally varying field (AVF) cyclotron at TIARA, National Institute for Quantum and Ragiological Science and Technology, Japan. The samples were irradiated with pulsed ion beams of 20 MeV H^+, 50 MeV He^{2+}, and 220 MeV C^{5+} to vary the LET. The pulsed beams were generated from continous beams using a primary and secondary choppers. The beam intensity was less than 10^4 ions/pulse, and the pulse repetition rate was less than 100 Hz. The details of the pulsed beam generation are described elsewhere [10].

The scintillation from the sample was detected with a photomultiplier tube (PMT; Hamamatsu R7400U). The detection signals were delivered out of the irradiation room to a digital oscilloscope (DSO; Tektronix DPO 7104) in a measurement room. The trigger signal of the DSO was supplied from the accelerator. Scintillation temporal profiles were obtained as the average of 1000 detection signals. The origin of the time axis was set at the peak time of the scintillation temporal profiles of a plastic scintillator, BC-400, in the same measurement condition. The overall time resolution of the measurement system was about 2 ns half width at half haximum. The details of the measurement system are described in our previous paper [11]. The scintillation spectra under the irradiation were also obtained with an optical multichannel detector (Ocean Optics, USB-4000).

3. Results and discussion

Fig. 1 shows the LET estimated with the SRIM code [12]. All the ions are estimated to be stopped within the samples 5-mm thick. The LET is estimated to be on the order of tens of eV/nm for 20 MeV H^+, and those for 50 MeV He^{2+} and 220 MeV C^{5+} are larger by one and two orders of magnitude, respectively.

Fig. 2 shows the scintillation spectra under irradiation of 220 MeV C^{5+}. Unfortunately, the comparison of the intensity is not feasible because the local beam intensity in the monitored spot cannot be measured. A broad band is observed with its center at around 450 nm for all the ions. These spectra are consistent with those under X-ray or gamma-ray irradiation [9], and the band is attributed to the emission of Ce^{3+} ions. This result indicates that the final stage of the scintillation is the radiative transition at Ce^{3+} ions even at LET.

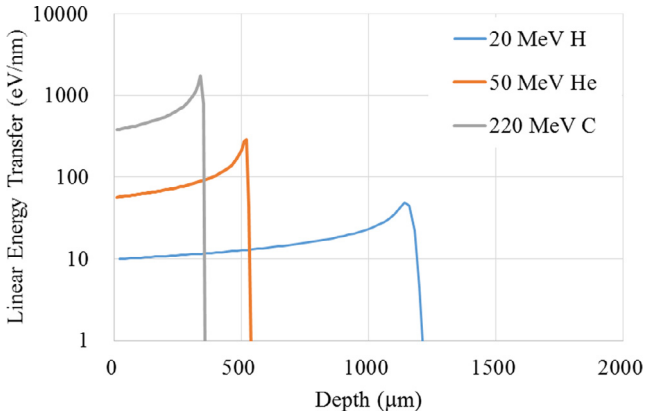

Fig. 1. Linear Energy Transfers estimated with the SRIM code.

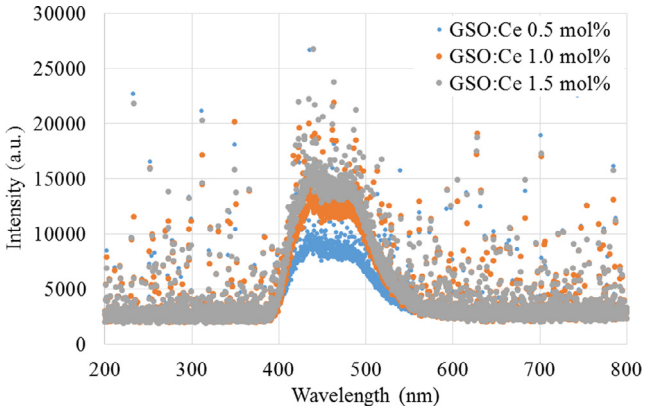

Fig. 2. Scintillation spectra under irradiation of 220 MeV C^{5+}.

Fig. 3 shows the rise in the temporal profiles of GSO:Ce with different Ce concentrations. The rise is significantly faster at higher LET. This is the same trend with those of a Li-glass scintillator and Ce-doped $LiCaAlF_6$ [5,6]. These results, combined with the result of lower light yield at high LET [13–15], indicate that the energy transfer process from the host to Ce^{3+} compete with the quenching owing to interaction between excited states in the host, similarly to the case of the Li-glass scintillator and Ce-doped $LiCaAlF_6$. In addition, the LET dependence is more significant for GSO:Ce with lower Ce concentration. Fig. 4 shows the decay part in the temporal profiles of GSO:Ce with different Ce concentrations. Except the decay for GSO doped with Ce at 0.5 mol% irradiated with 20 MeV H^+, the decay is quite similar for the largely different LET.

In the following, we discuss the dynamical aspect of the LET effects based on the energy transfer model in GSO:Ce proposed in a previous paper [16]. According to the proposed model, electron-hole pairs generated in the host are immediately captured at Gd^{3+} sites, and a relaxation within the Gd^{3+} ions leads to the ^6P_J excited state. Diffusion of the ^6P_J excited state and subsequent energy transfer to Ce^{3+} ions lead to scintillation. Initially, the energy is deposited in the host glass. The rise reflect the energy transfer process to the luminescence centers, i.e., Ce^{3+} ions. The difference in the rise for different LETs can be explained in terms of the difference in the excited states dynamics, which is caused by the excited states interaction, during the energy transfer. One is that the energy transfer is promoted at high LET. In this case, the light yield would be enhanced at high LET. The other is that the quenching is in competition with the energy transfer, and the fast rise is the result of quenching of the slow energy transfer component. In this case, the light yield would decrease at high LET. Actually, the light yield is lower at higher LET [13–15]. Hence, the fast rise at high LET is explained in terms of competition between the energy transfer and quenching. The latter is caused by the excited states interaction, possibly the interaction of the ^6P_J excited states at the neighboring Gd^{3+} ions, at high LET. At least, the influence of LET is less significant on the energy transfer to Ce^{3+} ions than on the quenching process prior to the energy transfer. The cause of the quenching is, unfortunately, speculative at present. One possible mechanism is the Auger-type annihilation of an excited state in which its excitation energy transferred to another excited state nearby, resulting in a reduction in the number of excited states.

Please cite this article in press as: T. Yanagida et al., Analysis of excitation density effects on the scintillation properties of Ce:Gd_2SiO_5 (GSO) crystals, Nucl. Instr. Meth. B (2017), http://dx.doi.org/10.1016/j.nimb.2017.04.001