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Effect of ion beam and neutron irradiations on the luminescence of polycrystalline Ce-doped $Y_3Al_5O_{12}$ ceramics

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A B S T R A C T

Polycrystalline ceramics of Ce-doped $Y_3Al_5O_{12}$ (Ce: YAG) is characterized as a candidate scintillator for a lost alpha particle measurement on a thermo-nuclear fusion experiments such as ITER. The luminescence intensity for polycrystalline Ce: YAG ceramics is investigated under MeV energy ion irradiation with the ion flux up to $\sim 6.7 \times 10^{16}$ m^{-2} s^{-1} and with the ion fluence of $\sim 1.0 \times 10^{19}$ m^{-2}. The luminescence has a good linearity up to ion flux mentioned above. In view of the fluence, the luminescence degradation is observed, but it is found to be a tolerable level. In neutron and gamma ray irradiation experiments at a fission reactor, the defect such as blobs is appeared in irradiated samples. It is found that the thermal annealing is effective for the recovery of the luminescence as well as the contraction of the defect.

1. Introduction

In burning plasma experiments in ITER, the loss of 3.5 MeV alpha particles produced by DT reaction causes a degradation of self-heating power. In addition, localized loss of alpha particles onto the first wall will give rise to serious problems. It is, therefore, important to understand alpha particle loss mechanism and to avoid these problems [1]. Scintillator lost ion probe (SLIP) is one of possible tools for lost alpha particle measurement in ITER. A scintillator is mounted in the head of SLIP system. Lost fast ions enter the SLIP detector through an aperture, and hit the scintillator, which produces the luminescence. The luminescence light is transmitted to outside of the vacuum vessel, and is detected by a photomultiplier array and a charged couple device. The information on the ion energy and pitch angles can be obtained from bright spots on the scintillator [2]. For the SLIP diagnostics, it is important to develop and characterize scintillation materials during and after hard irradiations of ions, gamma ray, and neutrons under the environment of first wall of ITER [3–5], where the SLIP will be mounted. The alpha particle flux and fluence up to 10^{16} m^{-2} s^{-1} and 10^{23} m^{-2}, the neutron flux and fluence $\sim 10^{18}$ m^{-2} s^{-1} and $\sim 10^{25}$ m^{-2}, and gamma ray flux and fluence 10^{18} m^{-2} s^{-1} and 10^{25} m^{-2} can be employed for design of system near the detector location for a year. The temperature of this location is supposed to be above 300 °C.

The hard irradiation environment was simulated experimentally to study the effect of ion beams and neutron irradiation on the luminescence of the polycrystalline ceramic of Ce: YAG as a scintillator in the ITER operation range.

2. Experimental results of irradiation test

As one of scintillation materials, the polycrystalline ceramic of 1 mol% Ce: YAG, was chosen, because the powder showed the better characteristics at high temperature [4] than conventional scintillation materials such as Ag-doped ZnS [6].

The transparent polycrystalline Ce: YAG was provided at the Institute of Laser Engineering in Osaka University, and is made from the powder. It was sintered at 1750 °C for 20 h. The polycrystalline Ce: YAG ceramics disk was 1.0 mm thickness and 10 mm in diameter with the density of 4.5×10^3 kg m^{-3}. Fig. 1 shows the spectra excited by He^+ beam and Hg lamp. In both cases the spectra have the peak near 560 nm at room temperature.

2.1. Ion irradiation

The scintillator was irradiated with ion beams of up to 300 nA at room temperature. The ion beam was limited to 6 mm in diameter using a diaphragm on the beam line. The luminescence spectra during 3.0 MeV H^+ and He^+ irradiations were measured by a spectrometer (Hamamatsu, PMA-11) at the dynamitron accelerator facility of Fast Neutron Laboratory (FNL) in Tohoku University. Fig. 2 shows the dependence of the luminescence intensity on the incident particle flux. Throughout this paper, the luminescence intensity corresponds to the value integrated over the wavelength from 450 to 800 nm. In both ion species, the luminescence

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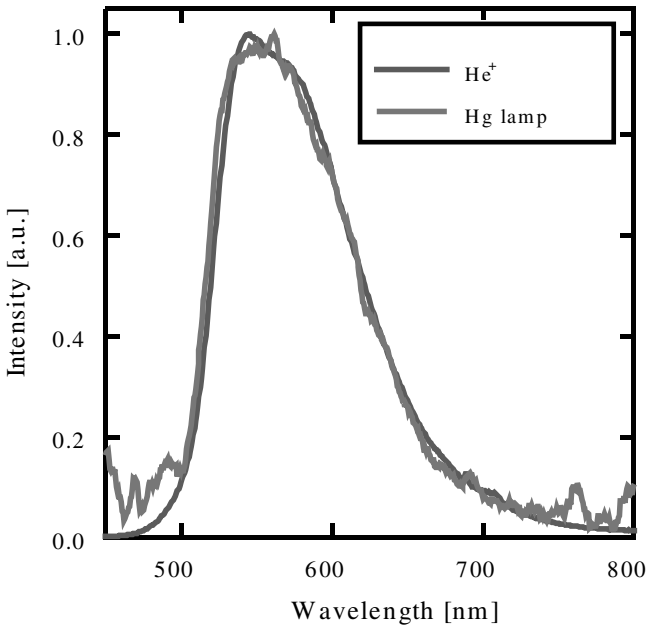

Fig. 1. The spectra excited by He^+ beam and Hg lamp.

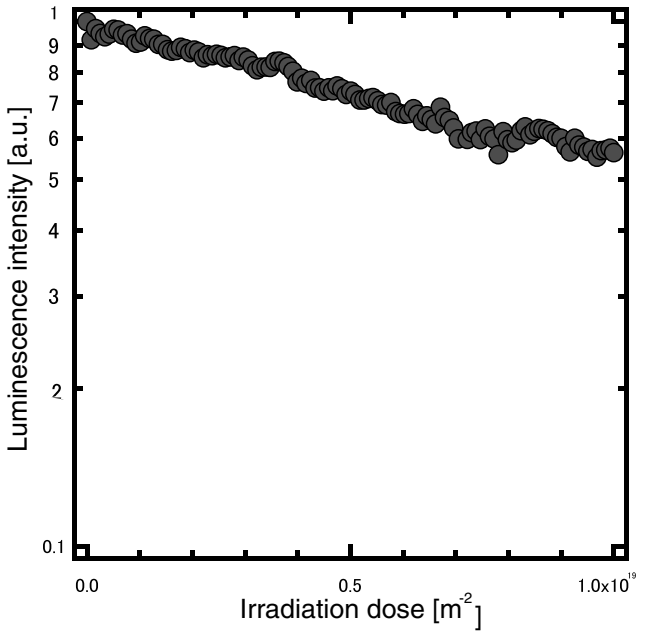

Fig. 3. The change of luminescence intensity by irradiating the He^+ beam with the energy of 3.0 MeV.

intensity increased linearly, when the ion flux goes up to $6.7\times10^{16}\ m^{-2}\ s^{-1}$, which simulated the environment of ITER for lost alpha diagnostics.

Fig. 3 shows the relation between the integrated luminescence intensity and the fluence for 3.0 MeV He^+ beam. The luminescence intensity decreased, and kept about 55 % of the intrinsic luminescence one after the irradiation at the maximum fluence.

2.2. Neutron and gamma ray irradiation

The polycrystalline Ce: YAG samples were irradiated with neutron and gamma ray at the Japan Research Reactor 3 (JRR-3) in Japan Atomic Energy Agency. The fast neutron flux was set to $1.6\times10^{16}\ m^{-2}\ s^{-1}$ for 12 h and $1.7\times10^{16}\ m^{-2}\ s^{-1}$ for 48 h, respectively at the sample temperatures of about 200 °C. After neutron irradiation, the samples were observed with a microscope and the luminescence characteristics of the samples were investigated. The samples were annealed at 200, 300, and 800 °C for an hour after irradiations to observe the recovery of the luminescence intensity with anneal, because it was reported that the annealing was effective to the recovery of irradiation damage [7].

The sample surface was observed with Digital Micro Scope at 3000-fold magnification. The irradiated samples found that the blobs, which might be defects or impurity precipitations, were

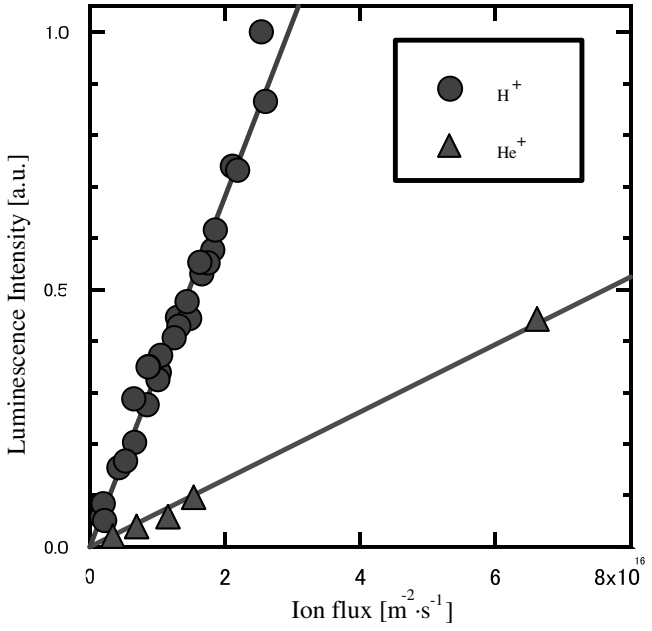

Fig. 2. Luminescence intensity of 1 mol% Ce: YAG by irradiating H^+ and He^+ beams with the energy of 3.0 MeV.

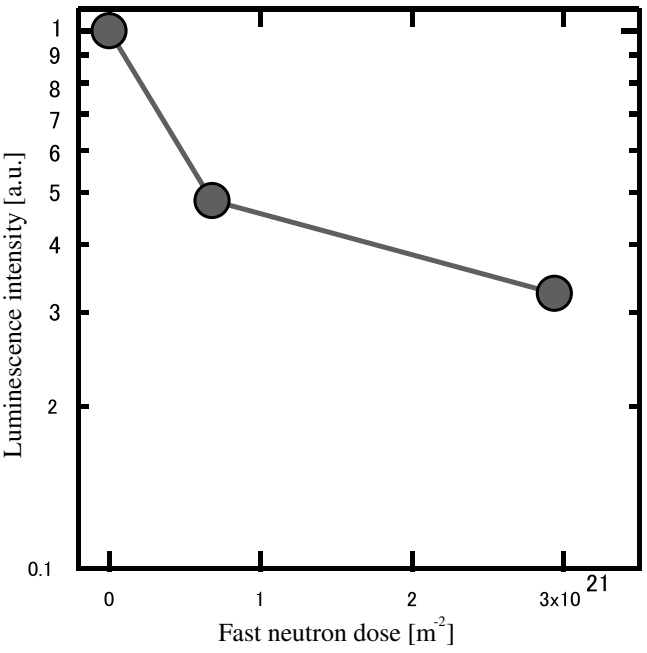

Fig. 4. Fast neutron irradiation effect on the integrated luminescence intensity. Irradiation duration was 12 and 48 h.

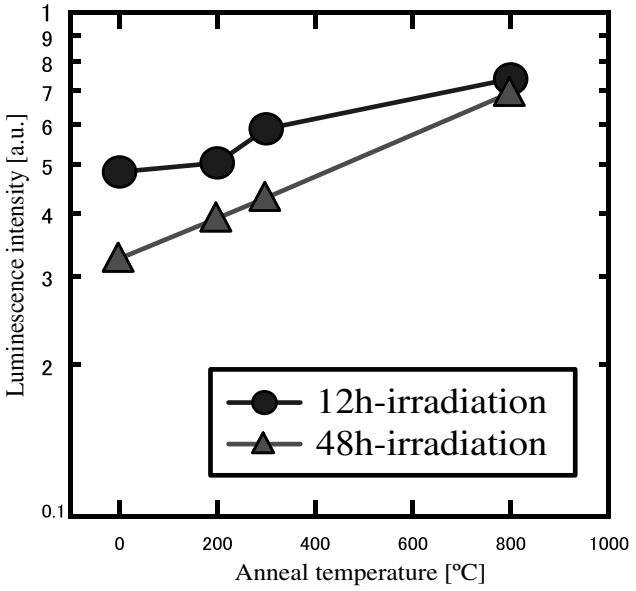

Fig. 5. Annealing effect on the luminescence intensity. Each sample was annealed for an hour at the controlled temperature.

observed everywhere apparently, while these blobs did not appear in the samples without neutron irradiation. The average cross section of blobs was about 2 μm^2. The number of blobs for the sample with 12 h irradiation was less than that with 48 h irradiation. However it was remarkable that 12 h irradiation samples had relatively large blobs whose average cross section was about 10 μm^2. The blobs almost vanished after annealing the samples at 800 °C.

The samples were irradiated with the excitation light of an Hg lamp to measure the change of luminescence spectrum. Fig. 4 shows the fast neutron irradiation effect on the luminescence intensity. The luminescence intensities integrated over the wavelength decrease to 50% in 12 h, and 28% in 48 h, though there were not much difference between two spectra. Fig. 5 shows the anneal effect on luminescence. Luminescence intensities of 12 h irradiation samples changed to 50%, 62%, and 76% of the original intensities for an hour anneal at the temperatures of 200, 300, and 800 °C, respectively. Similarly, the intensities of 48 h irradiation samples recovered to 42%, 46%, and 72% of the original intensities after an hour anneal at 200, 300, and 800 °C, respectively.

3. Summary and discussions

The luminescence intensity of the polycrystalline Ce: YAG was characterized during MeV ion beam and after neutron-gamma ray irradiations. It is clear that the polycrystalline Ce: YAG has the linear dependence of the luminescence intensity on the flux of MeV ion beams in the normal operation range of ITER for alpha particle loss diagnostics. As for the fluence durability, the polycrystalline Ce: YAG ceramic scintillator keeps 55% of the intrinsic luminescence intensity under the experimental conditions of ITER. We still need to extend the exposure time to a year operation. The neutron irradiation caused the decrease of luminescence intensity to 28% of the original one at the fast neutron fluence of 2.9×10^{21} m^{-2}. Though the fluence is still four orders of magnitude less than that for a year operation of ITER, it is found that the annealing is effective for the recovery of the radiation damage. The recovery to the intrinsic luminescence intensity can be expected for longer anneal time than an hour.

Several factors can be mentioned as to the degradation mechanisms of the luminescence intensity induced by ion beam irradiation. The charge state might be changed to Ce^{4+} by energy transfer from Ce^{3+} which plays as the luminescence center. Another factor is caused by the knock-out of oxygen atoms in YAG. Further experiments are required to explain the degradation mechanisms.

There is one of the possible explanations for the degradation of luminescence intensity induced by neutron irradiations. The impurity would be an absorber of Ce: YAG luminescence, because there are no changes in the luminescence spectrum shapes. It means that the luminescence center of doped Ce^{3+} does not change the energy level population. Therefore the degradation of the luminescence intensity is due to the change in the transparency by neutron and gamma ray irradiation, but not due to the change in population distribution of the luminescence center of doped ions.

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