Tritium measurement using a photo-stimulable phosphor BaFBr(I):Eu²⁺ plate

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Summary. Tritium measurement is indispensable for the fuel- processing systems of deuterium-tritium (DT)-fusion facilities. A new approach to detect tritium in regions deeper than the escape depth of beta-rays from tritium is being developed using an imaging plate (IP). The measurement principle of this approach is to observe bremsstrahlung X-rays induced by the tritium beta-rays. An IP made of europium-doped BaFBr(I), a photostimulated luminescence (PSL) material, is a two-dimensional radiation sensor. In the present study, the characteristics of this IP for measuring tritium by detecting bremsstrahlung X-rays, in particular a fading effect and the energy dependence of PSL sensitivities, are examined.

1. Introduction

Tritium measurement is indispensable for the fuel- processing systems of deuterium-tritium (DT) -fusion facilities. During the operation of fusion machines with D-T plasma, tritium accumulates in the plasma-facing components (PFCs). The amount of tritium that is handled in such facilities ranges from 10¹ to 10¹⁷ Bq. To improve the understanding of tritium accumulation in PFCs under plasma-wall interactions, various techniques such as the coring/full combustion method [1], beta-ray-induced X-ray spectrometry (BIXS) [2], a nuclear reaction analysis [3], a beta-ray analysis using a pin diode [4], and accelerator mass spectrometry [5, 6], have been used to measure the tritium inventory and distribution in these components. Recently, the tritium imaging plate (IP) technique has been successfully applied to determine the surface tritium distribution on plasma facing tiles [7–11]. An IP made of europium-doped BaFBr(I), a photostimulated luminescence (PSL) material, is a two-dimensional radiation sensor. This IP has many excellent properties for this purpose, including a high sensitivity, a wide dynamic range over five orders of magnitude, and a high degree of spatial resolution [12]. In addition, the IP can be used repeatedly by exposing it to visible

light between uses. The tritium IP is an autoradiographical technique relying on beta particles by using an IP sheet of BAS-TR (details are given below). Tritium emits beta particles with a maximum energy of 18.6 keV and an average of 5.7 keV. These have a range of about several micrometers in graphite and hence the technique is sensitive to tritium up to a depth of a few microns. However, the results obtained by the coring/full combustion method, which is used to determine tritium depth profiles, revealed that much of the tritium was concentrated in codeposition layers on the tiles and a large fraction (up to 61%) of it migrated into the bulk of the tiles [13]. This means that the tritium IP technique based on detecting the beta particles is unsuitable for use in regions deeper than the escape depth of beta-rays from tritium, and results in an underestimation of the amount of tritium. Besides, the technique might cause contamination of the IP plate with high levels of tritium during irradiation, and this contamination might spread from the plate to the plate reader when it is scanned.

We have developed a new approach to detecting tritium using the bremsstrahlung induced by tritium beta-rays with the IP [14]. The measurement principle of this approach to tritium detection is to observe bremsstrahlung X-rays generated by the interaction between the beta particles from tritium and matter, on the basis that X-rays penetrate materials much more easily than the weak beta-rays from tritium, which have a maximum energy of 18.6 keV. In the present study, the characteristics of the IP for measuring tritium by detecting bremsstrahlung X-rays it produces have been examined, in particular a fading effect and the energy dependence of PSL sensitivities. The fading effect in the IP after irradiation with bremsstrahlung X-rays from tritium was compared with that obtained after irradiation with the beta particles emitted from tritium. The dependence of the PSL sensitivities on energy was measured using monoenergetic X-rays.

2. Experimentals

2.1 Imaging plate and readout

A BAS-MS type-IP was used to detect bremsstrahlung X-rays from tritium. This IP consists of a 9-μm-thick polyethyleneterephthalate protective film and a 115-μm-

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Table 1. Structure of two types of imaging plates.

IP	BAS-MS	BAS-TR
Thickness of protective layer (μm)	9	0
Thickness of phosphor layer (μm)	115	50
Phosphor	$\text{BaF}(\text{Br}_{0.85} + \text{I}_{0.15})$	BaFBrI^a
Thickness of base (μm)	190	250
Thickness of ferrite layer (μm)	160	160

a: Ba (37.82%), F (5.23%), Br (22.01%), and I (34.94%).

thick photostimulable phosphor layer affixed to a 12- μm -thick plastic back layer and a 190- μm -thick polyethyleneterephthalate base layer [15]. It has a high sensitivity to photons. The BAS-MS type-IP can detect bremsstrahlung X-rays but not the beta particles emitted from tritium because of its thick protective film. To detect these beta particles, a BAS-TR-type IP without a protective surface layer was used. This IP consists of a 50- μm -thick phosphor layer affixed to a 250- μm -thick polyethyleneterephthalate layer as support and a 160- μm -thick ferrite layer. Both types of IPs were manufactured by FUJIFILM Co., Ltd. The differences in structures of these two types of IPs are listed in Table 1.

In the sensitive phosphor layer of an IP, ionizing radiation creates a large amount of trapped centers, which record information about the deposited energy and its position. The IP is read out using a He-Ne laser (633 nm) within the image reader. The laser light induces PSL (390 nm) from the positions of the trapped centers, which is detected *via* a photomultiplier tube. In this study, model FLA-3000 and BAS-5000 IP readers were used. Both image readers were fabricated by FUJIFILM Co., Ltd. They both have the same type of readout system, and are used with a spatial resolution of 100 μm and 8 bits of digital resolution dpi. The difference between the two readers is the IP conveyance system. The FLA-3000 can convey any size of IP sample to inside the reader, while the BAS-5000 cannot. To investigate the energy response of the BAS-MS-type IP, 3.0 cm $\times$ 5.0 cm pieces were cut from the PSL sheet. These pieces were read out by using the FLA-3000.

2.2 Irradiation and method

We used four small borosilicate glass tubes with a wall thickness of 0.088 mm, length of 6.36 mm, and diameter of 0.60 mm, filled with pure tritium gas of 12.5, 25, 50, and 100 MBq, respectively (manufactured by mb-microtec ag) as the tritium sources that generate bremsstrahlung X-rays. Ten strips of microscale-labelled polymer layers, which were developed for autoradiography (manufactured by GE Healthcare), were used as the beta-ray radiation source. Each strip contained approximately 2.96 kBq of tritium. The IPs were irradiated with beta rays or bremsstrahlung X-rays by placing the radiation sources directly on the IP. The temperature dependence of the fading effect of the latent image was measured for time periods from 0.05 to 380 h after 1 h

irradiation with beta rays or bremsstrahlung X-rays from tritium, at temperatures of 0, 30, and 50 $^{\circ}\text{C}$. The IPs were kept in an aluminum IP cassette inside an incubator with the temperature controlled to $\pm 1^{\circ}\text{C}$ during irradiation and for the time prior to reading the latent image. The detection limit was obtained by irradiating the IP with a borosilicate glass tube filled with 12.5 MBq of gaseous tritium for an irradiation time from 1 to 24 h. The IP was placed inside a 10-cm-thick lead outer shield during irradiation to reduce the effect of natural radiation. The energy response of the BAS-MS-type IP was measured by using 8.0, 10.0, 13.5, 16.0, and 18.6 keV monoenergetic X-ray beam sources. The experiments using the X-ray beam sources were performed on a beam line at the Photon Factory (PF) of the High Energy Accelerator Research Organization (KEK), Tsukuba, Japan. The PSL sensitivities of the IPs covered with aluminum filters of three different thicknesses, 0.1, 0.3, and 0.5 mm, were measured in order to investigate the variation of the energy response with filters present.

3. Results and discussion

3.1 Fading effect of the latent image

The IP has the defect of having a large fading effect. So it is very important to estimate this fading when using an IP as an integral-type detector. Fig. 1 compares the experimental results for the measured PSL values after irradiation with the tritium source generating bremsstrahlung X-rays using the BAS-MS and the beta-ray source using the BAS-TR in the range 0 to 50 $^{\circ}\text{C}$.

The fading-effect results were the same using both types of image readers, models FLA-3000 and BAS-5000. In Fig. 1, the measured PSL shows considerably larger fading effects using the BAS-TR than by using the BAS-MS. This result means that a large fading effect would accompany the detection of beta-rays from tritium using the BAS-TR. The BAS-MS-type IP shows a good fading characteristic, exhibiting only a small difference in the fading effect between 0 $^{\circ}\text{C}$ and 30 $^{\circ}\text{C}$. In all the fading curves in Fig. 1, however, the fading effect becomes stronger as the temperature increases. We found the relevant relation between the ambient temperature and PSL intensity [16] and established a method

Fig. 1. Comparison of the experimental results for the measured PSL values after irradiation with the tritium source generating bremsstrahlung X-rays using the BAS-MS and the beta-ray source using the BAS-TR in the range 0 to 50 $^{\circ}\text{C}$.

to develop a functional equation to correct for the fading effect [17]. The fading effect could be expressed as the sum of several exponentially decaying components. Because of its strong temperature dependence, we considered the fading effect to be a kind of thermal reaction, and the change of the reaction rate with temperature can then be expressed by the Arrhenius' equation [16, 17]. This is defined by the following relationship:

$$d \ln \lambda / dK = E / (RK^2) \quad (1)$$

where λ is the rate constant, E the activation energy, R the gas constant, and K the absolute temperature. By fitting the experimental results to the Arrhenius' equation, we developed a functional equation that includes two variables: elapsed time (t) and temperature (K). The fading equation after irradiation by bremsstrahlung X-rays using the BAS-MS is written as

$$\begin{aligned} (PSL)_{t,k} / (PSL)_{0,k} = & \\ & 0.348 \exp \left\{ -2.08 \times 10^{12} t \exp(-8.92 \times 10^3 / K) \right\} \\ & + 0.087 \exp \left\{ -9.89 \times 10^{10} t \exp(-8.69 \times 10^3 / K) \right\} \\ & + 0.374 \exp \left\{ -4.37 \times 10^{10} t \exp(-9.31 \times 10^3 / K) \right\} \\ & + 0.150 \exp \left\{ -2.41 \times 10^{10} t \exp(-9.54 \times 10^3 / K) \right\} \\ & + 0.041 \exp \left\{ -2.07 \times 10^9 t \exp(-9.53 \times 10^3 / K) \right\} \end{aligned} \quad (2)$$

Fig. 2 compares the calculated PSL density (PSL/mm²) from Eq. (2) with the experimental results, and indicates good agreement of both values in the range 0 to 50 °C and over all time periods. By using Eq. (2), we are able to correct the PSL values obtained at different elapsed times and/or at different temperatures and compare them.

Beta particles produce bremsstrahlung in passing through matter, especially if the absorbing medium is a high-Z material. The fraction of the beta energy that is converted to bremsstrahlung can be obtained by using empirical relations [18]. When the target is borosilicate glass, an energy yield fraction Y_i for the maximum energy of monoenergetic electrons is calculated to be approximately 10^{-4} . This rather

Fig. 2. Comparison of PSL density (PSL/mm²) calculated from Eq. (2) (line) and the experimental results (symbol). They are both in quite good agreement at all temperatures and over all time periods.

Fig. 3. Relationship between irradiation time (h) and PSL density obtained after irradiation with the glass tube filled with 12.5 MBq of tritium gas.

low yield fraction can be compensated to some degree by irradiating the IP with the source for much longer, utilizing the good fading characteristic of the BAS-MS-type IP. Fig. 3 shows an example of the relationship between irradiation time (h) and PSL density obtained after irradiation with the glass tube filled with 12.5 MBq of tritium. It shows excellent linearity and an equation obtained by linear regression is also shown in Fig. 3. Relationships among the PSL density of the background (BG) (left y-axis), the detection limit of tritium (right y-axis), and the irradiation time (h) are shown in Fig. 4. Error bars show one standard deviation for each value of the background. The detection limit was estimated from the equation obtained from the relationship between tritium radioactivity (Bq) and PSL intensity after irradiation with the glass tube filled with tritium gas of 12.5, 25, 50, and 100 MBq (not shown here) and three standard deviations from the mean PSL density of the background for each irradiation time. For 1 h of irradiation, the detection limit was estimated to be 970 kBq/cm². Longer irradiation periods increase the PSL intensity attributed to the background as well. To reduce this background effect, the IP was kept inside a lead outer shield during irradiation. The detection limit obtained was improved to approximately 90 kBq/cm² for a 24 h irradiation.

Fig. 4. Relationships between PSL density of background (left y-axis), detection limit of tritium (right y-axis), and irradiation time (h). Error bars show one standard deviation for each value of background.

3.2 Energy dependence of PSL sensitivities

Bremsstrahlung X-rays are produced by scattering due to the interaction between particles and the nucleus of an atom, so the energy of the bremsstrahlung can take any value from zero to the energy of the incident particle, and the energy spectrum varies depending on the atomic number and thickness of the target (or absorbing) material. In the IP technique using bremsstrahlung X-rays, PSL responses are affected by the variation of the energy spectrum depending on the thickness of the target (or absorbing material) and the energy dependence of the PSL sensitivities. Therefore, estimating the energy dependence of the PSL sensitivities and the configuration of the flat energy response is very important. The measured IP sensitivity (PSL per arbitrary unit) with and without aluminum filters of different thicknesses is plotted against the X-ray energy (keV) in Fig. 5. The result shows that the IP's sensitivity depends greatly on the X-ray energy, because the photostimulable phosphor is composed of elements with relatively high atomic numbers. The PSL intensity without an aluminum filter is quite low at 8 keV, indicating that most X-rays below 8 keV are attenuated by the thick protective film of the IP. It was also found that all of the PSL intensities with and without filters increase as the X-ray energy becomes higher. By combining the sensitivity data measured using aluminum filters 0.1, 0.3 and 0.5-mm thick, and without a filter, a constant PSL sensitivity of an IP per arbitrary unit independent of the X-ray energy can be obtained. By taking the weighted sum, Res_{sum} shown in Eq. (3) below, a response having a flat energy dependence can be obtained, as shown in Fig. 5:

$$Res_{sum} = 8.00 Res_{no\ filter} - 8.76 Res_{Al\ 0.1\ mm} - 2.00 Res_{Al\ 0.3\ mm} + 2.45 Res_{Al\ 0.5\ mm} \quad (3)$$

where $Res_{no\ filter}$, $Res_{Al\ 0.1\ mm}$, $Res_{Al\ 0.3\ mm}$, and $Res_{Al\ 0.5\ mm}$ are the IP sensitivities measured without a filter, and with aluminum filters 0.1, 0.3, and 0.5-mm-thick, respectively. The IP sensitivity obtained was constant to within $\pm 14\%$ deviation for X-rays with energies from 8.0 to 18.6 keV except at 13.5 keV, where Res_{sum} shows $\pm 25\%$ deviation.

Fig. 5. PSL sensitivities measured with and without aluminum filters of different thicknesses. The $\blacktriangle$ symbols show the weighted sum of PSL sensitivities measured with and without the different filters. The IP sensitivity was constant to within $\pm 14\%$ deviation for X-rays with energies from 8.0 to 18.6 keV except at 13.5 keV, where Res_{sum} shows $\pm 25\%$ deviation.

Fig. 6. Measured PSL densities and corrected PSL responses from Eq. (3) when different thicknesses of Ni foil in the range 5 to 25 μm were sandwiched between the glass tube containing 12.5 MBq of tritium and the IP. Error bars show 25% deviation for each corrected PSL value.

Fig. 6 shows the results of measured PSL densities when different thicknesses of nickel (Ni) foil in the range 5 to 25 μm were sandwiched between the glass tube containing 12.5 MBq of tritium and the IP, simulating migration of tritium into deeper regions of the nickel. PSL densities were measured with and without aluminum filters of different thicknesses in the range 0.1 to 0.5 mm. The relative PSL density was obtained by normalizing each PSL density to that without a nickel foil or aluminum filters (left y-axis). By combining the sensitivities measured using aluminum filters 0.1, 0.3 and 0.5-mm thick, and those without filters, PSL densities were corrected by using Eq. (3). The corrected PSL values are shown in Fig. 6 (right y-axis). Error bars show 25% deviation for each corrected PSL value.

Using Eq. (3), we can correct the energy dependence of the PSL sensitivities as shown in Fig. 6. In combination with the energy spectrum information (depth profiles), the amount of tritium in deeper regions can be quantified by this technique. These comprehensive results indicate that the tritium IP technique based on detecting bremsstrahlung X-rays can be a new method for the non-destructive monitoring of tritium migration into deeper layers.

4. Summary

The characteristics of an IP for the measurement of bremsstrahlung induced by tritium beta-rays were examined. A BAS-MS-type IP shows a better fading characteristic, exhibiting only a small difference in the fading effect between 0 $^{\circ}\text{C}$ and 30 $^{\circ}\text{C}$, compared to that obtained using the BAS-TR-type IP. A functional equation was developed to correct the fading effect observed in the BAS-MS. Both the calculated PSL values and the experimental results are in quite good agreement at all temperatures and over all time periods. Using this equation, we are able to correct the PSL values obtained at different elapsed times and/or at different temperatures and compare them. The rather low yield fraction of the beta energy that is converted to bremsstrahlung

can be compensated by irradiating the IP with the source for a much longer time. The detection limit was improved when the IP was kept inside a lead outer shield during irradiation to reduce the background effect. The dependence of the PSL sensitivities on energy was measured by using monoenergetic X-rays from 10 to 18.6 keV. The weighted sum of a few PSL responses with different thicknesses of aluminum filters was found to achieve a flat energy response with a PSL per arbitrary unit within $\pm 14\%$ deviation for X-rays with energies from 8.0 to 18.6 keV, except at 13.5 keV where Res_{sum} shows $\pm 25\%$ deviation. In combination with the energy spectrum information, the amount of tritium in deeper regions can be quantified by this IP technique using bremsstrahlung X-rays.

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