Tritium Measurement Using a Photo-Stimulable Phosphor BaFBr(I):Eu2+ Plate

IV. 2. Tritium Measurement Using a Photo-Stimulable Phosphor BaFBr(I):Eu^{2+} Plate

Ohuchi H.[1] and Hatano Y.[2]

[1]Graduate School of Pharmaceutical Sciences, Tohoku University
[2]Hydrogen Isotope Research Center, University of Toyama

Introduction

Tritium measurement is indispensable for the fuel-processing systems of deuterium-tritium (DT)-fusion facilities. 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. 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. We have developed a new approach to detecting tritium using the bremsstrahlung induced by tritium beta-rays with the IP. 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. In this 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 photostimulated luminescence (PSL) sensitivities. The fading effect after irradiation with bremsstrahlung X-rays from tritium was compared with that obtained after irradiation with the beta particles emitted from tritium.

Materials and Methods

A BAS-MS type-IP was used to detect bremsstrahlung X-rays from tritium and a BAS-TR-type IP (FUJIFILM Co., Ltd.) to detect beta particles. The differences in structures of these two are listed in Table 1. To read out the IP, an image reader fabricated by FUJIFILM Co., Ltd. was used. 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 (manufactured by GE Healthcare) were used as the beta-ray radiation source. Each strip contained approximately 2.96 kBq of tritium. 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°C. The IPs were kept in an aluminum IP cassette inside an incubator 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). 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.

Results and discussion

Figure 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°C. In Fig. 1, the BAS-MS-type IP shows a good fading characteristic, exhibiting only a small difference in the fading effect between 0ºC and 30ºC. In all the fading curves, however, the fading effect becomes stronger as the temperature increases. We found the relevant relation between the ambient temperature and PSL intensity[1)] and established a method to develop a functional equation to correct for the fading effect[2)]. 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

$$(PSL)_{t,k} / (PSL)_{0,k} =$$
$$0.348 \cdot \exp\{-2.08\times10^{12} \cdot t \cdot \exp(-8.92\times10^{3}/K)\}$$

$$+ 0.087 \cdot \exp\{-9.89\times10^{10} \cdot t \cdot \exp(-8.69\times10^{3}/K)\}$$
$$+ 0.374 \cdot \exp\{-4.37\times10^{10} \cdot t \cdot \exp(-9.31\times10^{3}/K)\}$$
$$+ 0.150 \cdot \exp\{-2.41\times10^{10} \cdot t \cdot \exp(-9.54\times10^{3}/K)\}$$
$$+ 0.041 \cdot \exp\{-2.07\times10^{9} \cdot t \cdot \exp(-9.53\times10^{3}/K)\} \quad (2)$$

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.

When the target is borosilicate glass, an energy yield fraction Yi for the maximum energy of monoenergetic electrons converted to bremsstrahlung is calculated to be approximately 10^{-4} [3]. This rather 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. 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. 2. 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/cm^2. 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/cm^2 for a 24 h irradiation.

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. 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. 3. The result shows that the IP's sensitivity depends greatly on the X-ray energy. 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. 4:

$$Res_{sum} = 8.00 \cdot Res_{no\ filter} - 8.76 \cdot Res_{Al0.1mm} - 2.00 \cdot Res_{Al0.3mm} + 2.45 \cdot Res_{Al0.5mm} \quad (3)$$

where $Res_{no\ filter}$, $Res_{Al0.1mm}$, $Res_{Al0.3mm}$, and $Res_{Al0.5mm}$ 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 ±14% deviation for X- rays with energies from 8.0 to 18.6 keV except at 13.5 keV, where Ressum shows ±25% deviation.

Figure 4 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. 4 (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. 4. 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.

References

1) Ohuchi H., Yamadera A., Nakamura T., Nucl. Instr. and Meth. **A450** (2000) 343.
2) Ohuchi H., Yamadera A., Rad. Meas. **35** (2002) 135.
3) Martin, J.E.: Physics for Radiation Protection, 2nd ed., Litges & Dopf Buchbinderei GmbH, Heppenheim (2006) 371.

Table 1. Structures of the 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	$BaF(Br_{0.85}+I_{0.15})$	BaFBrI[1]
Thickness of base (μm)	190	250
Thickness of ferrite layer (μm)	160	160

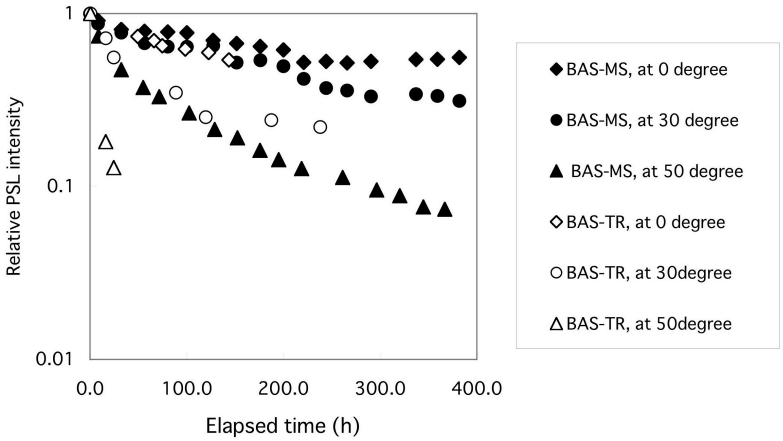

Figure 1. Comparison of the experimental results for the measured PSL values after irradiation with the tritium source generating bremsstrahlung X-rays using the BAS-MS and the beta-ray source using the BAS-TR.

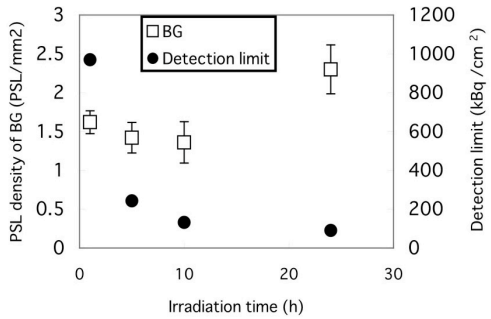

Figure 2. Relationships between PSL density of background (left y-axis), detection limit of tritium (right y-axis), and irradiation time (h).

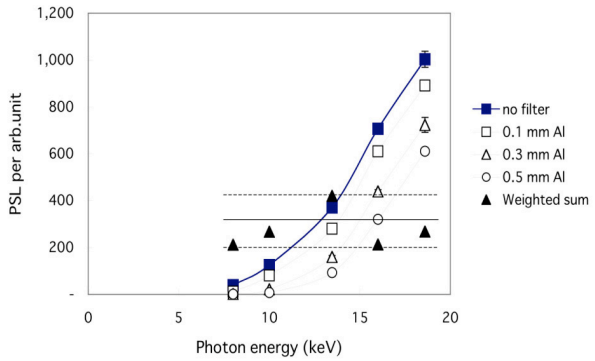

Figure 3. PSL sensitivities measured with and without aluminum filters of different thicknesses. The ▲ symbols show the weighted sum of PSL sensitivities measured with and without the different filters. The IP sensitivity was constant to within ±14% deviation for X- rays with energies from 8.0 to 18.6 keV except at 13.5 keV, where Ressum shows ±25% deviation.

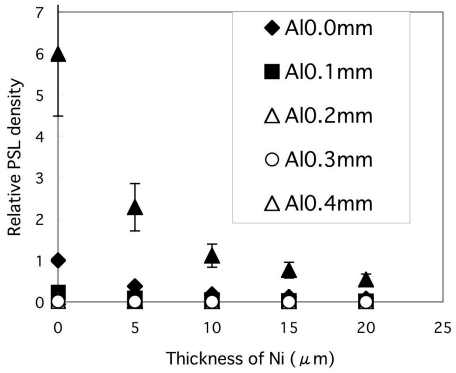

Figure 4. Measured PSL densities and corrected PSL responses from Eq. (3) when different thicknesses of Ni foil in the range 5 to 25 μm were sandwiched between the glass tube containing 12.5 MBq of tritium and the IP. Error bars show 25% deviation for each corrected PSL value.