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ARTICLE

Evaluation of imaging plate measurement for activated indium as fast-neutron detector in large radiation field

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The application of imaging plate (IP) method using indium foils to measure the distribution of fast neutron in large area was evaluated in this work. The radioactive nuclides of indium were individually quantified by high-purity germanium detector (HPGe) and chronological IP measurements. The detection efficiencies of each radioactive nuclide for various thicknesses of foil mylar polyester film were summarized. The scope of application for proposed method were clearly dependent on the flux ratio of fast and thermal neutron, limiting the applicable region only being close to the neutron source.

Keywords: imaging plate; indium; neutron; gamma-ray; beta-ray; HPGe

1. Introduction

Activation foil method has been widely used as the neutron detector in many radiation field such as nuclear fission and fusion research reactors and accelerator facilities, so on [1]. The indium (In) has the advantages of well-established database of the cross-sections to neutron with wide range of energy as well as the high cross-sections with neutrons. Especially, ^{115}In(n,n')^{115m}In reaction has the threshold energy of neutron, indicating that indium foils can be used as the fast neutron detector.

In the field of fusion plasma research, the understanding of neutron behaviors and neutron flux as well as its subsequent activation to reactor components is important since energetic neutrons were generated by the fusion reactions of hydrogen isotopes. The fusion research devices such as Large Helical Device (LHD) are equipped with many heating and diagnostic systems, the experimental building which holds that devices needs to be large. The actual size of the torus hall of LHD is W 75 × L 45 × H 40 m. The measurement technique applicable to evaluate the radioactivity of many foils with identifying the radioactive nuclides in one time lets one to survey the distribution of fast neutron fluxes in the torus hall of LHD easily.

IP method is one of the applicable technique to measure many foils in one time. IP can integrate the energy deposited from radiation, and release it as photo-stimulated luminescence (PSL) in measurements. Therefore, it is quite difficult to quantify the precise radioactivity with identifying radioactive nuclides in sample when several kinds of radioactive nuclides exist. The reactions of ^{115}In (major isotope of indium with the natural abundance of 95.7 %) with neutron result in ^{116m}In by thermal neutrons as well as ^{115m}In by fast neutrons. Also, minor isotope of indium as ^{113}In with the natural abundance of 4.3 % is also active to thermal neutron and transmutes to ^{114m}In. These radioactive nuclides release both of beta-rays and gamma-rays with different energies, and these complex radiations produce one active site in IP sheet. These characteristics make the measurement of fast neutron fluence difficult.

Therefore, the application of IP method with using indium foil for fast-neutron detection was evaluated in this study. The detection efficiencies of IP for quantifying three kinds of radioactive indium nuclides were determined. In addition, the dependencies of self-shielding of indium and outer-shielding were investigated to optimize the effective detection, especially for ^{115m}In.

2. Experimental

2.1. Neutron irradiation for In foils

Indium foils with the purity of 99.99 % were purchased from Nilaco Co. Three kinds of foils with different thicknesses as 0.1. 0.5 and 1.0 mm were investigated. These foils were cut into 1.0 mm$^{\phi}$ or smaller to encapsulate for neutron irradiation.

After foils were sealed into capsule made of

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polyethylene, they were introduced into the neutron activation system of LHD [2]. The capsule was transferred through pneumatic tubes by pressurized air to near the vacuum vessel of LHD where the deuterium plasma was generated for about 3 seconds. The capsule containing indium foils were irradiated by neutrons generated by deuterium fusion reaction in LHD for several times to integrate the radioactivity.

2.2. Measurement of radioactivity in foils by HPGe

Then, some pieces of foils were introduced into HPGe detector to quantify the radioactivity of each radioactive nuclide for calibrating the PSL values in IP measurements. The HPGe detector used in this work was of Canberra Industries, Inc. (Model: GX3018/CP5-PLUS-U). Because the detector is in a lead shield with the thickness of 100 mm, the background pulse counting rate due to external sources is low enough. Output pulses from preamplifier are fed into multichannel analyzer, DSA-LX of Canberra, based on advanced digital signal processing techniques, and data is analyzed on a personal computer. The data were acquired for 3600 s for each measurement.

For the calibration of the gamma-ray pulse counting rate to the radioactivity, the calibration curves were prepared in advance with using the multi gamma-ray sources in U8 container which contain several kinds of gamma-ray emitting radioisotopes. Also, these calibration curves were obtained for various height of multi gamma-ray sources to measure the correlation between the height of specimen and the calibration factor. In the calculation of the radioactivities of foils in this work, the calibration curve corresponding to the foil thicknesses, which can be deduced from the calibration curves obtained from the various height of multi gamma-ray sources, was used. It was assumed that the size of specimen does not influence on the calibration processes as both sizes of U8 container and foils were enough smaller than that of HPGe detector.

2.3. Evaluation of radioactivity in foils by IP

The IP sheet used in this work was BAS-SR 2040 manufactured by FUJIFILM. To identify the radioactive nuclides generated in foils, the chronological IP measurements were carried out. In these measurements, the foils were put on the IP for certain period to integrate the PSL values, and the PSL values were intermittently evaluated by image reader as Typhoon FLA 9500 of GE Healthcare. After a measurement, the IP sheet was exposed to intense light to remove the residual luminescence. The averaged PSL values were calculated by using a software of Image Quant TL developed by GE Healthcare. The background PSL value was subtracted by measuring the PSL values for the region of IP sheet away from foils. The chronological IP measurements were continued up to the PSL value being unchanged.

According to the HPGe measurements, the concentrations of radioactivity in In foils were slightly different although these foils were irradiated with neutron in the same capsule. Therefore, the following factor f was adopted in this work to precisely evaluate the influence of foil thickness on the detection efficiency in IP measurement.

$$f_k = \frac{I_k}{t_k - t_{k-1}} \left(\frac{L}{V} \frac{dN}{dt}\bigg|_{t=0} \right)^{-1}$$

Here, I, t, L, V and N indicate the average PSL [mm^{-2}], time [s], thickness of foil [mm], volume of foil [mm^3] and the individual amounts of radioactive isotopes of indium as ^{116m}In, ^{115m}In and ^{114m}In in a foil, respectively. The subscript of k is the number of measurement times.

In IP measurements, the mylar polyester film was used as outer shielding especially for beta-ray. The minimum thickness of mylar polyester film was 100 μm, and the desired thickness of film was produced by stacking this film to evaluate the shielding effects on detection efficiencies. For this evaluation, the following factor r was defined as follows.

$$r_k = \frac{f_k^{(s)}}{f_k^{(0)}}$$

The superscript of s in parenthesis just expresses the thickness of mylar polyester film.

3. Results and discussion

Figure 1 shows the gamma-ray energy spectra for indium foils with the thickness of 1 mm when elapsing several hours after neutron irradiation. As can be seen, many peaks by X-rays and gamma-rays appeared in the energy spectrum obtained after elapsing with 1 hour. However, many of them disappeared after elapsing with 8 hours. The intense gamma-ray peaks around 417, 1097 and 1294 keV marked with filled circle in Fig. 1, are typical gamma-ray peaks by ^{116m}In. The intensities of these peaks became to two orders of magnitude smaller after elapsing 8 hours. This result indicates that major radioactivity for indium foil just after neutron irradiation was ^{116m}In and it decayed preferentially with its shorter half-life as 54.3 min [3]. The energy spectrum obtained after elapsing with 8 hours also showed that the gamma-ray peak at around 336 keV marked with solid

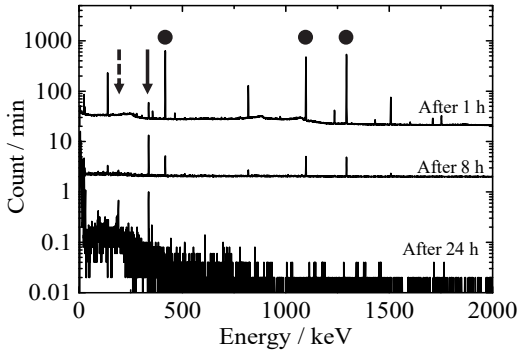

Figure 1. The γ-ray energy spectra for neutron irradiated indium foils after different elapsed times.

arrow became dominant. Also, the peak at around 190 keV marked with dashed arrow was prominent in the energy spectrum after elapsing with 24 hours although the peak at 336 keV still appeared. These peaks at around 190 and 336 keV were assigned to the gamma-ray by ^{114m}In and ^{115m}In which decayed with the half-lives as 49.5 days and 4.49 hours, respectively.

Figure 2 shows the results of chronological IP measurements for foils with different thicknesses. The thickness of mylar polyester film in these measurements were 100 µm. The elapsed time in Figure 2 indicates the half point of the beginning and stopping times for each integration of PSL value in IP measurement. The time of $t = 0$ means almost 3 hours past after neutron irradiation. Note that the f factor in this figure was deduced with using the amount of ^{116m}In for all measurement times. There were three stages on the decrement behaviors of radioactivity. In the stage 1 which is categorized by 0 - 500 min, the radioactivity was exponentially decreased. The radioactivity also decreased exponentially in the stage 2 which begins from around 500 min to around 1200 min although the exponential factor was much smaller than that of stage 1. Then, the radioactivity hardly showed any change in further elapsed time (stage 3). According to the half-lives of three kinds of radioactive nuclides of indium, the decrement behaviors of radioactivity in stages 1-3 were dominated by the decays of ^{116m}In, ^{115m}In and ^{114m}In, respectively. For stage 3, the decrement of radioactivity was hardly appeared as the half-life of ^{114m}In was much longer than the periods of chronological IP measurements carried out here.

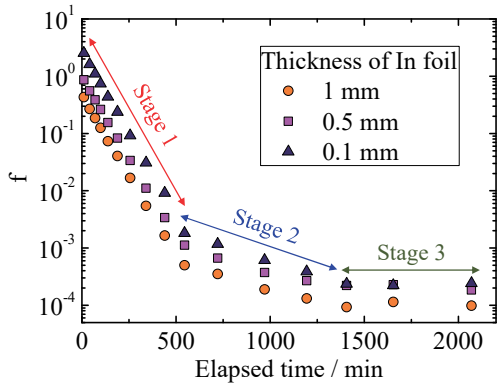

Figure 2. Time-dependent changes of f factor for indium foils with different thicknesses.

Figure 2 also suggests that the detection efficiency was related to the thickness of the indium foil. It can be found that thinner foil had the higher detection efficiency. In the integration process of PSL in IP sheet, the efficiency of energy deposition from radiation relates to the energy transfer rate which depends on the radiation, energy so on. Generally, beta-rays more effectively interact with solids compared to the case of gamma-rays due to its charge [4]. It can be expected that major radiation which contributes to the PSL value should be beta-ray. Taking the above processes of PSL

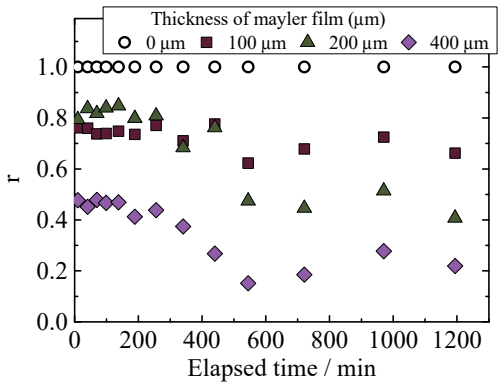

Figure 3. Changes of the detection efficiency of radiation from indium by the thickness of mylar polyester films.

integration on IP sheet into consideration, the higher detection efficiency for thinner foils would be caused by the self-shielding of beta-rays by indium foil itself. The thicker foils effectively stop the beta-ray to being out from foils, and subsequently interact with IP sheet, resulting in the lower detection efficiency. According to the recent work by Pu [2], the self-shielding of gamma-rays by indium foil was estimated to be negligible, supporting our interpretation for lower efficiency in thicker foils due to self-shielding of beta-rays.

The effects of outer-shielding are summarized in **Figure 3**. As found in Figure 3, the film worked to shield the radiation for both ^{116m}In and ^{115m}In. The r value for stage 2 showed that it almost proportionally depended on the thickness of film, and the shielding effectivity for ^{115m}In was higher than that for ^{116m}In. These differences on the shielding effectivities for the radiations from ^{115m}In and ^{116m}In were caused by many reasons. One of them would be relatively higher beta-ray energies and higher emission rates of gamma-rays for ^{116m}In. The other reasons would be lower emission rate of beta-ray and lower energies of gamma-ray for ^{115m}In. Anyway, the purpose of applying the mylar polyester films as outer-shielding was to decrease the contribution of ^{116m}In on PSL value, and to consequently emphasize that of ^{115m}In. However, it was found that the films shielded the radiation for ^{115m}In more effectively. Therefore, it should be difficult to use mylar polyester films to extract the radiation for ^{115m}In. The mylar polyester film itself is useful to stick many samples for chronological measurements, where many samples are repeatedly measured. For this application, the thinner thickness of mylar polyester film should be better.

The calibrations of PSL values to radioactivity were done for all tests carried out in this work and are summarized in **Table 1**. For the determination of these calibration factors, the exponential fitting of PSL values with elapsed time was conducted. The exponential factors obtained in this process were also shown in this table. The exponential factors indicate the decay rates for each radioactive nuclide. These factors in stages 1 and 2 were consistent with the decay rates for ^{116m}In and ^{115m}In, respectively, with the valid coefficients of

Table 1. Calibration factors for the conversion of PSL integration rate [cm^{-2} min^{-1}] to radioactivity [Bq cm^{-2}] for radioactive indium nuclides measured under various conditions.

Foil thickness (mm)	1.0	1.0	1.0	1.0	0.5	0.1
Mylar polyester film (μm)	0	100	200	400	100	100
Calib.factor for ^{116m}In	1.62	2.14	1.90	3.31	1.04	0.356
Exp. factor(min^{-1})	-0.0129	-0.0130	-0.0134	-0.0136	-0.0131	-0.0133
R^2	0.99	0.99	0.99	0.99	0.99	0.99
Calib.factor for ^{115m}In	6.15	12.1	19.8	71.0	5.98	1.55
Exp. factor(min^{-1})	-0.00269	-0.00254	-0.00234	-0.00252	-0.00257	-0.00266
R^2	0.98	0.96	0.95	0.97	0.94	0.98
Calib.factor for ^{114m}In		4.46				

determination. The lower calibration factors obtained here means the higher detection efficiency. These trends of detection efficiencies in Table 1 were consistent with the results as shown in Figures 2 and 3. Also, the calibration factor for ^{114m}In was determined assuming the decay rate of 9.7×10^{-7} min^{-1} which can be expected from the decay rate of ^{114m}In as $\ln(2) / t_{1/2}$.

As found in Figure 2, the quantification of ^{115m}In is interfered by ^{114m}In as well as ^{116m}In. The scope of application of IP method with indium foils for fast neutron detector depends on the neutron energy spectrum in the radiation field. Here, the simple assumption, where the two types of neutron with the single energies of 2.45 MeV (fast) and 0.025 eV (thermal) are irradiated into indium foil, was considered. The cross-sections of neutron capture reactions for ^{113}In(n,γ)^{114m}In, ^{115}In(n,γ)^{116m}In and ^{115}In(n,n')^{115m}In were assumed to be constants as around 12, 201 and 2 barn, respectively [5]. The calculated total integration rates of PSL values under different ratios of fast and thermal neutron flux are shown in **Figure 4**. The individual integration rates of PSL by ^{116m}In, ^{115m}In and ^{114m}In are also presented here to observe the contribution of these nuclides. The neutron irradiation for 1500 min, which is the saturation time for radioactivity of ^{115m}In, and subsequent stopping of irradiation is assumed in this figure. The cases in the flux ratio for fast to thermal neutrons of 1.0 and 0.1 with keeping thermal neutron flux constant are calculated. After stopping the neutron irradiation, the total PSL integration rate decreased by the decay of ^{116m}In, and became being dominated by the decay of ^{115m}In in the case of the fast and thermal neutron flux ratio of 1.0. This trend is almost consistent with the experimental results as found in Figure 2. On the contrary, the decay of ^{115m}In cannot be comparable with the total PSL integration rate in the case of the fast and thermal neutron flux ratio of 0.1, because the radioactivities of ^{116m}In and ^{114m}In became relatively higher in this case. Nevertheless, the quantification of ^{115m}In can be done by the subtraction of total PSL value by other nuclides, although this subtraction process will be much more difficult in higher thermal neutron flux field.

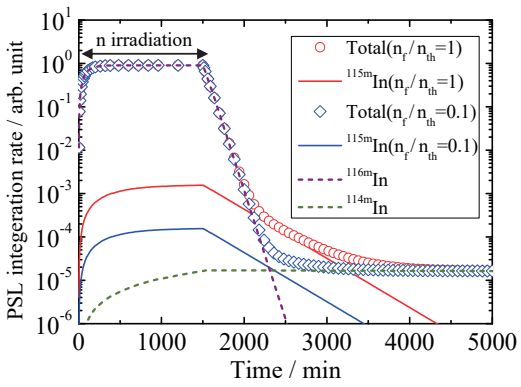

Figure 4. Prediction of PSL integration rate for indium under and after neutron irradiation with different the fast and thermal neutron flux ratios.

As the scope of application for the IP method with using indium foils as fast neutron detector, it was evaluated that this method should be applicable on the field where the fast neutron flux is relatively higher than that of thermal neutron. Also, the accumulation of ^{114m}In must be considered as ^{114m}In can integrate in foil for long time of neutron irradiation. In the fusion plasma, fast neutrons nearly monoenergetic are generated, then, these neutrons will lose their energy quickly by the interaction with components around the device. The application of a combination usage of IP and indium foils as fast neutron detector depends on those energy-loss processes of neutrons as expected from this work. It was considered that the region where the indium foils are applicable for fast neutron detector should only be close to the neutron source. Also, these foils should not be stored in that field more than 1 day to refrain the accumulation of ^{114m}In which will interfere the detection of ^{115m}In in IP measurements. One of the ways to improve the application of this method would be with using thermal neutron filter such as cadmium sheet. The cadmium has a large cross-section with thermal neutron, therefore, a cadmium sheet enveloping indium foils prevent the thermal neutrons approaching to indium foils. The generation of ^{116m}In and ^{114m}In in indium foils will be reduced by this method, consequently ^{115m}In will be more detectable.

4. Conclusion

The application of IP method using indium foils to measure the distribution of fast neutron in large area was evaluated, and following conclusions were obtained.

- The radioactivities of ^{114m}In, ^{115m}In and ^{116m}In in indium foil were individually measured by the HPGe and the chronological IP measurements.
- Relatively higher detection efficiencies were obtained with being the thickness of indium foil thinner due to self-shielding of beta-rays.
- The outer-shielding using mylar polyester film hardly showed a higher detection efficiency for ^{115m}In.
- The scope of application of proposed method were clearly dependent on the flux ratio of fast and thermal neutron, limiting the applicable region only being close to the neutron source.

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References

[1] L.C. Johnson, C.W. Barnes, H.H. Duong, W.W. Heidbrink, D.L. Jassby, M.J. Loughlin, A.L. Roquemore, E. Ruskov and J.D. Strachan, Cross calibration of neutron detectors for deuterium-tritium operation in TFTR, *Rev. Sci. Instrum*. 66 (1995), p. 894.

[2] N. Pu, T. Nishitani, M. Isobe, K. Ogawa, H. Kawase, T. Tanaka, S. Li, S. Yoshihashi and A. Uritani, In situ calibration of neutron activation system on the large helical device, *Review of Scientific Instruments* 88 (2017), p.113302. R.B. Firestone, Table of isotopes 8th edition, John Wiley & Sons, Inc. 1996.

[3] A.L. Gonzalez, H. Li, M. Mitch, N. Tolk and D.M. Duggan, Energy response of an imaging plate exposed to standard beta sources, *App. Radiat. Isot.* 57 (2002), pp. 875-882.

[4] K. Shibata, O. Iwamoto, T. Nakagawa, N. Iwamoto, A. Ichihara, S. Kunieda, S. Chiba, K. Furutaka, N. Otuka, T. Ohsawa, T. Murata, H. Matsunobu, A. Zukeran, S. Kamada and J. Katakura, JENDL-4.0: A new library for nuclear science and engineering, *J. Nucl. Sci. Technol*. 48(1) (2011), pp. 1-30.