Sensors and Materials, Vol. 29, No. 10 (2017) 1439–1445
MYU Tokyo

S & M 1437

Visualization of Radiation Dose Distribution Utilizing Radiophotoluminescence in Glass Dosimeter

Hidehito Nanto,* Yuka Yanagida,[1] Makoto Sugiyama,[1] Yasuhiro Koguchi,[1] Yohei Ihara,[2] Toshiyuki Iida,[3] Kikuo Shimizu,[3] Takuma Ikeguchi, Kazuki Hirasawa, Yoshinori Takei, and Takayoshi Yamamoto[1]

Advanced Materials Science R&D Center, Kanazawa Institute of Technology, 3-1 Yatsukaho, Hakusan, Ishikawa 924-0838, Japan
[1]Oarai Research Institute, Chiyoda Technology Co., 3681 Narita, Oarai, Ibaraki 311-1313, Japan
[2]EMF Japan Co. Ltd., 1-1-4 Kawachinagano, Osaka 586-0077, Japan
[3]Faculty of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

(Received April 5, 2017; accepted July 12, 2017)

Keywords: radiophotoluminescence, glass dosimeter, dose distribution, passive-type dosimeter

In the decommissioning of a nuclear power plant, especially for the decontamination of radioactive substances caused by the 2011 Fukushima nuclear power plant accident, which has affected reconstruction after the earthquake, dose evaluation and validation of the decontamination effect are very important. To prepare a novel passive-type dosimeter system, which can be applied to radiation dose monitoring and visualization of the two-dimensional distribution of ionizing radiation in a contaminated area, we tried to develop bead- and sheet-type glass dosimeters utilizing the radiophotoluminescence (RPL) phenomenon in Ag^+-doped phosphate glass. It is confirmed that the bead-type glass dosimeter with a diameter of about 50 μm exhibits an intense RPL. It is found that the sheet-type glass dosimeter, which is made of bead-type glasses, is useful for the monitoring of radiation dose in a wide dose range and the visualization of dose distribution.

1. Introduction

Silver (Ag^+)-doped phosphate glasses irradiated with ionizing radiation exhibit an intense emission of visible photons when excited by ultraviolet (UV) light. This phenomenon is called radiophotoluminescence (RPL). The RPL phenomenon in Ag-doped phosphate glass has, therefore, been applied to a passive-type (accumulation-type) radiation dosimeter.

When the glasses are exposed to ionizing radiation, electron–hole pairs will be induced in the phosphate glass. The electrons are captured into Ag^+ ions in the glass structure and then the Ag^+ ions change to Ag^0 ions. On the other hand, the holes are captured by PO_4 tetrahedra at the beginning of the migration and then the holes come to produce the Ag^{2+} ions due to interaction with Ag^+ ions over time. It has been reported that both Ag^0 and Ag^{2+} ions can be the luminescent centers in the phosphate glasses.[1–5] The RPL intensity is proportional to the amount of ionizing radiation. The RPL glass dosimeter has excellent characteristics such as homogeneous and stable radiation sensitivity, negligible fading, and reliable repeatability in radiation dose measurement. The RPL luminescence centers never disappear after reading out the accumulated information

*Corresponding author: e-mail: hnanto@neptune.kanazawa-it.ac.jp
http://dx.doi.org/10.18494/SAM.2017.1624

ISSN 0914-4935

by UV light again and again, unless the glasses are annealed at a high temperature of about 400 °C. This is the most different characteristic of the RPL phenomenon from that of the optically stimulated luminescence (OSL), which is used as one of the important processes for a passive-type dosimeter, that is, individual radiation monitoring. Therefore, these glasses have been widely used as passive-type personal dosimeters at many radiation facilities in Japan, France, and Germany.

We have tried to develop new bead- and sheet-type glass dosimeters that are usable from low to high dose conditions such as nuclear emergencies, especially for radiation dose monitoring near or inside the Fukushima Nuclear Power Plant-1 (NPP-1) in which a large amount and many kinds of radioactive materials were released by a catastrophic earthquake on March 11, 2011. In this study, two types of glass dosimeters, the bead type and the sheet type, are developed to adapt to the various environments in radioactively contaminated areas. The sheet-type glass dosimeter with a two-dimensional RPL reader is applied to the monitoring and visualization of radiation dose distribution.

2. Experimental Procedure

Two types of RPL glass dosimeters, the bead type and the sheet type, have been developed to adapt to various monitoring environments. The bead-type glass dosimeters with a diameter of about 50 μm were fabricated with a jet flame system, which was composed of a nozzle, a power feeder, and gas cylinders. Pulverized fragments of normal RPL glass were injected into the system together with fuel gas. The glass fragments were quickly heated up to about 3300 K in a jet flame. Glass beads were well formed in the cooling process in air, thanks to the surface tension and uniform heat transfer.(6,7)

A flexible RPL glass sheet can be made of the bead-type RPL glass dosimeters and is expected to be useful for two-dimensional radiation imaging. The bead-type RPL glass dosimeters are also expected to be effectively used for dose monitoring in radioactively contaminated areas.

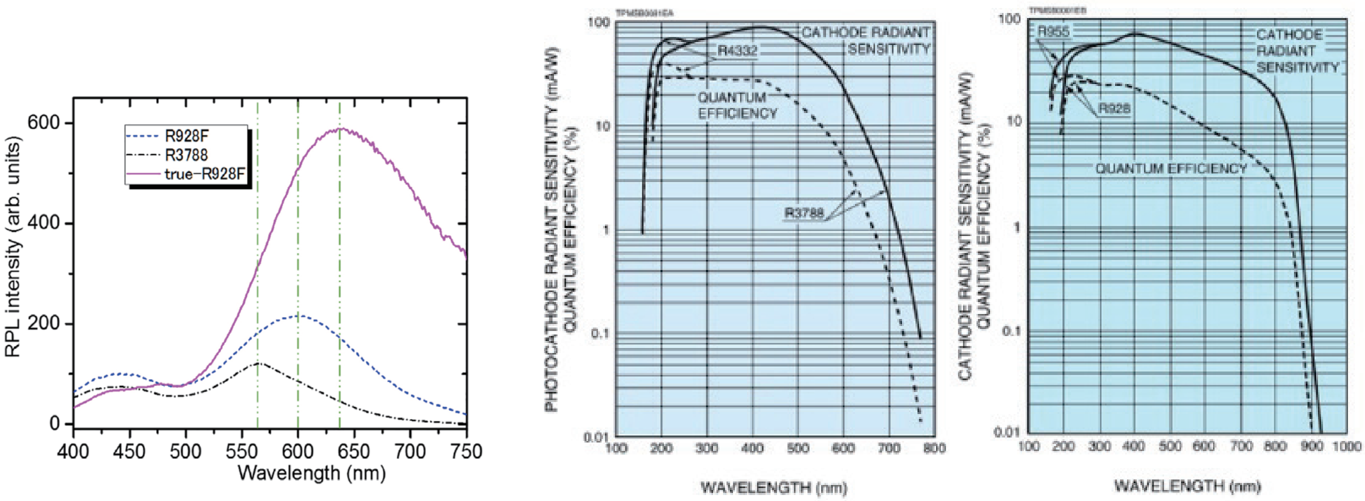
X-ray irradiation of the Ag^+-activated phosphate glass was carried out using an X-ray tube with a Cu target, operated at 30 kV and 20 mA. The emission and excitation spectra of RPL of the Ag^+-activated phosphate glasses were obtained using a Hitachi F-4500 fluorescence spectrometer at room temperature. The RPL spectra were corrected for the diffraction efficiency of the grating and the optical response of the photomultiplier.

Phosphate glass, the composition of which is the same as that of the commercially available glass dosimeter (GD-450, AGC Techno Glass Co., Ltd.), was used in this experiment. The weight composition of the Ag^+-doped phosphate glass was 31.55% P, 51.16% O, 6.12% Al, 11.00% Na, and 0.17% Ag. The glass is transparent from 300 to 800 nm in wavelength, which was measured with a Hitachi Spectrophotometer (F-4500).

3. Results and Discussion

Figure 1 shows typical RPL emission spectra of X-ray irradiated Ag^+-doped phosphate glass using a fluorescence spectrometer (Hitachi F-2010), which is equipped with a photomultiplier tube (PMT) R3778 or R928, supplied by Hamamatsu Photonics K.K.

We previously reported(1) that there are two emission bands, a blue band (peaked at about 460 nm) and a yellow band (peaked at 560 nm), in the RPL spectrum, which was observed using the PMT-R3778. It is seen from Fig. 1 that the RPL spectrum, which was measured using the



(a) (b)

Fig. 1. (Color online) (a) Typical RPL emission spectra of X-ray irradiated Ag-doped phosphate glass using a fluorescence spectrometer (Hitachi F-2010) equipped with a PMT R3778 or R928, supplied by Hamamatsu Photonics K.K. and (b) typical spectral response of PMT-R3778 (left) and PMT-R928 (right).

PMT-R928, is red-shifted in comparison with that measured using the PMT-R3778. This red shift of the RPL spectrum is ascribed to the difference in spectral response of the PMT.

The RPL emission images as a function of X-ray absorbed dose when the X-ray irradiated Ag^+-doped phosphate glass is excited using an UV black light of about 365 nm wavelength are shown in Fig. 2. It is seen that the RPL intensity, orange color emission to the human eye, increases with the X-ray absorbed dose. It was confirmed that the RPL intensity linearly increases with the absorbed dose of X-rays in the range from 0.01 to ~1000 Gy.[8,9] The RPL emission images of ^{60}Co-gamma-ray-irradiated bead-type RPL glasses with a diameter of about 50 μm are shown in Fig. 3. It can be seen that an intense RPL orange emission is observed from a homogeneous-shape bead-type dosimeter of Ag^+-doped phosphate glass.

Figure 4 shows photographs of the bead-type RPL glass exposed to ^{60}Co gamma rays. The bead-type RPL glasses with 35 mm diameter were set in petri dishes and covered with polystyrene plates of 0.8 mm thickness. At doses above 1 Gy, orange RPL could be observed with the naked eye. In addition, RPL was observed with a digital camera at doses above 0.5 Gy. This result strongly suggests that the bead-type glass dosimeters are applicable to the visualization of dose distribution of contaminated water, for example, in a nuclear power plant, by floating a plastic capsule with bead-type glass dosimeters in the contaminated water.

The sheet-type RPL glass dosimeters were made by kneading the mixture of bead-type glass (average diameter was about 50 μm) and an acrylic binder on an organic film. The acrylic resin film was used as a base film of organic materials. It was confirmed that the acrylic resin film excited with UV light did not exhibit luminescence, which does not disturb the RPL signal (orange luminescence). Figure 5 shows the RPL image [Fig. 5(b)] from an X-ray irradiated sheet-type glass dosimeter, which was shielded using a copper (Cu) mesh [Fig. 5(a)] when the sheet was iiradiated with X-rays. It can be seen that intense orange RPL emission was observed from the X-ray irradiated area.

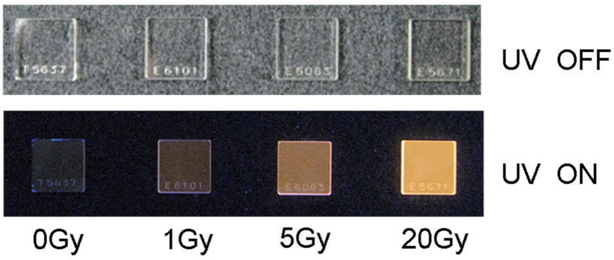


Fig. 2. (Color online) RPL emission images as a function of X-ray absorbed dose when the X-ray irradiated Ag-doped phosphate glass is excited using a UV black light.

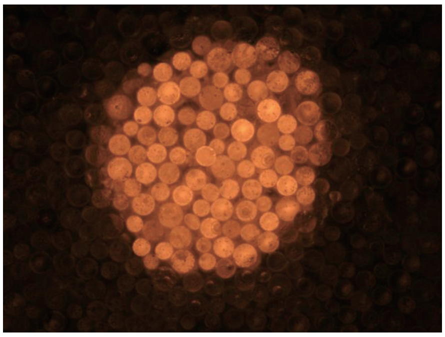


Fig. 3. (Color online) RPL emission image of ^{60}Co-gamma-ray-irradiated bead-type RPL glasses with a diameter of about 50 μm.

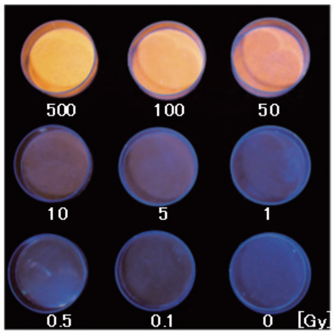
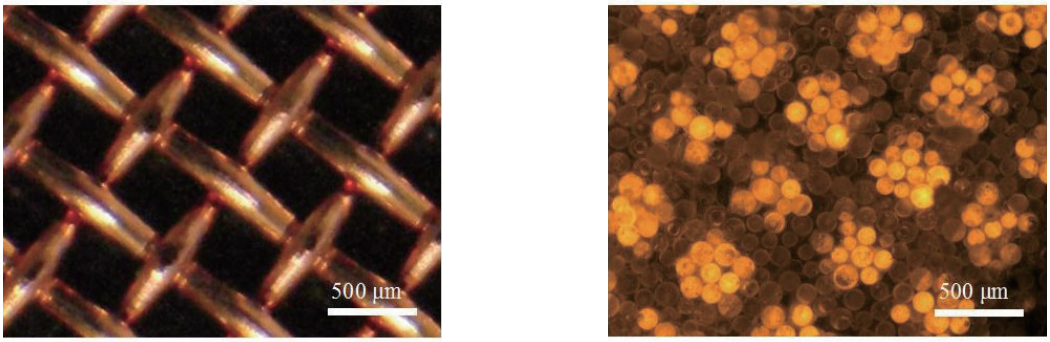


Fig. 4. (Color online) Photographs of bead-type RPL glass exposed to ^{60}Co gamma rays. The bead-type RPL glasses of 35 mm diameter were set in petri dishes and covered with polystyrene plates of 0.8 mm thickness.



(a) (b)

Fig. 5. (Color online) (a) RPL image from the sheet-type RPL glass dosimeter irradiated with X-rays and (b) photograph of a Cu mesh used for radiation shielding.

The pictures of the system for reading the two-dimensional radiation dose latent image written in a sheet-type glass dosimeter are shown in Fig. 6. The monitoring system is composed of a two-dimensional read-out apparatus and a laptop personal computer. To measure a two-dimensional RPL image, the RPL readout apparatus is composed of an x–y stage (A4 size: 195 × 327 cm^2) and a UV light emitting diode (LED). The process of obtaining a two-dimensional RPL image from the sheet-type glass dosimeter is shown in Fig. 7. The image is monitored by scanning the focused UV light on the irradiated sheet-type glass dosimeter. The view of the display during measurement is shown in Fig. 8. Figure 9 shows the two-dimensional dose distribution observed from the sheet-type glass dosimeter, which was uniformly irradiated with gamma rays. The irradiation was carried out on the circular area.

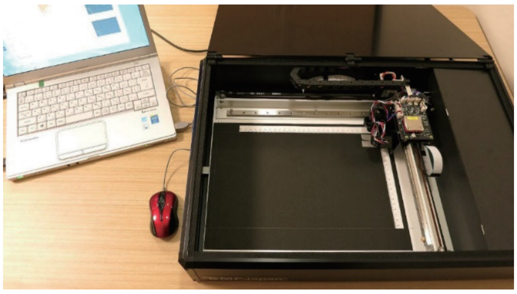


Fig. 6. (Color online) Photograph of the system for monitoring two-dimensional RPL image. The system is composed an x–y stage and a laptop personal computer.

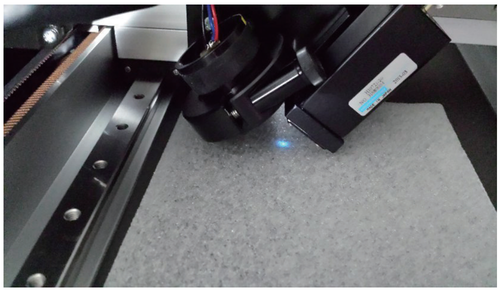


Fig. 7. (Color online) Process of obtaining a two-dimensional RPL image from the sheet-type glass dosimeter. The image is monitored by scanning the focused UV light on the irradiated sheet-type glass dosimeter.

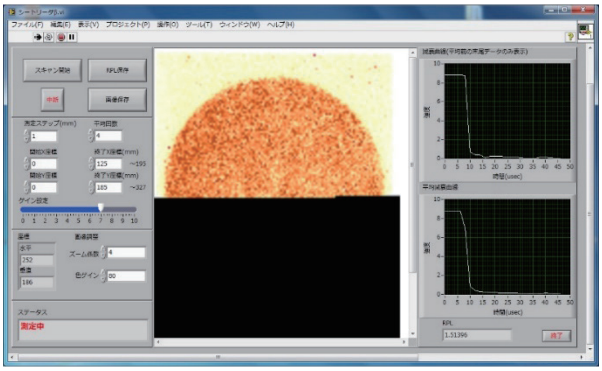
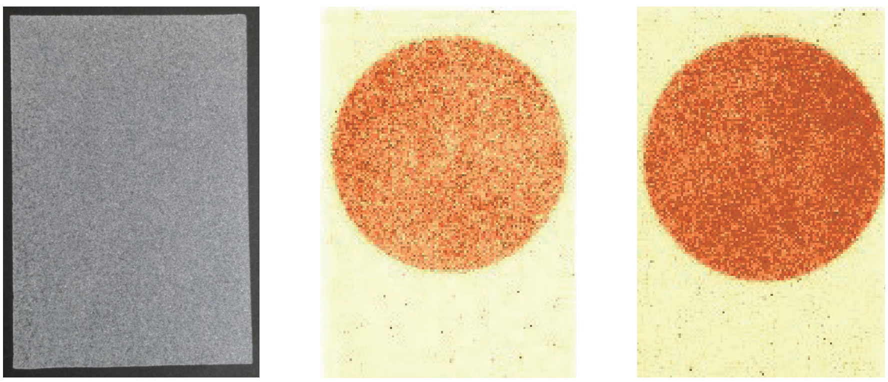


Fig. 8. (Color online) View of the display in a laptop personal computer when the sheet-type glass dosimeter was irradiated with about 1.5 Gy gamma rays from a Cs radioisotope.



(a) (b) (c)

Fig. 9. (Color online) RPL images obtained using the sheet-type glass dosimeter: (a) non-irradiated, (b) 100 mGy with gamma rays, and (c) 1 Gy with gamma rays. The irradiation was carried out on the circular area.

The dependence of the RPL intensity on the absorbed dose in the range from 0.01 to 1 Gy is shown in Fig. 10. The RPL intensity as a function of the absorbed dose of gamma rays in the range from 1 to 1000 Gy is also shown in Fig. 11. It can be seen from the figures that the RPL intensity is linearly increased with increasing absorbed dose, indicating that the sheet-type glass dosimeter can be used as a two-dimensional radiation dosimeter for the visualization of radiation dose distribution. This result strongly suggests that the sheet-type glass dosimeter can be used for the evaluation and visualization of radiation dose distribution. The research on the radiation monitoring using the bead- and sheet-type glass dosimeters in the contaminated area of Fukushima NPP-1 is in progress.

Figure 12 shows a simulation of radiation dose distribution in the case of using radiation protective gloves, which are made with a sheet-type glass dosimeter. The sheet-type glass dosimeter can be applied to the visualization and evaluation (extremity dosimetry) of the dose distribution of ionizing radiation, which was exposed to the hands of radiation workers at a

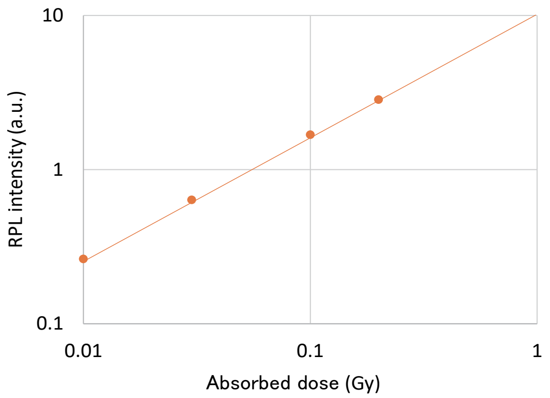


Fig. 10. (Color online) RPL intensity of the sheet-type glass dosimeter as a function of gamma rays irradiation dose in the range from 0.01 to 1 Gy.

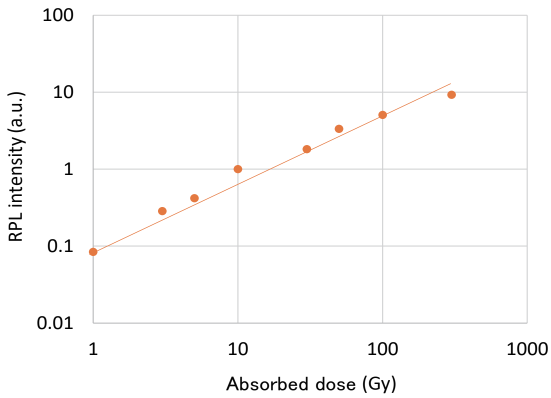


Fig. 11. (Color online) RPL intensity of the sheet-type glass dosimeter as a function of gamma rays irradiation dose in the range from 1 to 1000 Gy.

Fig. 12. (Color online) Simulation of radiation dose distribution in the case of using radiation protective gloves made with a sheet-type glass dosimeter.

contaminated area, for example, in the Fukushima nuclear plant. The research on the preparation and evaluation of the gloves sandwiched in the bead-type glass dosimeter between acrylic binders on an organic film is currently in progress.[10]

4. Conclusions

We have tried to develop novel bead- and sheet-type glass dosimeters that are usable under high-dose and high-temperature conditions such as nuclear emergencies, especially for radiation dose monitoring near or inside the Fukushima NPP-1, in which a large amount and many kinds of radioactive materials were released by a catastrophic earthquake on March 11, 2011. Two types of glass dosimeters, namely bead- and sheet-type, were developed to adapt to the various environments in radioactively contaminated areas. It was found that the sheet-type glass dosimeter with a two-dimensional RPL reader is useful for the visualization of radiation dose distribution.

Acknowledgments

The authors wish to express their gratitude to Prof. Toshio Kurobori of Kanazawa University for his fruitful discussion on RPL spectrum measurement and measurement of the RPL spectra using PTM-R3778 and PTM-R928. This study was partly supported by the System Development Program for Advanced Measurements and Analysis, Japan Science and Technology Agency (JST), Grant-in-Aid for Challenging Exploratory Research (15K13399) and the Cooperative Research Project of the Research Institute of Electronics, Shizuoka University.

References

1 Y. Miyamoto, K. Kinoshita, S. Koyama, Y. Takei, H. Nanto, T. Yamamoto, M. Sakakura, Y. Shimotsuma, K. Miura, and K. Hirao: Nucl. Instrum. Methods Phys. Res., Sect. A **619** (2010) 71.
2 Y. Miyamoto, Y. Takei, H. Nanto, T. Kurobori, A. Koyama, T. Yanagida, A. Yoshikawa, Y. Shimotsuma, M. Sakakura, K. Miura, K. Hirao, Y. Nagashima, and T. Yamamoto: Radiat. Meas. **48** (2011) 1480.
3 H. Nanto, Y. Miyamoto, Y. Takei, K. Hirasawa, and K. Endo: IEEJ Trans. Sens. Micromach. **133** (2013) 307 (in Japanese).
4 Y. Miyamoto, H. Nanto, T. Kurobori, and T. Yamamoto: Radiat. Meas. **71** (2014) 529.
5 T. Kurobori, Z. Wang, Y. Miyamoto, H. Nanto, and T. Yamamoto: Opti. Mater. **32** (2010) 1231.
6 F. Sato, N. Zushi, T. Maekawa, Y. Kato, I. Murata. K. Shimizu, T. Yamamoto, and T. Iida: Radiat. Meas. **68** (2014) 23.
7 M. S. Lee, Y. J. Rao, Y. H. Huang, J. H. Lee, S. K. Hung, T. R. Chen, and S. M. Hsu: Radiat. Meas. **46** (2011) 1009.
8 H. Nanto, Y. Yanagida, M. Sugiyama, Y. Ihara, T. Iida, K. Shimizu, T. Ikeguchi, K. Hirasawa, Y. Takei, and T. Yamamoto: Abstract of 18th Int. Conf. Solid State Dosimetry, Munuch (2016) IND-P-05.
9 Y. Miyamoto, M. Sugiyama, F. Sato, T. Iida, K. Shimizu, H. Nanto, Y. Ihara, T. Inada, N. Sato, and T. Yamamoto: Proc. 11th Int. Workshop on Ionizing Radiation Monitoring, Oarai (2015) p. 60.
10 F. Sato, N. Zushi, T. Maekawa, T. Sakiyama, N. Zushi, K. Shimizu, Y. Kato, I. Murata, T. Yamamoto, and T. Iida: Radiat. Meas. **85** (2016) 18.