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Original Article

Measurement of low energy beta radiation from Ni-63 by using peeled-off Gafchromic EBT3 film

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ABSTRACT

Ni-63 is pure beta source which emits low energy beta particles. The Ni-63 sources were fabricated to develop the beta-voltaic battery which converts decay energy into electrical energy for power generation. Activity distribution of the source was important factor of power producibility of the beta-voltaic battery. Liquid scintillation counter widely used for measurement of low energy beta emitters was not suitable to measure activity distribution. In this study, we used the peeled-off Gafchromic™ EBT3 film to measure the activity distribution of the Ni-63 source. Absorbed dose was increased proportionally to the source activity and exposure duration. The low energy beta particles could transport the energy into the active layer without the polyester protective layer. Also, Activity distribution was measured by using the peeled-off EBT3 film. Two-dimensional dosimetric distribution was suitable to measure the activity distribution. To use the peeled-off EBT3 film is user-friendly and cost-effective method for quality assurance of the Ni-63 sources for the beta-voltaic battery.

1. Introduction

Beta-voltaic batteries are power generator which converts radioisotope energy into electric energy [1–5]. A fundamental unit of beta-voltaic is composed of beta source and semiconductor, in which the beta particles from the source are collected in P–N junction of the semiconductor and a current is produced in the same way as the photo-voltaic battery. Since semiconductors are vulnerable to high energy radiation above 225 keV, low energy beta emitters are generally used for beta-voltaic batteries [5–7].

Nickel-63 (Ni-63) emits low energy beta particles ($E_{max} = 66.7$ keV, $E_{avg} = 18$ keV) with a long half-life (100.1 years). Due to the low energy particles and long half-life, Ni-63 is widely used for developing the beta-voltaic battery [3–5]. Since low energy beta particles have very short range in matter [8–10], the source was made as very thin plate to minimize self-absorption problem. In a previous study, we developed a small scale electroplating device to make Ni-63 planar source for the beta-voltaic battery [11–14]. The power producibility of the beta-voltaic battery is strongly related to the number of the beta particles emitting from the Ni-63 source. It is important to assure the Ni-63 source by measuring the beta particles. Liquid scintillation counter (LSC) [9], plastic scintillator [9], Si(Li) detector [10] or a charged particle detector with PIN diode sensor [15–17] are usually used to measure low energy beta emitters. Activity was measured with both LSC and the beta detector with PIN diode in previous study [15]. However, both method were not suitable for measuring activity distributions of the source.

Gafchromic™ EBT3 film is a film dosimeter with high spatial resolution which is commonly used for medical radiation treatment [18–21]. Recently, peeled-off EBT3 film was used to measure dose for alpha particle which had a very short ranges [21–23]. EBT3 film is composed of two 125 μm-thick protective polyester layers and one 28 μm-thick active layer inside. Peeled-off EBT3 film was made by removing one protective layer and the active layer was exposed to interact with alpha particle (see Fig. 1). The use of peeled-off EBT3 film demonstrates the feasibility to measure radiation particle of lower penetration. The average range of the 67 keV of beta particle is almost 0.007 $\text{g}\,\text{cm}^{-2}$ [8] and its average range is converted to 100 μm in the protective layer of EBT3 film. Therefore, most beta particles from Ni-63 are blocked in the protective layer.

In this study, we measure the radiation dose from the Ni-63 source with peeled-off EBT3 film to estimate the activity and the activity distribution of the Ni-63 sources. It is important to assure homogeneity of the Ni-63 source for quality control of the beta-voltaic battery. EBT3 film is known to have energy dependency at

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low energy region [24,25], but electron beam facilities in the MeV range are widely used for commercial or medical field but not in low energy range. The work of Jette Borg and the analysis of the data suggest to use Monte Carlo calculation for low energy beta dosimetry based on the agreement with the experimental data [10]. Therefore, radiation dose by the Ni-63 source is determined by MCNP6 calculation in this study.

2. Material and methods

2.1. Ni-63 sources for beta-voltaic battery

The Ni-63 sources were fabricated with a small batch electroplating device [14]. The fabrication condition was set to coat Ni-63 isotope as 2.5 μm thickness on to a natural nickel plate. The size of the source was 9 × 9 mm^2. Before electroplating, Ni-63 powder was dissolved in 0.4 M boric acid (H_3BO_3), 2 g/L saccharin, and a pH level of 4. The solution was also diluted with to make low activities. Three sources (No.1–3) were fabricated with the undiluted solution while other sources (No.4–7) were fabricated with the diluted solutions. These were unsealed sources without coating material on the surface and the surface should be intact for preventing contamination. Surface emission rate was measured with the beta detector and source activity was converted from the emission rates [15]. Details are described in 2.2.

2.2. Beta detector with PIN diode sensor

In order to detect the low energy beta particles, the beta detector system which consisted of a PIN diode sensor (S1223, Hamamatsu), a charge sensitive amplifier (CSA), a shaper and a pulse discriminator was developed in our previous study [15]. The CSA was developed with junction gate effect transistor input operational amplifier (JFET input OP-AMP, OPA-656, Texas instrument) and the CR-RC^2 shaper was used for the signal processing. The noise threshold value was 5 keV and it was enough to detect the beta particles from Ni-63. The dead time of the detector was determined as 7.46 μS by applying two-source method. Measured count rates was converted to true count rates by applying the dead time.

From the previous study, the true count rate was compared with the source activity measured by LSC (PerkinElmer, Tri-Carb 4910 TR). The correlation between the activity and the emission rate is shown in Eq. (1) ($R^2 > 0.99$). In this study, the electroplated Ni-63 sources of identical shape were measured with the beta detector (Table 1). Each measurement was conduct for 10 min.

$$\text{Surface emission rate (cpm)} = \text{activity (mCi)} \times 4160.9 + 253.62 \quad (1)$$

2.3. Measurement of Ni-63 with the peeled-off EBT3 film

Gafchromic™ EBT3 film consists of one active layer of 28 μm thickness and two protective layer (PL) of 125 μm that cover the active layer outside. The active layer contains diacetylene monomers that change from yellow to green by polymerization after irradiation [19]. Response of EBT3 film is expressed in the change of optical density (OD) which is due to polymerization of the monomers. The measured OD refers to the particular wavelength of the absorption spectrum.

One protective layer was peeled off using a cutter knife and the active layer could be exposed to the Ni-63 source without interfering material. Dose response of EBT3 film was known to be unaffected by the peeling-off process in previous study [23]. Fig. 2 shows the device to expose the film to the Ni-63 source. The device consisted of a source insertion part and a board to control exposure time. Each source was laid onto the insertion part, and peeled-off EBT3 film was exposed to the source for desired time. Because the Ni-63 source was unsealed, the source to target distance (STD) was 2 mm for preventing contact at the source surface.

The EBT films were scanned at 400 dpi (for dose measurement) or 4800 dpi (for activity distribution measurement) resolution using a color scanner (Expression 10000XL, Epson, Japan) before and after 24 h of radiation exposure [20]. The scanner was operated as positive film mode without colour correction function. The RGB scan value (SV) was extracted from each pixel in the scanned image. The red channel was read to measure optical density (OD) in this study, where OD was calculated by Eq. (2). To read red channel is commonly adopted at low dose region below 10 Gy for accuracy [26]. The uncertainty in dose estimation was within 10% by error propagation in the dose range [27].

$$\textit{Optical Density} = \log_{10}\left(\frac{SV_0}{SV}\right) \quad (2)$$

2.4. Dose calculation by MCNP6 simulation

MCNP6® code is a Monte Carlo radiation transport code developed by Los Alamos National Laboratory, USA. Beta particles from the Ni-63 were traced by MCNP6 code to calculate the energy deposition in EBT3 film. A mesh tally card was applied to the target volume to calculate distribution of energy deposition. Table 2 shows atomic composition and density for simulation. The uncertainty of the calculation is within 5%.

2.5. Measurement of activity distribution of Ni-63 source

A collimator with 11 × 11 circular holes of diameter ($\phi = 0.3$ mm) was fabricated in order to measure activity distribution of the sources (Fig. 4). The collimator was placed closely to the Ni-63 source and each hole admitted to pass the beta particles from the right below part. The peeled-off EBT3 film was laid on the collimator. STD (source to target distance) was set as 1 mm. The exposed films were scanned with 4800 dpi 24 h after exposure. The average OD of 10 × 10 pixels at the center of each hole was read to compare relativeness.

3. Results

3.1. Measurement of emission rates of the Ni-63 sources

The emission rate from the surface of the Ni-63 sources was measured by using the beta detector. The source activity was converted from true counts rate by applying Eq. (1). Activity of the Ni-

Table 1
Degree of dilution of the Ni-63 sources.

Source	1	2	3	4	5	6	7
Dilution	None	none	none	1/2 of source 3	1/4 of source 4	1/4 of source 5	1/4 of source 6

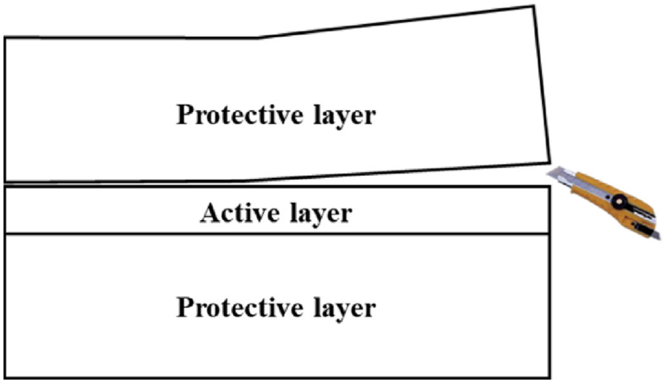

Fig. 1. Peeled-off EBT3 film by removing protective layer [22].

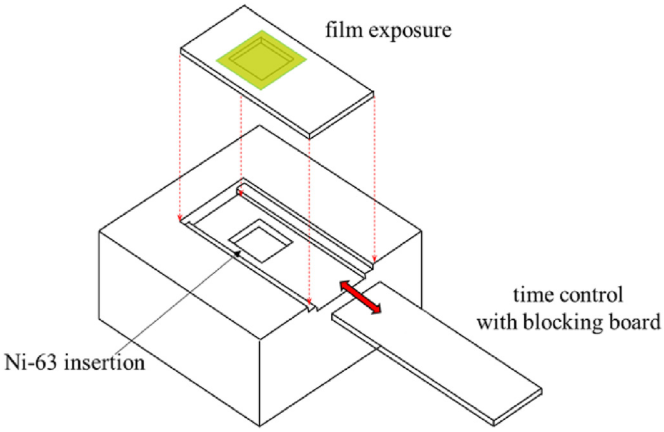

Fig. 2. The experimental device for EBT3 film to be exposed to the Ni-63 source for dose measurement.

63 source decreased as dilution was processed (see Table 3).

3.2. Measurement of radiation dose from the Ni-63 sources

The absorbed dose of the active layer in the configuration of Fig. 3 was calculated by MCNP6. Fig. 5 shows the distribution which the higher dose was delivered at central area and the lower dose was delivered at the edge of the film. Due to the 2 mm of STD, the central area of the film had higher possibility to interact with the particles from the source and it occurred the centralized dose distribution. The relatively uniform dose was delivered on the 1 × 1 mm^2 in the central area. For this reason, the target area of the film was set as 0.5 × 0.5 mm^2 in the central area. Two sources (No.4:12.744 mCi and No.5:3.317 mCi) were used to expose the peeled-off EBT3 film. Dose rate were 0.114 Gy/s with source No.4 and 0.0296 Gy/s with source No.5, respectively. Doses in the range of 0.5–10 Gy were exposed to the peeled-off EBT3 film by using each source. 24 h after exposing the film, OD measurement was conducted in the target area of interest. As shown in Fig. 6, the OD values of the film were approximated ($R^2 > 0.99$) to absorbed doses according to both equations (Eqs (3.1) and (3.2).).

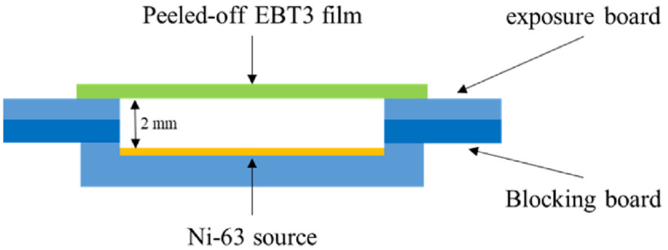

Fig. 3. Schematic diagram for exposure of peeled-off EBT3 film to low energy beta particles from Ni-63.

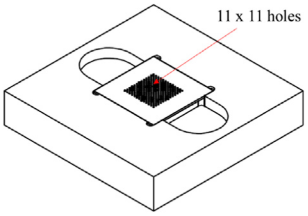

Fig. 4. The experimental device for measurement of activity distribution of the Ni-63 source.

$$Dose = 14.572 \times OD + 123.87 \times OD^2 \tag{3.1}$$

$$Dose = 11.763 \times OD + 139.59 \times OD^2 \tag{3.2}$$

Table 2
Atomic composition data for MCNP6 simulation.

material	Composition (Atom %)											Density (g/cm^3)
	H	He	Li	C	N	O	Na	Cl	Br	Ni-63	Cu	
Ni-63 coating										100		8.0
Active layer	58.3		0.8	29.6	0.1	10.8	0.1	0.2	0.1			1.2
Protective layer												1.39
device											100	8.96

Table 3
Activity estimation by measuring the surface emission rates with the beta detector.

Source	1	2	3	4	5	6	7
Measured Count rates (cpm)	53,800	57,600	57,000	38,100	12,700	4000	1200
True count rates (cpm)	89,800	100,900	99,300	53,300	14,100	4100	1200
Activity (mCi)	20.72	24.20	23.80	12.74	3.32	0.93	0.24

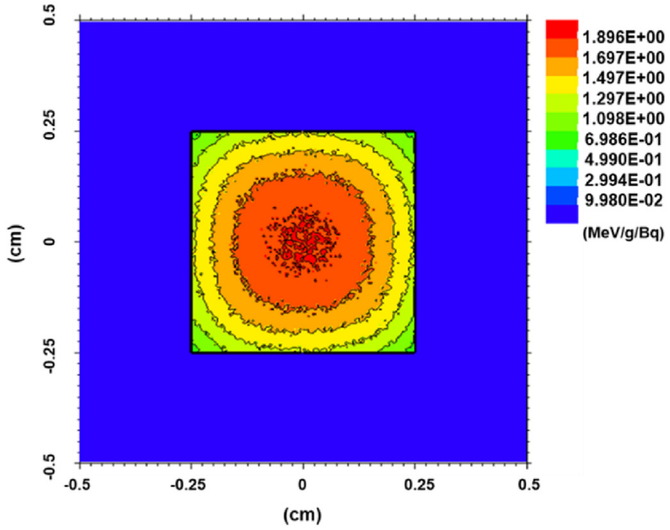

Fig. 5. The calculated dose distribution in the active layer in the configuration of Fig. 3.

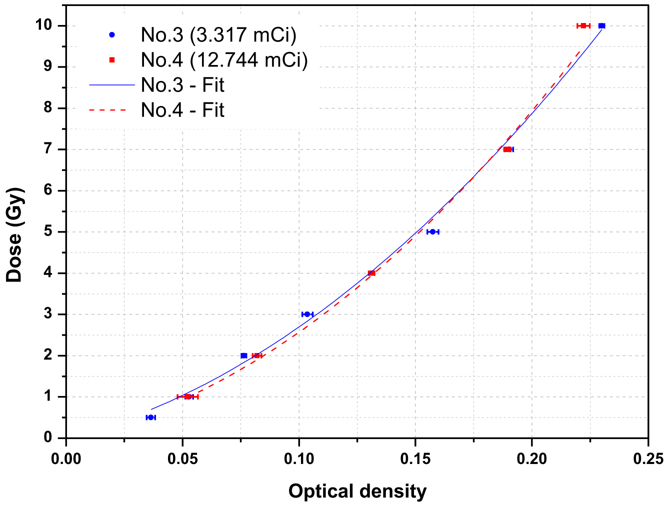

Fig. 6. Dose calibration curves fitted to the OD measurement with peeled-off EBT3 film to the No.3 (3.77 mCi) and No.4 (12.744 mCi) sources. The error bars indicate standard deviations of the mean OD value.

3.3. *Activity distributions of electroplated Ni-63 source*

The activity distribution of the Ni-63 source was measured by using the collimator in Fig. 4. Fig. 7 shows the scanned image of the film after exposure and intensity graph of the red channel value read from the image. The OD value was measured at the center of each hole. The radiation dose was converted from the OD value. The 11 × 11 measurement values were averaged and each relative value to the average is shown in Fig. 8. The relativity ranged from 70% to 118% of the average in source No.1, from 73% to 121% of the average in No.7, and from 73% to 115% of the average in No.8.

4. Discussion

4.1. *Applicability of peeled-off EBT3 film for low energy beta particle*

In this study, radiation dose of low energy beta radiation was measured by using peeled-off EBT3 film. First, we calculated the absorbed dose rates from the Ni-63 sources by MCNP6 simulation and two different sources (No. 4 & No. 5) were used to deliver radiation energy. In Fig. 6, we found that the net OD values of peeled-off EBT3 film were in good agreement with the absorbed doses. Therefore, we demonstrate that to use peeled-off EBT3 film is appropriate to measure dose from Ni-63. Since the absorbed dose is proportional to the source activity, we can estimate activity of the Ni-63 source without conducting LSC. Also, this technique can be applicable to measure other low beta emitters, such as C-14.

4.2. *Activity distribution of the Ni-63 sources*

The primary object to use the peeled-off EBT3 film is for measurement of activity distribution of the Ni-63 sources. Verification of homogeneity of the isotope is important for quality control of the beta-voltaic battery. The use of peeled-off EBT3 film provides two-dimensional distribution of the activity. The results shows that the areal activity varied from 70% to 121% of the average. This inhomogeneity might be caused during the fabrication of the Ni-63 sources. Electroplating in the small scale electroplating bath caused lack of uniformity [16].

5. Conclusion

To measure activity distribution of the electroplated Ni-63

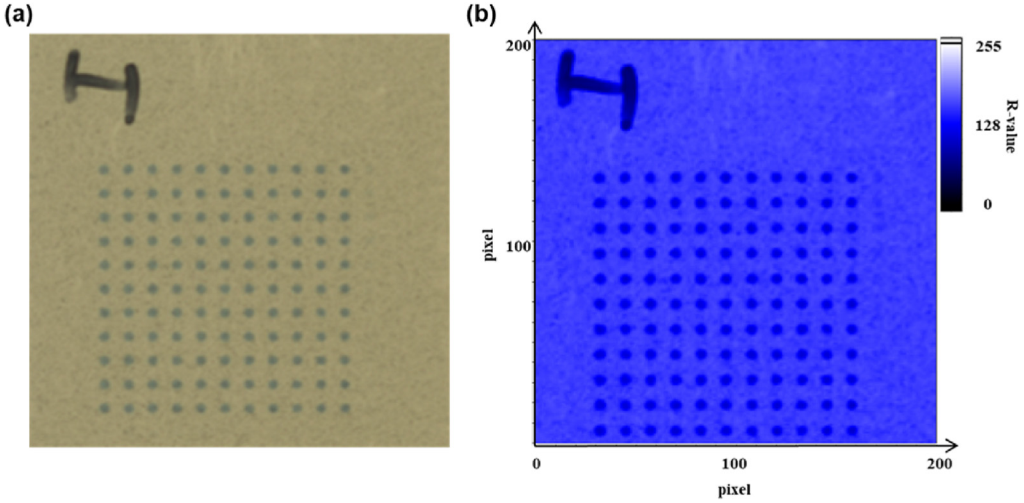

Fig. 7. (a) A scanned image of the peeled-off EBT3 film by using the collimator (11 × 11 holes) with source No.3, and (b) a image of red channel intensity of Fig. 7(a). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

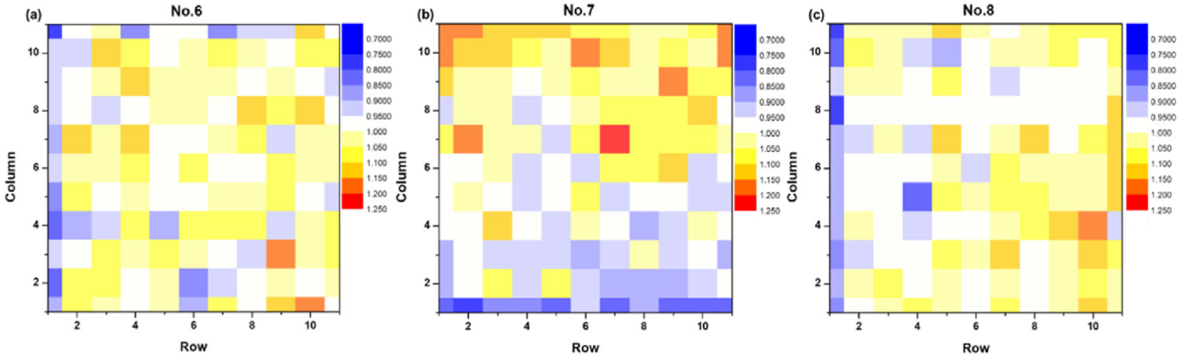

Fig. 8. The relative value to the average for measuring the activity distribution of the Ni-63 sources for the beta-voltaic battery (a) source No.1 (20.717 mCi), (b) source No.2 (24.191 mCi), (c) source No.3 (23.804 mCi).

sources was required for quality assurance of the beta-voltaic battery. A conventional method of LSC was not possible for the measurement. The peeled-off EBT3 film was used to measure radiation dose of the Ni-63 source. The OD was well corresponded to the absorbed dose in the peeled-off EBT3 film and source activity. Activity distribution of the source was measured with the peeled-off EBT3 film. Two-dimension distribution of the film was appropriate to measure activity distribution. The use of the peeled-off EBT3 film could provide simple and cost-effective method to assure quality of the Ni-63 sources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

[1] F.K. Manasse, J.J. Pinajian, A.N. Tse, Schottky barrier betavoltaic battery, IEEE Trans. Nucl. Sci. 23 (1976) 860–870.
[2] G.H. Miley, Direct Conversion of Nuclear Radiation Energy, U.I.U.C., 1970. No. TID-25672.
[3] H. Guo, A. Lal, Nanopower betavoltaic microbatteries, in: In *TRANSDUCERS'03. 12th International Conference on Solid-State Sensors, Actuators and Microsystems. Digest of Technical Papers (Cat. No. 03TH8664)* 1, IEEE Press, 2003.
[4] A. Krasnov, S. Legotin, K. Kuzmina, N. Ershova, A nuclear battery based on silicon p-i-n structures with electroplating 63Ni layer, Nucl. Eng. Technol. 51 (2019) 1978–1982.
[5] M.V.S. Chandrashekhar, I.T. Christopher, H. Li, M.G. Spencer, A. Lal, Demonstration of a 4H SiC betavoltaic cell, Appl. Phys. Lett. 88 (2006), https://doi.org/10.1063/1.2166699.
[6] G.C. Messenger, A summary review of displacement damage from high energy radiation in silicon semiconductors and semiconductor devices, IEEE Trans. Nucl. Sci. 39 (1992) 468–473.
[7] G.P. Summers, E.A. Burke, P. Shapiro, S.R. Messenger, R.J. Walters, Damage correlations in semiconductors exposed to gamma, electron and proton radiations, IEEE Trans. Nucl. Sci. 40 (1993) 1372–1379.
[8] J.E. Turner, Atoms, Radiation, and Radiation Protection, third ed., Wiley-VCH, Weinheim, 2012.
[9] H. Kang, S. Min, B. Seo, C. Roh, S. Hong, J.H. Cheong, Low energy beta emitter measurement: a review, Chemosensors (2020), https://doi.org/10.3390/chemosensors8040106.
[10] J. Borg, Dosimetry of low-energy beta radiation, Risoe Natl. Lab. (1996). No. RISO-R—907.
[11] J.J. Kim, Y.R. Uhm, K.J. Son, Fabrication of 63Ni layer for betavoltaic battery, in: Proceedings of the 15th IEEE International Conference on Nanotechnology, Rome, Italy, July 27-30, 2015.
[12] J.J. Kim, S.M. Choi, K.J. Son, J.T. Hong, Development of small-scale electroplating system for Ni-63 electroplating onto Ni foil, in: Transactions of the Korean Nuclear Society Autumn Meeting, Gyeongju, Korea, October 27-28, 2016.
[13] Y.R. Uhm, B.G. Choi, J.B. Kim, D.H. Jeong, K.J. Son, Study of a betavoltaic battery using electroplated nickel-63 on nickel foil as a power source, Nucl. Eng. Technol. 48 (2016) 773–777.
[14] J.J. Kim, S.M. Choi, G.J. Son, J.T. Hong, Manufacturing of Ni-63 sealed source for betavoltaic battery using the small-scale electroplating device, J. Radiat. Ind. 11 (2017) 173–179.
[15] W. Ji, J.B. Kim, Development of rapid beta detector using PIN diode to be used in quality control of Ni-63 beta-voltaic battery, J. Radioanal. Nucl. Chem. 330 (2021) 245–252.
[16] W.R. Wampler, B.L. Doyle, Low-energy beta spectroscopy using pin diodes to monitor tritium surface contamination, Nucl. Instrum. Methods Phys. Res.: Accel. Spectrom. Detect. Assoc. Equip. 349 (1994) 473–480.
[17] R.S. Willms, D. Dogruel, R. Myers, R. Farrell, A new solid state tritium surface monitor, Fusion Sci. Technol. 48 (2005) 409–412.
[18] M.J. Butson, K.N. Peter, T. Cheung, P. Metcalfe, Radiochromic film for medical radiation dosimetry, Mat. Sci. Eng. R. 41 (2003) 61–120.
[19] D. Slobodan, Radiochromic film dosimetry: past, present, and future, Phys. Med. 27 (2010) 122–134.
[20] D. Slobodan, N. Tomic, L. David, Reference radiochromic film dosimetry: review of technical aspects, Phys. Med. 32 (2016) 541–556.
[21] B. Mukherjee, Y.H. Gholami, U. Bhonsle, R. Hentschel, J. Khachan, A unique alpha dosimetry technique using Gafchromic EBT3® film and feasibility study for an activity calibrator for alpha-emitting radiopharmaceuticals, Br. J. Radiol. 88 (2015), https://doi.org/10.1259/brj.20150035.
[22] K.H. Lee, J.Y. Shin, E.H. Kim, Measurement of activity distribution in an Am-241 disc source using peeled-off Gafchromic EBT3 films, Appl. Radiat. Isot. 135 (2018) 192–200.
[23] C.Y.P. Ng, S.L. Chun, K.N. Yu, Quality assurance of alpha-particle dosimetry using peeled-off Gafchromic EBT3® film, Radiat. Phys. Chem. 125 (2016) 176–179.
[24] G. Massillon-JI, C.T. Sou-Tung, I. Doming-Muñoz, M.F. Chan, Energy dependence of the new Gafchromic EBT3 film: dose response curves for 50 kV, 6, and 15 MV X-ray Beams, I, J. Med. Phys. Clin. Eng. Radiat. Oncol. 1 (2012) 60–65.
[25] A. Rink, I.A. Vitkin, D.A. Jaffray, Energy dependence (75 kVp to 18 MV) of radiochromic films assessed using a real time optical dosimeter, Med. Phys. 34 (2007) 458–463.
[26] K. Duruer, D. Etiz, H. Yücel, Investigation of EBT3 radiochromic film response in a high-dose range of 6MV photon and 6 MeV electron beams using a three-color flatbed scanner, East. Eur. J. Phys. 3 (2020) 11–18.
[27] B.C. Ferreira, M.C. Lopes, M. Capela, Evaluation of an Epson flatbed scanner to read Gafchromic EBT films for radiation dosimetry, Phys. Med. Biol. 54 (2009) 1073.