A DOSIMETRY TECHNIQUE OF HIGH-ENERGY X-RAY IN MGy RANGE

Ju.V. Rogov, V.A. Shevchenko, I.N. Shlyakhov, B.I. Shramenko, A.Eh. Tenishev, A.V. Torgovkin, V.L. Uvarov, V.F. Zhiglo
National Science Center "Kharkov Institute of Physics and Technology", Kharkov, Ukraine
E-mail: uvarov@kipt.kharkov.ua

Possibility of a dosimetry method for high-energy bremsstrahlung radiation in the range up to MGy and higher by activation of a detector from indium via the ^{115}In (γ,γ′) ^{115m}In reaction with determination of absorbed dose on specific activity of the ^{115m}In isomer is investigated. To study such an approach, a method based on joint irradiation of a stack of thin targets is offered. Each target includes the foils from indium and molybdenum of natural isotopic composition, and also a standard dosimeter in the form of a plate from PMMA. Preliminary study of indium activation (with due regard to known data on the reaction cross section), detector thermal stability, yield of the reference reactions ^{92}Mo(γ,2n)^{90}Mo and ^{100}Mo(γ,n)^{99}Mo as well as absorbed dose in PMMA, was conducted by means of computer simulation. The measurement of specific activity of ^{115m}In, ^{90}Mo and ^{99}Mo was carried out jointly with the absorbed dose of in PMMA using the Harwell Red 4034 dosimeters. Sensitivity of the method against end-point energy of X-Ray in the range 8 to 71 MeV has been established.

PACS: 07.85.Nc; 41.50.+h; 82.80.Ej;87.53.Bn

INTRODUCTION

Nowadays, development of high-power X-ray sources intended for photonuclear isotope production [1], subcritical assembly control [2], etc. is carried out on the basis of high-current electron linacs. The output devices of such kind facilities are exposed to the photon flux with end-point energy up to 100 MeV at an absorbed dose rate of 10^5 Gy/s and higher. In the circumstances, the traditional methods of X-ray dosimetry appear to be of little use (see, e.g. Ref. [3]).

It is known, that photoactivation of the isomeric states in some nuclei is characterized by a low energy threshold. For example, the threshold of the ^{115}In(γ,γ′)^{115m}In reaction is 1078 keV [4]. The ^{115m}In isomer decays to the ground state with half-life $T_{1/2}$=4.48 h, emitting the gamma-quanta with energy 336.2 keV, which is convenient for detection. Owing to the low reaction threshold, the natural indium (the ^{115}In abundance makes 95.8%) can be activated by practically all the photons of the bremsstrahlung spectrum. This circumstance allows suggesting the presence of a relationship between the specific activity of ^{115m}In and the absorbed dose of X-ray.

In a number of studies, the ^{115}In(γ,γ′)^{115m}In reaction has been used for dosimetry in the γ-facilities with ^{60}Co sources [4, 5]. In that case, the activation of indium was realized by quasi-monochromatic photons with energy near the reaction threshold at a dose rate up to 10^2 Gy/s.

The present communication discusses the conditions of applicability of the method for high-dose dosimetry of the high-intensity X-Ray. So at the first stage, the sensitivity S of the proposed dosimetry technique as the dependence of the ratio of the ^{115m}In specific activity to absorbed dose in a standard dosimetry material (PMMA) contacting with the In-detector from electron energy E_0 (governing end-point energy of the bremsstrahlung spectrum) was studied. At irradiation with X-ray having end-point energy >>10 MeV, each target included also a foil from natural molybdenum both for verification of used calculation package, and for the control of activation mode against yield of the reference reactions ^{92}Mo(14.84%)(γ,2n)^{90}Mo and ^{100}Mo(9.63%)(γ,n)^{99}Mo on the stable molybdenum isotopes. It should be noted, that under those conditions ^{90}Mo can be produced only via the photonuclear channel, when in the ^{99}Mo yield the ^{98}Mo(24.13%)(n,γ)^{99}Mo reaction can contribute also. In its turn apart from γ-activation, ^{115m}In can be generated via ^{115}In(n,n′)^{115m}In reaction by photoneutrons, commonly accompanying the bremsstrahlung radiation (see e.g. [6]).

By means of computer simulation, a thermophysical analysis of the In-detectors with establishing in such a way their radiation lifetime was conducted at the second stage.

1. SIMULATION

For preliminary analysis of the photoactivation processes as well as absorption of radiation energy in the detectors, we used a simulation method based on a modified transport code PENELOPE-2008 [7]. The yield of photonuclear reactions was calculated through summation their microyields along all the trajectories of all the above-threshold photons in a target (a Step-By-Step technique [8]). Cross sections of the reference reactions on the ^{92}Mo and ^{100}Mo isotopes were taken from the database [9]. In the case of the ^{115}In(γ,γ′)^{115m}In reaction the situation turned out to be more complicated. Namely, the data on that cross section, reported in the different works, have shown considerable variations (Fig. 1).

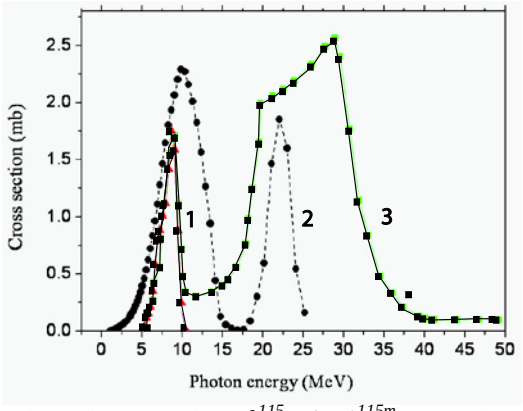


Fig.1. Cross section of ^{115}In(γ,γ′)^{115m}In reaction: 1 – [10]; 2 – [11]; 3 – [12]

Therefore, we calculated the ^{115m}In yield for different variants. At the same time, the absorbed dose rate in PMMA was calculated for each irradiation mode. Further the obtained data on the yield of ^{115m}In, and the reference reactions as well as the absorbed dose in PMMA were compared with the experimental results.

2. EXPERIMENT

2.1. For experimental study, the three electron linacs of NSC KIPT were used:

- LU-10 (E_0=8...12 MeV);
- EPOS (E_0=26 MeV);
- LU-40m (E_0=35...95 MeV).

Joint irradiation of the In-detectors with the PMMA dosimeters was conducted providing a condition of electronic equilibrium. In Fig. 2, the layout of the experiment carried out at the LU-10 Linac for determination a sensitivity of the proposed dosimetry technique at a low border of the studied X-ray energy range is shown. A bremsstrahlung converter C (a 2.4 mm thick tantalum plate encased in a copper housing and cooled with water) and a filter F (4 aluminium plates each 2 mm thick) were positioned at the exit window of the accelerator.

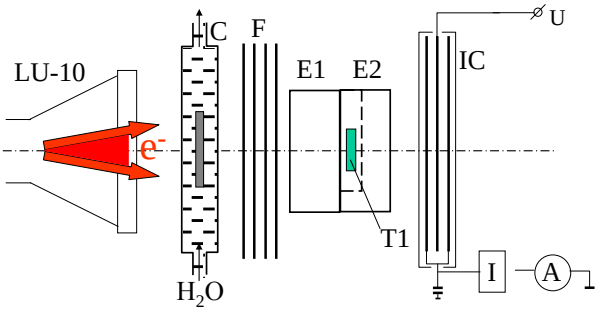


Fig. 2. Layout of output device at the LU-10 Linac

At a distance of 15 cm downstream the filter, a shield from polystyrene (the two plates E1 и E2 each of 50 mm thick) was disposed. A target T1 was placed in a well of the plate E2. The target comprised 2 dosimeters and 2 indium foils measuring 2×1×0.1 cm. Monitoring of the radiation flux on the target device was carried out using a free-air ionization chamber IC.

The accelerator operated in a mode with beam scanning. Similar conditions of the experiment were realized at the EPOS Linac providing electron energy 26 MeV.

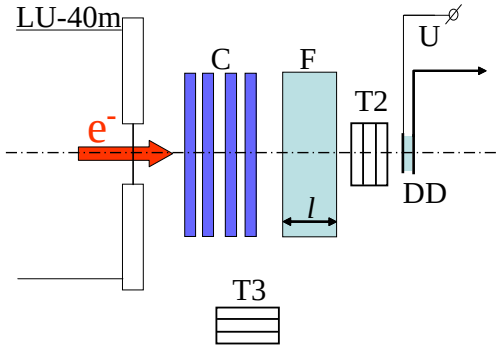


Fig. 3. Scheme of output device at LU-40m

In its turn, for target activating at the LU40m machine, a direct beam was used (Fig. 3). This time a converter corresponded the 4 tantalum plates each of 1 mm thick with 1 mm gaps for cooling. An aluminum cylinder F placed behind the converter was used as a filter. Its height was 5, 7 and 9 cm for electron energy of 35, 53 and 71 MeV respectively. The targets T2 and T3 included the similar sets of In-detectors, PMMA-dosimeters, and also molybdenum foils.

The targets were positioned at a distance of 20 cm from the converter at the beam axes and at right angle to it respectively. Just behind T2, a semiconductor detector on the basis of CVD was placed for monitoring the bremsstrahlung dose rate [13].

2.2. For the absorbed dose measurement, the Harwell Red 4034 (HR) detectors were used. They correspond the plates measuring 30×11×3 mm from dyed PMMA. The absorbed dose was determined on alteration of the detector optical density measured with a Shimadzu UV mini 1240 spectrophotometer using an established gauge dependence. The advantage of HRs is the possibility of determination the photon-induced absorbed dose at their exposure to the mixed γ,n-radiation [14].

2.3. A gamma-spectrometer was used for measuring the isotope composition and activity of the irradiated indium and molybdenum detectors. It consists of a Canberra HPGe detector, having relative detection efficiency of 20% and FWHM of 1.8 keV at 1332 keV. An In-Spector 2000 analyser with the Genie-2000 software was applied to read out the detector signal. Calibrating the spectrometer efficiency was performed with the use of a set of standard γ-sources in the photon energy range 59.5 keV (^{241}Am) to 1332 keV (^{60}Co). The γ-spectra of the irradiated Mo and In samples were measured by placing them at a distance of 250 mm from the detector surface. The assembly was surrounded with a 100 mm thick lead shield to reduce the background. Counting times were typically about 10 min. The ^{115m}In activity was determined against the line 336.25 кеV. The calculated self-absorption of the specific gammas in the samples was 4% for indium and less than 1% for molybdenum. The activity of all the isotopes at EOB was calculated with due correction for the decay, detection efficiency and self-absorption.

2.4. The measurements were conducted in the two stages. At the first, the targets were exposed for an established time to obtain the absorbed dose within the operating range of the HR-dosimeters (typically, up to 30 kGy). Thereafter, one from the activated In-detectors was transferred to the gamma-spectrometric station for measurement its specific activity against the ^{115m}In isomer and determination in such a way the sensitivity against the absorbed dose, measured with the HR-detectors. The other In-sample was remained in the target device for further exposure till a dose more than 100 kGy is achieved.

3. RESULTS AND DISCUSSION

3.1. In Fig. 4, the data on the dose rate and the yield of the reference reactions on the stable molybdenum isotopes in the target T2 are presented. It is seen, that the results of the simulation and experiment are in good agreement. It asserts both the sufficient accuracy of the measurements and the validity of the code used. The γ-spectra of the In-detectors irradiated at the various electron energy, are demonstrated in Fig. 5. There are no additional lines at E_0=71 MeV as compared with the spectrum at E_0=8 MeV. It is obvious also, that apart

from photonuclear channels, ^{116}In ($T_{1/2}$=55 min) is produced by the neutron capture. Some difference between relative intensity of the lines in the both spectra is connected with the different time of measurement after EOB. The most long-lived admixture is ^{114m}In ($T_{1/2}$=49 day) yielded from the ^{115}In(γ,n)^{114m}In reaction.

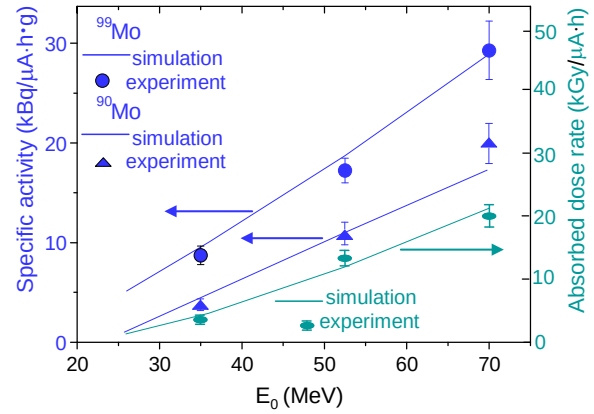


Fig. 4.Yield of the reference reactions and absorbed dose in PMMA vs electron energy

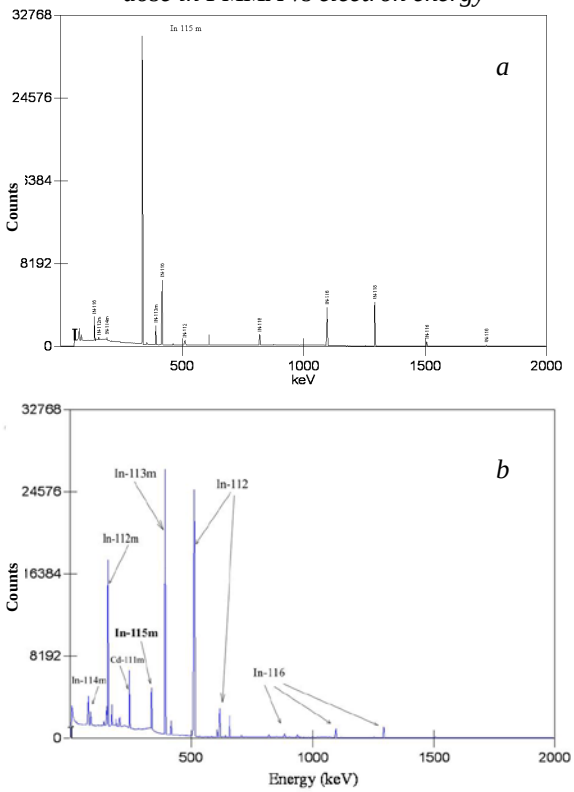


Fig. 5. Spectra of the activated In-detectors: a – E_0=8 MeV; b – E_0=71 MeV

3.2. Making use the experimental results on the specific activity of ^{115m}In and the absorbed dose in PMMA, the data on sensitivity S of the proposed dosimetry technique at an electron energy of 8 to 12 MeV (Fig. 6) and 26 to 71 MeV (Fig. 7) have been obtained. In the latter case, the various data on the cross section of the ^{115}In(γ,γ′)^{115m}In reaction were used.

3.3. The geometry of the output device with the two targets T2 and T3 given in Fig. 2 provided possibility of evaluating the contribution of the (n,n') channel to the ^{115m}In yield. Indeed, the target T2 is simultaneously exposed to the onward X-ray and quasi-isotropic photoneutron fluxes, whereas the target T3 to neutrons only. Considering equal distance between the converter and each the target, the neutron fluence acting on them was also nearly equal. The conducted measurements have shown that even at the highest value of the photoneutron flux corresponding to the electron energy of 71 MeV the contribution of the (n,n`) channel to the ^{115m}In activation does not exceed 4%. It can be explained by comparatively low photoneutron yield (by a factor of 10^{-3} relative to the bremsstrahlung photon flux) – [6].

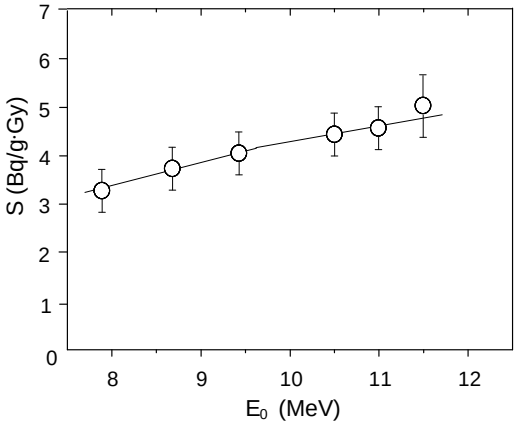


Fig. 6. Sensitivity of the In-detectors at E_0=8…12 MeV

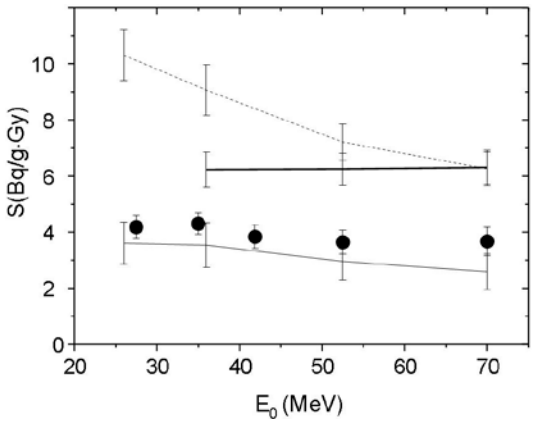


Fig. 7. Sensitivity of the In-detectors at E_0=26…71 MeV. (The data on cross section of the ^{115}I (γ, γ′)^{115m}In reaction were taken from the works: [10 - 12])

4. THERMOPHYSICAL ANALYSIS OF RADIATION RESOURCE OF In-DETECTOR

To establish an allowable maximum of the absorbed dose measured with the In-detector, as a criterion the condition of its heating by bremsstrahlung radiation up to a melting temperature T_m=156.4°C was chosen. The analysis was conducted for a detector in the form of rectangular plate measuring X=1 cm (width), Y=2 cm (height) and d=0.1; 0.05 cm (thickness). Providing the electron equilibrium condition, the distribution of the absorbed radiation power within the detector can be considered as homogeneous. It was suggested also, that cooling of the plate is the result of the convection and radiation processes at its surface. In Fig. 8, the temperature distribution along depth of a plate of 0.1 cm thick at a heating power of 1.5 W (~$1.5\cdot10^3$ Gy/s) obtained by a calculation technique is given. It is seen, that the temperature gradient is negligibly small in view of the high thermal conductivity of indium.

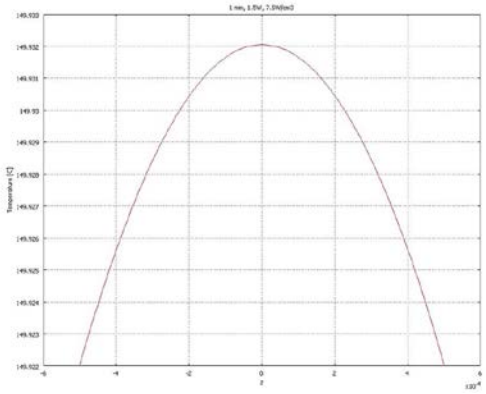
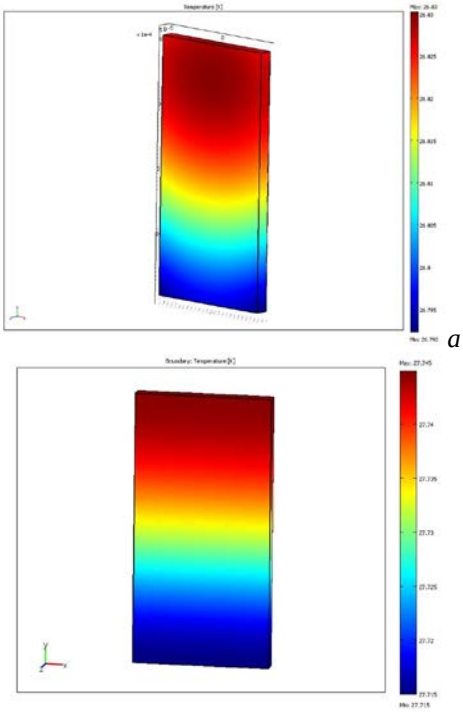


Fig. 8. Depth distribution of temperature along In-plate at a heating power of 1.5 W

The temperature field within the In-plate is demonstrated in Fig. 9. Vertically, the temperature increases slightly due to heating by the air convection. Presence of a round boundary in the temperature distribution at the homogeneous heat generation (see Fig. 9,a) becomes clear as the result of the heat extraction through the butt-end surfaces of the detector. Really, in case of their thermal isolation, the temperature distribution becomes even (see Fig. 9,b).



a

b

Fig. 9. Temperature distribution in the In-plate: a – at free butt-end surfaces; b – at thermal isolation of butt-end surfaces

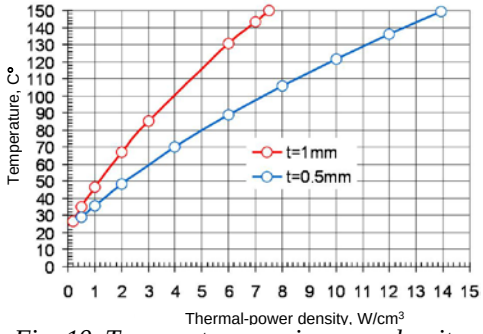


Fig. 10. Temperature maximum vs density of the heat generation

Dependences of the temperature maximum from density of the heat extraction in the plate at its thickness of 0.5 and 1.0 mm are given in Fig. 10. Just as it should be expected, in the latter case the allowable value of the absorbed power density (the dose rate) is by factor 2 less because the heat removal through the surface is practically equal in the both cases.

The heat conduction equation for a body with homogeneous heat generation within the volume has the appearance

$$\lambda \Delta T(\vec{r}) + q = 0\,, \qquad (1)$$

with boundary condition

$$\lambda \vec{n} grad T\big|_S = \alpha(T, \vec{\vec{r}})\big[T(\vec{r}) - T_0\big]\big|_S\,, \qquad (2)$$

where λ is a thermal-conductivity coefficient; Δ is the Laplace operator; α is a heat-transfer coefficient in the point $\vec{r}$ on the body surface; T_0 is air temperature;

$$\alpha(T) = \alpha_c(T) + \alpha_r(T)\,, \qquad (3)$$

where α_c, and α_r are heat-transfer coefficients for the convection and radiation respectively. It is seen from the data in Figs. 8 and 9, that the temperature alteration within the detector volume does not exceed 0.1% and so can be neglected. Then the maximum absorbed dose in the detector influential by its melting temperature T_m, makes

$$W_{\max} = \alpha(T_m)(T_m - T_0)S_d\,, \qquad (4)$$

where S_d is the area of the detector surface

Thus an allowable dose rate is given by expression

$$\dot{D}_{\max} = \frac{\bar{W}_{\max}}{\rho \cdot XYd}\,, \qquad (5)$$

where ρ – the density of the detector material (ρ_{In}=7.31 g/cm^3), or

$$\dot{D}_{\max} = \frac{2\alpha(T_m)}{\rho}(T_m - T_0)\left(X^{-1} + Y^{-1} + d^{-1}\right). \qquad (6)$$

Taking into account, that $d^{-1} >> X^{-1}$, Y^{-1}, the last expression can be reduced to the form

$$\dot{D}_{\max} = \frac{2\alpha(T_m)}{\rho \cdot d}(T_m - T_0)\,. \qquad (7)$$

At the given conditions of the detector cooling, α=22.8 W/m^2·K. In this case, at an irradiation time of about 1 h the resource of the In-detector makes $2 \cdot 10^6$ Gy (at the adiabatic mode) or ~10^7 Gy (at the heat removal by convection). Under these conditions, the linearity of the induced ^{115m}In activity with the time of the exposure is kept. That provides the conservation of sensitivity of the dosimetry system.

CONCLUSIONS

The undertaken studies have demonstrated that the ^{115}In(γ,γ′)^{115m}In reaction can be used for dosimetry of X-ray with end-point energy of up to 70 MeV and higher. The advantages of the proposed approach include: the ease of realization, the linearity with respect to the photon fluence, the independence from the dose rate, ambient conditions (pressure, temperature, humidity, neutron background), and also, the reusability of detectors. As the experiments have shown, the ratio of the ^{115m}In specific activity to absorbed dose in PMMA (the sensitivity of the method) increases from about 3 to about 5 Bq/g·Gy in the electron energy range 8 to 12 MeV. With the further

energy increase up to 71 MeV, the value of the ratio makes about 4 Bq/g·Gy and varies very slightly.

Under ordinary conditions of the In-detector application its one-time dose resource makes ~10^7 Gy. Providing a forced cooling, the allowable boundary of the dose measurement can be increased by 1 to 2 order of magnitude. A safe level of the In activity can be readily provided by appropriate choice of the detector weight.

As the results obtained with PMMA and molybdenum have demonstrated, the simulation technique based on the transport code PENELOPE-2008 makes it possible to calculate with a reasonable accuracy the absorbed energy (dose) of electron/photon radiation, and also the yield of the photonuclear reaction providing the correct description of its cross section. So the data on the $^{115}In(\gamma,\gamma')^{115m}In$ reaction cross section given in the work [11] provide the best agreement of calculations with the experimental results.

REFERENCES

1. S. Koscnielniak, N. Lockyer, L. Merminga. Electron Linac Concepts for the Production of Molybdenum-99 // *Proc. PAC 09*. 2009, Vancouver, Canada, v. 2, p. 1324-1326.
2. D.G. Naberezhnev, Y. Gohar, J. Bailey, H. Belch. Physics Analyses of an Accelerator-Driven Sub-Critical Assembly // *NIM*. 2006, 562A(2): p. 841-844.
3. ISO/ASTM 51608:2005 Practice for dosimetry in an X-ray (bremsstrahlung) facility for radiation processing.
4. J.-H. Chao, P.-Ch. Hsu, H.-M. Liu. Measurement of High-Dose Rates by Photon Activation of Indium Foils // *Appl. Rad. and Isot*. 2001, v. 55, p. 549-556.
5. A. Murataka, S. Endo, Y. Kojima, K. Shizuma. Dose Rate Estimation Around ^{60}Co γ-Ray Irradiation Source by Means of ^{115m}In Photoactivation // *J. Radiat. Res. Advance Publ*. 2010, p. 1-7.
6. T.V. Malykhina, A.A. Torgovkin, A.V. Torgovkin, et al. Study of Mixed X,n-Radiation Field under Photonuclear Isotope Production // *Problems of Atomic Science and Technology. Series "Nuclear Physic Investigations"*. 2008, № 5, p. 184-188.
7. F. Salvat, J.M. Fernández-Varea, and J. Sempau. "PENELOPE-2008, A Code System for Monte Carlo Simulation of Electron and Photon Transport" (OECD Nuclear Energy Agency, Issy-les-Moulineaux, France, 2008).
8. V.I. Nikiforov, V.L Uvarov. Development of the Technique Embedded into a Monte-Carlo Transport System for Calculation of Photonuclear Isotope Yield // *Nucleonika*. 2012, v. 57(1), p. 75-80.
9. IAEA – TECDOC – 1178. Vienna, IAEA, 2000.
10. V.S. Bokhinyuk, A.I. Guthy, A.M. Parlag, et al. Study of the Effective Excitation Cross Section of the ^{115m}In Isomeric State in the (γ,γ′) Reaction // *Ukr. J. Phys.* 2006, v. 51, p. 657-660.
11. Y.P. Gangrsky, V.M. Mazur. Scattering of γ Rays by the Nuclei and Excitation of the Isomeric States // *Phys. Elem. Part. Atom. Nucl*. 2002, v. 33(1), p. 158-199.
12. K. Kosako, K. Oishi, T. Nakamura, et al. Angular Distribution of Bremsstrahlung from Copper and Tungsten Targets Bombarded by 18, 28 and 38 MeV Electrons // *J. Nucl. Sci. Technol*. 2010, v. 47, p. 286.
13. V.E. Kutny, V.I. Nikiforov, O.A. Opalev, Yu.V. Rogov, A.V. Rybka, V.A. Shevchenko, I.N. Shlyakhov, V.E. Strelnitsky, A.Eh. Tenishev, V.L. Uvarov. Calibration of CVD-Diamond Based Dosimeter in High-Power Electron and X-Ray Radiation Fields // *Problems of Atomic Science and Technology. Series "Nuclear Physic Investigations"*. 2014, № 3, p. 162.
14. A.F. Fernandez, B. Brichard, H. Doms, et al. Gamma dosimetry using Red 4034 Harwell dosimeters in mixed fission neutrons and gamma environments // *IEEE Trans. Nucl. Sci*. 2005, v. 52(2), p. 505-509.

Article received 12.10.2015

МЕТОД ДОЗИМЕТРИИ ВЫСОКОЭНЕРГЕТИЧНОГО ТОРМОЗНОГО ИЗЛУЧЕНИЯ В МЕГАГРЕЙ-ДИАПАЗОНЕ

Ю.В. Рогов, В.А. Шевченко, И.Н. Шляхов, Б.И. Шраменко, А.Э. Тенишев, А.В. Торговкин, В.Л. Уваров, В.Ф. Жигло

Исследована возможность дозиметрии высокоэнергетичного тормозного излучения в диапазоне доз ~МГрей и выше путем активации индия по реакции $^{115}In(\gamma,\gamma')^{115m}In$ с определением поглощенной дозы по удельной активности изомера ^{115m}In. Для изучения такого подхода предложена методика, основанная на совместном облучении набора тонких мишеней. Каждая мишень включает фольги из индия и молибдена, а также стандартный дозиметр в виде пластины из ПММА. Методом моделирования проведено предварительное исследование активации индия с учетом известных данных по сечению реакции, тепловой стойкости детектора, выхода референтных реакций $^{92}Mo(\gamma,2n)^{90}Mo$ и $^{100}Mo(\gamma,n)^{99}Mo$ в молибдене, а также поглощенной дозы в ПММА. Выполнены измерения удельной активности ^{115m}In, ^{90}Mo и ^{99}Mo, а также поглощенной дозы тормозного излучения с использованием дозиметров Harwell Red 4034. Установлена зависимость чувствительности метода от граничной энергии тормозного излучения в диапазоне 8…71 МэВ.

МЕТОД ДОЗИМЕТРІЇ ВИСОКОЕНЕРГЕТИЧНОГО ГАЛЬМІВНОГО ВИПРОМІНЮВАННЯ В МЕГАГРЕЙ-ДІАПАЗОНІ

Ю.В. Рогов, В.А. Шевченко, І.Н. Шляхов, Б.І. Шраменко, А.Е. Тєнішев, О.В. Торговкин, В.Л. Уваров, В.Ф. Жигло

Досліджена можливість дозиметрії високоенергетичного гальмівного випромінювання в діапазоні доз ~МГрей і вище шляхом активації індію за реакцією $^{115}In(\gamma,\gamma')^{115m}In$ з визначенням поглинутої дози з питомої активності ізомеру ^{115m}In. Для дослідження такого підходу запропонована методика, що заснована на сумісному опромінюванні набору тонких мішеней. Кожна мішень включає фольги з індію і молібдену, а також стандартний дозиметр у вигляді пластини з ПММА. Методом моделювання проведено попереднє дослідження активації індію з урахуванням відомих даних з перетину реакції, теплової стійкості детектора, виходу референтних реакцій $^{92}Mo(\gamma,2n)^{90}Mo$ і $^{100}Mo(\gamma,n)^{99}Mo$ у молібдені, а також поглинутої дози в ПММА. Виконано вимірювання питомої активності ^{115m}In, ^{90}Mo і ^{99}Mo, а також поглинутої дози гальмівного випромінювання з використанням дозиметрів Harwell Red 4034. Встановлена чутливість методу залежно від граничної енергії гальмівного випромінювання в діапазоні 8…71 МеВ.