E18-2008-119

N. Baljinnyam, A. G. Belov, G. Ganbold*, Sh. Gerbish,
O. D. Maslov, V. N. Shvetsov

POSSIBILITY OF SOME RADIONUCLIDES PRODUCTION USING HIGH ENERGY ELECTRON BREMSSTRAHLUNG

Submitted to «Радиохимия»

*Nuclear Research Center of NUM, Ulan-Bator, Mongolia

Балжинням Н. и др. E18-2008-119

Возможность получения некоторых радионуклидов
с использованием тормозного излучения электронов
высоких энергий

Описан метод получения некоторых радионуклидов с использованием тормозного излучения электронов высоких энергий. Определены удельные активности и выходы фотоядерных реакций некоторых радионуклидов при облучении чистых фольг из платины, олова и обогащенного изотопа ^{118,112}Sn. Также оценены удельные активности и выходы фотоядерных реакций радионуклидов $^{117\,m}$Sn, ^{111}In и $^{195\,m}$Pt при использовании тормозного излучения электронов высоких энергий линейного ускорителя установки ИРЕН (ЛНФ, ОИЯИ).

Исходя из экспериментальных данных, полученных на ускорителе электронов — микротроне МТ-25 (ЛЯР ОИЯИ) с энергией $E_{e^-} < 30$ МэВ и мощностью 0,5 кВт, оценены выходы в фотоядерной реакции некоторых радионуклидов для линейного электронного ускорителя ($E_{e^-} = 75$ МэВ) установки ИРЕН (ЛНФ ОИЯИ).

Работа выполнена в Лаборатории нейтронной физики им. И. М. Франка и в Лаборатории ядерных реакций им. Г. Н. Флерова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2008

Baljinnyam N. et al. E18-2008-119

Possibility of Some Radionuclides Production Using
High Energy Electron Bremsstrahlung

The method of some radionuclides production using high energy bremsstrahlung of electron accelerators and determination of photonuclear reaction yield and specific activities for some radioinuclides is described. Photonuclear reaction yield and specific activities for some radioinuclides are determined for ^{117m}Sn, ^{111}In and ^{195m}Pt.

Based on the experimental data obtained at low energy ($E_{e^-} < 30$ MeV) electron accelerators as well as the microtron MT-25 (FLNR, JINR) with the power 0.5 kW, photonuclear reaction yields are estimated for some radionuclides for the linear electron accelerator ($E_{e^-} = 75$ MeV) of the IREN facility (FLNP, JINR) at irradiation of high purity platinum and tin metals.

The investigation has been performed at the Frank Laboratory of Neutron Physics and at the Flerov Laboratory of Nuclear Reactions, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2008

INTRODUCTION

At present, the requirements for an acceptable radionuclide are still considerably high. The nuclear reactors and cyclotrons are mainly used for the radionuclide production. However, they are not able to produce all the required types of radionuclides, therefore, electron accelerators as linac and microtron, that can produce radionuclides using the bremsstrahlung, are suitable complements.

Properties of the ^{117m}Sn radionuclide are acceptable for clinical and therapeutic use: the short half-life of 13.6 d is necessary to minimize the patient exposure, the gamma emission of photons of 158.4 keV (84%) for imaging, and abundance (116%) of low energy (127–129, 152 keV). Auger and conversion electrons for delivering a high radiation dose to sites of a bony metastatic disease [1, 2]. Also the platinum radionuclide ^{195m}Pt is used for cancer diagnosis and therapy.

The yields of a number of radionuclides produced by the bremsstrahlung beam irradiation were measured by different groups. Oka et al. [3] measured the yields of radionuclides induced by the (γ, n) reactions with the 20 MeV bremsstrahlung from the linac on 37 elements. Analogous measurements were performed at the Czech microtron by Randa et al. [4] with the 19 MeV bremsstrahlung beam. Detailed measurements were also performed by Gerbish and Thiep et al. [5] at the microtron in Dubna (FLNR, JINR) for practically all natural nuclides at different electron (14, 18 and 20 MeV) energies. Properties and activity yields of radionuclides obtained from photonuclear reaction data are based exclusively on experimentally obtained results for the majority of elements and are presented in book by Segebade et al. [6].

THE AIM OF THE EXPERIMENTS

In the present work we will try to give some information on the possibility of application of the bremsstrahlung beam of the linac of the new Intense REsonance

Neutron pulsed source (IREN) for fundamental and applied nuclear physics is being realized at the end of the 2008 at FLNP JINR, Dubna.

ACCELERATOR

The methods of multielemental photon activation analysis (MPAA) and radionuclide production will be developed at the linac of the IREN facility.

Some technical characteristics for the linac of the IREN facility are as follows:

- Maximum energy of electrons — 75 MeV,
- Peak current — 2.84 A,
- Pulse frequency — 50 Hz,
- Electron burst width (FWHM) — 140 ns,
- Power — 1.1 kW.

A water cooled tungsten or platinum target has been used for the bremsstrahlung converter. The temperature on the surface of the W or Pt disk of the target must be lower than $\sim 900\,^{\circ}$C (see Fig. 1).

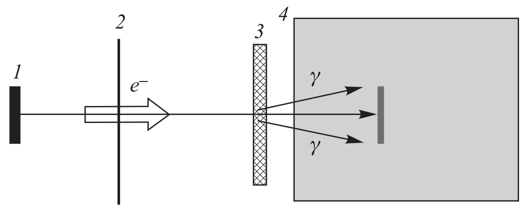


Fig. 1. Simple geometry of radiation target by bremsstrahlung. *1* — electron source; *2* — exit foil ($\sim 0.1 - 0.3$ mm thick Ti); *3* — converter (W or Pt plate ~ 2.5 and 5 mm); *4* — target

The preliminary methodical experimental investigation was carried out at the cyclic electron accelerator microtron MT-25 of FLNR, JINR. The microtron MT-25 was used as the bremsstrahlung source to determine specific activities and yields of the photonuclear reactions. The 0.57 kW power irradiation targets of the microtron MT-25 were operated with the electron beam current of 15μA and energy of 23.5 MeV. The results of methodical study are given in the present work.

EXPERIMENTAL PROCEDURE

To estimate the activity and yield of the ^{117m}Sn, ^{111}In, ^{195m}Pt radionuclide production, the natural pure tin, platinum and enriched ^{118}Sn (98.5%) isotope have been irradiated at the microtron MT-25 by the bremsstrahlung beam of 23.5, 22, 21, 20, 19 MeV with the current of 13–14.5 μA during 1–2 h at the radial distance of 2 cm from the tungsten target (4 mm thick). For determination of photonuclear reaction specific activity and yields of Sn, Pt isotopes several experiments have been carried out:

No.	Energy, MeV	Elements	Mass of sample, mg	Irradiation time, h
1	23.5	Au-1	5	1
		Zr-1	1.2	
		Sn-1	46.1	
		Sn-118 (9)	31.3	
		Cu-1	49.5	
2	22	Au-2	6.2	1.5
		Zr-2	0.6	
		Sn-2	19.2	
		Cu-2	48.8	
3	21	Au-3	6.9	1.5
		Zr-3	0.7	
		Sn-3	28.5	
		Cu-3	44.2	
		Pt-1	8.6	
		Sn-112	12	
4	20	Au-4	5.5	1.5
		Zr-4	4	
		Sn-4	14	
		Cu-4	35.8	
		Pt-2	8.4	
5	19	Au-5	1.2	1.5
		Zr-5	1	
		Sn-5	37.4	
		Cu-5	43.5	
		Pt-5	6.3	
6	24.2	Au-6	1.2	1.5
		Zr-6	1.1	
		Sn-6	37.4	
		Cu-6	39	
		Pt-6	6.3	
		Sn-118 (9)	29.7	

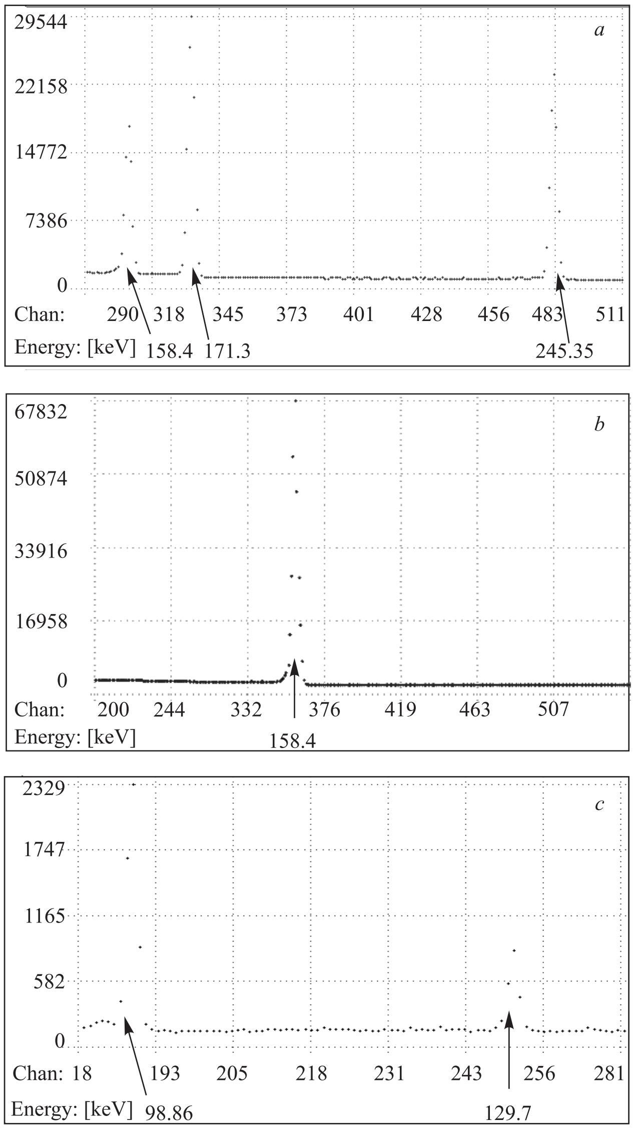
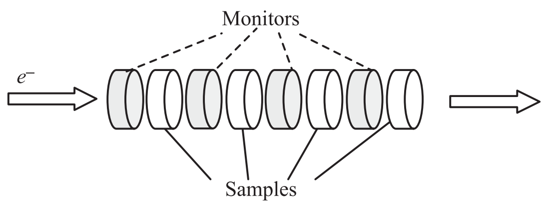


Fig. 2. *a)* A part of gamma spectrum of the irradiated natural pure Sn foil (19.2 mg). There are gamma lines: 158.4 keV for ^{117m}Sn and 171.3 and 245.3 keV for ^{111}In. *b)* A part of gamma spectrum of the irradiated enriched ^{118}Sn (98.5%) foil (29.7 mg). *c)* A part of gamma spectrum of the irradiated natural pure Pt foil (8.6 mg)

Typical irradiation scheme of the second experiment, disposition of samples and beam monitors on the bremsstrahlung of the microtron MT-25 are shown as follows:



Activity of the irradiated high purity natural platinum, tin and enriched ^{118}Sn samples and monitors were measured by HPGe detectors of the gamma spectrometer of FLNP, JINR with the energy resolution of 2 keV at gamma lines of 1332 keV for ^{60}Co radionuclide.

To the monitors of the high energy electron's bremsstrahlung flux are used pure metal foils of Au, Cu and Zr. Gamma lines of the radionuclides, which have been detected from samples and monitors [2, 4] are given in Table 1.

Table 1. Nuclear data of tin (Sn) and platinum (Pt) radioisotopes

Radionuclide and half-life	Main energy E_γ, keV	Intensity, %	Reaction	Reaction E_γ, MeV	Abundance of target, %
^{117m}Sn 13.6 d	158.40	84.00	^{117}Sn (γ, γ') ^{118}Sn (γ, n) ^{119}Sn $(\gamma, 2n)$	–0.32 –9.65 –16.13	7.57 24.01 8.58
^{117m}Sn 13.6 d	158.40	84.00	^{118}Sn (γ, n)	–9.65	98.50
^{111}In 2.83 d	171.29 245.35	91.00 94.00	^{112}Sn (γ, p)	–7.73	0.95
^{195m}Pt 4.1 d	98.86 129.74	11.01	^{195}Pt (γ, γ') ^{196}Pt (γ, n) ^{198}Pt $(\gamma, 3n)$	–0.26 –8.18 –21.60	33.80 25.20 7.19

Equations (1) and (2) for calculation of specific activity, yield and integrated cross section of the photonuclear reaction are as follows:

$$S = \Delta N \gamma \varepsilon = \phi_{\text{th}} \sigma_{\text{eff}} \frac{N_A \theta w}{M} \left(1 - \mathrm{e}^{-\lambda t_{\text{irr}}}\right) \mathrm{e}^{-\lambda t_d} \frac{\left(1 - \mathrm{e}^{-\lambda t_m}\right)}{\lambda} \gamma \varepsilon, \qquad (1)$$

$$\sigma_{\text{int}} = \frac{A_s M(E_{\max} - E_{\text{th}})}{N_A \Phi_{\text{th}}} \quad [\text{MeV} \cdot b], \tag{2}$$

where S — net peak area, N_A — Avogadro number (mol^{-1}), θ — isotopic abundance of the target isotope (%), γ — gamma-ray abundance (i.e., probability of the disintegrating nucleus emitting a photon of this energy, photons per disintegration), w — mass of the irradiated element (g), M — amotic mass ($\text{g}\cdot\text{mol}^{-1}$), ε — photopeak efficiency of detector (i.e., probability that an emitted photon of given energy will be detected to contribute to the photopeak in the spectrum), σ_{eff} — effective cross section (mb or 10^{-27} cm^2), ϕ_{th} — photon flux ($\text{cm}^{-2} \cdot \mu\text{A}^{-1} \cdot \text{s}^{-1}$), λ — decay constant, t_{irr} — irradiation time (s), t_{d} — decay time (s), t_{m} — measuring time (s), A_s — specific activity (Bq/mg), $E_{\max}$ — irradiation energy (MeV), E_{th} — reaction threshold energy (MeV).

Radionuclide of this class is therapeutically and diagnostically useful in skeletal imaging and for the radiotherapy of bone tumors and other disorders.

From the measured gamma-spectrum data the experimental photonuclear reaction activity and yield (see Fig. 2 *a, b, c*) have been determined for radionuclides ^{117m}Sn and ^{111}In, ^{195m}Pt. Their experimental values are shown in Table 2.

Table 2. Experimental results of specific activity and yield for some radionuclides

No.	$E\gamma$, MeV	Element	Radio-nuclide	Specific activities A, Bq/mg	Yields Υ, Bq/ [μA · mg · h]	Isomer's ratio R, [Y_m/Y_x]	Relative integrated cross section, [$\sigma^x/\sigma^{64\text{Cu}}$]
1	23.5	Sn-1	^{117m}Sn	8.17E+05	5.45E+04		0.44
			^{111}In	8.76E+06	5.84E+05	0.09	5.1
			^{113}Sn	7.01E+06	4.68E+05	0.116	3.46
			^{123}Sn	1.52E+06	1.01E+05	0.54	0.93
		Sn-118	^{117m}Sn	5.77E+05	3.85E+04		0.31
2	22	Sn-2	^{117m}Sn	3.14E+05	1.39E+04		0.28
			^{111}In	3.15E+06	1.40E+05	0.09	3.12
			^{113}Sn	2.62E+06	1.16E+05	0.12	2.01
			^{123}Sn	6.17E+05	2.74E+04	0.51	0.64
3	21	Sn-3	^{117m}Sn	3.02E+05	1.33E+04		0.40
			^{111}In	3.30E+06	1.47E+05	0.09	4.84
			^{113}Sn	2.60E+06	1.16E+05	0.115	3.12
			^{123}Sn	6.02E+05	2.68E+05	0.49	0.92
		Pt-1	^{195m}Pt	6.56E+05	2.92E+04		1.63
			^{191}Pt	4.46E+06	1.98E+05	0.15	10.77

Table 2 (continuation)

No.	$E\gamma$, MeV	Element	Radio-nuclide	Specific activities A, Bq/mg	Yields Υ, Bq/ $[\mu\text{A} \cdot \text{mg} \cdot \text{h}]$	Isomer's ratio R, $[Y_m/Y_x]$	Relative integrated cross section, $[\sigma^x/\sigma^{64\text{Cu}}]$
4	20	Sn-4	^{117m}Sn	2.75E+05	1.20E+04		0.41
			^{111}In	2.99E+06	1.30E+05	0.09	5.08
			^{113}Sn	2.33E+06	1.02E+05	0.12	3.18
			^{123}Sn	5.07E+05	2.21E+05	0.54	0.89
		Pt-3	^{195m}Pt	1.96E+05	8.55E+03	0.15	0.56
			^{191}Pt	1.30E+06	5.70E+04		3.62
5	19	Sn-5	^{117m}Sn	1.1E+05	3.77E+03		0.31
			^{111}In	1.25E+06	4.29E+04	0.08	4.12
			^{113}Sn	1.05E+06	3.59E+04	0.105	2.70
			^{123}Sn	2.49E+05	0.85E+04	0.44	0.84
		Pt-3	^{195m}Pt	4.04E+05	1.38E+04	0.15	2.23
			^{191}Pt	2.66E+06	0.91E+05		14.6
6	24.5	Sn-6	^{117m}Sn	9.73E+04	4.11E+03		0.07
			^{111}In	1.07E+06	4.51E+04	0.09	0.78
			^{113}Sn	8.69E+05	3.67E+04	0.11	0.54
			^{123}Sn	1.86E+05	7.86E+03	0.52	0.14
		Pt-6	^{195m}Pt	8.03E+05	3.39E+04	0.116	1.0
		^{191}Pt	6.95E+06	2.93E+05			0.84
		Sn-118	^{117m}Sn	5.43E+5	2.29E+4		0.06

DISCUSSION AND CONCLUSION

1. Results of the experiments showed the possibility of the ^{117m}Sn, ^{111}In, ^{195m}Pt radionuclide production using the bremsstrahlung of high energy electron accelerators. In case of irradiation of the natural pure tin, ^{117m}Sn and ^{111}In radioisotopes have been produced, which both are useful for medical purposes.

2. The experimental yield of photonuclear reaction is several times higher than when an enriched ^{118}Sn (98.5%) isotope is used in the target. From the gamma spectrum in Fig. 2, *a, b, c* one can see the gamma line of 158.4 keV of only one ^{117m}Sn radionuclide. The photonuclear reaction yields of the ^{111}In have been compared with the data determined in other works and they have sufficient coincidences.

3. The isomeric ratio of ^{117m}Sn was estimated from the experimental data of activities of 158.4 (^{117m}Sn); 391.7 (^{113}Sn) and 1089 (^{123}Sn) keV energy gamma lines. The isomeric ratios for ^{117m}Sn were estimated and determined: 0.114 ± 0.005 and 0.506 ± 0.038 for the radionuclides ^{113}Sn, ^{123}Sn, correspondingly. The isomeric ratio for ^{195m}Pt was determined: 0.141 ± 0.005.

4. The activities and yields of the ^{117m}Sn and ^{111}In radionuclides will be (10 times) increased at the bremsstrahlung beam of the linac of the IREN facility.

5. In Table 2 the calculated specific activities, photonuclear reaction yields and relative integrated cross section for the radionuclides (^{196}Au, ^{89}Zr, ^{117m}Sn, ^{111}In, ^{113}Sn, ^{123}Sn, ^{195m}Pt and ^{191}Pt) have been shown.

6. In Fig. 3 the histogramms of distribution for relative integrated cross section of the above-mentioned radionuclides in absorption energies of bremsstrahlung have been given.

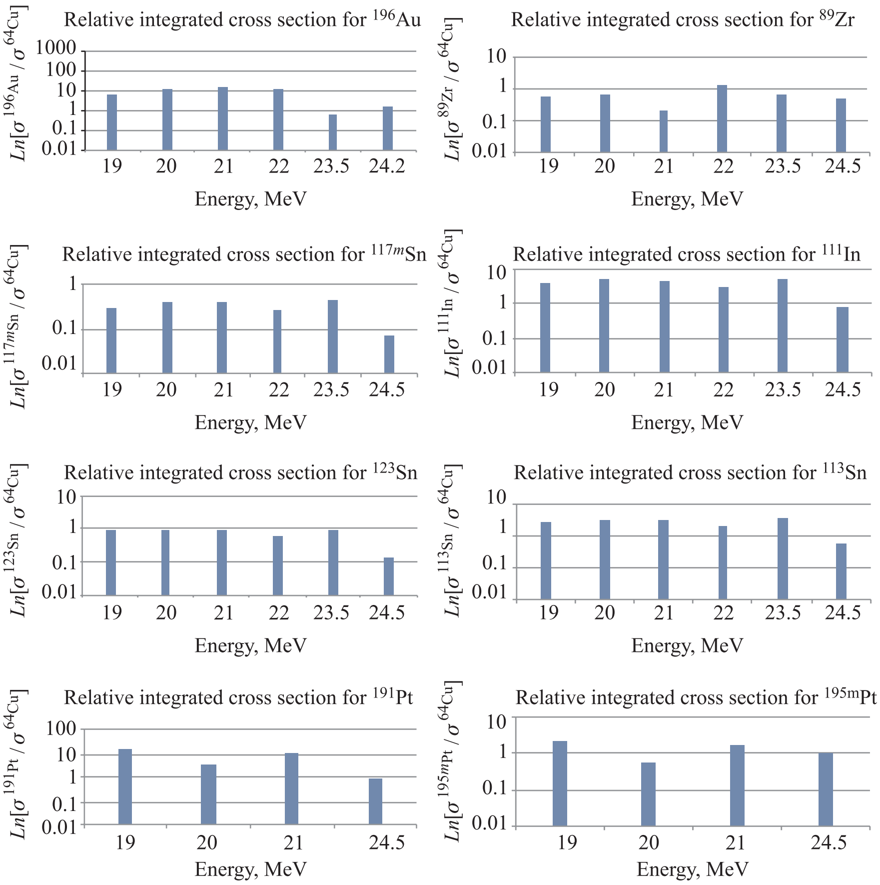


Fig. 3. The distribution of relative integrated cross sections $[\sigma^{x}/\sigma^{64\text{Cu}}]$ for some radionuclides at irradiation energies of bremsstrahlung

7. From the calculated integrated cross section the isomeric ratio can be determined and $\sigma(\gamma, n)/\sigma(\gamma, p)$ for Sn has been found: 0.088 ± 0.004.

8. From the experimental results one can deduce the possibility of production more useful radionuclides (^{117m}Sn, ^{111}In, and ^{195m}Pt) for nuclear medicine, science and technology using high-energy bremsstrahlung electron accelerators.

REFERENCES

1. *Srivastava S. C. et al.* Nuclear Medicine and Biology Advances. V. 2. / Ed. by C. Raynaud. Pergamon Press, 1983. P. 1635–1638.
2. *Srivastava S. C. et al.* // Clinical Cancer Research. 1998. V. 4. P. 61–68.
3. *Oka Y., Kato T., Nomura K., Saito T.* // Bull. Chem. Soc. Jap. 1967. V. 40. P. 575.
4. *Randa Z., Duchacek V., Hradil M.* // Jad. En. 1988. V. 34. P. 365.
5. *Gerbish Sh. et al.* JINR Commun. P6-91-123. Dubna, 1991.
6. *Segebade Ch., Weise H.-P., Lutz G. J.* Photon Activation Analysis. Berlin; N.Y.: Walter de Gruyter, 1988. P. 161–310.
7. *Novogorodov A. F. et al.* Simple Method of High Temperature Separation ^{111}In from Massive Tin Target // Radiokhimiya. 1987. V. 2. P. 254–258 (in Russian).
8. *Levin V. et al.* // Radiochem. Radioanal. Lett. 1981. V. 49, No. 2. P. 111.
9. *Malinin A. et al.* // Radiochem. Radioanal. Lett. 1983. V. 59, No. 4. P. 21
10. *Aksenov V. L. et al.* Proposal for the Construction of the New Intense Resonance Neutron Source (IREN). JINR Commun. E3-92-110. Dubna, 1992.
11. *Gerbish Sh. et al.* Application of the IREN Facility for Production of Radionuclide. JINR Commun. P18-2006-116. Dubna, 2006 (in Russian).

Received on August 18, 2008.

Корректор *Т. Е. Попеко*

Подписано в печать 27.11.2008.
Формат $60 \times 90/16$. Бумага офсетная. Печать офсетная.
Усл. печ. л. 0,93. Уч.-изд. л. 1,32. Тираж 265 экз. Заказ № 56420.

Издательский отдел Объединенного института ядерных исследований
141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/