PRODUCING THE PLANAR MULTIPHOTON SOURCES BY PHOTONUCLEAR TECHNIQUE: 1. A MODEL OF GAMMA-FLUORESCENT SOURCE

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The possibility is shown to produce the planar γ-sources with the two and more spectral bands in the photon energy range up to ~100 keV by activating K-lines of the characteristic x-ray radiation in the thin layers of the elementary substances by an external single-photon γ-source. A one-dimensional model of a combined planar source in the form of a stack of foils from the inactive and active materials has been developed. The model enables the analysis and optimisation of a source against its intensity and spectral composition. A variant of a production target for manufacturing at an electron accelerator the planar γ-sources on the basis of the isotopes ^{57}Co and ^{179}Ta is offered and investigated by computer simulation. It is shown, that from the viewpoint of yield of the isotopes, suitable for the use in absorptiometry, the photonuclear method is competitive as compared with the reactor and cyclotron technologies.

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INTRODUCTION

Single- and multi-photon γ-sources are widely used in up-to-date medical and industrial diagnostics, inspection check-up, spectrometry, etc. (see, e.g. [1, 2]). For instance, the diagnostics of the osteoporosis, the most widespread age-specific illness of the bone tissue, is based on measuring its mineral density by single- and dual- photon absorptiometry. As it was preliminary specified, the radiation with photon energy in the range 30…60 keV provides the most contrast differentiation between the soft and bone tissues in an introscopic image. So initially for the single photon absorptiometry, the ^{125}I isotope ($T_{1/2}$=59.4 day) having the most intensive spectral line 27.5 keV has been chosen, as well as ^{153}Gd (42 and 99 keV; 240.4 day) is used in the more accurate dual photon diagnostics [3, 4]. The both isotopes are produced by radiochemical extraction from the targets irradiated in reactor [5]. To obtain a sealed γ-source, a separated isotope product is hermetically encapsulated.

In the sequel, a method of dual absorptiometry based on the use of the X-ray tubes (DEXA-technique) has been developed (see, e.g. [4]). This time the spectrum of radiation is formed with specially chosen filters to provide the intensity maximum in the specified region. In turn, the peripheral quantitative computed tomography (pQCT) appears to be more informative [6]. That method uses the movable X-ray tubes with the filters, providing the spectral maximum near 60 keV. At the same time, obtaining in such a way the dual photon sources with the adequate operation life, high stability and uniformity of the radiation field, and also with the narrow spectral bands is rather knotty problem. In particular, this circumstance impedes realization of the dual photon variant of pQCT, which is promising for early recognition of the illness.

As known, the radionuclides decaying via the electron capture emit Auger electrons and photons of the characteristic X-ray radiation. Its high-energy part corresponds to the transitions of the orbital electrons on an empty K-state. The X-ray radiation is generated also at interaction of gammas with the bound atomic electrons as a result of photoeffect (see, e.g. [7]). Thus one can obtain a high-energy X-ray source either by selecting an appropriate radionuclide or by acting on an inactive (cold) material with the photons having energy higher than the K-line energy of that material. Their values lay in the range from 13 eV (hydrogen) to ~120 keV (uranium), i.e. just cover the range suitable for medical introscopy. The proton-redundant nuclei, decaying through the K-capture, can be generated in the photonuclear reactions realized at an comparatively inexpensive and safe electron accelerator. In this work, the conditions are studied for manufacturing the planar γ-sources with the specified two and more spectral bands by combining the layers of the cold materials and planar γ-sources produced by a photonuclear technique.

1. MAIN REACTIONS

^{57}Co (Eγ=122.1 and 136.5 кеV; $T_{1/2}$=271.3 day) is one of the most suitable γ-activator of the characteristic X-radiation. Therefore it is used in the form of quasi-point sources in the fluorescence elemental analysis [8]. The planar sources on the basis of this isotope with activity up to 740 MBq and measuring up to 700×500 mm (the flood sources) are applied in medicine for calibrating the gamma-ray chambers (see, e.g. [9]).

Commonly, ^{57}Co is produced on cyclotrons by the reactions ^{56}Fe(d,n)^{57}Co and ^{58}Ni(p,2p)^{57}Co [10]. This radionuclide can be generated also by a photonuclear technique in a target from nickel via the two channels simultaneously [11]:

$$^{58}\mathrm{Ni}(\gamma,\mathrm{p})^{57}\mathrm{Co},$$

$$^{58}\mathrm{Ni}(\gamma,\mathrm{n})^{57}\mathrm{Ni} \xrightarrow[35h]{\beta^+} {}^{57}\mathrm{Co}.$$

The first reaction has the threshold 8.2 MeV at a maximum of the cross section of about 60 mb, the second – 12.2 MeV and 23 mb, respectively [12].

A beam of the high-energy photons can be obtained at an electron accelerator as secondary bremsstrahlung radiation. For this, an intermediate target-converter from high-Z material is used. The radiation has a continuous spectrum with end-point photon energy corresponding to the electron energy E_0.

At the ^{57}Co photonuclear production with the use of a target from natural nickel (the ^{58}Ni abundance is 68.27%) and electron beam with energy higher than 20 MeV, the channels of the hot admixtures production are also revealed. Besides, the photoneutrons, generating as a result of the (γ,xn) processes in the elements of the accelerator exit devices, can also effect on a production target. The main reactions of the hot by-product generation in natural nickel are

$$^{58}\text{Ni}(\gamma,2n)^{56}\text{Ni} \xrightarrow[5.9\,day]{\varepsilon} {}^{56}\text{Co} \xrightarrow[77.3day]{\varepsilon} {}^{56}\text{Fe}_{\text{stab}};$$

$$^{58}\text{Ni}(\gamma,np)^{56}\text{Co} \xrightarrow[77.3day]{\varepsilon} {}^{56}\text{Fe}_{\text{stab}};$$

$$^{58}\text{Ni}(n,p)^{58}\text{Co} \xrightarrow[70.8day]{\varepsilon} {}^{58}\text{Fe}_{\text{stab}};$$

$$^{60}\text{Ni}(\gamma,np)^{58}\text{Co} \xrightarrow[70.8day]{\varepsilon} {}^{58}\text{Fe}_{\text{stab}}.$$

Those admixtures have considerably lesser half-life than ^{57}Co. So by proper target cooling, their relative contribution to the target activity can be reduced to the tolerable value.

^{179}Ta ($T_{1/2}$=665 day) is the one more isotope promising for medical diagnostics. Its principal K-lines are 54.07 keV (21.9%), 54.61 keV (12.6%), 62.98 keV (2.4%), 63.24 keV (4.7%), and 64.9 keV (1.6%) – [13]. This isotope can be produced by the reaction ^{181}Ta(γ, 2n)^{179}Ta with maximum of the cross section 180 mb and threshold 14.2 MeV. A target from natural tantalum (the ^{181}Ta abundance makes 99.99%) can be utilized – [14]. At the same time, ^{180}Ta is generated also under those conditions as the most active admixture via the reaction ^{181}Ta(γ,n)^{180}Ta with threshold 7.6 MeV. As a result of the decay ($T_{1/2}$=8.15 h), ^{180}Ta is transformed into the stable isotopes ^{180}Hf (86%) and ^{180}W (14%). The reaction $^{181}\text{Ta}(n,\gamma)^{182}\text{Ta} \xrightarrow[114.4day]{\beta^-} {}^{182}\text{W}_{\text{stab}}$ is induced also in tantalum by photoneutrons.

2. PHOTONUCLEAR ISOTOPE YIELD

The expression for the target activity A(t), produced by the end of the irradiation period t, can be presented in the form

$$A(t) = y_1 \lambda \frac{I_0}{e} tD(\lambda t), \quad (1)$$

where y_1 – is the yield of the new nuclei normalized to the one beam electron, λ – is their decay constant, I_0 – is the average electron beam current, $D(\lambda t)$ – is the coefficient of the target deactivation during exposure,

$$D(\lambda t) = \frac{1-\exp(-\lambda t)}{\lambda t}. \quad (2)$$

In case of a channel with the formation of an intermediate nucleus, its contribution $A'(t)$ to the activity of the target isotope by EOB can be determined from the expression

$$A'(t) = y_1' \lambda \frac{I_0}{e} t \frac{\lambda'}{\lambda' - \lambda}[D(\lambda t) - D(\lambda' t)], \quad (3)$$

where y_1' – is the normalized yield of the intermediate isotope, λ' – is its decay constant. So at a period of the nickel activation of t >>35 h, the contribution of the channel ^{58}Ni(γ,n)^{57}Ni→^{57}Co to the total yield of ^{57}Co can reach 24%.

The analytical expression for the normalized photonuclear yield of an isotope y_1 in a thick production target is presented in Ref. [15]. In particular, it is shown, that the effective isotope generation is provided at a target thickness of about free range $R_\gamma(E_\gamma^i)$ of the photons with energy E_γ^i, corresponding to the maximum of the cross section of i – reaction, where $R_\gamma(E_\gamma^i) = \left[\mu(E_\gamma^i)\right]^{-1}$, $\mu(E_\gamma^i)$ – is the mass attenuation coefficient of photons in the target material. So the volumetric distribution of the generated nuclei along the bremsstrahlung axes in a target layer of lesser thickness can be considered as quasi-homogeneous. Commonly, E_γ^i corresponds to the region of the giant dipole resonance and makes ~20 MeV. Thus in case of a target from nickel $R_\gamma(E_\gamma^i)$ ~1 cm.

3. GAMMA-FLUORESCENCE

Consider the type of radiation sources obtained by stacking the thin layers of the cold elementary substances and planar γ-sources with homogeneous volumetric distribution of the hot nuclei.

As known, the principal process of interaction of the gammas having energy up to 100 keV with substance is the photoeffect on bound electrons of the atoms, which probability increases with the grows the of binding energy [7]. The excitation of the atoms is removed by emission of characteristic X-ray (fluorescence), in which the relative contribution of K-lines makes from 84% (indium) to practically 100% (uranium) – [8]. Under such conditions, the yield of K-lines can be considered as proportional to the mass attenuation coefficient of gammas, given by the formula

$$\mu(E_\gamma, Z) = \sigma_{ph}(E_\gamma, Z) \cdot \frac{N_A}{\bar{A}_Z}, \quad (4)$$

where σ_{ph} – the cross section of the photoeffect, N_A – is the Avogadro number, $\bar{A}_\text{Z}$ – is the average atomic number of the substance.

Hereinafter, we will call the cold elements of a combined source, fluorescent under external irradiation, as *radiators*, while its hot elements as *activators*. We will identify also the combined radiation sources by the expressions like L+M*+…, where L, M,… – are the names of the chemical elements of materials in the order of their positioning relative to a radiation detector, while the mark * denotes the presence of γ-activity at the corresponding component of the source.

3.1. TWO-COMPONENT SOURCE

Let us consider a source of the L+M* type, comprising a radiator L and activator M* with thicknesses d_1 and d_2^*, respectively. Taking into account, that the lateral dimension of each element of the source considerably exceeds its thickness (the condition of planarity), the analysis of excitation and transfer of the radiation is being conducted using the one-dimensional approximation. Such an approach seems to be substantiated at least

at a source thickness of less or about the photon free range. So at a total activator's activity of A*, the flux of its gammas acting on the radiator L with due regard to the homogeneous depth distribution of hot nuclei, as well as to the self-absorption of their radiation, is defined by the expression

$$\Phi_{1,2}(E_\gamma) = \frac{I(E_\gamma)\cdot A^*}{2\mu_2(E_\gamma)\cdot d_2^*}\ 1-\exp\left[-\mu_2(E_\gamma)\cdot d_2^*\right], \quad (5)$$

where $I(E_\gamma)$ – is the quantum yield of the photons with energy E_γ, $\mu_2(E_\gamma)$ – is the mass attenuation coefficient of photons in the activator material. The subscript 1, 2 denotes the boundary between the first and second (L and M*) elements of the source. The analysis of the formula (5) shows, that in case of a planar activator, its optimum thickness, taking into account the self-absorption of radiation, makes ~ $[\mu_2(E_\gamma)]^{-1}$. That corresponds to the free range of the photons with energy E_γ. Their flux $\Phi(E_\gamma)$, leaving the radiator sideway the detector (a high-energy band of the source spectrum), amounts

$$\Phi\ (E_\gamma) = \Phi_{1,2}(E_\gamma)\exp\left[-\mu_1(E_\gamma)\cdot d_1\right], \quad (6)$$

where $\mu_1(E_\gamma)$ – is the mass attenuation coefficient of activating gammas in the radiator material.

In turn, the flux $\Phi_{1,2}(E_\gamma)$, acting on the radiator, excites in its volume characteristic X-rays with photon energy E_{x1}.The yield of the X-radiation from the source $\Phi(E_{x1})$, considering self-absorption in the radiator both activating gammas and induced fluorescent photons, makes

$$\Phi\ (E_{x1}) = \frac{\mu_1(E_\gamma)\ \exp\left[-\mu_1(E_\gamma)\cdot d_1\right]-\exp\left[-\mu_1(E_{x1})\cdot d_1\right]}{2\left[\mu_1(E_{x1})-\mu_1(E_\gamma)\right]}\Phi_{1,2}(E_\gamma), \quad (7)$$

where $\mu_1(E_{x1})$ – is the mass attenuation coefficient of the fluorescent photons, induced in the radiator, in its material.

As it follows from the expression (7), the maximum yield of the fluorescence provides a radiator with thickness

$$d_1^{\max} = \left[\mu_1(E_{x1})-\mu_1(E_\gamma)\right]^{-1}\ln\frac{\mu_1(E_{x1})}{\mu_1(E_\gamma)}. \quad (8)$$

The formula (8) gives the value of $d_1^{\max}$, that lays between the values of free range of the activating and fluorescent photons in the radiator material. This circumstance corroborates the validity of the developed model at the conditions of practical interest. At the same time, the formula (7) can overestimate the value of the X-ray yield beyond that region, since the offered one-dimensional model does not take into consideration the effects of photon scattering at a radiator thickness exceeding the value of their free range.

3.2. THREE-COMPONENT SOURCE

Now we will analyze a source of the L+M*+N type, including the two radiators: the main radiator of d_1 thickness and a complementary one, N, d_3 thick as well as an activator M* by d_2^* in thickness, placed between them. The fluxes of the activator radiation in both sides are equal, or $\Phi_{2,3}(E_\gamma)=\Phi_{1,2}(E_\gamma)$. The radiators can be manufactured either from the same material (to increase a low-energy band in a two-photon variant of the source) or from the different materials (if there is the necessity to create a third band in the spectrum).

Firstly, we will study the second variant as more general. In this case, the X-radiation with photon energy E_{x3} will be induced in the complementary radiator N. To reduce its self-absorption in the source, we will set a thin activator, that meets the condition $d_2^* << [\mu_2(E_{x3})]^1$, where $\mu_2(E_{x3})$ – is the mass attenuation coefficient of photons with energy E_{x3} in the activator material. The flux of the X-radiation at the boundary with activator, $\Phi_{2,3}(E_{x3})$, considering the absorption in this radiator both activating and fluorescent photons, makes

$$\Phi_{2,3}(E_{x3}) = \frac{\mu_3(E_\gamma)\left[1-\exp\ -\left[\mu_3(E_\gamma)+\mu_3(E_{x3})\cdot d_3\right]\right]}{2\left[\mu_3(E_\gamma)+\mu_3(E_{x3})\right]}\Phi_{2,3}(E_\gamma), \quad (9)$$

where $\mu_3(E_\gamma)$ and $\mu_3(E_{x3})$ – are the mass attenuation coefficients of photons with energy E_γ and E_{x3} in the material of the second radiator, respectively. The flux of this X-radiation from the source after its consecutive attenuation in the activator and L-radiator, amounts

$$\Phi(E_{x3}) = \exp\ -\left[\mu_2(E_{x3})\cdot d_2^* + \mu_1(E_{x3})\cdot d_1\right]\cdot\Phi_{2,3}(E_{x3}), \quad (10)$$

where $\mu_1(E_{x3})$ and $\mu_2(E_{x3})$ – are the mass attenuation coefficients of photons with energy E_{x3} in the materials of the first radiator and activator, respectively. In case of high-thick complementary radiator, $d_3 >> [\mu_3(E_{x3})+\mu_3(E_\gamma)]^{-1}$, the intensity of the third band in the spectrum is maximal, namely,

$$\Phi^{\max}(E_{x3}) = \frac{\mu_3(E_\gamma)\cdot\exp\ -\left[\mu_2(E_{x3})\cdot d_2^* + \mu_1(E_{x3})\cdot d_1\right]}{2\left[\mu_3(E_\gamma)+\mu_3(E_{x3})\right]}\cdot\Phi_{2,3}(E_\gamma). \quad (11)$$

If the both radiators have been fabricated from similar material ($\mu_1\equiv\mu_3$), the relative gain of the low-energy band, as compared with the two-component variant, makes

$$\frac{\Delta\Phi(E_{x1})}{\Phi(E_{x1})} = \frac{\mu_1(E_{x1})-\mu_1(E_\gamma)}{\mu_1(E_{x1})+\mu_1(E_\gamma)}\cdot\frac{\exp\ -\left[\mu_2(E_{x1})\cdot d_2^* + \mu_1(E_{x1})\cdot d_1\right]\cdot\left[1-\exp\ -\left[\mu_1(E_\gamma)+\mu_1(E_{x1})\right]\cdot d_3\right]}{\exp\left[-\mu_1(E_\gamma)\cdot d_1\right]-\exp\ -\mu_1(E_{x1})\cdot d_1}. \quad (12)$$

If the second radiator is thick ($d_3 >> [\mu_1(E_{x1})+\mu_1(E_\gamma)]^{-1}$, its contribution to the radiation of the source is maximal. So in a system Ta+Ni*+Ta at a thickness of the two first elements of 0.1 mm the addition of the second radiator can increase the yield of the tantalum fluorescence up to 64%.

The sources with the greater number of components and their various sequence can be analyzed in the same way. The value of the mass attenuation coefficient of the photons with any energy in various materials can be calculated, e.g., using a package XMuDat [16].

4. DESIGN OF PRODUCTION TARGET

In the report [17], a technique for joint production of the planar ^{179}Ta and ^{57}Co sources is described. The feature of a proposed target device lays in the usage of the tantalum simultaneously as a converter of the brems-strahlung radiation and a target for generation ^{179}Ta, as well as for photonuclear activating the nickel resulting in the ^{57}Co generation.

It should be noted, that the free range of photons with energy above the reaction threshold of 14.2 MeV in tantalum makes ~1 cm. This is by two orders of values higher than the free range of the Ta characteristic photons. As it was shown above, the latter value determinates the optimal thickness of the source-see formula (8).

The geometry of the accelerator exit device providing the maximal yield of both the target isotopes and minimal self-absorption of their radiation in the obtained γ-sources was determined by means of computer simulation. The target device corresponds a two-layer cylinder with the central passage for cooling water. A tape from tantalum 0.1mm thick by 180 cm in length and coiled into a roll by 23.5 mm in outward diameter forms an outside of the cylinder. A similar tape from nickel, coiled also into a roll, forms an inner layer of the cylinder. To provide even distribution of the surface activity and absorbed dose, the electron beam is scanned along the moving line of the target-cylinder with its simultaneous rotation about the axis of the target. For a comparison, the data on productivity of the known technologies for manufacturing isotopes used in the absorptiometry as well as the results of calculation of joint ^{179}Ta and ^{57}Co capacity on the basis of an electron accelerator are given in Table. It is seen, that photonuclear technology realized even at an accelerator with rather ordinary parameters (40 MeV; 250 μA) is competitive.

Capacity of various techniques of isotope production (100 h irradiation run)

Isotope	Installation	Parameters	Reaction	Yield, GBq	Reference
^{125}I	Reactor	$5\cdot10^{13}$ n/cm^2·s	^{124}Xe(n,γ)^{125}Xe→^{125}I	14.0	[5]
^{153}Gd	Reactor	$6\cdot10^{13}$ n/cm^2·s	^{152}Gd(n,γ)^{153}Gd	0.13	[5]
^{57}Co	Cyclotron	20→15 MeV 250 μA	^{58}Ni(p,2p)^{57}Co	10	[18]
^{179}Ta + ^{57}Co	Electron accelerator	40 MeV 250 μA	^{181}Ta(γ,2n)^{179}Ta ^{58}Ni(γ,p)^{57}Co ^{58}Ni(γ,n)^{57}Ni→^{57}Co	3.7 + 8.2	[17]

CONCLUSIONS

A model has been developed for analysis of the multi-photon sources obtained by mixing the radiation of the thin γ-active foils, produced at an electron accelerator, and the induced characteristic X-radiation in the thin layers of the cold elementary substances, contacting with the active foils. The model makes it possible to calculate the intensity of the bands of the combine sources as well as their optimization with regard to the intensity and spectral composition using the available data on the mass attenuation coefficients of gammas in the materials of the source components.

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ПОЛУЧЕНИЕ ПЛАНАРНЫХ МУЛЬТИФОТОННЫХ ИСТОЧНИКОВ ФОТОЯДЕРНЫМ МЕТОДОМ: 1. МОДЕЛЬ ГАММА-ФЛУОРЕСЦЕНТНОГО ИСТОЧНИКА

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Показана возможность получения планарных γ-источников с двумя и более спектральными полосами в диапазоне энергий фотонов до ~100 кэВ путём активации К-линий характеристического рентгеновского излучения в тонких слоях простых веществ внешним однофотонным γ-источником. Разработана одномерная модель комбинированного планарного источника в виде набора тонких слоёв из неактивных и активных материалов. Модель обеспечивает возможность анализа и оптимизации источника в отношении интенсивности и спектрального состава излучения. Предложен и исследован методом компьютерного моделирования вариант технологической мишени для наработки на ускорителе электронов планарных γ-источников на основе изотопов ^{57}Co и ^{179}Ta. Показано, что в отношении выхода изотопов, пригодных для использования в абсорбциометрии, фотоядерный метод является конкурентоспособным по сравнению с реакторными и циклотронными технологиями.

ОДЕРЖАННЯ ПЛАНАРНИХ МУЛЬТИФОТОННИХ ДЖЕРЕЛ ФОТОЯДЕРНИМ МЕТОДОМ: 1. МОДЕЛЬ ГАММА-ФЛУОРЕСЦЕНТНОГО ДЖЕРЕЛА

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Показана можливість одержання планарних γ-джерел з двома і більше спектральними смугами в діапазоні енергій фотонів до ~100 кеВ шляхом активації К-ліній характеристичного рентгенівського випромінювання в тонких шарах простих речовин зовнішнім однофотонним γ-джерелом. Розроблена одномірна модель комбінованого планарного джерела у вигляді набору тонких шарів неактивних та активних матеріалів. Модель забезпечує можливість аналізу та оптимізації джерела щодо інтенсивності та спектрального складу випромінювання. Запропоновано та досліджено методом комп'ютерного моделювання варіант технологічної мішені для напрацювання на прискорювачі електронів планарних γ-джерел на основі ізотопів ^{57}Co і ^{179}Ta. Показано, що відносно виходу ізотопів, придатних для використання в абсорбціометрії, фотоядерний метод є конкурентоспроможним у порівнянні з реакторними і циклотронними технологіями.