DETECTORS AND NUCLEAR RADIATION DETECTION

METHOD FOR EXPRESS DETERMINATION OF MEDICAL RADIONUCLIDES ^{99m}Tc, ^{67}Cu CONCENTRATION USING SPECTROMETER BASED ON Si PLANAR DETECTOR

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The method of express analysis provides the measurement of radiation spectra of medical radionuclides by a detection module based on an uncooled silicon planar detector and a "scintillator-silicon photosensor" type module. A spectrometric device for express analysis of radionuclide concentration has been developed and its prototyping has been carried out, confirming the operability of the device. The spectrometer analyzer is powered from the USB port of the PC (including a laptop) that provides the possibility of autonomous operation. Test measurements were carried out using the prototype of the spectrometer and X-ray sources, as well as spectral distributions of the radiation of real samples of ^{99m}Tc in a special container (glass ampoule). For a Si-PIN-detector with a thickness of 300 μm, the emission spectrum consists of a line of ^{99m}Tc with an energy of 140.5 keV and two peaks of the characteristic X-ray radiation (CXR) of technetium. The calculation in GEANT4 shows the matching of the ^{99m}Tc activities obtained from the registration of CXR and the main line. A measurement of the technetium concentration both by the 140.5 keV line and on the CXR radiation was proposed that significantly increases the speed of the data accumulation.

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INTRODUCTION

Modern nuclear medicine uses radioactive isotopes in the form of radiopharmaceuticals, which allow carrying out various diagnostic and therapeutic procedures [1, 2]. The most important radionuclide used in 80…90% of the procedures is ^{99m}Tc. This isotope is formed during the decay of the parent isotope ^{99}Mo [3, 4]. The ^{99m}Tc isotope is separated by physicochemical means and used as a radioactive label in various chemical compounds. A perspective isotope is also ^{67}Cu. This isotope has a short half-life and low-energy emission lines what reduces the radiation dose on the patient's body.

The NSC KIPT developed the physical and technological basis for the production of ^{99}Mo/^{99m}Tc [3, 4] and ^{67}Cu [5, 6] using the bremsstrahlung of an electron accelerator. The obtained real pharmaceutical drug ^{99m}Tc was contained in glass ampoules with a wall thickness of ~ 1 mm. Due to the fact that the obtained pharmaceuticals are of different activity, the production of such drugs requires the determination of the concentration of the medical radionuclide in a short time. For the express analysis of the medical radionuclides ^{99m}Tc and ^{67}Cu concentration a spectrometer analyzer with interchangeable detection modules was developed in NSC KIPT. One of the detection modules based on an uncooled planar silicon detector, the second module is a scintillator-photodetector system.

The spectrometer-analyzer was powered from the USB port of the PC (laptop) that provides the possibility of autonomous operation.

1. RADIATION LINES ^{99m}Tc, ^{67}Cu

The half-life of ^{99m}Tc is 6.01 h, the energy and relative intensity of its main lines of gamma and X-ray radiation show in Table 1 [7, 8]. The radiation of technetium consists of the main line of 140.5 keV (the intensity of the quantum yield per decay 89%) and several lines of the CXR technetium. The K-lines of the CXR technetium arise after the internal conversion of electrons from the K shell and total quantum yield is of ~ 8.3% of the main line intensity 140.5 keV. K_α lines have a yield intensity per decay of 6.25%, K_β – 1.157%.

Table 1

Energy and intensities of the main lines of gamma and X-rays for ^{99m}Tc

Gammas from ^{99m}Tc (6.01 h)		
E_γ (keV)	I_γ (%)	
140.511	89	
X-rays from ^{99m}Tc (6.01 h)		
E_γ (keV)	I_γ (%)	Assignment
18.251	2.15	Tc K_{a2}
18.367	4.10	Tc K_{a1}
20.599	0.333	Tc K_{b3}
20.619	0.644	Tc K_{b1}
20.789	0.00357	Tc K_{b5}
21.005	0.146	Tc K_{b2}
21.042	0.0302	Tc K_{b4}

The half-life of ^{67}Cu is 61.83 h. The ^{67}Cu emission spectra also consists of several lines (decay mode β^-) and is presented in Table 2.

Table 2

Energy and intensities of the main lines of gamma and X-rays for ^{67}Cu

Gammas from ^{67}Cu (61.83 h)		
E_γ (keV)	I_γ (%)	
91.266	7.0	
93.311	16.1	
184.577	48.7	
X-rays from ^{67}Cu (61.83 h)		
E_γ (keV)	I_γ (%)	Assignment
8.616	1.95	Zn K_{a2}
8.639	3.84	Zn K_{a1}
9.572	0.239	Zn $K_{\beta3}$
9.572	0.464	Zn $K_{\beta1}$

The low-energy CXR is strongly absorbed by the walls of the glass ampoule and is not suitable for express analysis of the drug. Therefore, the detection of ^{67}Cu is detected by gamma radiation lines. The lines with E_γ = 91.266 and 93.311 keV are detected by the Si-PIN detector and line 184.577 keV – by the system of CsI(Tl) scintillator – Si-PIN-photodiode.

2. SPECTROMETER-ANALYZER FOR EXPRESS ANALYSIS OF MEDICAL RADIONUCLIDES CONCENTRATION ^{99m}Tc AND ^{67}Cu WITH REPLACEABLE DETECTION MODULES

The sealed modules of uncooled planar silicon detectors and read-out electronics developed and fabricated at the NSC KIPT showed high stability when using in high-energy physics, nuclear physics experiments, devices for monitoring the concentration of elements, and in medical diagnostic devices [9 - 11].

In Fig. 1 two types of detection modules used in the present work are presented, an uncooled silicon PIN detector (on the left) and a detection system CsI(Tl) scintillator – a silicon PIN-photodiode (on the right).

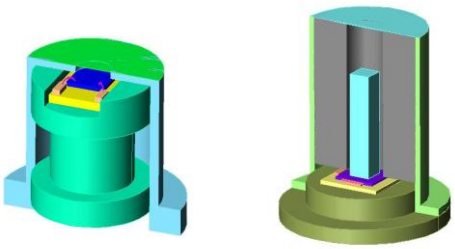

Fig. 1. Two types of detection modules: the uncooled Si-PIN detector (on the left) and the detection system CsI(Tl) scintillator – Si-PIN-photodiode (right)

A spectrometric channel based on an uncooled Si-PIN detector of 300 μm thickness provides detection of radiation in the energy range E_γ = 5...150 keV with a resolution of FWHM = 1…1.2 keV. Radiation with an energy E_γ > 50 keV is more efficiently registered with the help of detection systems consisting of a scintillator CsI(Tl) and a Si-PIN-photodiode. In [12 - 14] the efficiency of quanta registration of different energies for detecting modules based on an uncooled planar Si-detector, as well as a scintillator-photodetector system, was considered.

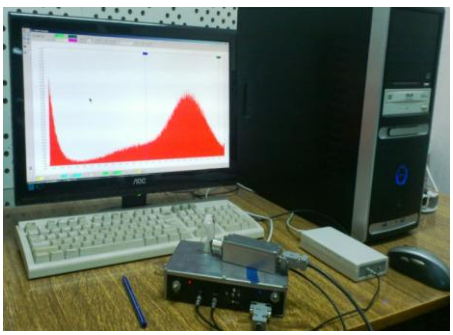

Fig. 2. Prototype of the spectrometer-analyzer for the express analysis of the ^{99m}Tc activity consists of sealed detector, preamplifier, module with a spectrometric amplifier, ADC converter and PC

In Fig. 2 spectrometer-analyzer prototype for the fast analysis of the concentration of ^{99m}Tc is shown. The monitor shows the spectrum accumulation.

Fig. 3 shows the experimental emission spectrum of a real pharmaceutical preparation ^{99m}Tc, placed in a glass ampoule with a wall thickness of ~ 1 mm, obtained by a Si-PIN-detector of 300 μm htickness.

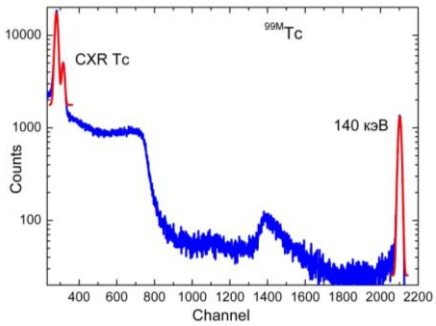

Fig. 3. Experimental emission spectrum of the real pharmaceutical preparation ^{99m}Tc, obtained by the Si – PIN-detector of 300 μm thickness (blue curve). Red curves – the fitting of CXR peaks (K_α, K_β) and 140.5 keV

The emission spectrum consists of a main line of 140.5 keV, Compton distribution (49.8 keV – edge), and two CXR lines of technetium (with energy K_α = 18.36 and K_β = 20.6 keV). K_α lines have a yield intensity per decay of 6.25%, K_β – 1.157% (Table 1). Each emission line has its own absorption coefficient in media and its registration efficiency in Si detector. The calculation in GEANT4 shows the agreement of the ^{99m}Tc activities obtained from the registration of CXR and the main line. A measurement of the technetium concentration both by the 140.5 keV line and on the CXR radiation was proposed, what significantly increases the speed of the data accumulation.

For experimental measurements the ratio of K_α / K_β was ~ 5.15…5.35 (the estimate from Table 1 gives a ratio of ~ 5.4), and the ratio of the sum of CXR quanta (K_α + K_β) exceeded the sum of the quanta at 140.5 keV peak at 15.2 times, what is close to the simulation results for the glass of 1 mm thickness.

The dependence of drug activity over the time was measured. Figs. 4, 5 shows the experimental values of drug activity measured at different times (red squares) and the estimated activity rating with allowance for decay (blue line).

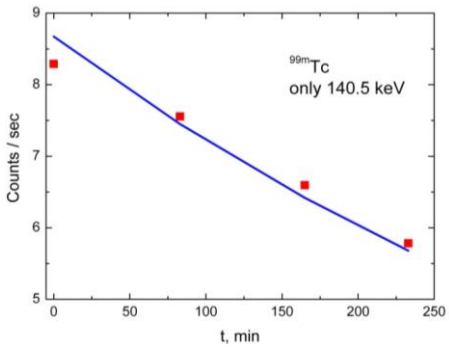

Fig. 4. Experimental values of the activity of the drug for the 140.5 keV line, measured at different times (red squares) and calculated activity with allowance for decay (blue line)

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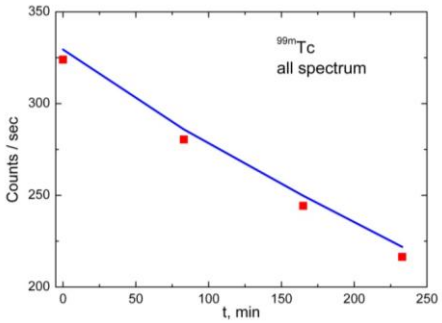

Fig. 5. Experimental values of drug activity (the entire spectrum is summed in the interval ΔE = 10…140.5 keV) measured at different times (red squares) and calculated activity with allowance for decay (blue line)

These two measurements showed the same decrease in activity 1.45 times in 233 minutes. The speed of accumulation the entire spectrum in the energy range ΔE = 10…140.5 keV exceeds the speed of dialing along the line 140.5 keV by about 37 times. Calculations in GEANT4 give approximately the same ratio of the registered integral spectrum to the baseline. This opens the possibility of calibrating the spectrometer over the integral spectrum and significantly increases the speed of the analysis.

3. CALCULATIONS OF THE INTENSITY OF THE MAIN GAMMA LINE AND X-RAY LINES IN GEANT4

Fig. 6 shows the visual representation in GEANT4 of the trajectories of gamma and X-ray quanta (green lines) for a system: ampoule with liquid (cylinder, yellow), glass (red) and detector (Si, 300 μm, blue). For simplicity, the trajectories of quanta are shown directed to the area of the detector are normal to its surface. Quanta arise randomly in the volume of a liquid.

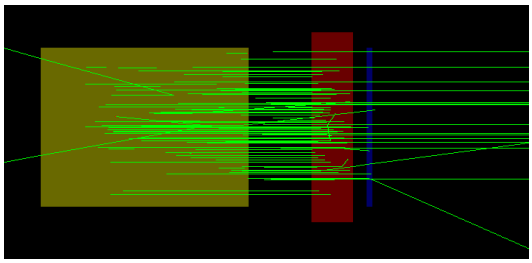

Fig. 6. Visual presentation in GEANT4 of the trajectories of gamma and X-ray quanta (green lines) for a system: ampoule with liquid (cylinder, yellow), glass (red) and detector (Si, 300 μm, blue)

A feature of the passage of low energy quanta through thin layers of matter is their strong absorption. The trajectories of quanta in Fig. 6 are rectilinear, but some of the quanta are absorbed in the glass and liquid, and the quanta passing into the Si detector are only partially registered in it. In GEANT4 the relative intensities of the main gamma and X-ray lines registered in Si, 300 μm for ^{99m}Tc for various glass ampoule thicknesses are calculated. The results are presented in Table 3.

Table 3

Energy and relative intensities of the main lines of gamma and X-ray radiation registered in Si, 300 μm for ^{99m}Tc for various glass thicknesses of the ampoule

E_γ	0.2 mm	0.5 mm	1 mm	2 mm
18.36	24.79	18.83	11.52	5.41
20.6	3.95	3.27	2.33	1.43
140.5	1	1	1	1

As one can see from the data for 1 mm of glass, the calculated increasing of the value of CXR quanta over the main peak is ~14, what is close to the experimentally measured value of ~15.2.

The results of calculations in GEANT4 of the relative intensities of the main emission lines of ^{99m}Tc for various plastic materials of the exit window with a thickness of 0.5 mm are presented in Table 4.

Table 4

Energy and relative intensities of the main lines of gamma and X-ray emission registered in Si, 300 μm for ^{99m}Tc for various materials of the output window, thickness 0.5 mm (GEANT4)

E_γ	Teflon	Plexiglass	Kapton
18.36	28.34	30.81	30.74
20.6	4.40	4.69	4.65
140.5	1	1	1

In the case of a thin plastic window, the increasing of the number of CXR quanta over the main peak is ~ 35 times.

Note that excitation of CXR Tc and Mo by the main line 140.5 keV is also possible. For the provisional thickness Tc and Mo ~ 100 μm, the yield of CXR (Kα + Kβ) in the full angle does not exceed 0.7% of the intensity of the main line for Tc and 0.6% for Mo.

The line Kβ CXR Tc with an energy of 20.6 keV may re-emit, exciting CXR Mo. This increases the possible amount of Mo CXR (K_α + K_β) up to ~ 0.62% of the intensity of the main line in solution Mo 50% / Tc 50% and thickness ~ 100 μm. One should take into account that K-lines of CXR Tc from the process of internal conversion of electrons together give a quanta yield of ~ 8.3% of the intensity of the main line of 140.5 keV. In such an estimate, the distorting effect of additional CXR of technetium and molybdenum is ~ 1.32 / 8.3 = 16%. In pure solution the increase in CXR Tc is ~ 0.7 / 8.3 = 8%.

An analysis of the experimental spectrum gives the CXR lines exactly to the energies 19.35 and 20.6 keV. The K_α peak of the Mo line of 17.47 keV would be located 13 channels to the left. Such a difference would be fixed by the detection system. In addition, the ratio K_α / K_β for Tc would change noticeably. This fact means that the amount of residual Mo is at least an order of magnitude smaller than the case considered.

4. MEASUREMENT OF THE ^{99m}Tc ACTIVITY WITH A DETECTION MODULE BASED ON THE SCINTILLATOR-PHOTODETECTOR SYSTEM

The emission lines of the sources ^{241}Am, ^{57}Co, ^{99m}Tc were measured by the detecting system of the scintillator CsI(Tl)-Si-PIN photodiode.

Fig. 7 shows the results of gamma-ray spectrum measurements of the ^{241}Am source – line with E_γ = 59.54 keV and ^{57}Co, E_γ = 122 keV. The size of the scintillator CsI(Tl) is 5×5×10 mm, Si-PIN-photodiode is 5×5×0.3 mm.

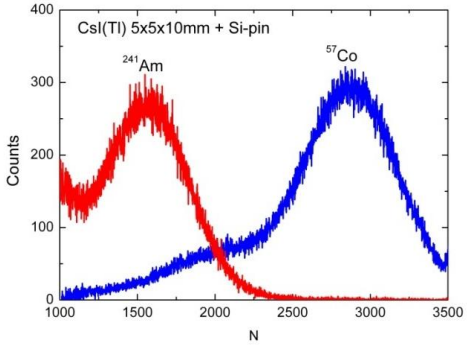

Fig. 7. Gamma spectrum of the ^{241}Am source – E_γ = 59.54 keV line and ^{57}Co, E_γ = 122 keV

Fig. 8 shows the results of gamma-ray spectrum measurements of ^{99m}Tc the 140.5 keV line. The size of the scintillator CsI(Tl) is 2×2×10 mm, Si-PIN-photodiode is 2×2×0.3 mm. Calculation of the drug activity was carried out according to the number of counts in the peak ^{99m}Tc. The processing is performed by fitting the peak of 140.5 keV.

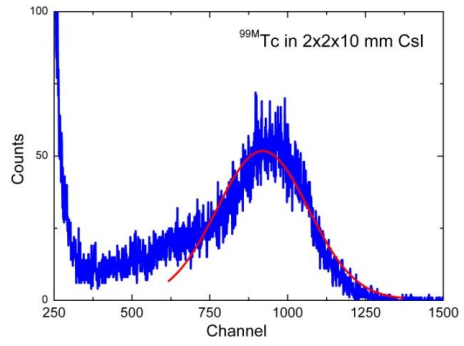

Fig. 8. The experimental gamma-ray spectrum of ^{99m}Tc, the 140.5 keV line (blue curve). The red curve is the fitting of the 140.5 keV peak in ORIGIN8

Comparative measurements of drug activity by three spectrometers were performed: by High Purity Cooled Germanium Detector (HPGe) Canberra, by Si-PIN-detector and CsI(Tl)-Si photodiode system. Taking into account the decrease in the ^{99m}Tc emission intensity with time, all the results practically coincide. For example, for 1 ml of the drug: HPGe gives the activity result 25,900,000 Bq; 300 μm Si-PIN photodiode – 25,351,000 Bq in 4π, 2×2×10 mm^3 CsI(Tl) – Si-PIN-photodiode – 25,971,000 Bq in total solid angle 4π. All the results are calculated to the end of the bombardment (EOB) time.

CONCLUSIONS

A spectrometer analyzer with replaceable detection modules was developed in NSC KIPT for the express analysis of medical radionuclides ^{99m}Tc, ^{67}Cu concentration. One of the detection modules is based on an uncooled planar Si-PIN-detector (300 μm), the second one is based on a detection system CsI(Tl) scintillator – Si-PIN-photodiode. The spectral distribution of radiation of real samples of the pharmaceutical preparation ^{99m}Tc in various experimental geometry was measured by two detection systems.

The emission spectrum consists of a main line of 140.5 keV, Compton distribution (49.8 keV – edge) and two CXR lines of technetium (with energy K_α = 18.36 and K_β = 20.6 keV).

The calculation in GEANT4 shows the agreement of the ^{99m}Tc activities obtained from the registration of CXR and the main line.

A measurement of the technetium concentration both by the 140.5 keV line and by the CXR radiation was proposed what significantly increases the speed of the data accumulation.

In the case of a thin glass or plastic window, the increasing of the value of CXR quanta over the main peak is ~ 35 times.

Comparative measurements of drug activity by three spectrometers were performed: HPGe, Si-PIN-detector and CsI(Tl) – Si-photodiode system. Taking into account the decrease in the ^{99m}Tc emission intensity with time, the results practically coincide.

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СПОСОБ ЭКСПРЕССНОГО ОПРЕДЕЛЕНИЯ КОНЦЕНТРАЦИИ МЕДИЦИНСКИХ РАДИОНУКЛИДОВ ^{99m}Tc, ^{67}Cu С ИСПОЛЬЗОВАНИЕМ СПЕКТРОМЕТРА НА ОСНОВЕ Si-ПЛАНАРНОГО ДЕТЕКТОРА

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Методика экспресс-анализа предусматривает измерение спектров излучения медицинских радионуклидов детектирующим модулем на основе неохлаждаемого кремниевого планарного детектора и модулем типа «сцинтиллятор – кремниевый фотосенсор». Разработано спектрометрическое устройство для экспресс-анализа концентрации радионуклидов и проведено его макетирование, подтверждающее работоспособность устройства. Питание спектрометра-анализатора выполнено от USB-порта ПК (в том числе, ноутбука), что обеспечивает возможность автономной работы. Проведены тестовые измерения с использованием рабочего макета спектрометра и источников рентгеновского излучения, а также исследованы спектральные распределения излучения реальных образцов ^{99м}Tc в специальной упаковке (стеклянной ампуле). Для Si-PIN-детектора толщиной 300 мкм спектр излучения состоит из линии ^{99m}Tc с энергией 140,5 кэВ и двух пиков характеристичного рентгеновского излучения технеция. Расчет в GEANT4 показывает согласие активностей ^{99m}Tc, полученных по регистрации ХРИ и основной линии. Предложено измерение концентрации технеция не только по линии 140,5 кэВ, но и по излучению ХРИ, что существенно увеличивает скорость набора данных.

СПОСІБ ЕКСПРЕСНОГО ВИЗНАЧЕННЯ КОНЦЕНТРАЦІЇ МЕДИЧНИХ РАДІОНУКЛІДІВ ^{99m}Tc, ^{67}Cu З ВИКОРИСТАННЯМ СПЕКТРОМЕТРА НА ОСНОВІ Si-ПЛАНАРНОГО ДЕТЕКТОРА

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Методика експрес-аналізу передбачає вимір спектрів випромінювання медичних радіонуклідів детектуючим модулем на основі неохолоджуваного кремнієвого планарного детектора і модулем типу «сцинтилятор − кремнієвий фотосенсор». Розроблено спектрометричний пристрій для експрес-аналізу концентрації радіонуклідів і проведено його макетування, що підтверджує працездатність пристрою. Живлення спектрометра-аналізатора виконано від USB-порту ПК (в тому числі, ноутбука), що забезпечує можливість автономної роботи. Проведено тестові вимірювання з використанням робочого макета спектрометра і джерел рентгенівського випромінювання, а також досліджені спектральні розподіли випромінювання реальних зразків ^{99m}Tc у спеціальній упаковці (скляній ампулі). Для Si-PIN-детектора товщиною 300 мкм спектр випромінювання складається з лінії ^{99m}Tc з енергією 140,5 кеВ і двох піків характеристичного рентгенівського випромінювання технеція. Розрахунок у GEANT4 показує узгодження активностей ^{99m}Tc, отриманих по реєстрації ХРВ і основної лінії. Запропоновано вимірювання концентрації технеція не тільки по лінії 140,5 кеВ, а й по випромінюванню ХРВ, що істотно збільшує швидкість набору даних.