JOINT INSTITUTE FOR NUCLEAR RESEARCH
Frank Laboratory of Neutron Physics
Group neutron activation analysis IREN

IREN (SOURCE OF RESONANCE NEUTRONS) USAGE FOR ACTIVATION ANALYSIS. AUTOMATION SYSTEM FOR GAMMA SPECTRA MEASURING.

Supervisor: PhD Andrey Yurievich Dmitriev

Student: Usmanov Temur Mamasolievich

Participation period:
November 14 – December 25

Dubna, 2021

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Introduction.

Activation analysis (radio activation analysis) - a method of qualitative and quantitative elemental analysis substances based on the activation of atomic nuclei and the study of the formed radioactive isotopes(radionuclides). The substance is irradiated with nuclear particles (thermal or fast neutrons, protons, deuterons, α-particles, etc.) or γ-quanta. Then the species is determined, i.e. serial number and mass number, the resulting radionuclides by their half-lives $T_{1/2}$ and radiation energies E, which are tabulated. Since nuclear reactions leading to the formation of certain radionuclides are usually known, it is possible establish which atoms were the original.

Activation analysis is the most common nuclear physics method determination of the composition of the substance.

Activation analysis was first carried out by D. Hevesy and G. Levy (Hungary, 1936) and A.A. Grinberg (USSR, 1940).

Basics of the method.

Neutron activation analysis is a nuclear-physical method for determining the composition of matter, based on the activation of atomic nuclei using neutrons and the study of radioactive radiation arising from the excitation of atomic nuclei.

Neutron activation analysis is a sensitive multi-element analytical method for qualitative and quantitative analysis of almost all elements. The NAA was discovered in 1936 by Hevesy and Levy, who found that samples containing certain rare earth elements became highly radioactive upon contact with a neutron source. This observation led to the use of induced radioactivity to identify elements. NAA differs significantly from other spectroscopic methods of analysis in that it is based not on electronic transitions, but on nuclear transitions. For NAA analysis, the sample is placed in a suitable irradiated object and bombarded with neutrons. This creates artificial radioisotopes of the elements present in the object. After irradiation, artificial radioactive isotopes decay with the emission of particles or, more importantly, gamma rays.

Analysis stages.

1. **Sample preparation.** Before irradiation, the samples undergo the necessary preparation stages (cleaning, washing, drying, grinding, etc.) and are packed in plastic bags or aluminum foil.

2. **Irradiation.** Samples ready for irradiation, together with standard samples and neutron flux monitors, are delivered to the irradiation position, where they are activated by neutrons.

3. **Measurement of activity.** After irradiation, the samples are repackaged in clean (not irradiated) polyethylene containers and placed on detectors to measure gamma radiation spectra. Spectral measurements of the same sample can be repeated at different time intervals.

4. **Processing of gamma radiation spectra** for studying and standard samples, as well as neutron flux monitors. Processing consists in finding peaks in the spectrum, their energy and area. The set of peaks in the spectrum is used to determine the type of radionuclide, and, consequently, the element present in the sample under study. The peak area is used to calculate the activity of the corresponding radionuclide, which is proportional to the concentration of this element.

5. **Calculation of the concentrations** of elements in the samples. Calculating can be done by absolute and relative methods. When calculating by relative method, the activities of the same radionuclides in the sample and the standard are compared and, taking into account the known values of the concentrations of elements in the standard, the concentrations of elements in the sample are calculated. The neutron flux gradient is taken into account based on the data from the spectra of the flux monitors.

6. If necessary, **statistical processing** of the measurement results is carried out.

The activity of the resulting isotope changes during irradiation, storage, and measurement as:

$$A = \frac{S\, t_{meas\,real}\, \lambda\, e^{\lambda t_d}}{m_s\, \varepsilon\, \gamma\, t_{meas\,live}\, U_f \left(1 - e^{-\lambda t_{irr}}\right)\left(1 - e^{-\lambda t_{meas\,real}}\right)} \quad (1)$$

in which t_d – the decay time, s; t_{irr} – the irradiation time, s; $t_{meas\ real}$ – the real measurement time, s; $\lambda = \frac{\ln 2}{T_{1/2}}$ – the decay constant, s^{-1}, in which $T_{1/2}$ – the half-life, s; m_s – the sample mass, g; ε – the efficiency of the detector; γ – the yield of the i-th gamma line; S is the total absorption peak area for the line with energy $E_{\gamma i}$ in the measured gamma-ray spectrum, the number of counts; U_f – the factor to convert the activity from the number of decays per time unit to μCi, U_f = 37000; $t_{meas\ live}$ – the live measurement time, s.

From the NAA theory it is well known that the mass fraction *C*, *mg/kg*, of the required element in the sample can be calculated by absolute method by the formula:

$$C = \frac{S(E_{\gamma i})\, M\, \lambda\, e^{\lambda t_d}}{m_s\, N_A\, \varepsilon(E_{\gamma i})\, \gamma(E_{\gamma i})\, \theta \left(\sigma_{th}\, \Phi_{th} + I_{res}\, \Phi_{res}\right)\left(1 - e^{-\lambda t_{irr}}\right)\left(1 - e^{-\lambda t_{meas\,real}}\right)} \quad (2)$$

in which M – the molar mass of the desired element, g/mol; N_A – the Avogadro's number, mol^{-1}; $\varepsilon(E_\gamma)$ – the efficiency of the detector; $\gamma(E_{\gamma i})$ – the yield of the i-th gamma line; θ – the isotopic abundance in the natural mixture; σ_{th} – the capture cross section for thermal neutrons, barn; Φ_{th} – the thermal neutron flux, $(cm^2\ s)^{-1}$; I_{res} – the resonance integral, barn; Φ_{res} – the resonance neutrons flux at the energy 1 eV, $(cm^2\ s)^{-1}$.

Gamma activation analysis.

Activation by high-energy gamma quanta has a number of advantages over activation by thermal neutrons. This is the high representativeness of the analysis due to the possibility of analyzing samples of large portions, the absence of the self-shielding effect, high productivity and rapidity of determinations, since during activation, nuclides with a short half-life are mainly formed, and increased selectivity due to the threshold nature of photonuclear reactions.

Most often, for activation, radiation with energy of γ-quanta up to 30 MeV is used. The value of the threshold energy of quanta, at which a nuclear reaction occurs, depends on the element. Rock-forming elements have a higher activation threshold than most non-ferrous, rare and noble metals of interest to practice, which allows due to rational the choice of the energy of γ-quanta significantly reduce the influence of the matrix.

Electron accelerators of various types are usually used as sources of activating radiation: betatrons, microtrons, linear accelerators; for activation, bremsstrahlung radiation arising from the bombardment of heavy metal targets with electrons is used [1].

IREN (source of resonance neutrons).

The Joint Institute for Nuclear Research, the Laboratory of Neutron Physics created a Resonance Neutron Source (IREN) – a basic facility for solving problems of fundamental and applied nuclear physics.

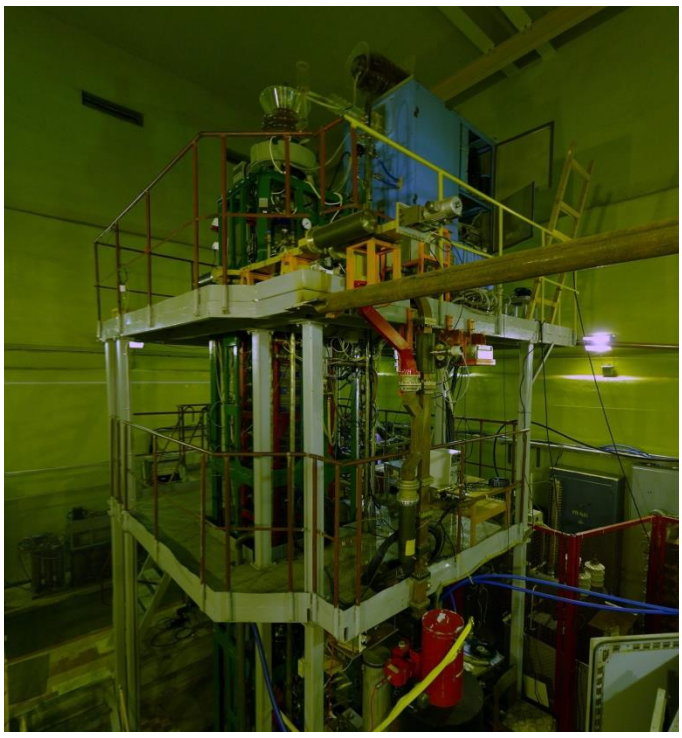

Fig 1. Source of resonance neutrons (IREN).

The full-scale research complex IREN includes a linear electron accelerator at energies up to 80 MeV with a beam power of 0.5 kW, target made of tungsten, a beam infrastructure with measuring pavilions, as well as technological, control, protective and auxiliary ones systems. According to its characteristics, the full-scale IREN complex will put this installation on a par with the best neutron sources of this class (GELINA, Belgium and ORELA, USA).

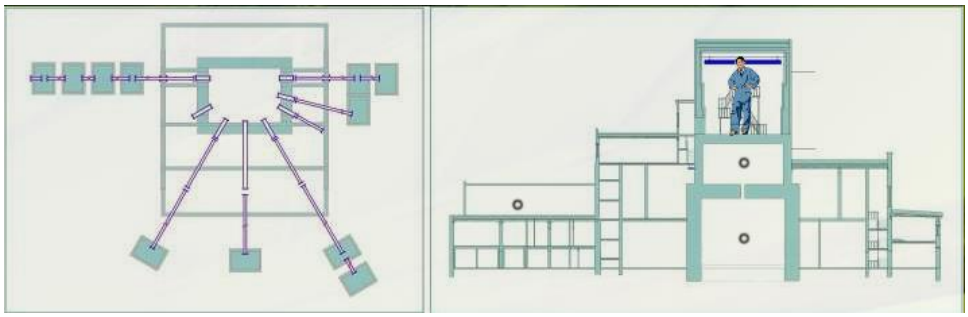

Fig 2. Scheme of the IREN facility.

The IREN research facility is was created for such type of experiments [2].
Fundamental nuclear physics:

- search for violation of time invariance with polarized resonance neutrons and polarized target;
- study of parity violation in neutron induced reactions (fission, (n,α), (n,p), (n,γ));
- electromagnetic structure of the neutron;
- quantum aspects of neutron induced fission;

- phase transitions in excited nucleus - chaos and order in quantum systems.

Applied nuclear physics:

- study of elemental composition by neutron activation, prompt gamma and gamma activation analysis;
- nuclear data for astrophysics;
- nuclear data for technology;
- isotope analysis with resonance neutrons.

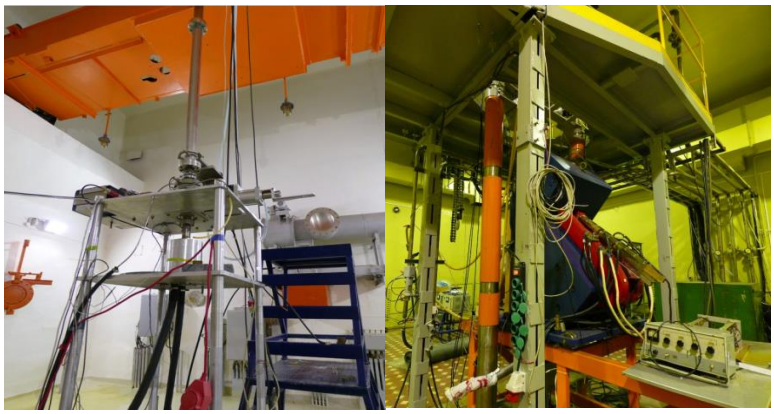

Fig 3. Source of resonance neutrons (IREN).

Tab 1. Parameters of the IREN.

Maximum current (A)	3
Repetition rate (Hz)	50
Electronic pulse duration (ns)	100
Electron energy (MeV)	80
Beam power (kW)	0,4
Reproduction factor	1
Neutron yield (n/s)	10^{11}

Determination of the neutron fluxes at different operating modes of the IREN.

The IREN facility is an intense pulsed source of resonance neutrons, which is a combination of a linear electron accelerator and a target made of tungsten.

Since the flux of neutrons emitted from the target is non-uniform, it is necessary to estimate at what height of the target the neutron flux is the highest. To determine the fluxes, flux monitors with well-known capture cross sections and resonance integrals are irradiated. Copper is suitable as such an element. It is possible to separate thermal and resonance neutrons using cadmium, since it has a large thermal neutron capture cross section. Five pairs of monitors Cu (in a cadmium

protection) – Cu (without a protection) were used. The experiment was carried out twice: first, the facility operated at a pulse frequency of 25 Hz (beam current - 1.7 A, neutron yield - 1810000), then - 50 Hz (beam current - 2.2 A, neutron yield - 4000000) with the following characteristics of the facility: pulse duration - 10 ns, power - 500 W; electron energy - 80 MeV.

The fluxes were calculated using the 1345 keV copper line using the formulas:

$$\Phi_{res} = \frac{S_{\gamma 2} \cdot M \cdot \lambda \cdot e^{\lambda t_{d2}}}{m_2 \cdot N_A \cdot \gamma \cdot \varepsilon \cdot \theta \cdot I_{res} \left(1 - e^{-\lambda t_{irr}}\right) \cdot \left(1 - e^{-\lambda t_{meas}}\right)} \quad (3)$$

$$\Phi_{th} = \frac{x - \Phi_{res} \cdot I_{res}}{\sigma_{th}} = \frac{r}{\sigma_{th}} \left(S_{\gamma 1} \cdot \frac{e^{\lambda t_{d1}}}{m_1} - S_{\gamma 2} \cdot \frac{e^{\lambda t_{d2}}}{m_2} \right) \quad (4)$$

$$r = \frac{M \cdot \lambda}{N_A \cdot \gamma \cdot \varepsilon \cdot \theta \cdot \left(1 - e^{-\lambda t_{irr}}\right) \cdot \left(1 - e^{-\lambda t_{meas}}\right)} \quad (5)$$

in which $\boldsymbol{\Phi_{th}}$ – the thermal neutron flux, $(cm^2\ s)^{-1}$; $\boldsymbol{\Phi_{res}}$ – the resonance neutrons flux at the energy 1 eV, $(cm^2\ s)^{-1}$.

Before irradiation of the flux monitors, the following sample preparation was carried out: the surfaces of the copper and cadmium samples were cleaned with alcohol and dried in air. Further, the pairs of Cu without a cadmium ptotection - Cu in a cadmium protection were compiled. The samples were sealed in a polyethylene film and placedat a distance of 1, 3, 5, 7, and 9 cm from the upper edge of the moderator.(Fig. 4).

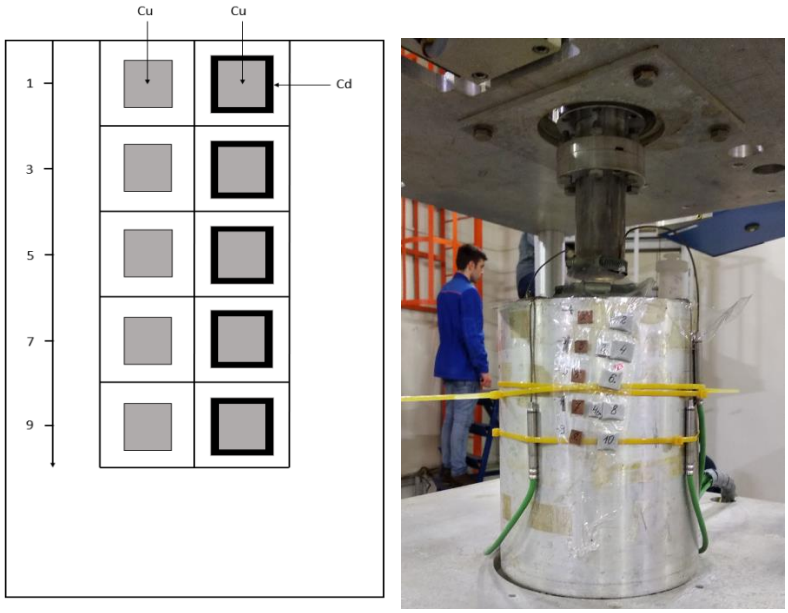

Fig. 4. Scheme of the experiment (left); real view (right).

The samples were irradiated at a frequency of 25 Hz for about 14 hours, at a frequency of 50 Hz for about 5 hours. For the collection of spectrometric data, an automatic system for measuring the spectra of induced activity, developed at the FLNP JINR, was used [3], [4]. The spectra were measured with a Canberra GC4018 HPGe detector with a resolution of 2.1 keV for the ^{60}Co gamma line with an energy of 1332.5 keV. The spectra were processed using the GENIE-2000 program.

Tab. 2 Fluxes of thermal and resonance neutrons at 25 and 50 Hz.

frequency, Hz	h, sm	mass of Cu (g)	mass of Cu in Cd-protection (g)	$\Phi_{thermal}$, 10^7(n/(sm^2·s))	$\Phi_{resonance}$,10^7(n/(sm^2·s))
25	1	0.340	0.340	2.57 ± 0.077	1.00 ± 0.032
	3	0.326	0.340	5.69 ± 0.16	1.32 ± 0.041
	5	0.336	0.339	7.46 ± 0.21	1.29 ± 0.040
	7	0.343	0.350	6.64 ± 0.19	1.25 ± 0.039
	9	0.338	0.345	5.30 ± 0.16	1.12 ± 0.036
50	1	0.236	0.220	7.17 ± 0.22	2.42 ± 0.077
	3	0.228	0.246	17.2 ± 0.47	3.14 ± 0.097
	5	0.240	0.252	20.6 ± 0.55	3.57 ± 0.11
	7	0.233	0.234	19.6 ± 0.55	3.34 ± 0.10
	9	0.238	0.235	17.8 ± 0.94	3.04 ± 0.097

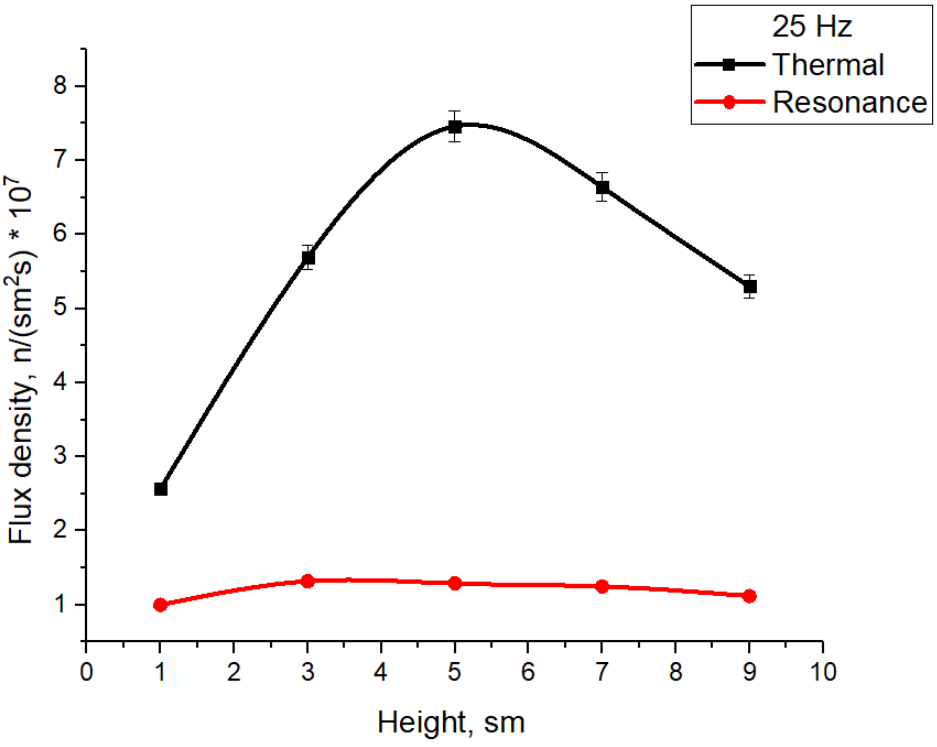

Fig.5. Fluxes of thermal and resonance neutrons depending on the height of the target at 25 Hz.

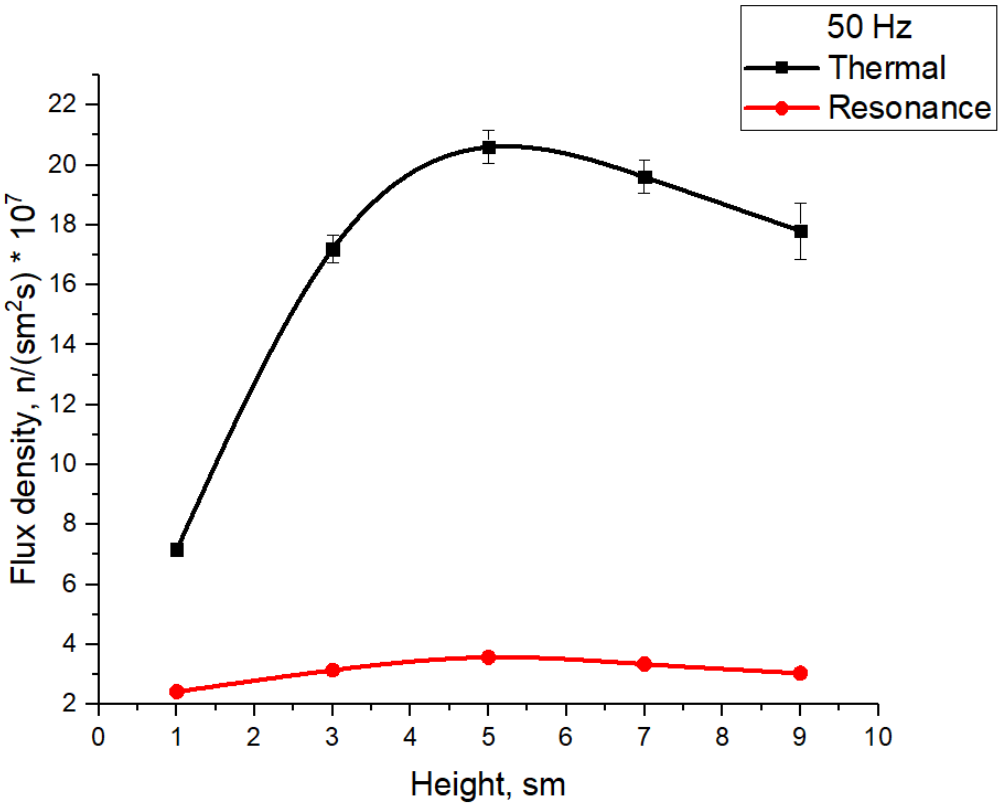

Fig 6. Fluxes of thermal and resonance neutrons depending on the height of the target at 50 Hz.

The masses of the monitors used and the received fluxes at a frequency of 25 and 50 Hz are presented in Table2 and Fig. 5, Fig. 6.

Experimental methods for determining the electron flux at the IREN facility.

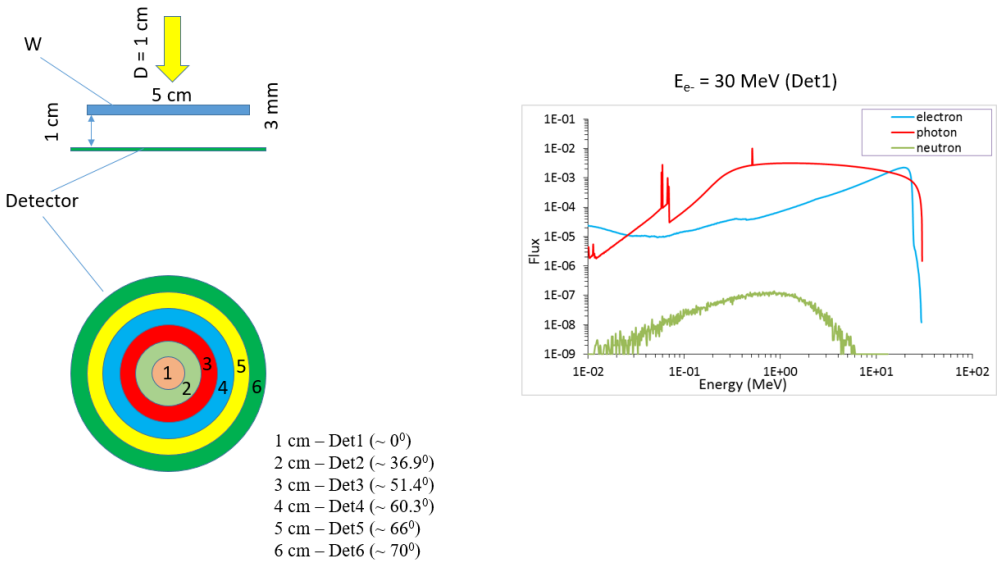

Fig 7. Schematic diagrams for determining the electron flux. Zh. Khushvaktov's experiment.

Tab 3. The number of secondary neutrons per electron incident on a tungsten converter.

E(MeV)	Det1 (0,79 sm^2)	Det2 (2,36 sm^2)	Det3 (3,93 sm^2)	Det4 (5,50 sm^2)	Det5 (7,07 sm^2)	Det6 (8,64 sm^2)	Общее (4π)
30	2,31E-5	5,12E-5	5,24E-5	4,39E-5	3,44E-5	2,71E-5	6,37E-4
40	3,04E-5	6,64E-5	6,86E-5	5,68E-5	4,48E-5	3,52E-5	8,25E-4
50	3,51E-5	7,73E-5	7,99E-5	6,57E-5	5,15E-5	4,07E-5	9,53E-4
60	3,96E-5	8,60E-5	8,79E-5	7,27E-5	5,70E-5	4,50E-5	1,06E-3

Tab 3. The number of bremsstrahlung photons per electron incident on a tungsten converter[5].

E(MeV)	Det1 (0,79 sm^2)	Det2 (2,36 sm^2)	Det3 (3,93 sm^2)	Det4 (5,50 sm^2)	Det5 (7,07 sm^2)	Det6 (8,64 sm^2)	Общее (4π)
30	1,26	1,30	0,63	0,33	0,197	0,128	1,97
40	1,74	1,50	0,59	0,29	0,167	0,108	2,27
50	2,16	1,58	0,51	0,24	0,141	0,091	2,48
60	2,53	1,60	0,45	0,21	0,125	0,082	2,68

Tab 4. Neutron flux in the IREN facility.

I (A)	t (s)	e (Cl)	E	n		k	k(4π)	S(sm2)	Flux (n/sm2*s)	Flux(4π)
			30 MeV				6.37E-04			1.19E+09
					Det1(~ 0)	2.31E-05		0.79	54825949.4	
					Det2(~ 36.9)	5.12E-05		2.36	40677966.1	
3	1.00E-07	1.60E-19		1.88E+12	Det3(~ 51.4)	5.24E-05		3.93	25000000.0	
					Det4(~ 60.3)	4.39E-05		5.5	14965909.1	
					Det5(~ 66)	3.44E-05		7.07	9123055.2	
					Det6 (~ 70)	2.71E-05		8.64	5881076.4	
3		1.60E-19	**40 MeV**				8.25E-04			1.55E+09
					Det1(~ 0)	3.04E-05		0.79	72151898.7	
					Det2(~ 36.9)	6.64E-05		2.36	52754237.3	
					Det3(~ 51.4)	6.86E-05		3.93	32729007.6	
					Det4(~ 60.3)	5.68E-05		5.5	19363636.4	
					Det5(~ 66)	4.48E-05		7.07	11881188.1	
					Det6 (~ 70)	3.52E-05		8.64	7638888.9	
3		1.60E-19	**50 MeV**				9.53E-04			1.79E+09
					Det1(~ 0)	3.51E-05		0.79	83306962.0	
					Det2(~ 36.9)	7.73E-05		2.36	61414194.9	
					Det3(~ 51.4)	7.99E-05		3.93	38120229.0	
					Det4(~ 60.3)	6.57E-05		5.5	22397727.3	
					Det5(~ 66)	5.15E-05		7.07	13658062.2	
					Det6 (~ 70)	4.07E-05		8.64	8832465.3	
3		1.60E-19	**60 MeV**				1.06E-03			1.99E+09
					Det1(~ 0)	3.96E-05		0.79	93987341.8	
					Det2(~ 36.9)	8.60E-05		2.36	68326271.2	
					Det3(~ 51.4)	8.79E-05		3.93	41937022.9	
					Det4(~ 60.3)	7.27E-05		5.5	24784090.9	
					Det5(~ 66)	5.70E-05		7.07	15116690.2	
					Det6 (~ 70)	4.05E-05		8.64	8789062.5	

Automatic system for measuring spectra of irradiated samples.

The hardware and software complex [3], [4], [6], [7] developed for the mass multi-element NAA at FLNP JINR uses an automatic system for measuring the spectra of induced activity, which operates with the simultaneous use of up to four original sample changers (SCh), serving the corresponding number of high-purity germanium detectors.

Each SCh consists of a two-axis linear module and a disc with 45 cells for containers with samples.

The linear module is fixed with aluminum profiles over two metal tables with adjustable legs. A rotating disk with samples is installed on one of the tables, and a Dewar flask with a detector is under the other. The detector head goes through a hole in the table top and sits above the table top. On the surfaces of both tables, there are wells made of lead blocks designed to provide biological protection.

The sample disk is driven by a stepper motor. On one side of the motor, the shaft, which is fixed in the bearing, is connected to the disk shaft by means of an adapter coupling. On the other hand, an incremental encoder is installed on it, with the help of which the selection of a cell with a sample is controlled. The initial cell of the disc is fixed using an electromagnetic sensor.

Fig 8. General view of automatic sample changer.

Moving containers from the disk to the detector and back along the horizontal and vertical axes is carried out using a linear movement module. Each axis is equipped with an incremental encoder for determining the position of the carriage, as well as two end and one reference encoders. The containers are gripped with a spring-loaded mechanical device. The exchange of information between the control software "Measurements" and linear movement modules and/or electric motors of the disk with samples is carried out using controllers.

Spectrometers.

After irradiation, the samples are repackaged in clean polyethylene containers. And they are placed on the detector, where the spectrum of γ-radiation is measured.

The spectrometers use CANBERRA detectors with the following characteristics:

1. Relative efficiency 100%, resolution 2.1 keV for ^{60}Co energy 1332 keV.
2. Relative efficiency 70%, resolution 2.3 keV for ^{60}Co energy 1332 keV.
3. Relative efficiency 40%, resolution 1.8 keV for ^{60}Co energy 1332 keV.

All spectrometric electronics are also made by CANBERRA. Spectrometers are based on digital processor DSA-LX, LYNX and DSA-1000,

Since the final step in the work of the Genie-2000 program is the activity of isotopes in the samples under study, the additional software "Concentration" was developed to calculate the concentrations of elements by the relative NAA method [8], [9].

Literature and links.

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