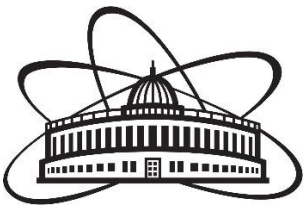

JOINT INSTITUTE FOR NUCLEAR RESEARCH
Frank Laboratory of Neutron Physics

FINAL REPORT ON THE START PROGRAMME

Characteristics of ionizing radiation fields near the surface of the water moderator during the shutdown of the IBR-2 reactor

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Abstract

The technical shutdown of the IBR-2 reactor caused the need to clarify the characteristics of ionizing radiation fields of the irradiation facility of channel №3. Low fluxes and chronic exposures (~10^6 s) make it possible to study the stochastic effects of radiation exposure on biological systems. In this paper, we consider the physical foundations of the methods used and the results of measurements of neutrons and gamma rays near the surface of the water moderator of the IBR-2 reactor.

Introduction

Ionizing radiation detectors are used to find ionization effects or excitation of atoms that result from the recorded radiation getting into the sensitive volume of the device. When radiation goes through the working substance, it partially or completely loses its energy in various processes of interaction with electrons and atomic nuclei. Afterwards the energy absorbed by the detector transforms into an electrical impulse that is recorded by read out electronic.

The type of detector that will used in the measurement is mainly determined by the type of recorded radiation and the task at hand (particle counts or determination of its energy, arrival time, coordinates of a hit in the detector). The most commonly used types of detectors are bubble chamber, gas counters, photographic emulsion, scintillation and semiconductor detectors. Particles with an electric charge, when going through the working substance, expend their energy due to ionization and excitation of atoms, bremsstrahlung and Cherenkov radiation. Gamma quanta and neutrons are non-ionizing radiation since they do not have an electric charge and are detected through secondary particles that result from their interaction with the nuclei of the detector. Interaction of photons with the matter results in production of an electron through photo effect, Compton scattering by the atomic electron or in production of an electron-positron pair. The study of neutron radiation is of particular interest, because the nature of the interaction of neutrons with substance strongly depends on the magnitude of their kinetic energy, and varies irregularly from the nuclear composition of the target.

Neutrons with kinetic energy greater than 100 keV are called fast, ones with kinetic energy less than 100 keV are called slow. Slow neutrons are divided into intermediate neutrons (with energies ranging from 1 to 100 keV), resonance neutrons (0.5 – 1 keV), thermal neutrons (0.005 – 0.5 eV), cold neutrons (10^{-7} – 0.005 eV) and ultracold neutrons (<10^{-7} eV). Most experiments on the study of the radiation resistance of materials work precisely with fast neutrons due to their high ability of defect formation in the condensed matter. The wavelength of thermal (λ = 0.1–1 nm) and cold (λ = 1–4 nm) neutrons used in structural research and their energy (En = 0.1–100 meV) correspond to common interatomic distances in crystal lattice and liquids and characteristic excitation energies. Thus, the same source of neutrons allows us to study both the structure and dynamics of a substance.

Since neutrons have no charge, they don't interact with electron shell of atoms, interacting only with nuclei. Unlike protons, which cannot interact effectively with the nucleus at low energies due to the Coulomb barrier, neutrons, even at low

energies, are able to approach the nucleus at a distance of the order of the radius of action of nuclear forces. Because of this neutron radiation is widely used in the studies of the characteristics of materials, in particular, the properties of condensed matter are studied using neutronografic methods. The results of these studies are of paramount importance in solving complex scientific problems about the structure of matter, its magnetic properties in materials studies and biology, for determining the properties of nanomaterials [1]. Determination of the flux density is carried out by indirect methods due to their large value of it.

Neutron activation analysis

Neutron activation analysis (NAA) is a method for determining the elemental composition of a sample and the densities of neutron fluxes of corresponding energies, based on measuring the characteristics of radioactive nuclei excited by neutron capture.

The main reaction of the interaction of neutrons with the substance under study, which results in the formation of unstable nuclei, is a radiation capture:

$$n + {}^{A}_{Z}X \rightarrow {}^{A+1}_{Z}X + \gamma$$

After the neutron is captured by the nucleus, it goes into an excited state, which is removed in a short time by emitting a gamma quantum. The ground state of the formed ${}^{A+1}_{Z}X$ nucleus may also be unstable, then β^- or β^+ decay will occur during a characteristic time. If the decay occurs to the excited state of the finite nucleus, then characteristic gamma quanta are emitted. These gamma quanta are detected by a detector with a good energy resolution and acceptable efficiency, their energy and activity are determined. According to these data and the radiative capture cross-section known from the literature, the mass of the element ${}^{A}_{Z}X$ contained in the sample is determined.

The main contribution to the formation of radioactive nuclei is made by thermal and resonance neutrons, since the neutron capture cross-section varies according to the law 1/υ.

The formula by which one can calculate the mass of the desired element in the sample is as follows:

$$m_x = \frac{N_\gamma(E_\gamma) \cdot M \cdot \lambda \cdot e^{\lambda t_d}}{N_A \gamma \varepsilon(E_\gamma) \theta (\sigma_{th}\Phi_{th} + \mathrm{I}_{res}\Phi_{res})[1 - \exp(-\lambda t_{irr})][1 - \exp(-\lambda t_{meas})]}, \quad (1)$$

where λ is the decay constant, M is atomic weight of the desired element, θ is the isotope abundance in natural mix, $N_\gamma(E_\gamma)$ is the area of the measured line, γ is the probability of emitting this line, $\varepsilon(E_\gamma)$ is efficiency of the detector, t_{irr} is irradiation time, t_d is the time after irradiation before the measurement begins, t_{meas} is the time of measurement, Φ_{th} is the thermal neutron flux, σ_{th} is the thermal neutron capture cross-section, $\mathrm{I}_{res} = \int_{E_{Cd}}^{\infty} \frac{\sigma(E)}{E} dE$ is the resonance integral, i.e. value characterizing interaction between resonance neutrons and the target nuclei, Φ_{res} is the density of the thermal neutron flux at an energy of 1 eV.

To quantify the abundance of unknown elements, it is necessary to determine the fluxes of thermal and resonance neutrons. To do this, together with the sample, indicators (in our work, this is gold) are irradiated, i.e. elements with well-known cross-sections. Indicators of the same type are irradiated in a Cd shell (since it is an excellent thermal neutron absorber) and without it under the same conditions.

Then the flows are calculated using the following two formulas:

$$\Phi_{res} = \frac{N_{\gamma 2} \cdot M \cdot \lambda \cdot e^{\lambda t_{d2}}}{m_2 \cdot N_A \cdot \gamma \cdot \varepsilon \cdot \theta \cdot I_{res} [1 - \exp(-\lambda t_{irr})][1 - \exp(-\lambda t_{meas})]}, \quad (2)$$

$$\Phi_{th} = \frac{X - \mathrm{I}_{res} \Phi_{res}}{\sigma_{th}} = \frac{r}{\sigma_{th}} \left(N_{\gamma 1} \frac{\exp(\lambda t_{d1})}{m_1} - N_{\gamma 2} \frac{\exp(\lambda t_{d2})}{m_2} \right), \quad (3)$$

where m_2 is the mass of the indicator irradiated with the Cd shell and m_1 is the mass of the indicator without the Cd shell; common factor $r = \frac{M \cdot \lambda}{N_A \cdot \gamma \cdot \varepsilon \cdot \theta [1 - \exp(-\lambda t_{irr})][1 - \exp(-\lambda t_{meas})]}$.

Thus, we can measure the thermal and resonance neutron fluxes using indicators and a filter.

Semiconductor detector

The operation of a semiconductor detector is similar to the operation of an ionization chamber, but in comparison with it, this type of detectors has a number of advantages [2]. Firstly, the energy that needs to be expended by the flying particle to form an electron-hole pair in a semiconductor detector is significantly less than in a gas-filled detector. Secondly, the solid-state working volume provides a higher braking capacity, which makes it possible to register high-energy and weakly-ionizing particles. Thirdly, electrons and holes have comparable mobility, which makes it possible to exclude the dependence of the amplitude of the output signal on the place of passage of the particle.

In a semiconductor detector, a sensitive region is created in which there are no free charge carriers. Once in this region, the ionizing particle transfers part of its energy to the electrons, after which the latter move from the valence band of the semiconductor crystal to the conduction band. Under the influence of voltage applied to the electrodes, electrons and holes begin to move along the field to the anode and cathode, respectively. The number of electron-hole pairs generated is proportional to the deposited energy. The movement of the charge, according to the Shockley–Ramo theorem [3], forms a current pulse in the reading electrodes, which in turn is proportional to the moving charge.

Silicon or germanium is most often used as a working substance in such detectors. Germanium detectors are more difficult to operate, but they have greater recording efficiency: the thickness of the sensitive zone of the detector should contain the mean free path of the detected particles, while it depends on the density of the medium and its atomic number, the values of which are higher for germanium (the density of silicon is 2.33 g/cm^3, atomic number is 14; the density of germanium is 5.32 g/cm^3, atomic number is 32). The gamma-ray spectrometers most commonly

used in laboratory measurements are made of germanium, since the cross-section of the interaction of the gamma quantum with the medium, depending on the type of interaction, is proportional to Z^5 (photoelectric effect), Z (Compton effect), or Z^2 (pair generation). A necessary condition for the operation of a germanium detector is its cooling to cryogenic temperatures, otherwise the thermal current will create too much noise, which will significantly reduce the resolution of the detector. This is primarily due to the small size of the band gap ($g = 0.67\,eV$), because the probability of an electron passing into the conduction band depends on the temperature as follows: $p \sim e^{-g/kT}$. Cooling to the temperature of liquid nitrogen reduces the thermal excitation of valence electrons, so that only interaction with gamma radiation can give the electron the energy necessary to cross the band gap and reach the conduction band. To maintain such low temperatures, a cryostat is used..

In this work, a coaxial HPGe detector of the GC10021 model from Canberra was used (see Fig.1 and Fig.2) [4]. The energy range of the detected gamma radiation for such an installation is from 40 keV to 10 MeV, and the relative recording efficiency is 100%.

The detection unit consists of the following main components:

− The detector itself, which is a crystal made of high purity germanium (HpGe)

− A cryostat with a cooling system

− A preamplifier

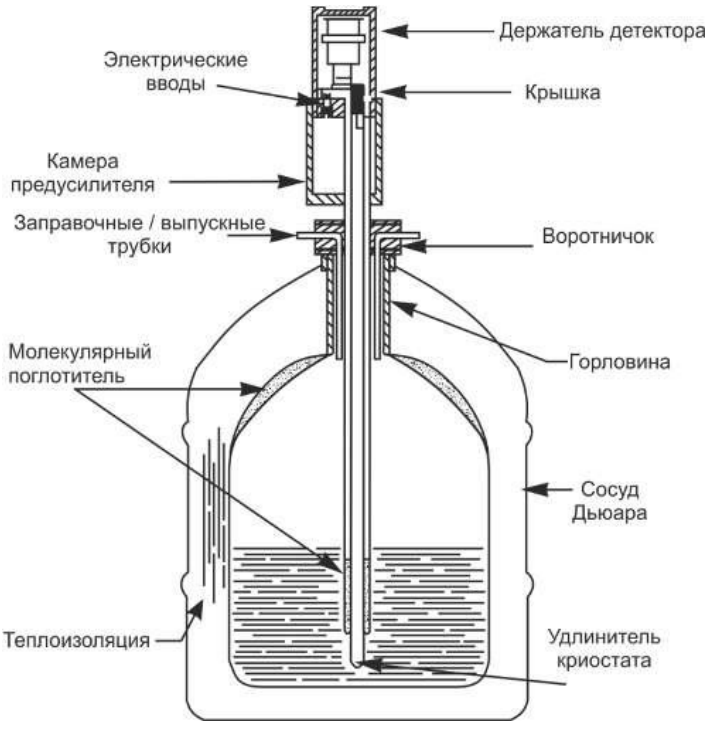

Figure 1. Appearance and design of detection units in a vertical submersible cryostat with nitrogen cooling.

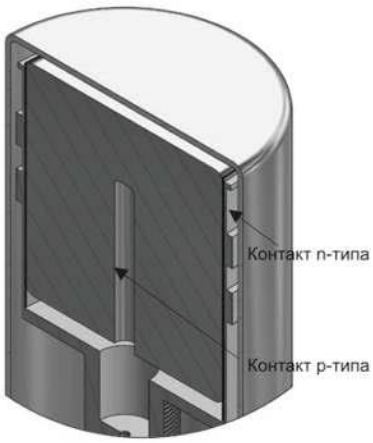

Figure 2. Section of the detector

The detector is a semiconductor diode having a P-I-N structure, where I is a depletion region, sensitive to ionizing radiation, P and N are electrodes collecting the formed charge carriers. When the reverse bias voltage is applied to the diode, the

electric field expands the depletion region for almost the entire volume of the crystal. A charge-sensitive preamplifier converts the collected charge into a voltage pulse. The charge accumulates on the feedback capacitor, and the voltage on it increases proportionally. After the pulse is completed, the capacitor is discharged through a feedback resistor, and the voltage on it decreases until the next current pulse in the detector. The voltage pulse, the amplitude of which is proportional to the energy of the gamma quantum, is amplified and transmitted to the pulse amplitude analyzer, where the distribution of the number of pulses by their amplitudes is constructed. These distributions are then transmitted to the control computer for further processing.

The obtained spectra are processed in GENIE-2000 software [5, 6], which allows searching for peaks and determining their area. Next, the analysis of the obtained data is performed, which consists in the identification of radionuclides by half-lives, energies and outputs of gamma lines present in the obtained spectrum. After identifying the peaks, the program automatically calculates the activity of nuclides, taking into account the calibration for energy and efficiency under the specified measurement conditions.

Radiachromic dosimetry

In addition to neutrons, the IBR-2 reactor is a source of gamma quanta. Due to the fact that the energy of the emitted gamma quanta is small for gamma-ray activation analysis, the absorbed doses from the passed gamma quantum flux are determined. The radiachromic dosimetry method is used to determine absorbed doses of gamma radiation. This method is based on chemical processes occurring in the active layer of the film under the action of ionizing radiation, leading to irreversible changes in the color of the dosimeter film, the depth of which depends on the amount of absorbed dose.

The value whose change characterizes the absorbed dose is the optical density (OD) – a measure of the opacity of a substance for incident light, defined as the decimal logarithm of the ratio of the intensity of incident light to the intensity of the transmitted light $OD = \ln I_0/I$.

Dosimeters are read, that is, their OD is determined using a photometer or a spectrophotometer. To convert the readings of the change in the optical density of the film into the absolute absorbed dose, a calibration line is required. In this work, a two-wave photometer FWT-92 was used (see Fig. 3), consisting of a miniature incandescent lamp, narrow band three-cavity band pass filters, and a sensitive silicon photovoltaic cell to measure the OD. The device can read the film at two wavelengths of 510 nm and 600 nm to cover the range of radiachromic dosimeters. The range of readings is from 0.001 OD to 3.0 OD [7].

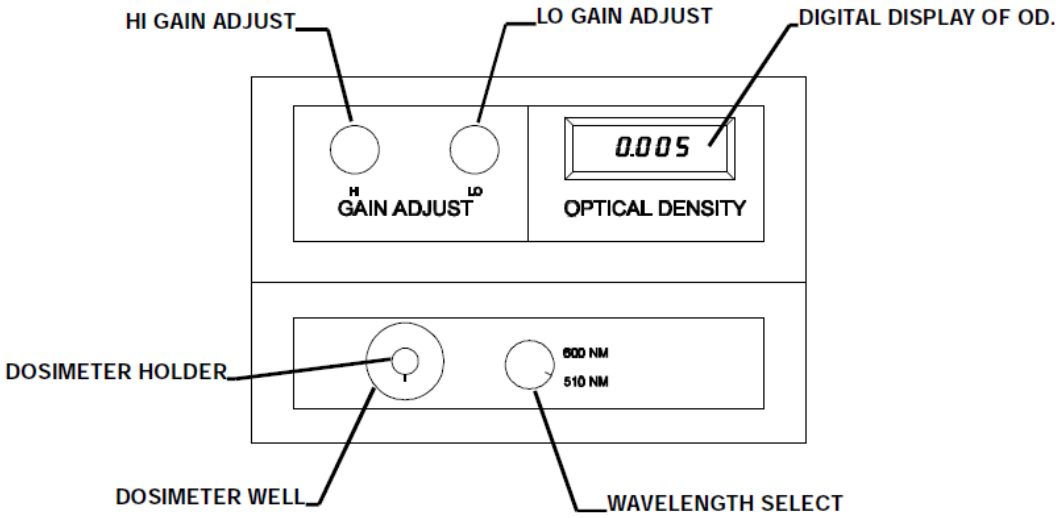

Figure 3. The appearance and basic controls of the FWT-92 photometer.

The results were taken from radiachromic thin-film dosimeters of the FWT-60 series, which, under the influence of gamma quanta with a wavelength of less than 350 nm, change from a colorless to a colored state. The dosimeter contains in the nylon matrix the leuco-dye hexa(hydroxyethyl) paraosaniline nitrile [8]. The linear dimensions of such a film are 1 cm × 1 cm and a thickness of about 50 microns. Important advantages of these dosimeters are the linear relationship between the optical density and the absorbed dose in a wide 8 dose range, insignificant influence on changes in environmental conditions and ease of handling even after irradiation.

Experimental setup

The irradiation facility designed to study the radiation resistance of materials is located in the zone of channel No. 3 of the IBR-2 pulse reactor and consists of two parts: a transport I-beam and a massive cylinder (see Fig. 4).

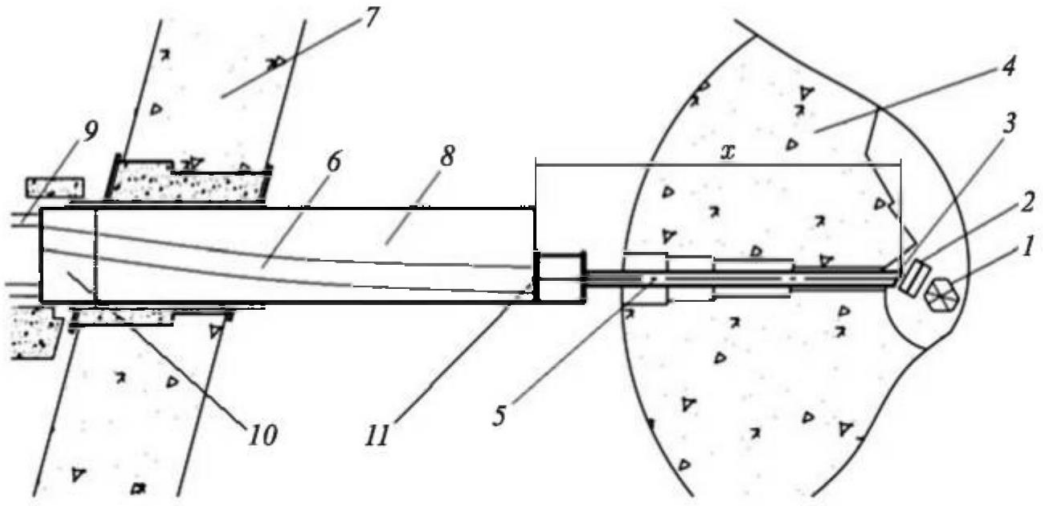

Figure 4. Experimental setup for radiation research on the IBR-2 beam No. 3 [9]: 1 — IBR-2 core; 2 —Water moderator – WM-303; 3 — end of the straight section (I-beam) for the installation of test samples; 4 — first biological shield; 5 — straight section (x = 3 m); 6 — curved section (pipe 4,87 meters long); 7 — second biological shield; 8 — steel cylinder with water; 9 — rail track; 10 — gate; 11 — initial point of the curved pipe.

The transport I-beam (5) is located closest to the surface of the water moderator near the core from the side of the reactor beam No. 3 and is a supporting

structure in the form of a beam made of AMg6 alloy (contains a minimum amount of impurities, and its gamma activity quickly decreases after irradiation) with a length of 3 m. The pipe of the curved section (6) with an internal diameter of 160 mm has a length of 4.8 m and a radius of curvature of 25 m. The pipe is located inside a cylinder (8) with an external diameter of 800 mm, filled with water, which plays the role of biological protection. The end of the cylinder is equipped with a special gate (10) with a thickness of about 1 m, which covers the pipe and is used to protect against direct neutron beam and gamma radiation during irradiation of samples. When the reactor is not working, the gate opens and there is access to the curved pipe for installing of samples.

The closest (initial) point of the pipe (11) to the core, when the installation is located at the minimum possible distance from the WM-303, equal to 40 mm, is at a distance of about 3 m, the farthest is at 7.8 m. The samples are placed in metal containers and attached to the transport I-beam, which is connected to the massive part of the irradiation unit. The latter moves along the rail track (9) using an electrically driven mechanism, adjusting the distance from the samples to the surface of the water moderator.

Results of the experiment

During the experiment, gold samples with and without cadmium protection were irradiated, as well as 10 sets of radiachromic dosimeters arranged in 10 cm increments starting from the WM. Irradiation occurred for 12 days on channel No. 3 of the switched-off IBR-2 reactor (the average reactor power during the cycle was 0 kW).

The spectrum measurement from activated gold was carried out on the Canberra GC10021 spectrometer, and the measurement of dosimeters was carried out using the FWT-92 photometer, the principles of operation of which are described above.

1. Absorbed dose of gamma radiation

Dosimeters were located along the I-beam (5) (Fig. 4). Starting from the minimum possible distance to the WM, three dosimeters were located every 10 cm, the optical density of which was measured on a photometer before and after the experiment. According to the difference in optical density before and after irradiation, the duration of irradiation and the already known calibration of the photometer, the powers of absorbed doses of gamma radiation were found. The graph of the obtained dependence is shown in Fig.5.

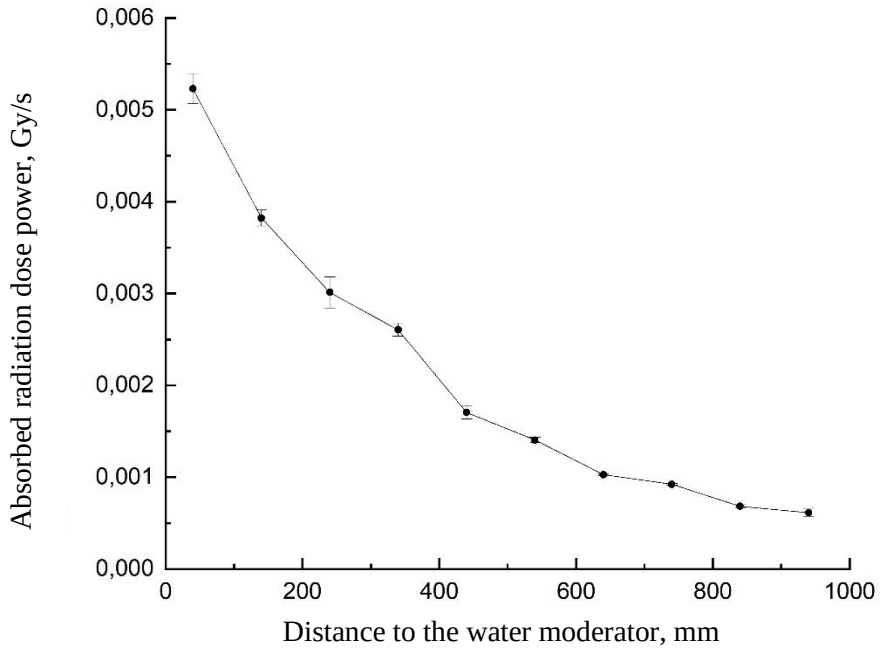

Figure 5. The dependence of the absorbed dose of gamma radiation on the distance to the WM at reactor power of <W>=0 кВт.

The determined values of the absorbed dose rate are in the range of $5 \cdot 10^{-3} - 0.6\ Gy/s$. The dependence behaves like an exponent to a negative degree, and is well approximated by it (χ^2 is 0.97).

2. Thermal and resonance neutron flux densities

The indicators were placed at the point closest to the WM (3). After irradiation, activation spectra (Fig.6) were taken from them in turn using the Canberra HPGe detector.

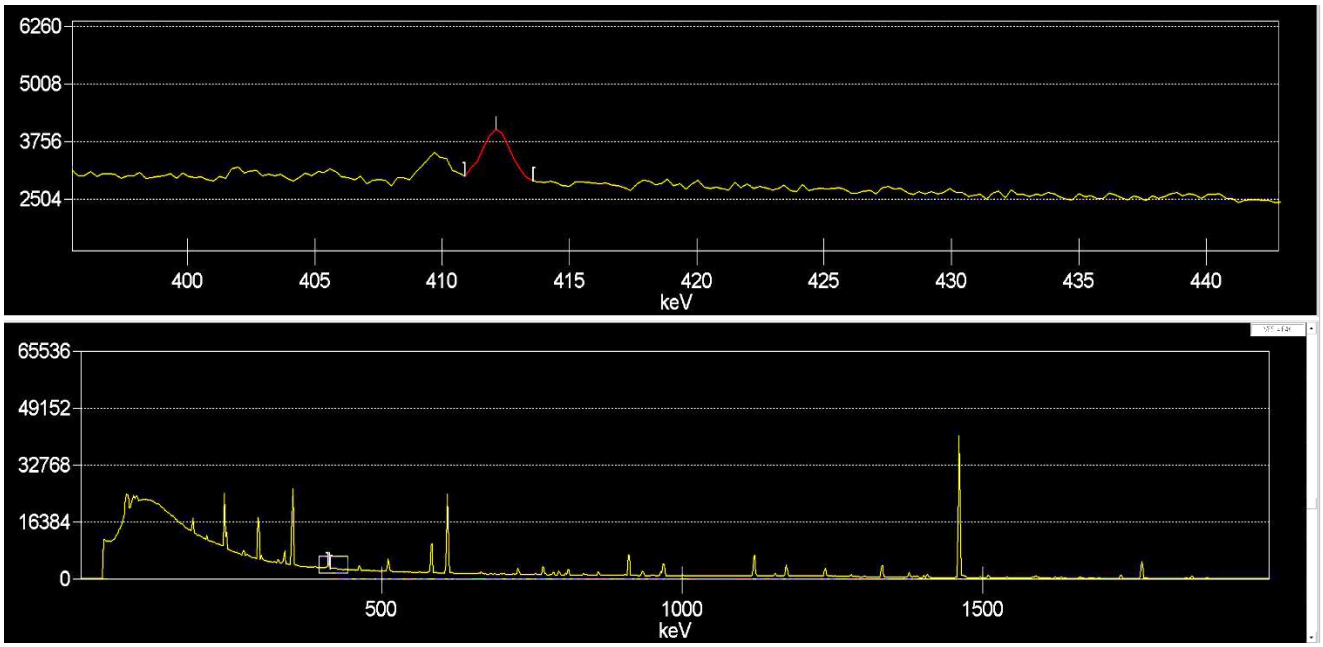

Figure 6. The spectrum obtained after irradiation of the gold indicator (bottom), the gold line on an enlarged scale (top).

Further, from these spectra, the area of the peak of the gold-198 isotope and the time of its measurement were determined using the Genie-2000 program. Knowing

these values for a gold sample with and without cadmium protection, we can calculate the values of the flux densities of resonance and thermal neutrons by the formulas (2) and (3), respectively.
The calculation results are presented in Table 1.

Sample	Area of the peak	Neutron flux density, $n/cm^2 \cdot s$
^{197}Au+Cd	4258 ± 195	$\Phi_{res} = 148 \pm 7$
^{197}Au	4305 ± 234	$\Phi_{th} = 499 \pm 129$

Table 1. Results of calculations of resonance and thermal neutron fluxes.

So, the neutron flux density near WM of channel No. 3 of the switched-off IBR-2 reactor have an order of $\sim 10^2\ n/cm^2 \cdot s$, the thermal neutron fluxes are greater than the resonance neutron fluxes after passing the WM. The large error in these results is primarily due to the short time of irradiation and measurement of satellites. The errors of the calculated values consist of statistical errors in determining the areas of gold peaks, inaccuracies in measuring the time intervals of irradiation, exposure and measurement, as well as errors in determining the mass of the sample.

Summary of the work

As a result of the work:

1. The features of interaction with the substance and methods of registration of non-ionizing radiation were studied,
2. The experiment was set up to measure ionizing radiation fields near WM-303 of the switched-off IBR-2 reactor ($\langle W \rangle = 0\ kW$),
3. The technique of radiachromic dosimetry on the example of FWT dosimeters and photometer, as well as gamma-ray spectrometry using the Canberra spectrometer and Genie-2000 software has been mastered,
4. The methods of neutron activation analysis and radiachromic dosimetry were applied in practice,
5. The power values of the absorbed dose rate of gamma radiation ($5 \cdot 10^{-3} - 0.6\ Gy/s$) and the flux densities of resonance ($\Phi_{res} = 148 \pm 7\ n/cm^2 \cdot s$) and thermal ($\Phi_{th} = 499 \pm 129\ n/cm^2 \cdot s$) neutrons were obtained.

I want to thank my mentor from the FLNP, Maria Petrova, for the time spent explaining the material, for all possible help in the process of working and conducting the experiment, and for important comments to my report. Special thanks to Elena Karpova for timely informing and arranging excursions to JINR laboratories. I would also like to express my gratitude to the entire START team for the opportunity to come and work at JINR.

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