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= RADIOBIOLOGY, ECOLOGY, AND NUCLEAR MEDICINE

Pneumatic Transport System REGATA-2 for Automation of Activation Analysis at the IREN Facility, FLNP JINR

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Abstract—The IREN facility at the Frank Laboratory of Neutron Physics (FLNP) of the Joint Institute for Nuclear Research (JINR) is used for experiments to determine elemental composition of various samples by neutron activation analysis (NAA). A pneumatic transport system (PTS) REGATA-2 was implemented to automate the delivery of containers with samples to the irradiation position and back. The article describes the characteristics of the PTS, its adaptation for operation at the IREN facility, and also plans for subsequent modernization and future prospects. After the assembling of the system, the optimal parameters were chosen for irradiating the typical samples, and a calibration was created for a high-purity germanium detector, which let to consider the volume nature of the samples. Samples of archaeological ceramics from Kazakhstan were studied using the PTS. The features of the IREN facility let to use not only a neutron, but also a gamma-producing target. The first experiments on the qualitative determination of elements by gamma-activation analysis (GAA) were carried out, and the prospects for using the facility for these purposes were confirmed. The creation of the PTS let to automate the process of irradiation samples and increase the efficiency of research in NAA and GAA, especially for short-lived isotopes. It also reduces the negative impact of radiation on the human body.

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COMPONENTS OF THE REGATA-2 PNEUMATIC TRANSPORT SYSTEM

The IREN facility [1] uses the LUE-200 electron accelerator and let to obtain neutrons or gamma quanta depending on the type of target. The electron energy currently reaches 120 MeV. The facility operates in a pulsed mode with a burst frequency of 25 or 50 Hz at an average current of 5 μ A.

The neutron-producing target is a cylinder made of a wolfram alloy with a diameter of 40 mm and a height of 100 mm, placed inside an aluminum tank with a diameter of 160 mm, through which distilled water is pumped from a closed loop to cool the target and slow down neutrons. The gamma-producing target is a 3 mm thick plate of the same alloy.

The REGATA-2 pneumatic transport system was developed in the Development and Implementation Base – Physics of the Bulgarian Academy of Sciences for the applied use of the neutron or photon fluxes. The project for the placement of the PTS in the rooms of the IREN facility was created at JSC VNIPIET.

The main purpose of the PTS is to deliver the container with the sample to the irradiation position and back. The return delivery time should be kept to a minimum to allow research on short lived isotopes. It is also important that the irradiation time is limited only by the operation time of the accelerator.

Figure 1 shows the main components of the PTS. The system consists of a box with a loading and unloading station. A polyethylene transport channel with a length of about 40 m departs from the box. The transport channel is divided by splitters into three channels that enter the target hall and end with channels for irradiation made of stainless steel. Two channels, N1 and N2, are designed for irradiation in a neutron flux, channel G let to work with gamma quanta. The channels are attached to the truss where the neutron moderator is located.

A polyethylene transport container with a height of 42.5 mm, an inner diameter of 17.4 mm, an outer diameter of 24 mm is moved along the transport channels using compressed air. For the stable operation of the PTS, the developer recommended a compressor with a 500 liters receiver with a working pressure of 5 bar. During commissioning it was found that a receiver of 80 liters is enough for stable operation of the system. The waste air from the transport system is removed to a special ventilation.



Fig. 1. General scheme of the REGATA-2 pneumatic transport system at the IREN facility.

During commissioning, the problem of magnetic valve coil overheating during long irradiation (over 30 min) of the sample was revealed. Overheating of this coil did not allow to return the container in the normal mode. The problem was solved by rewriting the control program. Now the irradiation time is not limited.

The box is located on a frame (Fig. 2), inside which the controllers, some pneumatic components and the storage are located. The storage is used for temporary placement of highly active containers. It is an aluminum disk with six cells. The disk is surrounded on all sides by lead protection 50 mm thick. The current cell number is visible through the information hole. There are two holes above the storage—inlet and outlet. The outlet is used to remove the container with a special grip. The storage is controlled by a remote control located above the frame door.

There are the following status sensors in the loading and unloading station:

(1) An optical position sensor that detects the presence of the container in the loading and unloading station.

(2) An inductive sensor of the chamber door position gives a permissive signal for operation in case the door is closed.

(3) An inductive sensor of the position of the manual valve for moving the sample into the storage. In the normal state, the manual valve must be closed, otherwise this sensor will not give a permission signal for operation.

The control panel is located next to the box. It contains cables for the transmission of all control signals of the PTS. A separate LED indicates sufficient compressed air pressure. Three conditions must be performed to send the container with the sample to the irradiation position: there is the container in the chamber, the chamber door and the manual valve are closed. The experimenter selects the required channel



Fig. 2. Box with loading and unloading station.



Fig. 3. The dose control post.

for irradiation using the buttons N1, N2 or G, which is signaled by the corresponding LEDs. When the sample moves through the transport channels, the LEDs change their color. Turning on the last LED indicates that the container has reached the selected irradiation position. When returning, the container stops at the dose control post to measure the induced activity. Then the experimenter moves it to the loading and unloading station. The container can be removed from the station for further work, or sent to the storage.

The transport system is controlled by two ZELIO SCHNEIDER controllers. The transport system is controlled by the SR3 controller with the extension module "C", the storage—by the SR2 controller. The controllers are located in the electrical cabinet together with the terminal strips. Signal cables from the PTS sensors are connected to the terminal strips. The ladder logic programming language Ladder diagram (LD) is used for programming controllers. Two main components can be distinguished in this language: a contact and a coil. The contact can be normally closed and normally open. Logical chains are made up from these contacts, the results of which are transferred to target variables called coils.

A dose control post is located above the box (Fig. 3). Its main part is a stopper, which automatically closes the pipeline after the container is launched into the irradiation position and stops each irradiated sample when it returns to the box. A dosimeter was installed near the stopper to measure the induced activity, the readings of which are displayed. Based on these data, the experimenter decides on further actions with the sample: extract it for work or send it to the storage. A separate sensor signals the presence of the container in the stopper. Initially, a reflective optical sensor was used, similar to those located on the splitters and in the target hall. The reflective sensor worked unstably due to the fact that the sample moved very quickly in this area, and was replaced by a barriertype sensor, for which additional mounting parts were made in the FLNP Workshop.

The transport channel branches into three channels using splitters before entering the target hall (Fig. 4). These splitters are pneumatic "arrows" that determine the further path of the container. The splitters are driven by pneumatic cylinders. Next to the cylinders are optical sensors. The readings of these sensors are displayed on the control panel, which allow the experimenter to monitor the current position of the container. The first splitter branches off the pipeline that leads to the horizontal irradiation channel G. The second one separates the vertical channels N1 and N2.

Three transport channels enter through the holes in the wall into the target hall (Fig. 5), where they are fixed on supporting columns. The transport channels end with irradiation channels made of stainless steel. Channels for irradiation are positioned close to the neutron moderator.

The authors needed to solve the problem with the container presence sensor at the irradiation position. All used types of sensors failed in short periods of time, when placed close to the place of irradiation. It was proposed to give a signal with a reflective optical sensor at the moment when the container is about one and a half meters from the irradiation position. The transit time of the container along this section is very short compared to the whole route. Changes were made to the program of the container motion control controller, according to which the air supply continued for one second after the response of the described sensor. The FLNP Design bureau developed a special module to install the sensor, where the sensor and the reflecting element are placed. The operation of the created structure showed excellent results, including



Fig. 4. Splitters.



Fig. 5. Target hall.

in terms of radiation resistance: the first sensor worked for more than six months.

The irradiation channels are fixed on an adjustable structure called a truss. The Bulgarian truss was not put into operation due to some reasons, so the design for fixing the channels was modified for the rack used. This rack did not have a supporting structure, which made it impossible to securely fix the channels for irradiation. Especially for this, the FLNP Design Bureau developed a support structure, which was implemented in the FLNP Workshop. This construction fixes the irradiation channels N1 and N2 in such a way that the irradiated sample in the container is at a height of 5 cm from the target surface (Fig. 6).

The above mentioned value was determined experimentally. It was necessary to find the point of maximum energy release because the flux of neutrons emitted from the target is irregular along the height of the moderator. For this, the fluxes of thermal and resonance neutrons were determined as follows: flux monitors were prepared and irradiated—copper in cadmium shell and without it. These monitors were placed on the surface of the moderator at a height of 1, 3, 5, 7, 9 cm from the top edge and irradiated at the burst frequency of 25 Hz for about 14 h. The second set of monitors was irradiated at the burst frequency of 50 Hz for about 5 h.

Table 1 shows the highest thermal neutron flux is observed at the height of 5 cm from the upper edge of the moderator. At this height, the N1 and N2 channels were positioned for neutron activation analysis.



Fig. 6. Fixing channels for irradiation to the rack.

AN EXPERIMENT TO DETERMINE THE QUALITATIVE AND QUANTITATIVE ELEMENTAL COMPOSITION OF SAMPLES USING NEUTRON ACTIVATION ANALYSIS

As a result of commissioning, the PTS was created with the following characteristics:

• the average delivery time of the container with the sample to the irradiation position is 10 s, the return of the container takes about the same time;

• air is released from the pipeline within 50 s after the container arrives at the irradiation position or at the stopper. Therefore, the minimum irradiation time is about one minute;

• pipeline length—about 40 m;

• the maximum mass of the container with the sample is not less than 13 g. The mass of the empty container with a lid is approximately 6 g.

These characteristics let to use the PTS for carrying out NAA on short-lived isotopes with a half-life of about one minute. A series of experiments was carried out at the IREN facility for choosing the optimal parameters for irradiation of typical samples, taking into account the specifics of neutron fluxes. Five sets were prepared from one sample, consisting of 3, 5 and 7 g subsamples. Samples of all sets were irradiated for 10, 20, 30, 40 and 50 min.

The obtained spectra were processed using the GENIE-2000 program [2]. It was necessary to identify such irradiation parameters at which it is possible to determine the maximum number of short-lived iso-

Burst frequency, Hz	Height, cm	Mass of Cu, g	Mass of Cu in the shell of Cd, g	$\Phi_{\rm therm},$ $10^7 {\rm cm}^{-2} {\rm s}^{-1}$	$\Phi_{\text{resonance}},$ $10^7 \text{ cm}^{-2} \text{ s}^{-1}$	
25	1	0.340	0.340	2.57 ± 0.08	1.00 ± 0.03	
	3	0.326	0.340	5.69 ± 0.16	1.32 ± 0.04	
	5	0.336	0.339	7.46 ± 0.21	1.29 ± 0.04	
	7	0.343	0.350	6.64 ± 0.19	1.25 ± 0.04	
	9	0.338	0.345	5.30 ± 0.16	1.12 ± 0.04	
50	1	0.236	0.220	7.17 ± 0.22	2.42 ± 0.08	
	3	0.228	0.246	17.2 ± 0.5	3.14 ± 0.10	
	5	0.240	0.252	20.6 ± 0.6	3.57 ± 0.11	
	7	0.233	0.234	19.6 ± 0.6	3.34 ± 0.10	
	9	0.238	0.235	17.8 ± 0.9	3.04 ± 0.10	

Table 1. Thermal Φ_{therm} and resonace $\Phi_{\text{resonance}}$ neutrons fluxes

PNEUMATIC TRANSPORT SYSTEM REGATA-2

	Mg ²⁷ Mg		Al ²⁸ Al		Si ²⁹ Al		Ca ⁴⁹ Ca		V ⁵² V		Mn ⁵⁶ Mn		U ²³⁹ U	
	$T_{1/2} = 9.5 \text{ min}$		$T_{1/2} = 2.2 \text{ min}$		$T_{1/2} = 6.6 \text{ min}$		$T_{1/2} = 8.7 \text{ min}$		$T_{1/2} = 3.7 \text{ min}$		$T_{1/2} = 2.6 \text{ h}$		$T_{1/2} = 23.5 \text{ min}$	
	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%	mg/kg	%
e-01	5110	11	157000	5.7	275000	3.9	13600	23	38.5	15	130	7.3	7.44	19
e-02	4310	11	147000	5.7	287000	4	15300	18	35.5	16	79.4	8.2	6.45	19
e-03	2850	11	77200	6	217000	4	15900	20	91.9	13	522	6.6	5.58	19
e-04	3240	11	88800	5.7	246000	4.1	13400	21	105	13	617	6.3	6.99	19
e-05	2890	11	102000	6	237000	4	9200	24	75.4	13	407	6.7	5.32	19
e-06	2680	11	104000	6.1	269000	4	26900	19	86.3	13	417	6.8	5.32	24
e-07	3140	11	158000	6.1	252000	4.2	19200	21	122	14	481	6.7	5.05	20
z-01	3480	11	97100	6	362000	3.9	11 500	24	132	15	786	6.5	9.49	27
z-02	2670	11	98900	6.4	331000	4	23 500	21	159	11	854	6.7	9.19	26
z-03	2770	11	80700	6.3	265000	4	15100	22	130	12	283	7.3	8.02	26
z-04	3890	11	112000	6	315000	3.9	18 2 0 0	20	271	11	745	6.1	7.97	27
z-05	3830	11	56400	6	243000	3.9	12500	21	168	12	355	7	11.1	26
z-06	4040	11	159000	6	420000	3.9	16300	27	207	12	961	6.3	11.9	26
z-07	2840	11	89500	6.4	319000	4	11 500	23	142	13	566	6.9	8.61	26
z-08	3720	11	124000	6	339000	3.9	13 500	28	212	12	883	6.9	12.1	28
z-09	5160	11	327000	6	484000	4	31 000	20	213	20	407	8.2	14	28
z-10	4130	12	177000	6	437000	3.9	18000	21	138	16	528	7.5	13.6	27

Table 2. The elemental composition of archaeological ceramic samples from Kazakhstan

topes. At the same time, it was important to minimize the number of long-lived isotopes for more comfortable processing of the spectra. From Table 2 it can be seen the mass has practically no effect on the number of isotopes obtained. The most acceptable results are shown by samples irradiated for thirty and forty minutes. A choice was made in favor of forty-minute irradiation due to the greater number of determined shortlived isotopes. Further, changes were made to the container movement control program. The sample began to return automatically after forty minutes without pressing the return button.

Due to the volume of the samples did not allow them to be considered as point sources, a new efficiency calibration was created for the HPGe detector Canberra GC-10021. This calibration is for the counting geometry of the container in which the samples are irradiated and measured for heights of 2.5, 5, 10, and 20 cm. Efficiency calibrations based on point reference spectrometric sources of gamma radiation were taken as a basis. It was necessary to move from these calibrations to volume source calibrations. For this, the ANGLE program from AMETEK [3] was used, which allow to transfer efficiency between different geometries. The program let to take into account the parameters of the detector used for measurements. It is also possible to describe the geometry of the container in which the source is located, take into account the degree of its filling and the material of the sample.

Figure 7 shows plots of calibration curves. The efficiency calibration for the height of 10 cm from the detector surface for the point source is highlighted in blue. Red color—the efficiency calibration for the same height for the volume source. The source used here is a soil type sample, 15 mm high, placed in the polyethylene container described above.

An experiment was carried out for determine the quantitative elemental composition of the samples after creating volume efficiency calibrations. Investigations of ceramics samples found on the territory of Kazakhstan was carried out on short-lived isotopes. Seventeen samples, four standard samples and flux monitors were irradiated sequentially for forty minutes each. The spectra were processed using the GENIE-2000 program after irradiation. As a result, seven elements were found. The calculation of mass fractions for these elements was carried out using the "Mass fractions" software [4, 5]. Table 2 contains results of calculation. In the header lines of the table, in addition to the elements, isotopes are indicated, according to which the mass fractions of elements were calculated, as well as their half-life. These data show that the PTS can be successfully used for research the elemental composition on short-lived isotopes. Earlier, studies of the



Fig. 8. ⁶¹Cu isotope, the 282 keV line of the cuprum sample.

elemental composition on long-lived isotopes were carried out at the IBR-2 reactor (FLNP JINR) with the same samples, so the experiment significantly supplemented the picture of the elemental composition of ceramic products.

GAMMA-ACTIVATION ANALYSIS: FIRST EXPERIMENT

The first steps were taken in gamma-activation analysis using the PTS. Monoelement samples were taken for irradiation: copper and aluminum foil, and also the samples, whose elemental composition is known: the NIST 50C standard and a plaster's fragment of the Saviour Church on Nereditsa (12th century) from Veliky Novgorod. The investigated sample was placed in the polyethylene container, fixed with a cotton swab, and delivered to the gamma-producing target using the PTS. Irradiation was carried out at different burst frequencies: 2, 5, and 10 Hz with an energy of 50 MeV. Also, the amount of the sample was varied: 0.1, 0.5, and 1 g. The goal of the experiment was a qualitative elemental analysis.

The copper sample was irradiated first. Lines of the ⁶¹Cu isotope were detected when analyzing the spectra. This isotope was obtained as a result of the γ -2*n* reaction from the natural isotope ⁶³Cu [6].

Next, the aluminum foil was irradiated. The spectrum contained lines of two isotopes: ²⁹Al and ²⁸Al. These isotopes could not have been produced by





Fig. 10. ⁵²V isotope peak of the 50C standard.

gamma reactions. Unfortunately, fast neutrons collide with the sample in addition to gamma radiation, which appear due to the bremsstrahlung hitting the tungsten target. This aspect complicates the gammaactivation analysis.

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Then the 50C standard was irradiated, which is a steel chip, basic including W, Cr, V and C. Peaks of the isotopes Ta, Cr (Fig. 9) and V (Fig. 10) were found on the spectrum. Peaks of some other isotopes were also detected, among which only ¹⁸⁵Ta, ⁴⁹Cr and ⁵⁷Ni can be confidently attributed to the results of gamma reactions. The rest were obtained by irradiating the standard with fast neutrons most likely. ¹⁸⁵Ta was obtained from the γ –*n*,*p* reaction from the natural isotope ¹⁸⁶W.

⁴⁹Cr and ⁵⁷Ni were obtained by the γ -*n* reaction from the natural isotopes ⁵⁰Cr and ⁵⁸Ni, respectively.

Next, the plaster's fragment from the Saviour Church on Nereditsa was irradiated. The elemental composition of this sample was determined earlier using neutron activation analysis. Isotopes ⁴³K, ⁵³Fe (Fig. 11), and also isotopes ²⁹Al, ⁴⁷Ca, ¹²⁶Cs, ¹³⁷Nd, ⁴⁷Sc, ^{89m}Zr, ⁵⁶Co, ⁶⁴Co and ⁷⁵Zn were detected.

Of this group, only the appearance of ⁴³K, ⁵³Fe, and ^{89m}Zr isotopes can be confidently attributed to the results of gamma reactions. ⁴³K appeared by the reaction γ –*n*,*p* from the natural isotope ⁴⁴Ca. ⁵³Fe and ^{89m}Zr – by the γ –*n* reaction from the natural isotopes ⁵⁴Fe and ⁹⁰Zr, respectively.



Fig. 11. Peaks of ⁴³K and ⁵³Fe isotopes of plaster's fragment from the Saviour Church on Nereditsa.

As a result of the experiment, four samples were irradiated, in three of which isotopes were found that appeared using gamma reactions. The results showed that gamma-activation analysis is possible at the IREN facility, but for its implementation it is necessary to create special software for calculation of the elements mass fractions. In addition, it is necessary to solve the problem caused by the influence of thermal neutrons, which also activate the sample.

MODERNIZATION OF THE PNEUMATIC TRANSPORT SYSTEM REGATA-2

Currently, the authors are working on the modernization of the PTS. The original project did not allow simultaneous neutron irradiation of containers with samples in different channels. Nowadays, the program code is being updated accordingly to enable the irradiation of several containers simultaneously. Also, work is underway to replace the electromechanical control panel with a sensor one and implement the possibility of exchanging information with the database of neutron activation analysis [7].

CONCLUSIONS

The REGATA-2 PTS was put into operation at the IREN facility. The PTS includes two channels for irradiating samples with neutrons and one with gamma quanta. The PTS let to carry out experiments to determine the elemental composition of samples from short-lived isotopes with the half-life of approximately one minute or more. Samples of archaeological ceramics found on the territory of Kazakhstan were investigated using the PTS. Also, the first steps were taken to carry out gamma-activation analysis. The PTS is being upgraded.

It is planned to continue carrying out neutron activation analysis, and also further experiments on the implementation of gamma-activation analysis.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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