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ANALYTICAL INVESTIGATIONS AT THE **IBR-2** REACTOR IN DUBNA

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INTRODUCTION

Instrumental neutron activation analysis (INAA) is continuing to be the most sensitive method for multi-element analysis along with the relatively new analytical techniques: inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS). Each of these techniques has their own merits and problems. A comparison of precision and detection limits for elements to be determined by different methods should be made with «real» samples, as the result of the analysis depends strongly on the elemental composition of the sample.

Pulsed fast reactor IBR-2, equipped with pneumatic system REGATA for INAA, provides activation with thermal, epithermal and fast neutrons. Thermal NAA takes advantage of the high intensity of neutrons available from the thermalization of fission neutrons and the large thermal neutron cross sections for most isotopes. Epithermal NAA (ENAA) is a useful extension of INAA in that it enhances the activation of a number of trace elements relative to the major matrix elements. Epithermal is taken to be neutrons with energies from the Cd «cut-off» of 0.55 eV up to approximately 1 MeV. At $E<0.55$ eV, the spectra of the thermal neutrons with and without the Cd coat are shown in **Fig. 1**. In general, the activation cross sections of the matrix elements of environmental samples are inversely proportional to the neutron energy ($1/v$ low).

The trace elements also follow this general trend but many of them (rare earth elements in particular) have large activation cross section (resonance integrals) at specific energies in the epithermal energy region [1].

REGATA EXPERIMENTAL SETUP

The layout of the experimental setup called REGATA for INAA at the reactor IBR-2 is given in **Fig. 2**. It consists of four channels for irradiation (Ch1-Ch4), the pneumatic transport system (PTS) and three gamma-spectrometers located at three special rooms on the ground floor of the reactor IBR-2. The main parameters of the irradiation channels are presented in **Table 1**. The channels Ch3, Ch4 are cooled by water, and the channels Ch1, Ch2, connected with the pneumatic transport system, are cooled by air. That is why the temperature in channels Ch3 and Ch4 is lower than the temperature in channels Ch1 and Ch2 in spite of the greater neutron flux density. The time of sample irradiation in channels Ch3, Ch4 depends on the operation cycle duration of the reactor and is equal to 10-12 days, as a rule. The irradiation channels Ch1 and Ch2 are the same, but Ch1 is Cd-coated. Each channel consists of two concentric pipes made from stainless steel. They are placed into an aluminum box filled with biological shield. Samples are transported through one of the pipes («flight pipe») of 28 mm in diameter. Compressed air flows through the second pipe.

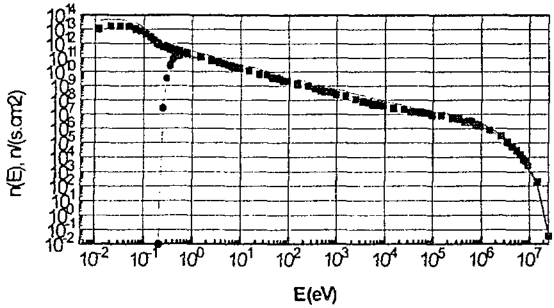

Fig. 1. Neutron energy spectra in irradiation channels CH1(■) and CH3 (curve) [2].

Table 1. The main characteristics of the irradiation channels

Irradiation site	Neutron flux density (n/cm^2 s)×10^{12}			T °C [3]	Channel diam., mm	Channel length, mm
	Thermal	Resonance	Fast			
Ch1	Cd-coated	3.31	4.32	70	28	260
Ch2	1.23	2.96	4.10	60	28	260
Ch3	Gd-coated	7.5	7.7	30-40	30	400
Ch4	4.2	7.6	7.7	30-40	30	400

Samples are irradiated in transport containers made of polyethylene, teflon and aluminum (**Fig. 3**). The irradiation time of a polyethylene container is limited by radiation and temperature resistance of polyethylene and is equal to 30 minutes, both for Ch1 and Ch2 at 2 MW power of the reactor. A teflon container is used for irradiation up to 5.0 h. Aluminum containers are used for longer irradiation. The internal volume of the polyethylene container is equal to approximately 4 cm^3, and the volume of the aluminum container is 1.5 times lager.

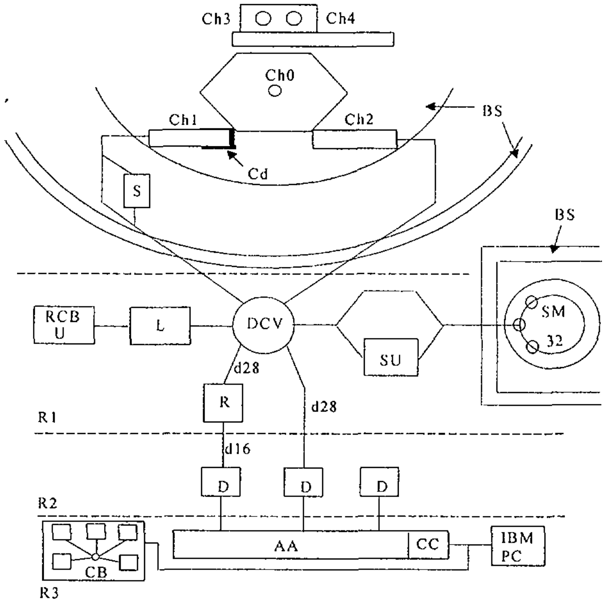

Fig. 2. The scheme of the REGATA experimental setup.

Ch1-Ch4 –irradiation channels, S- intermediate storage, DCV- directional control valves, L- loading unit, RCB- radiochemical glove-cell, U- unloading unit, SU- separate unit, SM- storage magazine, R- repacking unit, D- Ge(Li) detector, AA- amplitude analyser, CB- control board, CC- CAMAC controller, R1-R3- the rooms where the system is located.

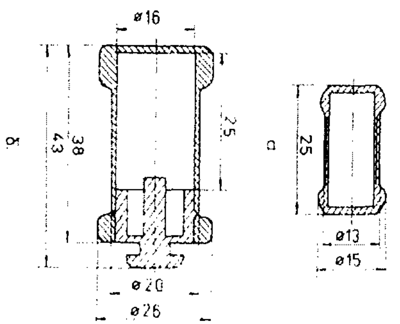

Fig.3. Polyethylene transport container and inner capsule for a sample

Up to 7 containers can be simultaneously loaded in each channel for long irradiation. The neutron flux density is controlled by monitors (Au, Zr, etc.) **Fig. 4** shows that the reduction of the neutron flux density along the channel length is significant, up to 30-50%. It should be taken into consideration while data processing.

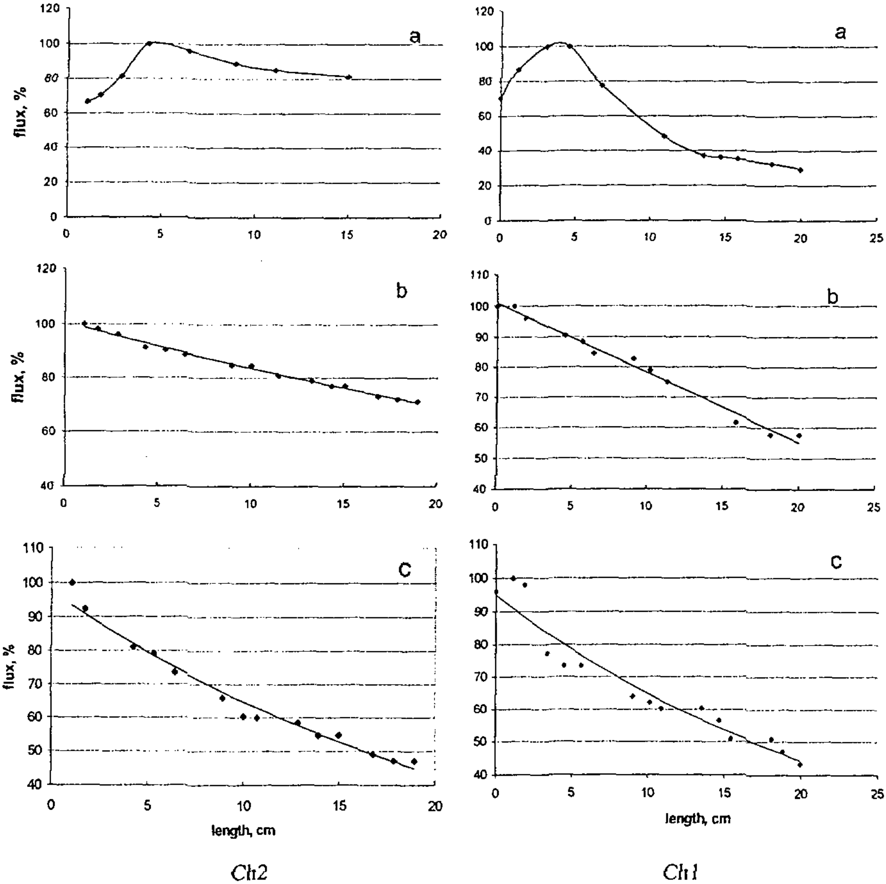

Fig. 4. The distribution of neutron flux density along the length of the channels Ch1 and Ch2 [4].
a – thermal neutrons; **b** – epithermal neutrons; **c** – fast neutrons.

The pneumatic system transports containers by compressed air (at 3-6 atm. pressure). All its units are located far from the reactor core (50-60 m). The polyethylene containers are transported to irradiation site for 7-20 s, the aluminum containers flight faster – (3-7) s. The intermediate storage (S) (for up to 15 containers) is used to reduce the activity of aluminum containers after long irradiation. It is located between two rings of the biological shield of the reactor. There is also a magazine to store highly activated samples with 32 cells (SM). It is surrounded with the biological shield made of lead and concrete blocks.

There are three hot cells for material science studies equipped with a set of instruments and devices to measure the mechanical characteristics of samples.

When short-lived isotopes are determined, a small polyethylene capsule is placed into a transport container. There is a special unit (R) to extract a small capsule from a transport container.

PTS has loading (L) and unloading (U) units to load and to extract containers from the system. To provide radiation safety, the unloading unit is placed into a glove-cell. All devices of the pneumatic system are equipped with photosensors and end-switches to indicate the container position in the system and to correct operation of all mechanisms. Acoustic detectors placed on the «flight pipe» of irradiation channels behind the first ring of a biological shield allow one to determine the time of a container arrival and departure accurately. There are many noise pulses during the flight of a container in the «flight pipe». Each noise pulse clears the irradiation timer. The knock of the capsule at the bottom of the channel forms the last clearing pulse. After this pulse the irradiation timer starts counting the time of irradiation. Such positioning of acoustic detectors protects them from radiation damage and therefore prolongs their lifetime.

Personal computer controls the pneumatic transport system and amplitude spectrometers. All units of gamma-spectrometers and counting electronics are made in CAMAC at JINR. The software developed at FLNP JINR for peak search, peak fitting and nuclide identification routines is used for processing the amplitude spectra [5].

The main trends of investigations performed at the Department of activation analysis and radiation research, FLNP JINR, are summarized below.

ANALYTICAL INVESTIGATIONS AT IBR-2 REACTOR

Instrumental neutron activation analysis Conventional INAA	*Epithermal neutron activation analysis ENAA*	*Cyclic neutron activation analysis CNAA*

Environmental Studies

- Air pollution studies (**air filters**)
- Biomonitoring atmospheric depositions (**moss, lichens**)
- Assessment of the environmental situations (**waste dumps, oil accidents**, aquatic pollution (**Moscow Sea, Oka River Basin, Lake Baikal**)

Material Science

- **High purity materials** (aluminium, semiconductors – silicon, germanium, gallium arsenid)
- **Biotechnologies:** (i) biochemical leaching of elements by aerobic and anaerobic bacteria; (ii) Se-containing pharmaceuticals based on blue-green algae *Spirulina Platensis*)
- **Construction materials** in the problem of decommissioning of nuclear power plants

The on-going projects and projects under preparation at the Department of activation analysis and radiation research are listed in the **Table 2**.

Table 2.

No	ON-GOING INTERNATIONAL PROJECTS
1	Biomonitoring of heavy metals and radionuclides in Chelaybinsk Region, South Ural, Russia: temporal and spatial trends (Russia, Norway) (IAEA grant)
2	Heavy metal atmospheric deposition in Bulgaria (Grant of the Plenipotentiary of Bulgaria)
3	Atmospheric deposition study in Poland (Grant of the Plenipotentiary of Poland)
4	Heavy metal atmospheric deposition in Romania (IAEA grant, JINR's share 50%)
5	Biomonitoring of some industrial areas in Slovakia (Grant of the Plenipotentiary of Slovakia)
6	Biomonitoring in Yugoslavia around Belgrade and Novi Sad (Serbia) and mountainous area (Bosnia) (Pilot project)
7	Heavy metal atmospheric deposition in the South Korea (moss, lichens, bark) (Pilot project)
8	Atmospheric deposition study in China (Protocol on collaboration)
9	Air Pollution Studies in the Greater Cairo Area, Egypt (Agreement with the Atomic Energy Authority of Egypt)
10	Workplace monitoring and occupational health studies at some selected phosphate fertilizer plans using nuclear and related analytical techniques in Russia, Uzbekistan, Poland, Romania, the Netherlands, Denmark (Grant COPERNICUS)
11	Workplace monitoring and health related studies at phosphate fertilizer plant in Voskresensk, Russia (IAEA grant)
12	Workplace monitoring and health related studies at phosphate fertilizer plant in Gdansk, Poland (Grant of the Plenipotentiary of Poland)
13	Biotechnology and biochemistry of Se-containing pharmaceuticals based on blue-green algae *Srirulina Platensis* matrix for use in practical medicine (Pilot project JINR-Georgia) (Submitted to Rockefeller Foundation)
14	Biotechnology applied to rocks and poor ores in Georgia for extraction of radioactive and rare earth elements (ggrant ISC)
15	Development of the Analytical Complex REGATA at IBR-2 (Grant of the Academy of Sciences of Czechia)
	PROJECTS UNDER PREPARATION
1	Assessment of the environmental situation in the deltas of two great rivers of the world: Volga and Nile (European Union Commission on Mediterranean projects)
2	Quality and toxic safety of food products grown in contaminated areas under strong anthropogenic impact of heavy metals and radionuclides (INTAS)
3	Monitoring of radionuclides and heavy metals in the Northern Kazakhstan, Russia, Mongolia and China Affected by Semipalatinsk Nuclear Test Site (World Bank)

During the last decade the main activity of the Department of AA and RR is focused on studying air pollution in the framework of several national and international programmes and projects as seen from above. Since the year of 2000 jointly these projects are recognized as the first priority project REGATA (named after the experimental setup and possibly be read as abbreviation: **R**ussian-**E**uropean **GA**te **T**o **A**sia) «*Atmospheric Deposition of Heavy Metals in Some Industrial Areas of Russia, Bulgaria, Poland, Romania, and Slovakia Studied by the Moss Biomonitoring Technique and Employing Nuclear and Related Analytical Techniques, and GIS Technology*» of the Theme-Plan of JINR.

ENAA FOR AIR POLLUTION STUDIES

Epithermal neutron activation analysis (ENAA) has certain advantages over conventional instrumental activation analysis for many trace elements in terms of improvement in precision and lowering of detection limits, reduction of high matrix activity. The technique is based on the fact that some elements have isotopes with resonances in the epithermal neutron region. The ratio of resonance activation integral/thermal neutron cross-section (I_0/σ_0) is of the order of 0.5 for nuclides without resonance in epithermal neutron region and it may be as high as 100 in other cases. This means that the radionuclide distribution originating from epithermal activation may deviate strongly from that apparent when the whole reactor spectrum is employed.

There are two challenging areas of air pollution studies: analysis of airborne particulate matter and analysis of the biomonitors of atmospheric deposition. Air filter analysis is an area, where INAA hardly can be replaced by any non-nuclear analytical technique because of a small total mass of aerosol, collected on a filter [6-7].

The dominant part of air pollution studies is based on the use of the moss biomonitoring technique. A moss have no developed root system, that is why it takes nutrients almost exclusively from the atmosphere. Widely-spread in the Western Europe [8], this technique at the present is applied to air pollution studies in some industrial areas of Russia (the Siberia, the Kola peninsula [9] the South Ural [10], Central Russia (Tula region [11] and Moscow region [12]) and in the country-members of JINR (Poland [13] and Romania [14-15]) as well as in non-members (Norway [16]), China [17].

The combination of epithermal and conventional activation analysis on IBR-2 reactor, supplemented by atomic absorption spectrometry for the elements Pb, Cd, and Cu, allows one to determine about 45 elements of the periodic table in moss. The results of these investigations are presented in the form of tables, diagrams, graphs and, using GIS (geographical information systems) technology, in the form of colored contour maps for each element.

The multivariate statistics (principle component analysis) is applied to obtain information on the character and origin of the pollution sources. A competitive technique, inductively coupled plasma emission mass spectrometry (ICP-MS), demonstrates a satisfactory result for about 55 elements in moss. In 7 cases (Sc, As, Sb, Hf, Ta, Au, Th) ENAA is judged to be the preferred technique , whereas ISP-MS seems preferable in 5 cases (Sr, Sn, Ba, Ce, Nd). Four elements (Cl, Br, I, Se) could only be determined well by ENAA, whereas the reverse situation applied for another 14 elements (Li, Cu, Ga, Ge, Y, Cd, Te, Pr, Dy, Ho, Er, Tl, Pb, Bi).

It should be added that NAA technique not requiring sample dissolution, therefore has a great advantage if the total concentration is the aim of the analysis. The choice of either multi- element technique thus depends on the purpose of the investigation and the priority of elements.

CONCLUSION

The REGATA experimental setup allows to carry out multi-element analysis of many sample types in different areas of applied studies using conventional and epithermal neutron activation. Because of a high resonance neutron flux of IBR-2 reactor ENAA is especially advantageous for determining many rare-earth elements and heavy metals due to their high resonance activation integral/thermal neutron activation crosssection ratio. Thus this technique allows one to determine about 45 chemical elements in air pollution studies.

The development of the GIS (geographical information system) technology for the examined areas with interfaces to the already existing international Geographical Information Systems: ARCO-INFO, MAP-INRO, GIS-INTEGRO, GREENVIEW, and others, allow us to present the results of our study in a simple and illustrative way to a wide variety of users from different fields of science and practices.

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REFERENCES

[1] M.V.Frontasyeva, E.Steinnes, Epitermal neutron activation analysis for studying the environment. Proc. Int. Symposium on Harmonization of Health Related Environmental Mesurements Using Muclear and Isotopic Techniques (Hyderabad, India, 4-7 November, 1996), IAEA 1997, p. 301-311.

[2] V.F. Peresedov, A.D. Rogov, Simulation and analysis of neutron energy spectra from irradiation channels of the IBR-2 reactor. *J. Radioanal. and Nucl. Chem., Letters*, 214(4), 277-283 (1996).

[3] V.M.Nazarov, S.S.Pavlov, E. Herrera, M.V.Frontasyeva, Recent developments of radioanalytical methods at the IBR-2 pulsed fast reactor. *J. Radioanal. and Nucl. Chem.*, 167 (1993)11-21.

[4] V.M. Nazarov, S.S. Pavlov, V.F. Peresedov, M.V. Frontasyeva, Channels for irradiation and pneumatic system at IBR-2. *JINR Rapid Communications*, No.6-85, Dubna, 1985, p.37-41; Development of activation analysis at IBR-2 reactor. In Proceedings “Activation Analysis at Nuclear Reactors”, D14-88-833, Dubna, 1988, p. 6-22 (in Russian).

[5] T.M. Ostrovnaya, L.S. Nefedyeva, V.M. Nazarov, S.V. Borzakov, L.P. Strelkova. Software for INAA on the basis of relative and absolute methods using nuclear data base. In Proceedings “Activation Analysis in Environment Protection”, D14-93-325, JINR, Dubna, 1993, p. 319-325.

[6] V.F. Peresedov. Atmospheric aerosols and possibilities of their analysis for elemental content at the IBR-2 reactor. *JINR Rapid Communications*, No. 5[85]-97, Dubna, 1997, p.43-50.

[7] M.V. Frontasyeva, A.B. Ramadan, T.Ye. Galinskaya. Weekly cycles of element pollutants in air of the Greater Cairo Area (Egypt) studied by neutron activation analysis. Book of Abstracts, International Aerosol Conference, 26-30 June, 2000, Moscow, Russia.

[8] Ake Ruehling and Eiliv Steinnes. Atmospheric Heavy Metal Deposition in Europe 1995-1996, Nord. 1998 : 15, pp.67.

[9] V.M. Nazarov, M.V. Frontasyeva et al. Resonance neutrons for determination of elemental content of moss, lichens and pine needles in atmospheric deposition monitoring. *J.Radioanal. and Nucl.Chem.*, Vol. 192, No.2 (1995) p. 229-238.

[10] M.V. Frontasyeva, E. Steinnes, S.M. Lyapunov, V.D. Cherchintsev, L.I. Smirnov. Biomonitoring of heavy metal deposition in the South Ural Region: some preliminary results obtained by nuclear and related techniques. *Preprint of JINR*, E14-99-257, Dubna, 1999; *J.Radioanal. and Nucl.Chem.*, No. 2 (2000) (in press).

[11] M.V. Frontasyeva, Ye.V. Yermakova, E. Steinnes. Reliability of mosses (*Hylocomium Splendens, Pleurozium Schreberi and Calliergon Geganteum*) as biomonitors of heavy metal atmospheric deposition in Central Russia. Frank Laboratory of Neutron Physics Annual Report-1999, JINR, 2000-81, p. 178-179.

[12] M.V. Frontasyeva, S.S. Pavlov, L.P. Strelkova, E. Steinnes, N.V. Kirpichnikova, A.V. Bogdanov. How Konakovo power plant affects the environment? In the Book of Abstracts, VIII International Seminar on Interaction of Neutrons with Nuclei (Dubna, May17-20, 2000). E3-2000-71, p. 127.

[13] M.V. Frontasyeva, K. Grodzinska, E. Steinnes. Atmospheric deposition of heavy metals in two of the most polluted areas in the world: the copper basin in Poland and the South Ural mountains in Russia. Int. Conf. Modern Trends in Activation Analysis, MTAA-10, Bethesta, Maryland, USA, April, 1999, p. 117.

[14] A. Lucaciu, M.V. Frontasyeva, E. Steinnes et al. Atmospheric deposition of heavy metals in Romania studies by the moss bimonitoring technique employing nuclear and related analytical techniques and GIS technology. *J.Radioanal. and Nucl.Chem*, Vol. 240, No. 2 (1999) 457-458.

[15] O. Stan, A. Lucaciu, M.V. Frontasyeva, E. Steinnes. New Results from Air Pollution Studies in Romania, *Preprint of JINR*, E14-2000-126, Dubna, 2000.

[16] M.V. Frontasyeva, E. Steinnes. Epithermal Neutron Activation Analysis of Mosses Used to Monitor Heavy Metal Deposition Around an Iron Smelter Complex. *The Analyst*, vol. 120, No. 5 (1955) p. 1437-1440.

[17] O.A. Stan, Zh.H. Zhang, M.V. Frontasyeva, E. Steinnes. Selection of appropriate moss biomonitors for studying atmospheric elemental deposition in China. Frank Laboratory of Neutron Physics Annual Report-1999, JINR, 2000-81, p. 181-183.

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