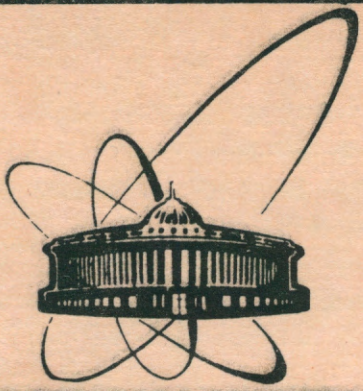

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D14-91-395

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RECENT DEVELOPMENT OF RADIOANALYTICAL METHOD AT IBR-2 PULSED FAST REACTOR OF THE JINR

Submitted to International Conference
"Modern Trends in Activation Analysis",
16-20 September, 1991, Vienna, Austria

1991

Introduction

A many year experience acquired in the use of the neutron activation analysis at the IBR-2 reactor [1] of LNPh.JINR has shown the following directions to be most productive:

1. Multielement NAA using mainly resonance neutrons to investigate the distribution of trace elements (including toxic) in environmental materials and agricultural products [2,3,4].

2. NAA to independently identify trace or impurity elements in the bulk and over the surface of pure and high-purity materials such as Si, Ge, Al, Be, V, Cu, Nb, SiO_2, Al_2O_3, MgO, BeO, polymers and organic solvents [5,6].

3. NAA of geological samples aimed at the determination of rare-earth elements along with other trace elements, which are of interest for geochemistry and geology [7].

4. Analysis of a gold content at all stages of gold search and enrichment in a wide range of its concentration from 10^{-4} up to 10^{-12} g/g regardless of natural particle size distribution.

5. Use of prompt γ-rays following neutron capture for the determination of a stoichiometric composition of various compounds (e.g. ceramics, HTSC) and alloys with SD no worse than 2-5 % [1].

6. Concentration analysis of impurity or trace elements in synthetic and natural crystals like corund, spinel, granite, beryl, topaz, aimed at the study of the impurity atoms influence on the formation of radiation (neutron) dye centres.

7. Study of slow and fast transfer processes of water, hydrogen, organic liquids in catalysts, sorbents, cements and concretes by means of static and dynamic radiography methods and also by using gamma-rays following neutron capture in hydrogen.

Experimental installations for NAA and Radiography Analysis

In order to conduct the studies outlined in the **Introduction** the IBR-2 reactor has four sites equipped for sample irradiation, their transportation, intermediate storage, repacking and measurement. There

is also a many purpose neutron beam-line on the reactor. Table 1 summarizes main parameters of the irradiation sites near the core of the IBR-2 reactor.

Table 1.

The physical and technical parameters of irradiation sites

Irrad. sites	Neutron flux density ($n/cm^2 s$) 10^{12}			Temperat. at irrad. site, ° C	Container diam., mm	Max. number of containers	Transport-system
	thermal λ=1.8 Å	epitherm. > 0.55 eV	fast >0.5 MeV				
P1	Cd coat	0.23±0.3	1.4 ±0.2	70	28	7	pneum
P2	0.54±0.1	0.12±0.01	0.64±0.1	60	28	7	atic
P3,P4	4.0 ±0.5	0.4 ±0.04	4.2 ±0.5	30-40	30	10	mechan.

The internal height of the container is 40 mm. Polyethylene, teflon and aluminum containers are used. The analysis is performed in one of the three possible regimes in dependence on the material analyzed and the elements to be identified.

Table 2

Regime	Container material	Time		
		irradiation	decay	measurement
1	polyethylene	5 -10 m	2- 5 m	5 -15 m
2	teflon	0.5- 3.0 h	10-30 h	0.5- 2.0 h
3	aluminum	3 -10 d	3-10 d	0.5- 6.0 h

When short-lived (within second range) isotopes are studied, one can use the cyclic NAA [1]. The transportation time from the irradiation position to the measurement site is 10 - 15 s. This kind of analysis is used mainly for the experimental determination of Sc, Se, Rh, As, In, Dy, Yb and Hf concentrations in environmental and biological samples. The cyclic NAA could be potentially used for a search of Hf in zirconium.

The gamma-ray spectrometry of samples is performed by using

Ge(Li) detectors having the resolution 2.5-3 keV for Co^{60}. The detectors were calibrated using standard solid or point gamma-sources. The element concentration was determined by absolute comparison and relative comparison methods. As the multielement standards, MES, we used both Soviet and international standards, and the JINR made ones on the basis of SiO_2 and Al_2O_3 chromatographic powder products of the MERCK firm. The NAA results were processed by the programs developed at the JINR for the IBM PC/AT computer.

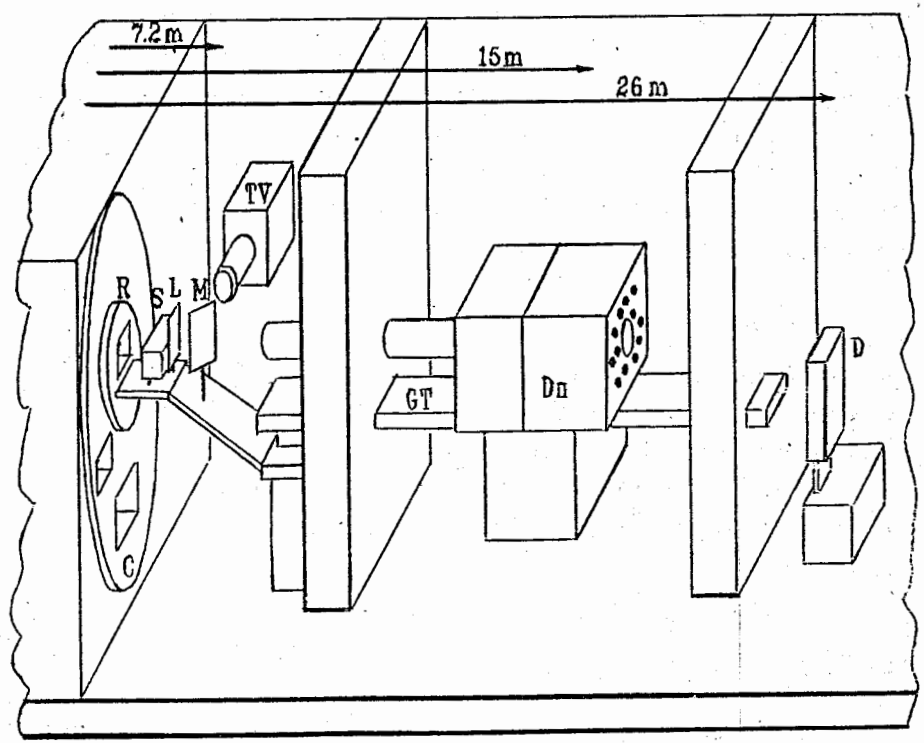

Fig.1. Equipment lay-out.

C - changeable collimator; RC - rotating collimator, S - sample, L - convertor ($ZnS + {}^6LiF$); M - mirror, TV - vidicon with TV camera, GT - mirror neutron guide tube, D_n - fast neutron detector ($\varepsilon \sim 40$ %); D - Ge(li) and Si-detectors

In the neutron beam-line there are three positions at 7.2,15 and 27 m from the reactor core equipped with beam-shaping devices and a set of detectors. Their disposition on the beam-line is shown schematically in Fig.1. These instruments are used for static and dynamic neutron radiography and the elemental analysis with prompt neutron capture reactions (n,α), (n,p),and (n,γ) at the 27 m site and with (n,f) at the 15 m site.

Usual static neutron radiography studies are carried out by the time of flight method on all the above sited flight paths, while dynamic radiography studies just on the 7.2 m path with the possibility to vary the incident neutron energy from 1.0 up to 10^{-3} eV. Modern video-equipment together with the ZnS(Ag)+^{6}LiF converter allow one to obtain a stable black and white or color image of the object in one reactor flash with the effective pulse duration for thermal neutrons 1.5 ms at the site and the frequency 5 s^{-1}. The fast neutron pulse duration at half maximum is 250 μs.

The beam parameters at the 7.2 m site

Average thermal neutron flux density	-	2 10^{8} n/cm^{2}s
Ratio L/D at full open shiber	-	720/20
Ratio L/D at half-close shiber	-	720/4
Average dose rate of fast gamma-rays	-	0.26 g/m
Average fast neutron flux density (E= 1.3 MeV)	-	8.4 10^{8} n/cm^{2}s
Ratio of gamma-rays to neutrons dose rate P_γ / P_n	-	0.25

With a 4 cm Pb filter installed, the ratio P_γ/P_n decreases down to 0.02 following a threefold decrease in thermal and fast neutron flux density. A fast automated (15 s) change of collimators is possible to produce beam cross-sections of 5 x 5, 10 x 10 и 15 x 20 cm^2. The 15 m site is equipped with a mechanical collimator which having transmitted thermal neutrons with the neutron density on sample 2.4 10^{7}n/cm^{2}s helps to reduce the fast neutron background between pulses by a factor of 10^{4} ÷ 10^{5}. To the 27 m site a pure thermal neutron beam is guided via a

curved Ni-coated glass mirror tube 20 m long with a rectangular cross section 15 mm by 150 mm, the thermal neutron (λ > 2.0 Å) flux density is 2.5 10^{6} n/cm^{2}s. No neutrons with wave-lengths less than 1.5 Å are practically supplied.

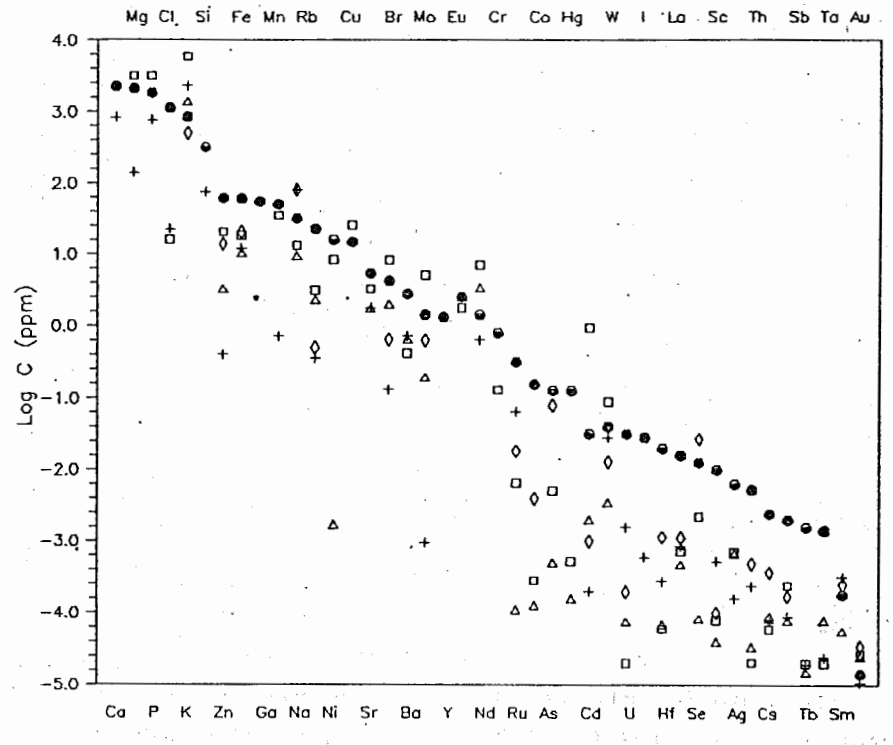

Fig.2. Element concentration distributions in rye (●), potato starch (+), buckwheat (□), wheat (Δ), rice (◇). ⊙ - detection limit

EXAMPLES OF NAA APPLICATION AT IBR-2 REACTOR

1. Fig.2 demonstrates the results of multielement analysis of corn using resonance neutrons. Concentration distributions of the main and trace elements in rye, wheat, buckwheat and potato starch were obtained for samples of up to 2 g. Weight three regimes of analysis reported in Table 2 were used. Standard deviations, SD, of absolute concentrations were found to lie in the interval from 5 to 30 %.

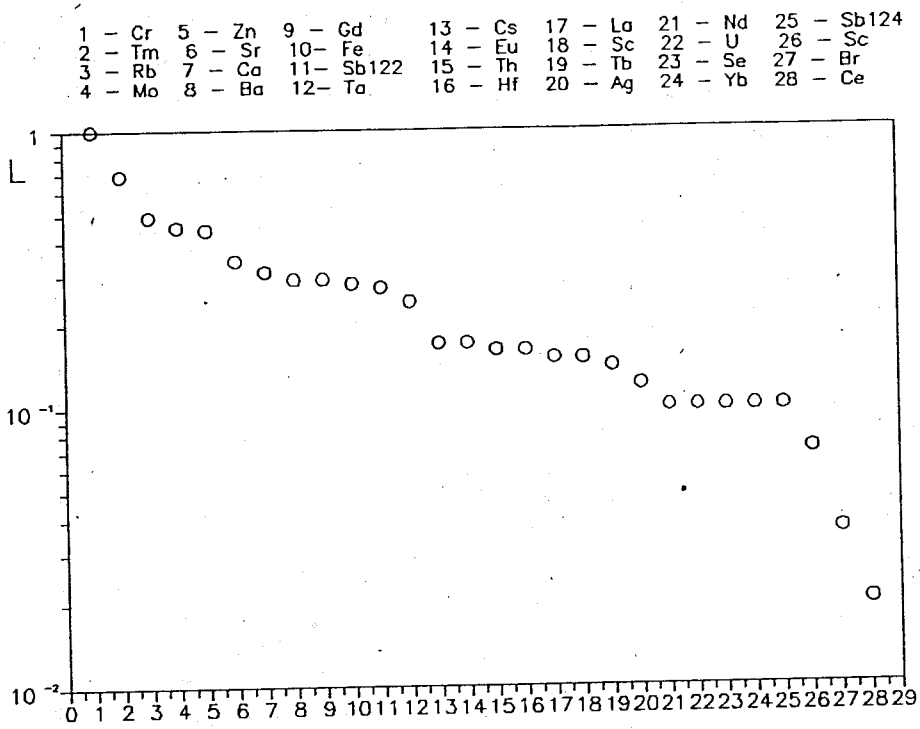

Fig.3

This multielement analysis of corn had the aim to find ecologically pure corns for children's food production. In the large scale analysis of corns from different districts of this country it is planned to analyze ashes after corn has been burnt at high temperature, with preliminary determined volatility for constituent elements and their complexes. Fig.3 shows volatility values for some elements in rough rice corn. Larger volatility values for Br in comparison with the other elements and smaller weights of ash samples make the INAA much easier. In an analogous manner the other agricultural plants, herbs, moss, lichens and mushrooms can be analyzed.

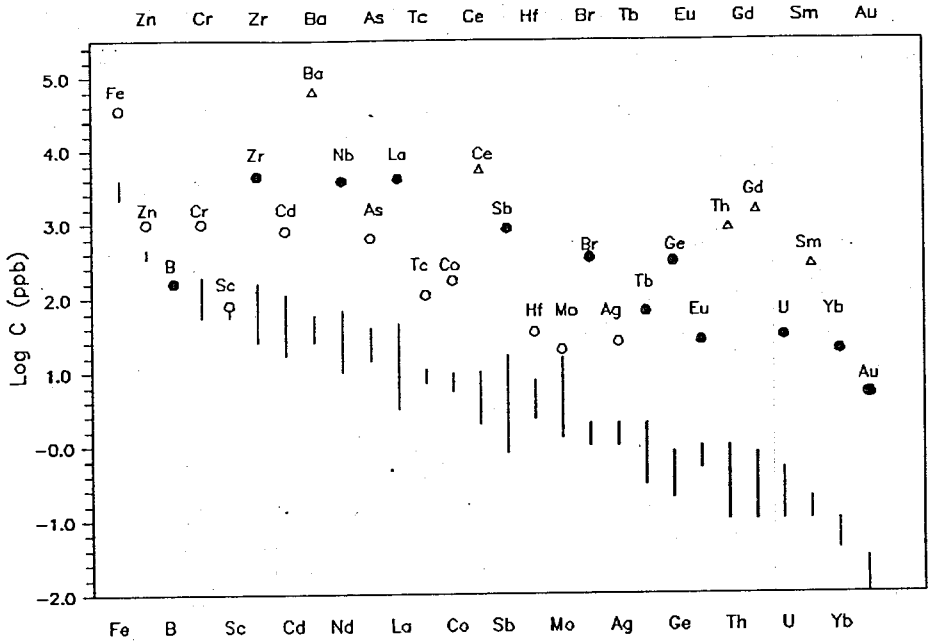

Fig.4.

| - concentration along a pure area of 60 cm length;

o - concentration along a contaminated area of 10 cm length (the purification rate is 0.1 - 0.3)

● - the same, at the purification rate of (0.01 - 0.1)

Δ - the same, but the purification rate is 0.001 - 0.01

2. Fig.4 illustrates the use of resonance neutrons in the INNA of high-pure aluminum obtained by zone melting. The range of element concentrations obtained is determined by the change in element concentration along the pure part of the ingot. The results illustrated were obtained by measuring 6 g samples for 7 days of irradiation and exposure time. Boron concentration was determined with the help of the $^{10}B(n,\alpha)^{7}Li$ reaction by measuring 478 kev γ-rays on the neutron beam

The results of the INNA of chromatographically pure fine powders of SiO_2 and Al_2O_3 are shown in Fig.5. When analyzing high-pure materials their surface pollution is removed by means of usual acids or solvents

after irradiation. As a rule the detection limits are given only for the elements found on the surface.

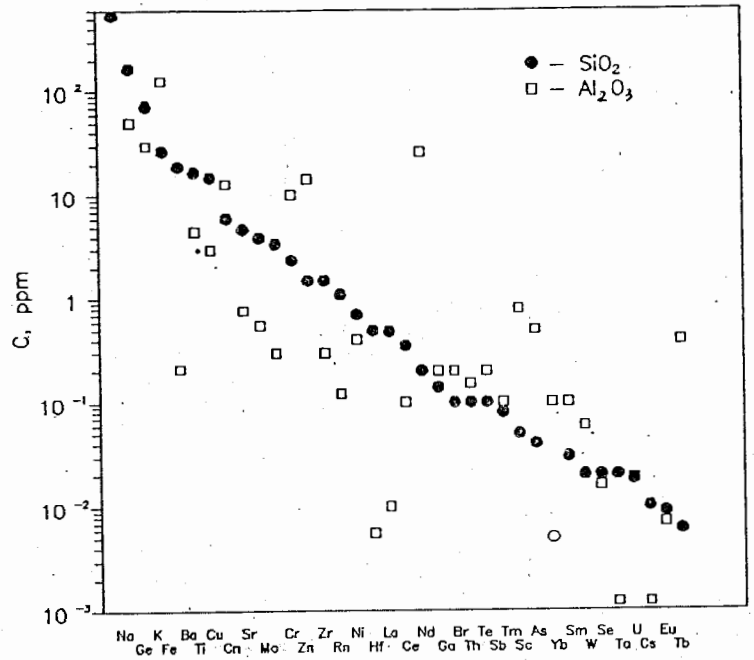

Fig.5

Table 3 summarizes the detection limits obtained in three regimes of the INAA with resonance neutrons for 4 g samples of high-pure silicon.

Table 3

Group of elements	C, ppb SD = 50%
Au	0.0003
Sm Sb Ir	0.002
Tb Lu Ta Th U	0.006
Eu Tm Yb Sc Mn As Br Ag	0.03
La Se Mo Cd Hf W	0.10
Ce Na Ni Rb In Pt Hg	0.30
Nd Gd Co Ga Ru Te Ba	1.00
Cr Zn Sn	2.00
Sr Zr	15.00
Fe	70.00

Practically all the elements mentioned, but iridium, are usually found on the surface with SD no worse than 30 %.

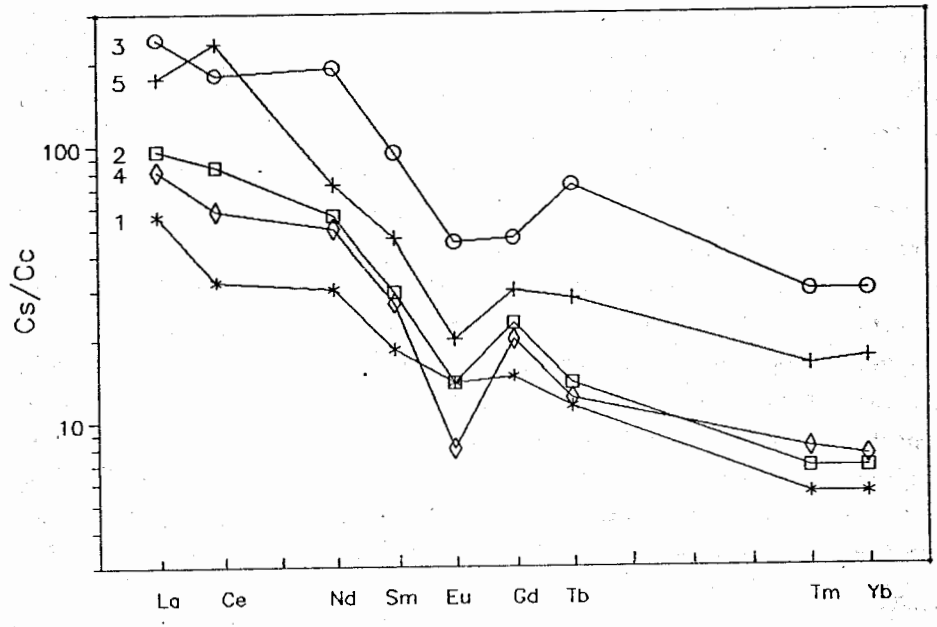

Fig.6. C_s and C_c are the concentrations of elements in the sample and chondrites, respectively.
1 - averaged C_s/C_c values at the depth of 1648 and 1808 m, 4 - the same at the depth 2311 and 2421 m, 2 - C_s/C_c at the depth of 1993 m; 3 - the same at the depth of 2160 m; 5 - C_s/C_c for the SL-1 standard sample

3. As the example of the INAA application in geology the variation diagrams of concentration distributions of rare-earths, normalized to their content in chondrites, are given. These diagrams were obtained from the study of the element distribution, including rare-earths, along oil wells in one of districts of Cuba. A significant increase in REE content 300 m from the oil pool at the 2400 m depth attracts one's attention (diagram 3). Besides, along distances of just 150 -160 m before and behind this maximum the rare-earth content decreased by several times (diagrams 2 and 4). The same investigation has also yielded

the data on the distributions of Si, Ca, Fe, Mn, Al, K, Na, Sr, V, Cl, Zn, Ba, Mo, As Rb, as their concentrations decreased from 10^5 up to 10^{-1} ppm along the 2421 m well.

4. It is for several years already the JINR group in collaboration with the Institute of Geology of the Academy of Sciences of the USSR have been carrying out investigations by INAA and other nuclear methods of the effect of fertilizers and wastes on the environment pollution with REE's [4]. The results are reviewed in the contribution to this conference by Dr. M.V.Frontasyeva and co-authors: "The Effect of Phosphorus Fertilizers on the Environment".

5. During the last two years our scientists together with the Institute of Theoretical and Experimental Physics (Moscow) and GSI (Darmstadt) were measuring, using the NAA methods the sputtering cross-sections of Au atoms for their interaction with heavy ions [7]. In these experiments a 0.4 μ foil was bombarded by heavy ions and the Au atoms emitted in result were collected on a carbon layer of about 1 μ thick. The number of Au atoms deposit on the collector is obtained with NAA using resonance neutrons. Fig.7 shows the dependence of the

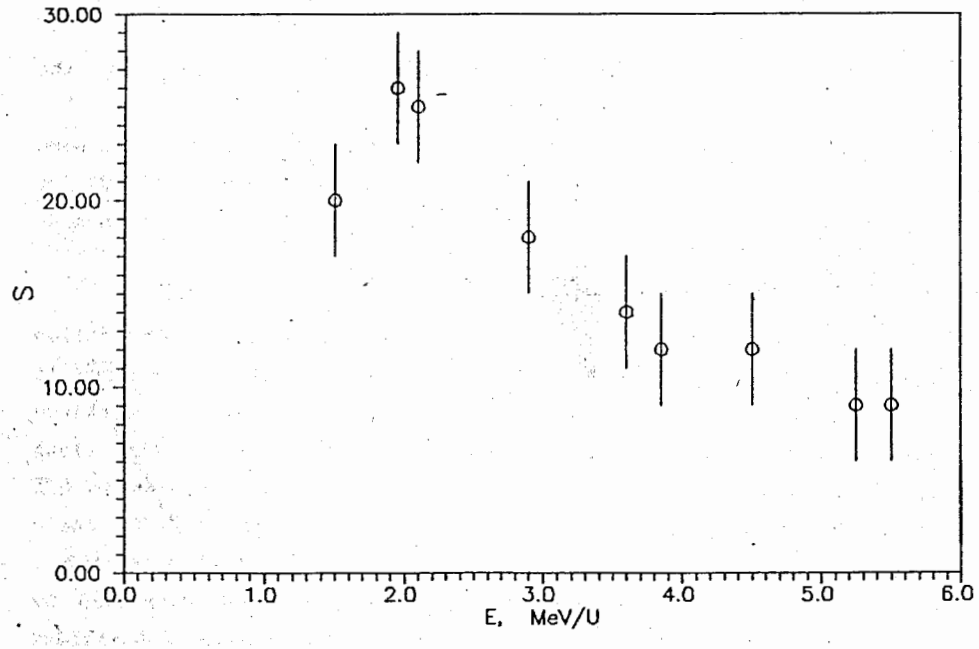

Fig.7. S is the cross-section in atoms/ion (relative units)

sputtering cross-section of Au (in atoms/ion) on the energy of ions (in MeV). For the U ions with the energy 5.5 MeV/U the absolute cross-section value was obtained to be (12 ± 2) atoms/ion.

6. In the Laboratory of Neutron Physics work continues on the use of isotopic sources of neutrons for express determination of nitrogen content in natural and synthetical materials by measuring prompt γ-rays following thermal neutron capture in nitrogen. The instrument "Azot", based on this principle has been designed and tested in industrial conditions, to measure the protein content in corns and mixed feed. The instrument uses the Cf neutron source, which produces 5 10^7 n/s. 2.223 MeV γ-rays from hydrogen and 10.828 MeV from nitrogen are registered by the NaJ(Te) detector measuring 150 x 100 mm^3. The material (8-10 l in volume) under measurement plays at the same time the role of the neutron moderator in the instrument. Here to compensate the influence of the change in material density and composition the number of registered quanta from nitrogen nuclei is normalized to the thermal neutron flux density in the sample and to the number of registered quanta from hydrogen. Standard SD deviation in protein detection is in the range from 8 up to 70 % during one 5 min run and satisfies the requirements of the Kjehldahl method.

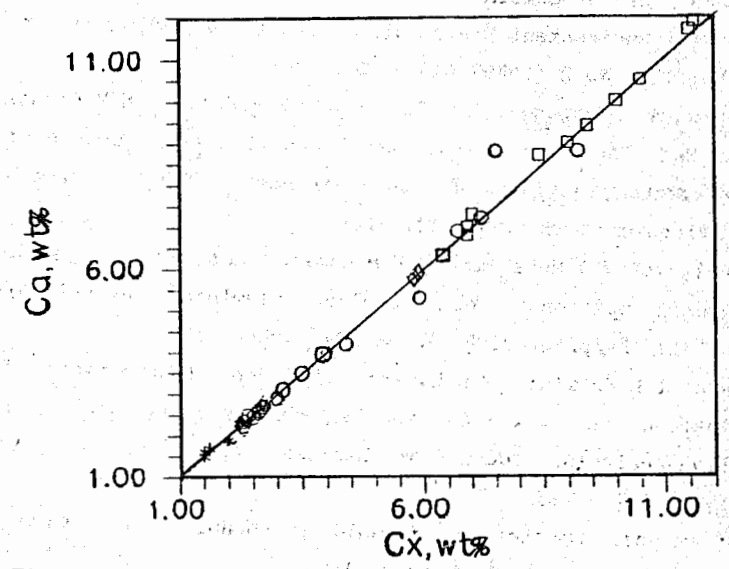

Fig.8

Fig.8 shows the ratio C_a/C_x, where C_a is the nitrogen concentration in the sample, C_x is the same, but measured by the Kiehldahl method. This ratio is calculated using one and the same calibration for all the materials from rice to protein vitamin concentrate and fish flour. Some points are outside the error corridor which could be explained by poor free-flowing capacity of the given products. The instrument comprises devices to control the chamber loading and to measure the material density by neutron absorption.

The given examples of the application of the radioanalytical methods based on the use of neutrons show, that their improvement and development would promote their successful use in both fundamental and applied research.

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Received by Publishing Department
on October 2, 1991.

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Д14-91-395

Современные направления развития радиоаналитических методов на импульсном быстром реакторе ИБР-2 ОИЯИ

Обсуждается опыт использования радиоаналитических методов, включая НАА, на импульсном быстром реакторе ИБР-2 ЛНФ ОИЯИ. Приводятся физико-технические характеристики экспериментальных установок, используемых для НАА и радиографии. Даются конкретные примеры применения резонансных нейтронов в контроле окружающей среды, в геологии нефти, в многоэлементном анализе продуктов питания и особо чистых материалов, в ядерной физике. Сообщается о работах по применению изотопных источников нейтронов для экспрессного определения содержания азота в природных и синтетических материалах.

Работа выполнена в Лаборатории нейтронной физики ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1991

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D14-91-395

Recent Developments of Radioanalytical Methods at IBR-2 Pulsed Fast Reactor

The experience of the use of radioanalytical methods, including NAA at IBR-2 pulsed fast reactor of the JINR, is discussed. Physical and technical parameters of the experimental installation designed for NAA and radiography are given. The detailed examples of the application of resonance neutrons to the control of the environment in the geology of oil, in multi-element analysis of food products and superpure materials as well as in nuclear physics are reviewed. The works on the application of the neutron isotopes sources for express determination of nitrogen content in original and synthetic materials are introduced.

The investigation has been performed at the Laboratory of Neutron Physics, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1991