POSSIBLE APPLICATIONS OF NEUTRON ACTIVATION ANALYSIS AT THE RB REACTOR FOR THE ENVIRONMENTAL MONITORING

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ABSTRACT

This paper presents the possibilities of performing neutron activation analysis in Yugoslavia for the purpose of environmental pollution monitoring. Neutron activation analysis (NAA) is an established method for the determination of trace elements in a broad range of samples. It is non-destructive method used in a routine manner with gamma-ray spectrometry for various applications in biology, environmental toxicology, radiation assessment, mineral exploration, trace elements monitoring regarding human health studies. As a complex, powerful analytical tool, it might give results even when other analytical methods fail. NAA has been performed at the Nuclear Engineering Laboratory (NET), the VINČA Institute of Nuclear Sciences, for various applications in the reactor physics research. The samples are irradiated in the RB research reactor, which is the zero-power, bear, heavy water critical facility utilising three fuel element types. The reactor system is very flexible and allows for different core configurations, resulting in various neutron fields, with energy spectra ranging from thermal to fast (in the coupled fast-thermal system HERBE). There are several vertical and horizontal experimental channels available for sample's irradiation. The achievable thermal neutron flux is approximately 10^7 n/(cm^2 s) per 1 W of fission power. We believe the RB reactor could be successfully used for neutron activation analysis of trace elements in the environmental samples, comprising short-lived isotopes. Although the flux level and restricted irradiation time pose a certain limitation to NAA applications at the RB reactor, it can be compensated by the specific methodology advancement. That involves overcoming the main sources of error in the instrumental NAA evaluation methodology which utilises generally valid k_0, and Q_0 factors. In particular, the computational accuracy of required nuclear parameters (e.g. neutron flux distribution, effective nuclear cross sections) can be increased and evaluation method improved. Knowledge and experience of NET Laboratory in implementation/modification of nuclear data libraries and

corespondent data processing computer codes, as well as implementation/development of radiation transport calculations codes, give us prospect to go a step ahead the commonly used instrumental NAA. At this moment, the main disadvantages of the RB rector use for considered NAA applications are the lack of measurement instrumentation and necessary supplementary equipment and tools. With certain instrumentation upgrade and acquiring of updated nuclear data libraries and codes, it would be possible to found the NAA service facility for wide range of applications in environmental studies of stable and radioactive contaminants in atmospheric, terrestrial and aquatic environments, and human tissue samples.

Key words: nuclear reactor, irradiation, neutron flux, neutron activation analysis, environmental monitoring

1. Introduction

This paper presents the prospects of performing neutron activation analysis in Yugoslavia for the purpose of environmental pollution characterisation and monitoring.
Neutron activation analysis (NAA) is non-destructive method used in a routine manner with gamma-ray spectrometry for the trace elements determination in a broad range of samples. It allows for simultaneous determination of many elements in analysed sample. As a complex, powerful analytical tool, it might give results even when other analytical methods fail. Other instrumental multielement methods of trace analysis which are in competition with NAA are XRF (X-ray fluorescence spectroscopy, including total XRF), OES-ICP (optical emission spectroscopy with inductively coupled plasma), MS-ICP (mass spectroscopy with ICP) and SS-MS (spark source mass spectroscopy). NAA is superior to other techniques for trace elements analysis in the cases were volatile elements, such as the halides, are present and may be lost during dissolution or chemical processing.
Since the NAA utilises signals that are related to the atomic properties of the nucleus, the results are not affected by the chemical or physical state of the elements. The method is well described by physical laws and selectivity is unambiguous for all elements. The combination of nuclear properties such as decay constant and the energies and intensities of the γ radiation is uniquely characteristic for each radionuclide. These facts contribute to a high degree of accuracy that make NAA well acknowledged for analysis related to various fields, including the certification of reference materials /1/.

2. Basic principles

Analytical method applied to obtain results of NAA utilises activities of the (n,γ) activation products, the neutron fluxes and the required nuclear parameters. The presence of a particular element in the irradiated sample is determined from it's γ activity. Activity of a nuclide is computed from its γ -lines pattern and their intensities (contained in the code library), folded with detector's efficiency function which is energy-dependent.
The widely used evaluation methodology utilises generally valid analytical k_0, and the pertinent Q_0 factors /2/ (applicable to all nuclear reactors and irradiation facilities). This

was possible by treating separately the contributions of thermal and epithermal neutrons. These factors also provide set of equations which could be easily converted into a computerised and largely automated procedure for the analytical results evaluation. De Corte and Simonits have measured accurate k_0, and the pertinent Q_0 values for nearly all elements important for NAA /2/. Zirconium is introduced as a double neutron flux monitor: for simultaneous measurement of thermal & epithermal flux.

2.1 Precision and accuracy

Irradiation and radiation detection conditions had to be adapted with respect to materials to be analysed and elements to be determined.
Samples of well defined, regular shape (cylindrical disks optimal) enables reproducibility better than 0.6%, even when trace element contents is in ng/g range.
Main sources of error are:

- neutron flux gradients,
- γ ray self-absorption (especially in large samples),
- γ - γ coincidence occurring with γ ray counting.

Detection limits are valid for the particular element in a given sample analyzed under the stated experimental conditions, and must not be considered as general or typical detection limits of the method. However, concentrations as low as ng/g can be measured /3/.

3. Applications

NAA has various applications in material science, chemistry, biology, radiation assessment, mineral exploration, medicine and other fields.

- **Medicine**

Trace elements monitoring regarding human health include both normal values measurement and specific disorders (diabetes, multiple sclerosis) determination.

- **Mineral exploration**

NAA is convenient method because dissolution of rock sample is not required prior to analysis. It appears to be the most sensitive method for direct determination of gold (for concentrations down to 10μg/kg), and rare earth elements (La->Lu) tracing.

- **Industry**

Industrial production of high purity materials requires measurement of impurities - trace elements at ng concentrations /4/. High purity quartz glass, mainly used for optical purposes, contains trace elements that reduce light transmission at certain wavelengths. This might be very important for using glass in specific environments, and therefore is necessary to determine the quality and quantity of impurities. High purity aluminium used in microchips manufacturing must not contain uranium and thorium over certain limits, which are less than 1ng/g or even 0,1ng/g .

NAA can be applied to particular material for cross-checking of analytical results obtained by other methods in industrial analytical laboratories, or to compare materials from different producers. It also may be employed as analytical method in the purification procedure development, in quality control of pre-products and products, and their relationship with element concentrations in the rooting zone. Plants which selectively accumulate the element of interest are used for that purpose.
Analysis of plastic and carbons used in food industry is of significant importance as well.

- **Bio-geological exploration**

Exploration of mineral deposits is based on a quantitative study of element accumulation in plants

- **Environment**

NAA is used for pollution characterisation and monitoring of soils, dust, sea sediments, air. Determination of heavy metal toxicity, i.e. the presence of heavy metals (As, Hg) in various environmental and food samples, is among major interests for environmental protection.
Additionally, relationship of these values with human levels in hair, blood and urine can be examined. Hair accumulates toxic metals and therefore is good environmental monitor for 20 to 25 elements that can be measured.

- **Radiation assessment**

The environmental impact of radionuclides released from nuclear power plants can be assessed by:

- determination of ^{99}Tc & ^{129}I with very high sensitivity,
- estimation of radionuclides uptake by food corps,
- evaluation of transfer factors of radioactive fallout nuclides from soils to plants.

NAA can also help to detect a certain types of undeclared activities violating nuclear non-proliferation and arms control regulations. Environmental samples (river or lake water, soils) taken in the vicinity of nuclear installations are analysed to determine any possible change in the natural uranium content and isotopic composition. Highest sensitivity and selectivity are necessary for both uranium assay and isotopic ratio U^{235} / U^{238} determination. NAA is 105 times more sensitive in the determination of uranium compared to gamma or alpha spectrometry. Uranium contents of 1ng are detectable in environmental sample when NAA is applied.

- **Waste characterisation and monitoring**

All around the world there are disposals of hazardous chemical and radioactive waste. Some of them have been disposed for a long period of time. The condition of containers and even it's contents might be susceptive, and consequently, handling them problematic. The NAA is the most convenient, non-destructive method for analysis, where containers do not have to be decomposed in any way prior to analysis.

- **DU monitoring**

Depleted uranium ammunition has been used during last decade in several conflicts in the world. The environmental impact of depleted uranium depends on the specific situation where DU ammunitions are used and the physical, chemical, and geological characteristics of the environment affected. The main potential hazard associated with DU ammunitions is the inhalation of the aerosols created when DU ammunitions hit an armoured target. In the long term, the exposure pathways that become more important are ingestion of DU incorporated in drinking water and the food chain through migration from the soil or direct deposition on vegetation. It has also been estimated that a large fraction of DU ammunitions fired from an aircraft probably miss their intended target. The majority of these projectiles will be buried at various depths under the surface of the ground and even in buildings.
Therefore, to assess the environmental impact of depleted uranium, it is necessary to take various samples (soil, water, biological samples) from affected areas. Samples should be analysed to determine whether or not DU is present, and to estimate the possible doses from different sources in that environment.

4. The RB reactor

NAA has been performed at the Nuclear Engineering Laboratory (NET), the "Vinca" Institute of Nuclear Sciences, for various applications in the reactor physics research. The samples are irradiated in the RB research reactor /5/, operated by the NET Lab.
The RB is the only available irradiation facility in Yugoslavia. It is the zero-power, bear, heavy water critical facility utilising three fuel element types: natural uranium, the 2% enriched metal uranium and the 80% enriched uranium dioxide fuel elements. Reactor system is very flexible and allows for different core configurations, resulting in various neutron fields ranging from thermal to fast (in the coupled fast-thermal system HERBE).
Several vertical and horizontal experimental channels are available for sample's irradiation. Achievable thermal neutron flux is approximately 10^7 n/(cm^2 s) per 1W of fission power, and neutron fields' characteristics are given in Table 1.

Table 1 Characteristics of the neutron fields at the RB reactor

Neutron field at RB reactor	Field's position in reactor	Experimental space		Neutron flux [$cm^{-2}s^{-1}$] at 1W		Dominant neutron spectrum
		Diameter [mm]	Length [m]	Thermal (< 0.465 eV)	Fast (> 0.8 MeV)	
Horizontal experimental channel (HC)	RB core & reflector	17	2.0	$1\ 10^7$	$5.3\ 10^5$	thermal
Vertical experimental channel (VC)	RB core & reflector	110	2.3	$1\ 10^7$	$5.3\ 10^5$	thermal

The RB reactor could be successfully used for NAA of trace elements in the environmental samples, comprising short-lived isotopes. Although the flux level and restricted irradiation time pose a certain limitation to NAA applications at the RB reactor, we believe it can be compensated by the specific methodology advancement.
The NET Laboratory has gained certain knowledge and experience in implementation /modification of nuclear data libraries and corespondent data processing computer codes, as well as implementation/development of radiation transport calculations codes. That gives us prospect to increase computational accuracy of nuclear parameters required for NAA (e.g. neutron flux distribution, effective nuclear cross sections) and to improve evaluation method. The most important improvements that can be introduced are:

- accurate calculation of the neutron flux energy dependence, taking into account fine neutron cross section structure, i.e. resonances and resonant self-shielding effects;
- effects of flux perturbation introduced by the sample (spatial depression of flux at the sample's irradiation position);
- taking into account γ self-absorption in the sample (important for large samples).

4.1. Present status

At this moment, the main disadvantages of the RB rector use for considered NAA applications are the lack of measurement instrumentation and necessary supplementary equipment and tools. With the respect to inquiries this Conference poses to achieve its goal, we here quote what are our needs to successfully perform NAA at the RB reactor:

- new high resolution HPGe detector
- upgrade of measurement electronics (MCA, signal processing instrumentation)
- supplementary equipment (primary adequate rabbit system)
- updated gamma spectrum evaluation codes and gamma libraries
- organise and establish collaboration with other "Vinca" Institute Labs to provide proper sampling and sample preparation.

5. Conclusion

Providing above requirements fulfilled, it would be possible to arrange the NAA service facility at the RB research reactor, available for users (scientists, analytical service departments in industry, academics) involved in environmental characterisation and monitoring. Analysis of stable and radioactive contaminants in atmospheric, terrestrial and aquatic environments, biological and human tissue samples, can be performed by neutron activation analysis at the "Vinca" Institute of Nuclear Sciences.

References

1. "Quality aspects of research reactor operations for instrumental neutron activation analysis", IAEA-TECDOC-1218, International Atomic Energy Agency, Vienna, 2001

2. F. De Corte et all "A Compilation of $k_{0,Au}$ Factors and Related Nuclear Data for 112 Radionuclides of Interest in Neutron Activation Analysis", INW/KFKI Interim Report, Institute for Nuclear Sciences, State University< Gent (Belgium), and Central Research Institute for Physics, Budapest (Hungary), 1986

3. G. Erdtmann et all , Trace and Microprobe Techniques, Vol. 6, pp 337, 1988

4. K.P. Egger, V. Krivan, Analytical Chemistry, Vol. 323, pp 257, 1986 (Na)

5. "HERBE - Finalni sigurnosni izveštaj", IBK-NET-54, Vin~a, 1991