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ACTIVATION ANALYSIS OF ENVIRONMENTAL SAMPLES USING THE MT-25 MICROTRON OF THE FLNR

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Abstract: Instrumental neutron and gamma activation analysis of coal and combustion products, determination of platinum content in natural samples by radiochemical gamma activation analysis and high-sensitive track method of thorium determination has been studied with the use of the MT-25 microtron.The optimal conditions for complete elemental analysis of coal and combustion products, isolation and determination of platinum and thorium are recommended.

Keywords: Microtron, Instrumental neutron activation analysis, Instrumental gamma activation analysis, Coal, Track method, Platinum, Thorium.

1. Introduction

The main purpose developing nuclear-physical methods of analysis at the Laboratory is to work out sufficiently sensitive methods of elemental analysis using the simplest and accessible source of radiation - a compact electron accelerator - MT-25 microtron. It serves as a rather intensive source of both gamma-rays and neutrons, which is sufficient for developing a number of methods for the gamma-ray, neutron activation and radiochemical analysis using high-resolution gamma-spectrometry[1].

The choice of the method and means of its realization play an important role for the success of analysis. The microtron was employed to develop and test the technique of determination of a number of elements on different samples.

We'll touch upon any methods, developed at our laboratory.

One of the technique which has found practical use is the technique determining the element contents in coal and combustion products such as fly ash, bottom ash, which is formed in large amounts and contains many elements. and are one of the main sources of environmental pollution[2,3].

The use of instrumental version of the neutron and gamma activation analysis which belong to the most sensitive ones at the analysis of the majority of noble elements in the case of platinum is very limited[4-6]. Radiochemical neutron and gamma activation analysis are the most perspective ones for platinum determination in natural samples.

The present particular attention is given to the determination of long-lived actinides element contents in environmental samples. Published date on contribution of various radionuclides to human

irradiation dose point on to the significant potential hazard of thorium. Therefore it is necessary to know it mobility in biosphere, accumulation in plants, bioorganisms and behavior in the human body including farm produce.

According to different authors, the global content of thorium in the environment varies in the range of 10^{-12}-10^{-4} Bq kg^{-1} in soils and sediments 10^{-7} Bq l^{-1} in see water[7, 8]

Low specific radioactivity of thorium leads to difficulties in radiometric measurements. Usually for this purpose rather expensive method are used, based on neutron activation analysis (NAA), which nevertheless need prior purification from macro- and microcomponents. Detection limits for highly effective techniques are as follows: 10^{-9} g/g: α-spectrometry, 10^{-11} g/g: NAA[9-12]. Besides, it is known that thorium content can be measured by track methods without isolation componenets, however, the detection limits is not lower than 10^{-9} g/g[12].

The development of chemical techniques, including ion exchange, liquid membranes makes it possible to achieve a high preconcentration and purification degree for thorium from solutions of different nature as well as leaching liquors [11].

We have investigated the possibility determination of the ultra-small thorium concentration by the track method with the use (γ,f) reaction.

2. Experimental

2.1. The MT-25 microtron.

The microtron has the next parameters:

Energy range 10-25 MeV
Pulsed beam current 20 μA
Gamma-ray flux 10^{14} photon·s^{-1}
Thermal neutron flux $8 \cdot 10^{8}$ n·cm^{-2}·s^{-1}
Epithermal neutron flux $8 \cdot 10^{7}$ n·cm^{-2}·s^{-1}
Power consumption 20 kW

The electrons accelerated are extracted from the microtron to a stopping target to produce gamma-rays (*Fig. 1*) or are directed into a uranium-berillium convertor to produce neutrons. The uranium-berillium convertor is placed into the middle of a graphite cube with a 120 cm side., which is the main neutron moderator (*Fig.2*).

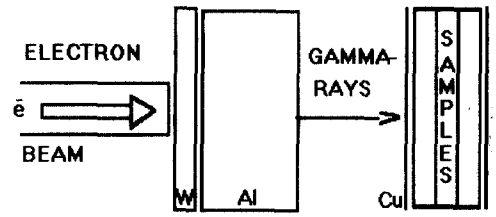

Fig.1. Scheme of irradiation by gamma-rays.

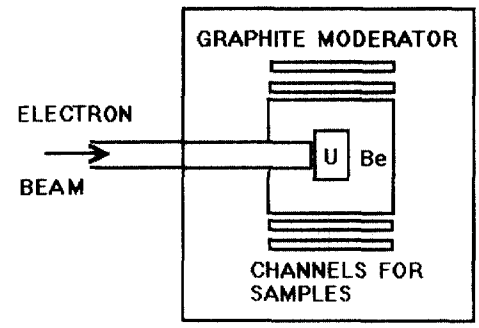

Fig. 2. Graphite cube and uranium-berillium convertor.

2.2. Instrumental activation analysis.

In this study coal samples were analyzed from some coal-fields of the Russia. 25-35 g of each of the coal samples were placed in the cassettes and were analyzed for gold. Samples were placed in two channels around an uranium-beryllium converter and were irradiated with photoneutrons for 10 h. The flux density of the epithermal neutrons in the central part of the channel was the $8 \cdot 10^{7}$n·cm^{-2}·s^{-1} The beam current was of 15 μA with an electron energy of 25 MeV. The use of epithermal neutron for activation allows to decrease by one order of magnitude the background affect due to the thermal neutron activation of coinconmitant elements (sodium, manganese and etc). The detection

limit of gold determination was of about 0.03 g/g for an irradiation time of 10 hours and for 10 min measuring time for each sample. 250 sumples per day can be irradiated simulltaneously.

The contents of trace elements in coal and combustion products were determined with the help of instrumental gamma-activation analysis (IGAA). The coal combustion on the laboratory plant was carried out[13]. There were 15 experiments, in each one 10 kg of coal were burnt. After of the experiment the plant was dismantled, the surface of all the parts of the gas way was cleaned from deposited fly ash. The bottom ash and fly ash were taken from two power plants (Ulan-Bator, MPR). 3-4 g of samples were placed in polyethylene cassettes. A packet consisting of 10 cassettes with samples and a standard sample was irradiated by gamma-rays for 4 h. The electron energy was 25 MeV.

Measurements the unduced activity were performed with the help of a HPGe detector,thin and coaxial Ge(Li) detectors. Each sample was measured from 5 to 60 min depending on the exposure time before the measurement

2.3. Radiochemical analysis.

The samples with standards were irradiated 5 hours by the gamma-quanta beam of the microtron with the energy up to 25 MeV and the average current ≈15 μA. After irradiation the samples containing platinum were dissolved in aqua regia. The solution was evaporated to dryness and taken up in concentrated hydrochloric acid to remove all nitrate derivatives. Then the residue was extracted by the solid extractant (SE 50% TOPO}[14]. The time of the radiochemical procedure was 1 hour. Each sample was measured from 15 to 60 min depending on the exposure time before the measurement.

2.4. Track method.

The solution with the known thorium content was deposited on a 175 μm thick polyethylenterephthalate foil (lavsan) detectors and evaporated. The lexan detector was placed above the foil and fixed.

A packet consisting of 10-15 samples was irradiated by photons of the microtron with an initial energy of 25 MeV and at 15 mA beam current for 4 hours. The detectors were chemically etched in 6N NaOH at 60°C for 3 hours [15]. The fission fragment tracks was observed and counted in an optical microscope at a magnification of (100-300)X.

3.Results and Discussion

Instrumental activation analysis. The use of the microtron permits to determine more than 40 elements in coal which are of interest for industry and ecology with the 10^{-3}-10^{-5}% detection limit.

At combustion of coal the summary mass of fly ash was 1.5-2.2% from the initial coal. The enrichment factors $K=C_{fly\ ash}/C_{coal}$ for some elements are completed in *Table 1*.

Table 1

Enrichment factors $K=C_{fly\ ash}/C_{coal}$

Element	K	Element	K
Zn	35	Re	190
Ga	25	Os	125
As	40	Hg	230
Se	20	Tl	15
Mo	30	Pb	35
Cd	16	U	1.5

The elements contents in coal, bottom ash and fly ash on heat power plants (MPR) as well as the distribution of the elements during a combustion of brown coal (Poland) on the laboratory plant is shown in *Fig.3*.

From the received of results can draw a conclusion about the principal possibility to model the behavior of elements on a heat power plants during the combustion of coal on the laboratory plant.

By the coal combustion used as an energy source in heat power plants (HPP) the elements were concentrated in bottom ash, fly ash or gas phase

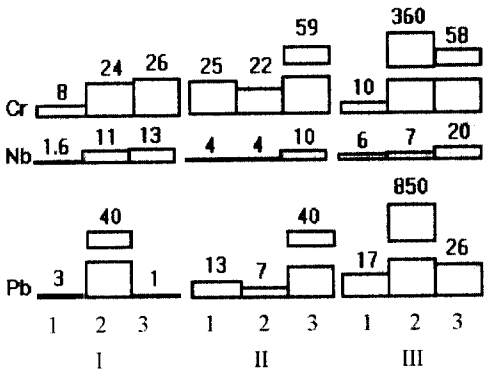

Fig. 3. The content of elements in coal, fly ash, bottom ash, g/t.
I-HPP-1, II-HPP-2 (MPR), III-the laboratory plant, brown coals (Poland).
1-coal, 2-fly ash, 3-bottom ash.

depending on their physical and chemical properties. The coal combustion led to a decrease on the sample mass. This can be used for a decrease of the detection limit for some trace elements[12].

Radiochemical analysis.

The most intensive peak is the peak with the energy of 77 keV carrying to isotope Pt-197 ($T_{1/2}$ = 18 h). This isotope was used for platinum determination.In the *Table 2* the results of platinum determination in the reference samples are given.The platinum determination limit for instrumental gamma activation version was 100 μg and in the case for radiochemical version - 0.1 μg.

At radiochemical analysis the background is minimum in the region of 77 keV and the principal path of decrease

Table 2. The results of platinum determination in the reference samples

No	Platinum content, g/g	
	Certified value	Radiochemical method
1	8.60±0.30	6.80±0.40
2	2.55±0.34	1.50±0.12
3	1.80±0.20	1.70±0.15

for the platinum detection limit is an increase of the irradiation time and the beam intensity. While increasing them twice the detection limit will be 2.5 $\cdot 10^{8}$ g/g.

Track method. The dependence of track density upon the thorium content is given in *Fig.* 4 and may be used to provide a reliable concentration.

The mean variation in the background tracks on the lexan material had remained between 0 and 3 tracks during the 10 experiments.The same results was obtained for the mean reagent blank background. For individual samples we used a detection limit equal to 2 σ above the mean of the background on the lexan which we take equal 3 tracks. In this case the detection limit is 3 track, which is equivalent to $4 \cdot 10^{-13}$ g.

By using higher photon fluence up to 10^{19} photon·cm^{-2} and solid state detectors with a very low uranium content $\leq 10^{-14}$ g/g it is possible to reach the sensitivity of 10^{-14} g of thorium in specimens.

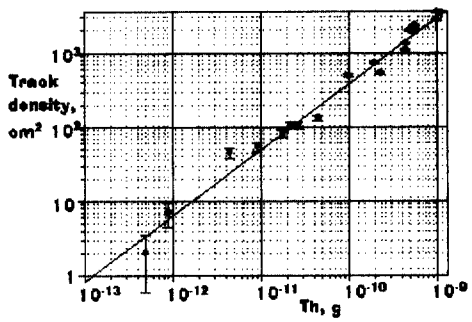

Fig. 4. Track density in dependence on the Th contents.

The chemical elements analyzed by using the MT-25 microtron is presented in *Table 3.*

Conclusion

1. The use of the microtron permits to determine more than 40 elements in coal and more 57 elements in environmental samples which are of

interest for industry and ecology with the 10^{-3}-10^{-5}% detection limit.

2. The platinum determination limit for instrumental gamma activation version is 100 μg and in the case for radiochemical version - 0.1 μg.

3. The detection limit of thorium for track method is $4 \bullet 10^{-13}$ g.

At present a technique of preconcentrating gold, noble metals , uranium, thorium and plutonium from natural samples for their quantitative determination in submicro- and subpicogram amounts activation analysis using the microtron is being developed at our laboratory.

Table 3

The chemical elements analysed by using the electron accelerator - the MT-25 microtron.The detection limit is given in g/g.

H $1 \cdot 10^{-4}$							**He**		
Li	**Be**	**B**	**C**	**N** $1 \cdot 10^{-7}$	**O** $1 \cdot 10^{-6}$	**F**	**Ne**		
Na $2 \cdot 10^{-4}$	**Mg** $2 \cdot 10^{-4}$	**Al** $2 \cdot 10^{-5}$	**Si** $1 \cdot 10^{-3}$	**P**	**S**	**Cl** $1 \cdot 10^{-6}$	**Ar**		
K $2 \cdot 10^{-4}$	**Ca** $2 \cdot 10^{-4}$	**Sc** $8 \cdot 10^{-7}$	**Ti** $1 \cdot 10^{-4}$	**V** $5 \cdot 10^{-6}$	**Cr** $5 \cdot 10^{-6}$	**Mn** $4 \cdot 10^{-7}$	**Fe** $1 \cdot 10^{-3}$	**Co** $1 \cdot 10^{-6}$	**Ni** $1 \cdot 10^{-6}$
Cu $1 \cdot 10^{-5}$	**Zn** $5 \cdot 10^{-6}$	**Ga** $1 \cdot 10^{-5}$	**Ge** $1 \cdot 10^{-5}$	**As** $5 \cdot 10^{-7}$	**Se** $1 \cdot 10^{-5}$	**Br** $5 \cdot 10^{-6}$	**Kr**		
Rb $2 \cdot 10^{-6}$	**Sr** $2 \cdot 10^{-7}$	**Y** $2 \cdot 10^{-6}$	**Zr** $6 \cdot 10^{-7}$	**Nb** $1 \cdot 10^{-6}$	**Mo** $1 \cdot 10^{-6}$	**Tc**	**Ru**	**Rh**	**Pd**
Ag $1 \cdot 10^{-6}$	**Cd** $5 \cdot 10^{-6}$	**In** $1 \cdot 10^{-7}$	**Sn** $1 \cdot 10^{-5}$	**Sb** $5 \cdot 10^{-7}$	**Te** $1 \cdot 10^{-6}$	**I** $3 \cdot 10^{-6}$	**Xe**		
Cs $5 \cdot 10^{-7}$	**Ba** $5 \cdot 10^{-6}$	**La**	**Hf**	**Ta** $5 \cdot 10^{-8}$	**W** $1 \cdot 10^{-7}$	**Re** $7 \cdot 10^{-7}$	**Os**	**Ir**	**Pt*** $1 \cdot 10^{-7}$
Au $2 \cdot 10^{-8}$	**Hg** $5 \cdot 10^{-7}$	**Tl** $7 \cdot 10^{-7}$	**Pb** $2 \cdot 10^{-6}$	**Bi**	**Po**	**At**	**Rn**		
Fr	**Ra**	**Ac**	**Ku**	**Ns**	**106**	**107**	**108**	**109**	**110**

Ce $1 \cdot 10^{-6}$	**Pr**	**Nd** $2 \cdot 10^{-6}$	**Pm**	**Sm** $5 \cdot 10^{-8}$	**Eu** $1 \cdot 10^{-7}$	**Gd**	**Tb**	**Dy**	**Ho**	**Er** $1 \cdot 10^{-7}$	**T m**	**Yb**	**Lu**

Th $5 \cdot 10^{-8}$ $5 \cdot 10^{-13}$	**Pa**	**U** $5 \cdot 10^{-8}$ $2 \cdot 10^{-13}$	**Np** $1 \cdot 10^{-13}$	**Pu** $8 \cdot 10^{-14}$	**Am**	**Cm**	**Bk**	**Cf**	**Es**	**Fm**	**Md**	**No**	**Lr**

E
(γ, n; n, γ)
(γ,f;)*

*- radiochemical method.

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