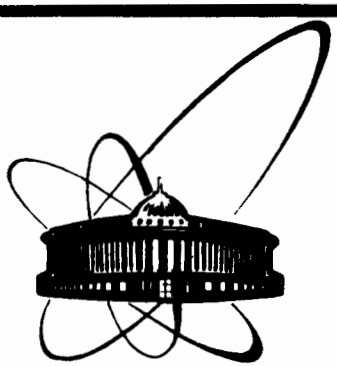

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DETERMINATION OF FISSILE ELEMENTS BY PHOTOFISSION DELAYED NEUTRON COUNTING

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INTRODUCTION

The delayed neutron activation analysis (DNAA) of uranium and thorium is a well-known method [1]. It is based on the utilization of fission of the nuclides by thermal or fast neutrons and can be performed rapidly with a simple counting system. The detection limits (10^{-7} - 10^{-8}) g/g were obtained when the samples were irradiated in reactors by a neutron flux of 10^{12} n/cm^{2}s. Few papers on the use of photofission delayed neutron activation analysis (PDNAA) with intense sources of hard bremsstrahlung (LINAC or MICROTRON), mainly for nondestructive nuclear material assay, have been published [2-5]. The electron accelerator has many evident advantages, such as a lower cost, compactness, a wide range of applications and comparatively simple operator manipulations. Besides, they can be switched off at any moment and subsequent work may be carried out without complicated precautions against radiation hazard. Recently much effort has been devoted to using the electron cyclic accelerator-microtron for gamma and neutron activation analysis [6-9]. Less attention is being paid to PDNAA with a strong bremsstrahlung beam. The present work aims at complementing the practical scope of its application in the analysis of samples with small contents of uranium or thorium.

EXPERIMENTAL

Photoactivation and Delayed Neutron Measurements

The scheme of the experimental arrangement is shown in Fig.1. Irradiations were performed on the microtron MT-22 with a maximum bremsstrahlung energy of 20.5 or 18 MeV. 30 - 40 g samples were placed into 25 mm ϕx80 mm vinilite containers, samples and standards were irradiated for 1 min, let decay for 5s or 15s and measured for 1 min. The transfer of the samples was performed with the aid of a pneumatic post. The counting station with 18 percent efficiency for fission neutron spectrum contained seven ^{3}He counters of type SNM-18 connected in parallel and placed in a cylindrical plexiglass block 300 mm ϕx550 mm. The moderator was shielded against neutrons from external sources by a 0.5 mm Cd shield. A time analyser with microprocessor of type MIKAM-2 [10] was used. The channel widths was chosen to be 0.1 s.

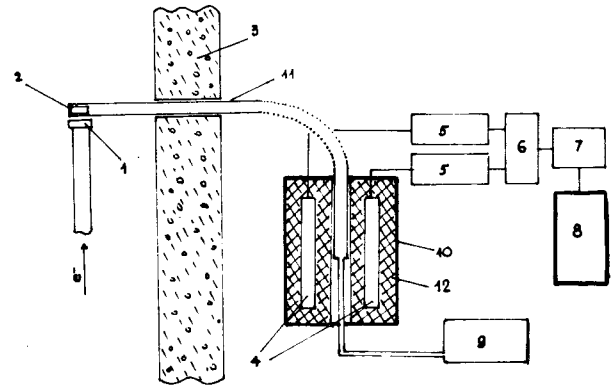

Fig. 1. The experimental arrangement: 1 - target, 2 - sample container, 3 - concrete shielding, 4 - neutron counters, 5 - preamplifier, 6 - pulse-height discriminator, 7 - amplifier, 8 - time amalyzer, 9 - compressor, 10 - Cd shielding, 11 - pneumatic sample tube, 12 - plexiglass.

The main characteristics of the microtron MT-22 are as follows:

- the maximum energy of the electron beam	20.5 MeV;
- mean electron current	10-12 μA;
- bremsstrahlung intensity at a distance of 1 m from target	6000 roentgens per minute;
- target	Ta, cooled.

Interferences

The determination of U and Th is complicated by the formation of ^{17}N by the reaction $^{18}O(\gamma, p)^{17}N$ with a 15.9 MeV threshold. This neutron emitter has a 4.17 s half-life. The interference can be avoided by activating with lower photon energy. In this case, as the main part of the giant resonance of the excitation function of photofission for U and Th lies in the 8 - 18 MeV energy range, the yield of the (γ, f)-reaction changes insignificantly [10].

Two samples of distilled water were irradiated at 20.5 and 18 MeV electron energies for estimating the oxygen influence. The results show that 1 g oxygen gives 125 counts per min in the first case and 8 counts in the second. It is clear that the oxygen interference cannot be neglected at high energy. It can

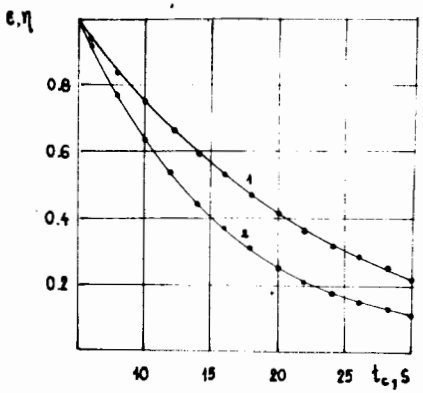

Fig. 2. The sensitivity of uranium analysis ϵ (curve 1) and relative oxygen contribution η (curve 2) as a function of cooling time t_c. ϵ and η are in relative units.

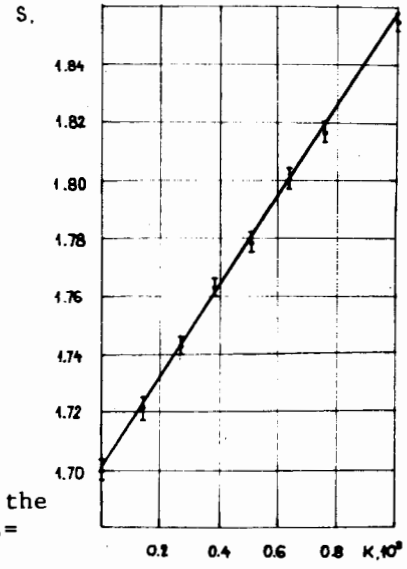

Fig. 3. Quantity S as a function of the relative oxygen content; t_1 = 5 s, t_2 = = 15 s.

be avoided by increasing the cooling time to let oxygen activity decrease, but a part of the useful information could be lost then. Figure 2 illustrates the behaviour of analysis sensitivity ϵ for uranium and the relative oxygen contribution η as functions of cooling time t_c. The value of η is determined as the ratio of counts due to oxygen neutrons to the total number of the delayed neutrons registered. In Fig. 1 η and ϵ are in relative units. At t_c = 30 s, ϵ decreases ≈5 times, whereas η decreases by a factor of ≈10.

The oxygen contribution will be easy to correct for by using the following equation

$$S = \frac{m_o N_{o1} + m_{u1}}{m_o N_{o2} + m_u N_{u2}} = \frac{k N_{o1} + N_{u1}}{k N_{o2} + N_{u2}}, \qquad (1)$$

where m_o, m_u are amounts of oxygen in g and uranium in mg, respectively; N_{o1}, N_{o2} are counts per g of oxygen with cooling times t_1 and t_2, respectively; N_{u1}, N_{u2} - counts per mg of uranium with cooling times t_1 and t_2, respectively; k is ratio of the amounts of oxygen and uranium, $k = \frac{m_o}{m_u} \times 10^3$ g/g.

The function S represents the ratio of the total number of counts registered with cooling time t_1 to the one registered with t_2. It depends on k whereas N_{o1}, N_{o2}, N_{u1}, N_{u2} are constants for the given experimental conditions. From Eq. (1) we can determine k and in this way take into account the oxygen contribution. Figure 3 shows S as a function of k in the region of $k \leq 1$.

RESULTS AND DISCUSSION

Uranium gives 2.0 counts per mg; and thorium, 1.2 counts when the samples were irradiated at a mean electron flux of 11 μA and energy of 18.0 MeV. Analysis results for some solid samples and solutions are compiled in Table 1.

Table 1

The U and Th concentrations (g/g) determined by PDNAA and other methods

No.	PDNAA		Chemical	
	U	Th	U	Th
1	$(2.67\pm0.03)\times10^{-4}$	-	$(2.60\pm0.05)\times10^{-4}$	-
2	$(1.20\pm0.05)\times10^{-5}$	-	$(1.20\pm0.1)\times10^{-5}$	-
3	-	$(2.3\pm0.1)\times10^{-5}$	-	$(2.4\pm0.2)\times10^{-5}$
4	-	$(3.4\pm0.3)\times10^{-6}$	-	$(3.7\pm0.4)\times10^{-6}$ *
5	-	$(2.8\pm0.7)\times10^{-7}$	-	2.5×10^{-7} *

* Gamma activation analysis (GAA)

The content values obtained using the PDNAA, the chemical method, and the GAA generally agree well. Table 2 shows the results of analyses of samples with fixed uranium content and different concentrations of oxygen. Good results were obtained when oxygen was taken into account.

The important sources of errors are the counting statistics. The cyclic measurement technique was used for raising the reliability of the results. It is difficult to determine small U(Th) contents in the presence of Th(U) by PDNAA. Nevertheless, for sufficiently large amounts of them one may use the method of kinetic functions proposed by J.Keepin /12/ for this purpose. The kinetic functions $S_{+t/\Delta}$ and $S_{-t/\Delta}$ for ^{238}U and ^{232}Th were determined to check this possibility.

Table 2

The uranium content determined with and without oxygen correction 10^{-4} g/g

oxygen content, percent	without oxygen correction	with oxygen correction
0	2.67±0.03	2.67±0.03
13.3	2.84±0.05	2.66±0.16
16.6	2.89±0.05	2.67±0.16
20.1	2.94±0.05	2.67±0.16
23.3	2.98±0.05	2.67±0.16
27.0	3.02±0.05	2.68±0.16

The discriminative ratio obtained in our experiment, $(S_{-f}/\Delta)_{Th}/(S_{+f}/\Delta)_U = 1.19$ with $f = 10$ s, $\Delta = 1$ s, $t_c = 5$ s, agrees well with the value given in ref. /13/.

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Определение делящихся элементов путем регистрации запаздывающих нейтронов фотоделения

Рассматривается применение циклического электронного ускорителя - микротрона для определения содержания урана и тория путем регистрации запаздывающих нейтронов фотоделения. Образцы облучались тормозными γ-квантами микротрона с граничной энергией 20,5 и 18,0 МэВ. В первом случае предлагается метод исключения интерференции кислорода. Порог определения урана равен $4 \cdot 10^{-7}$ г/г, а тория - $6 \cdot 10^{-7}$ г/г.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

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Determination of Fissile Elements by Photofission Delayed Neutron Counting

The application of a cyclic electron accelerator-microtron for photofission delayed neutron activation analysis of U and Th is considered. The samples are irradiated by microtron 20.5 and 18 MeV bremsstrahlung. In the first case a simple method for account of the oxygen interference is developed. The detection limits are 4 x 10^{-7} g/g for uranium and 6 x 10^{-7} g/g for thorium.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

Prep h. Dubna 1982