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NEUTRON FLUX MEASUREMENT OF (γ, n)-REACTIONS ON NUCLEI OF ZIRCONIUM

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Abstract. *This paper investigates the probability of activation of zirconium atoms under irradiation of Bremsstraglung gamma-rays with maximum energy of 24 MeV. The neutron flux of (ɤ, n)-reaction on nuclei of zirconium was measured using the method of activation detectors. It is suggested that the activation of zirconium atoms and the isomeric transitions of zirconium isotopes are the main cause of radiation defects in materials containing zirconium.*

Key words: *Zirconium, neutron flux, isomer transition, target of titanium dioxide, irradiation*

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1. INTRODUCTION

There are two reasons why the research of neutron flux measurement of (γ, n) –reactions on zirconium nuclei is relevant. Firstly, zirconium has lately become very popular in radiochemistry and nuclear medicine [1-7]. The second reason is that this work is continuing research into the effects of ionizing radiation on zirconium silicate sorbents. From a scientific point, the influence of Bremsstrahlung gamma-rays on such a sorbent is very interesting because zirconium is a well-known radiation-resistant structural metal [1]. Previous research has showed the effect of Bremsstrahlung gamma rays on the surface parameters of microporous amorphous zirconium silicate and on its ability for sorption. The ability of zirconium silicate to absorb Sr2+ ions from an aqueous solution increased noticeably after irradiation by 24-MeV Bremsstrahlung gamma rays, but clear correlation between changing the surface parameter and the increase in sorption was not found [2].

However, this effect was not observed when zirconium silicate was irradiated by electrons 1 MeV and 7 MeV. Also other sorbents such as zeolite or titanium phosphate after irradiated by gamma rays had not such effect too.

Therefore, we made a conclusion that the cause of increasing sorption of Strontium ion is the presence of Zirconium in the structure of the sorbent and high energy gamma rays, which initiate a photonuclear reaction.

During irradiation of Zr nuclei by gamma-quanta with energy greater than 8 MeV, their activation can occur.

We made a suggestion that the main mechanism of influence on sorbent properties is the formation of a radiation defect due to the IT decay of Zr nuclei after the Zr(ɤ, n) reaction.

Table 1. Some important zirconium isotopes

Isotope	Energy, keV	Half-life	Type of decay	Daughter isotope
^{88}Zr		83.4 days	EC	^{88}Y
^{89}Zr	897 909	78.4 hours 3.27 days	β+ IT(93.77%)	^{89}Y ^{89}Zr
^{89m}Zr	587	4.16 min	β+(6.23%)	^{89}Y
^{90}Zr (51.45%)		**stable**		
^{90m1}Zr	2319	809.2 ms	IT	^{90}Zr
^{90m2}Zr	3589.4	131 ns	IT	
^{91}Zr (11.22%)		**stable**		
^{91m}Zr	3167	4.35 ms	IT	
^{92}Zr(17.15%)		**stable**		

The samples which contained Zr in their structure (a few grams) were irradiated using a Betatron (electron accelerator) with a maximal energy of the γ-quanta of 24 MeV, a flux density of γ-quanta of 10^8 γ-quanta/cm²s μA (0.5 MeV), while angular distribution was the same, and the optimal dimensions

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of the samples were 2x3 cm. The energy distribution of the Bremsstrahlung γ-quanta is described in Table 2 and Figure 1.

Table 2. Energy distribution of Bremsstrahlung γ-quanta of the electron accelerator (Betatron B-25) in Uzhgorod National University

E γ, MeV	The content of γ-quanta, %
5	28.273
7	18.56
9	13.35
11	10.16
13	8.04
15	6.535
17	5.379
19	4.388
21	3.37
23	1.92
25	0

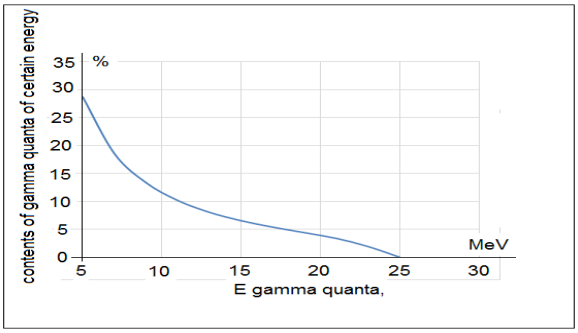

Figure 1. Energy distribution of Bremsstrahlung γ -quanta of Betatron B-25 in Uzhgorod National University

The objective of this work is to determine the possibility of the described processes by measuring the neutron flux in reaction (ɤ, n) in Zirconium nuclei under irradiation in our laboratory conditions.

2. Experimental Technique

Experience in experimental nuclear physics dictates that the target structure (crystalline or amorphous) has a certain influence on the final result of irradiation. Most amounts of gamma-quanta flow through the target or fall on the target and scatter.

So, the crystal structure of irradiated samples explains the occurring processes much easier. Neutrons, which are formed in the (ɤ, n) reaction can also be scattered and take place in (n, ɤ) or other reactions, etc. These considerations demonstrate the need for research targets with crystalline structure, certain interatomic distances and angles, and regular matrix structure. Zirconium silicate does not possess such characteristics.

Two targets, based on titanium dioxide, were made for this purpose. In one of the targets, 40 percent of titanium atoms was replaced with zirconium atoms. These substances had a crystal structure, were in the form of powder, and were placed in plastic containers 3cm x 2cm in size. The distance from the samples to the Bremsstrahlung target of the accelerator was 1 meter, as described in [2,3]. A short scheme of the experiment can be seen in Figure 2.

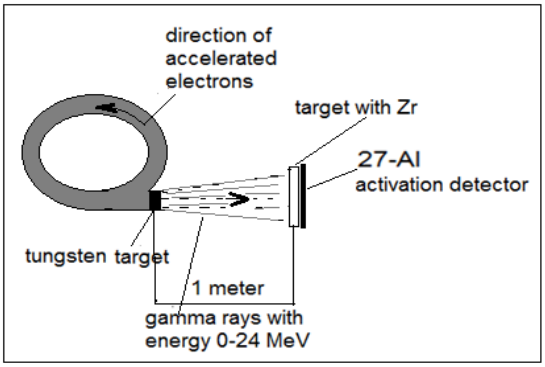

Figure 2. Scheme of targets and activation detector situated

The target was irradiated by the gamma quanta for 30 min. The time interval between the end of irradiation and the start of measurements – 'wait time' – was also 30 min. Aluminum and reaction 27Al (n, p) 27Mg were selected as activation detectors. All collectors were situated close to the target (Fig. 2). Neutrons from the reaction (ɤ, n) bombarded 27-Al and initiated the reaction ^{27}Al (n, p) ^{27}Mg in the activation detector. Isotopes of ^{27}Mg were identified by the scintillated spectrometer with NaI(Tl) with the AkWin program, calibrated to standard gamma sources of ^{137}Cs and ^{60}Co. Also, its pulses could be registered by the pulse analyzer G-M. The decay scheme of ^{27}Mg can be seen in literature [8].

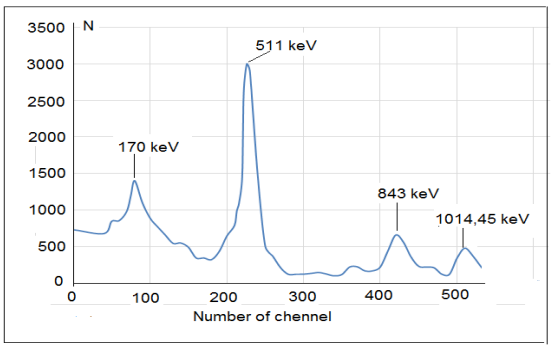

Figure 3. Experimental gamma spectrum of activation detector

According to the literature, we can observe peeks with the energy of 843 keV, 170.68 keV and 1014.45keV.

If we consider the reaction ^{27}Al (n, α)^{24}Na, we will have peeks with the energy of 996 keV, 1338 keV and more than 4000 keV. Moreover, the half-life of ^{24}Na is 14 hours, so irradiation for 30 min is not enough to activate this isotope [8]. The peak of 511 keV (Fig. 3) belongs to ^{26}Al after the reaction 27Al (ɤ, n) ^{26}Al, which emits β^+ particle [9].

Gamma rays can pass through the target and not initiate the photonuclear reaction. This can be shown by the presence of a peak of 511 keV. Therefore, in our opinion, the crystal structure of samples is important, because all processes of scattering will occur in the crystalline structure in the same way. The compound of

crystal titanium dioxide ($ZrO_{2(40\%)}TiO_{2(57\%)}AsO_{4(3\%)}$) was used as a zirconium-containing target. In the second experiment, the compound ($TiO_{2(97\%)}AsO_{4(3\%)}$) was used as a target without zirconium. Both compounds were synthesized by original methods at the Vasyl Stefanyk Precarpathian National University (Ivano-Frankivs'k, Ukraine) and had a crystal structure and the same mass; they differed only in terms of zirconium in the structure. The cross section of the (γ, n)-reaction on zirconium nuclei was greater than that on the nuclei of Oxygen, Titanium, and Arsenic [9] – Figure 4.

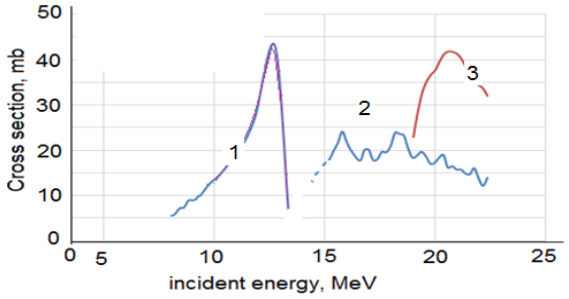

Figure 4. Cross section of 1 – 40 Zr 91 (ɤ,n); 2 – 22Ti (ɤ,n); 3 – 33As (ɤ,n). The plots built using data taken from reference [9]

Cross section of ^{90}Zr (ɤ,n) ^{89}Zr production is 0.0686±0.0031 barn, and ^{16}O (ɤ,n) 0.6±0.12 mb [9].

Photonuclear reaction can occur by gamma quanta with the energy of more than 8 MeV. The energy of a neutron, which was emitted from the target, can be calculated by using the equation $E_n = Eɤ - E_b$

E_n – energy of neutron; E_b – bond energy of neutron in nuclear.

Therefore, the maximum energy of a neutron can be less than 14 MeV. If we take Fig. 1 into account, we can see that a more common diapason of neutron energy will be 4-12 MeV.

So, a target that is made of modified titanium dioxide (ZrO_2 $_{(40\%)}TiO_{2(57\%)}AsO_{4(3\%)}$) was used for neutron production via the thin-target Zr (γ,n) reaction and quantitative determination of neutron flux. Aluminum and the ^{27}Al (n, p) ^{27}Mg reaction were selected as activation detectors.

Table 3. Nuclear reactions that are the basis of the aluminum activation detector

Nuclear reaction	Preva-lence, %	Half-life	σ, mbarn	En, MeV
$^{27}Al(n,p)^{27}Mg$	100	10.2 min	26(5)	4-12
$^{27}Al(n, \alpha)^{24}Na$	100	15 h	135	14

The difference in the number of pulses of aluminum collectors, which were behind the target, was caused by ^{27}Mg radiation generated by the influence of neutrons from the $^{90}Zr(ɤ,n)^{89}Zr$ reaction.

Experimental evidence of this fact would mean that the main change in the properties of sorbents is isomeric transitions of zirconium isotopes [10, p. 206].

Experiment was conducted in order to prove the presence of a photonuclear reaction in described conditions. Zirconium by ɤ, n - reaction proceeds in the isomer state. The main causes of chemical changes in isomeric transitions are processes of electron internal conversion and occurrence of vacancies in inner shells of atoms [10, 11]. Then, as a result of internal conversion of electrons, an atom of Zirconium leaves its cell and generates radiation defects. This is the main mechanism of radiation changes of the surface and sorption properties of Zirconium silicate.

3. Measuring the neutron flux

In the activation method, the registration of the neutron flux is done by measuring the radioactivity of the product's radiative capture (n, ɣ) or other reactions caused by neutrons.

$^{90}Zr + ɤ = {}^{89}Zr + {}^{1}n + {}^{27}Al = {}^{27}Mg + p$

or

$^{90}Zr(ɤ,n)^{89}Zr$ $^{27}Al(n,p)^{27}Mg$ $T_{1/2}$ Mg-27 = 10 min

If the activation detector is thin, the neutron flux determines the amount of radioactive nuclei, which are formed in 1 second [8, 12].

$$R = \sigma_a \varphi_n n_x V \qquad (1)$$

n – amount of isotop's nuclei in 1 cm^3 of target

φ_n – neutron flux

V –volume of sample, cm^3

Cross section of interaction, σ_a – barn, φ_n *1/cm² s* - neutron flux.

If the detector was irradiated during the time t, its activity measured from moment t_1 to t_2 and N pulses were registered, we would get an expression for the neutron flux density

$$R = \frac{N}{\varepsilon\tau\left(1-e^{-t/\tau}\right)\left(e^{-t_1/\tau}-e^{-t_2/\tau}\right)} \qquad (2)$$

ε –the effectiveness of the counter of the equation the expression for the neutron flux density

$$\varphi_n = \frac{N}{\delta_n V \varepsilon\tau\left(1-e^{-t/\tau}\right)\left(e^{-t_1/\tau}-e^{-t_2/\tau}\right)} \qquad (3)$$

where

$$\tau = 1/\lambda = T_{1/2}/\ln 2$$

4. Results

The main results are shown in figures 5, 6 and in Table 4.

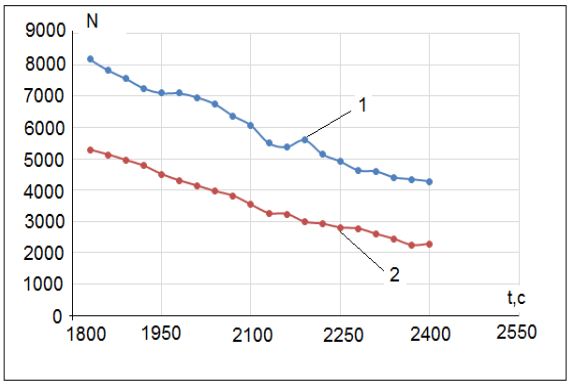

Figure 5. The number of pulses from aluminum with 1 - $ZrO_{2\,(40\%)}TiO_{2(57\%)}AsO_{4(3\%)}$ used as a target; 2- target of $TiO_{2(97\%)}AsO_{4(3\%)}$; t,c – time interval between the end and the start of radiation measurements; N – number of pulses (measured by a G-M detector with the applied voltage of -400 V)

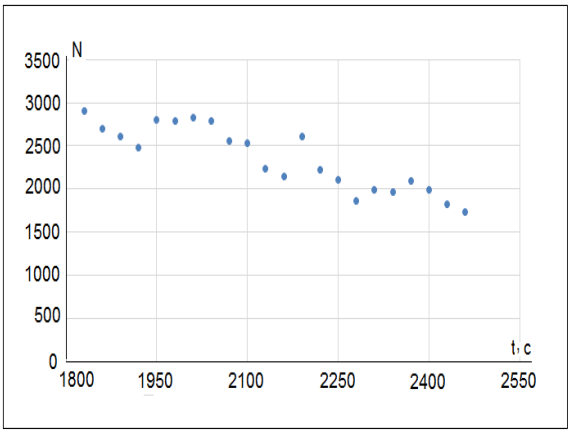

Figure 6. The number of pulses from 27Al (n, p) produced by neutrons from (γ, n) reaction on irradiated Zr sample

Table 4. Density of neutron flux in reaction Zr (γ, n) which is calculated with different 'wait time'

neutron flux × 10^6 n/cm²s	'wait time', s	standard deviation × 10^6	%
0.1263	1800	0.0285	21
0.1637	2100		
0.1077	2400		

The arithmetic mean of the density of neutron flux in the reaction Zr (γ, n) is φ=0.1325 10^6 n/cm²s.

The main changes in the properties of sorbents which contained Zr in their structure under irradiation of high energy Bremsstrahlung gamma-rays are predetermined by isomeric transitions of Zirconium isotopes.

These research studies are characterized by the relative simplicity of calculations and affordable equipment. This is their advantage. The use of such substances as targets, taking into account their strict crystalline structure, simplifies calculations in the study of photonuclear reactions very much.

These investigations make processes of interaction between sorbents and high energy gamma quanta clearer. The new information about the reaction Zr (γ, n) can be useful in the research of radiation resistance of sorbents containing zirconium in their structure. It is essential to consider the impact of generated neutrons and isomeric transitions of the zirconium atoms on the structure of materials. Perhaps, by using these experimental data, we can develop a method for modifying the surface of zirconium silicate. Our results may be useful in the study of radiation-resistant materials that can be used in high background radiation. In continuation of these studies, it would be possible to offer a new alternative method for obtaining the isotope ^{89}Zr, which is very popular in nuclear medicine for use in PET [13].

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