SORPTION INVESTIGATION OF FISSION FRAGMENTS OF URANIUM NUCLEI BY TITANIUM PHOSPHATE IN WATER SOLUTION

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The interaction of titanium phosphate (two modifications) and absorbate with the radioactive microadditives in the water solution was investigated using gamma spectrometry of fission fragments. The sorption of ^{93}Y, ^{97}Nb , ^{98}Zr, ^{99}Mo, ^{132}Te, ^{133}I, ^{143}Ce ions was investigated. The fragments were obtained in thermal neutron induced fission of uranium-235. To provide the reaction under study the M-10 microtron located at the Department of Nuclear Physics (Uzhhorod National University), was used.

From the environmental point of view the problems of water and air purification from different pollution agents, in particular from radionuclides, are of high priority. Various sorbents are used for this purpose [1]. The investigation of their properties as well as working out new experimental techniques are of major importance. A great attention to the sorption of radioactive fragments of the heavy nuclei fission in water solution was paid. Such sorption process can be used as one of chemical techniques for extraction and collection of the fission fragments at the investigations of their cummulative and especially independent yields under different conditions of the experiment. Therefore the investigations like this promote for the search and usage of sorbents for solving ecological problems.

In the present work the preliminary results of the absorption studies of the ^{235}U fission fragments (^{97}Zr, ^{143}Ce, ^{99}Mo, ^{97}Nb ^{135}Xe) by titanium phosphate in water solution are described. The radioactive fragments were formed at thermal neutron fission of ^{235}U. With this purpose four targets of 90%-enriched uranium oxide with the thickness of 1.75 mg/cm^2 and 5 cm diameter each were installed in the vessels with water 3 cm apart from the water surface. These vessels were situated very close to the Be target which was installed ~15 cm apart from the brake target of the microtron and was surrounded by a polyethylene moderator. Electrons, accelerated in the microtron up to 7-8 MeV, emitted hard gamma-rays hitting the brake target, which caused the reaction $^{9}Be(\gamma,n)^{8}Be$ in the Be target. The emerged neutrons were moderated in Be target and polyethylene and subsequently caused $^{235}U(n,f)$ fission reaction hitting the uranium target. The fragments were partly accumulated in water. The water from all vessels was poured together and divided into several portions 20 ml each in two hours after the irradiation of the target block in microtron beam. One of them sorbent was not added to. Each other portion contained titanium phosphate (~1g) of two modifications with Ti/P ratio 1 and 0,8, respectively. The time intervals of sorption were 1, 2, 4 and 16 hours under static conditions and at room temperature. After sorption the water was poured out into another vessel. The γ-spectra of such solution were measured by Ge(Li) spectrometer. The content of certain fragments was determined from the intensity of the characteristic maximum in γ-emission spectra. Immediately after irradiation the measured intensity

spectral maximum S is related to the number of fragments N by a relation

$$N = \frac{S}{\varepsilon}(e^{-\lambda t_1} - e^{-\lambda t_2}),$$

Here ε is γ-ray registration efficiency with account of the measurement geometry, t_1 and t_2 are the start- and stop-time of γ-spectrum measurement, respectively. t=0 corresponding to the end of the irradiation process.$\lambda = \ln 2/T_{1/2}$ is decay constant of the radionuclide, $T_{1/2}$ is the half-life of the nuclide. If half-life of the precursor was comparable to that of the fragments of interest, the contribution of the precursor decay was done.

The efficiency of sorption is determined as

$$C = \frac{So - S}{So},$$

S_0 is the sum of pulses from the sample without contact with the sorbent with correction made for the decay effect; S is the similar quantity for the sample having been put into contact with the sorbent. The results of the measurement for the fragments of ^{235}U which show the sorption effect are given in Table 1.

Since we deal with the microquantities of fragments, whose migration in water is studied poorly, the comparison of the regularities of the sorption process for macro- and microquantities of fission fragments is important. With this purpose the sorption efficiency of Zr ions was determined using complexonometric titration [3], based on creation of stable complex of Zr ion and black eriochrome T at high salt concentration (2n HCl) with the mole ratio 1:1. This technique allows determining up to 1000 mg of Zr in the volume of 100 ml with error less than 1%. The results of this measurement are given in Table 1 as well. It is seen that the values obtained by using both of these techniques (gamma-spectrometry and complexonometry) are similar however the values obtained by gamma-spectrometric technique are somewhat smaller.

Table 1. Physical and sorption characteristics of the ^{235}U fission fragments obtained by gamma-spectrometric ($C1_\gamma$, $C2_\gamma$) and complexonometric titration ($C1_t$,$C2_t$)

fragment	$T_{1/2}$, min	branch ratio	$T_{sorption}$, min	$C1_\gamma$ Ti/P=1	$\Delta C1$	$C2_\gamma$ Ti/P=0.8	$\Delta C2$	$C1_t$ Ti/P=1	$C2_t$ Ti/P=0.8
Ce-143	1980	41.3	60	0.62	0.43	0.29	0.18		
			240	0.57	0.23	0.53	0.22		
			960	0.60	0.33	0.70	0.30		
Xe-135	545	90.6	60	0.18	0.01	0.10	0.006		
			240	0.30	0.06	0.22	0.005		
			960	0.44	0.30	0.24	0.160		
Mo-99	3961	90.1	60	0.51	0.38	0.27	0.230		
			240	0.32	0.13	0.13	0.049		
			960	0.33	0.08	0.37	0.070		
Nb-97	72.1	100	60	0.50	0.20	0.32	0.12		
			240	0.47	0.09	0.52	0.09		
			960	0.63	0.21	0.75	0.30		
Zr-97	1020	94.6	20					0.66	0.68
			40					0.70	0.70
			60	0.55	0.21	0.38	0.17	0.70	0.71
			120					0.71	0.79
			240	0.48	0.08	0.57	0.10	0.71	0.81
			960	0.68	0.24	0.78	0.21		
			1440					0.81	0.85

The correlation coefficients between the sorption values for two modifications of titanium phosphate obtained by using both of these techniques are r_1(Ti/P=1)=0.936 and r_2(Ti/P=0.8)=0.828, respectively. Therefore, the obtained values r_1 and r_2 indicate on the quantitative relation between the sorption characteristics obtained by both of these techniques. Thus, the preliminary results of the investigation indicate the possibility of the gamma-spectrometric technique to be used for qualitative and/or quantitative analysis of the sorbent properties at the sorption of the radioactive fragments formed at trans-uranium elements fission.

References

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ДОСЛІДЖЕННЯ СОРБЦІЇ УЛАМКІВ ПОДІЛУ ЯДЕР УРАНУ ФОСФАТОМ ТИТАНУ У ВОДНОМУ РОЗЧИНІ

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Методом гамма-спектрометрії уламків поділу досліджено взаємодію фосфату титану (двох модифікацій) і абсорбату з радіоактивними мікродомішками у водному розчині. Вивчено сорбцію іонів ^{93}Y, ^{97}Nb , ^{98}Zr, ^{99}Mo, ^{132}Te, ^{133}I, ^{143}Ce. Уламки було отримано в результаті розпаду урану-235, викликаного тепловими нейтронами. Для забезпечення досліджуваної реакції використано мікротрон М-10 кафедри ядерної фізики Ужгородського національного університету.