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Measurement of the ^{242g}Am ($T_{1/2}$= 16.02 hour) fission cross section

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Abstract. There was performed a cycle of ^{242g}Am fission characteristics measurements with the use of IRT-MEPhI reactor. The ^{242g}Am isotope was produced in the course of irradiating ^{241}Am targets in the experimental channel with the density of neutron flux $\approx 2 \cdot 10^{13}$ 1/(cm^2 ·s). Then, with no isolation of americium-242 from the irradiated targets the time dependencies of induced fission fragments yield from the targets were measured. Moreover the irradiated americium targets undertook irradiation in another channel of IRT reactor with the density of thermal neutron flux $\approx 5.6 \cdot 10^{10}$ 1/(cm^2 · s).

The fission fragments were registered with the aid of dielectric track detectors. In each time dependence of fission fragments an exponent with $T_{1/2}$ = 16.02 hour is available; the cross section of ^{242g}Am fission by thermal and resonance neutrons was determined by the intensity of induced fissions with the given half-life. The quantity of ^{242g}Am nuclei in each of irradiated targets was determined basing on accumulation of ^{242}Cm, daughter product of ^{242g}Am decay.

Based on the experimental results for the ^{242g}Am nuclide, the evaluations of both the fission cross section by thermal neutrons and the fission resonance integral were obtained.

1 Introduction

The ^{242g}Am isotope can play an important part in the process of ^{241}Am transmutation in intense fields of thermal neutrons. However, the data on cross section of ^{242g}Am formation and cross sections of ^{242g}Am interactions with neutrons are extremely few. Sufficiently reliable experimental data on radiation capture cross section are given in publications of G. Fioni et al. only [1,2]. The value 2100 barn of ^{242g}Am fission cross section by thermal neutrons given in all libraries of evaluated data is based on theoretical estimations [3].

The authors made an attempt to measure cross section of ^{241}Am(n,γ)^{242g}Am reaction as well as cross sections of ^{242g}Am fission by thermal and super-cadmium neutrons.

In the investigations of fission characteristics of short-lived odd-odd nuclei undertaken in VNIIEF the nuclides of ^{232}Pa and ^{238}Np were produced in (p,n) reactions; metal plates of ^{232}Th and ^{238}U were irradiated by accelerated protons [4]. The irradiated surface of plates was etched, and the corresponding nuclide was isolated using radiochemical methods. However, to get samples of ^{242g}Am this method is too difficult.

Thus, in the work described we have tried to get sufficiently enough number of ^{242g}Am nuclei in ^{241}Am(n,γ) reaction with further measurements of ^{242g}Am nuclide characteristics without its isolation from the irradiated target of ^{241}Am.

2 Characteristics of neutron fields

The investigations were performed on IRT MEPhI reactor (MEPhI – Moscow Engineering and Physics Institute, Moscow) [5]. For irradiation there were used VEC-25 (Vertical Experimental Channel) and VEC-28 channels.

To measure the flux density and spectral characteristics of neutrons in these channels there was applied a set of neutron activation detectors (DNA). The samples from this set were irradiated in the channels at the given reactor power with no filters and with cadmium filters. Accordingly, there were determined the density of thermal (maxwellian) neutron flux F_{th} and cadmium ratio L_{Cd} by gold ^{198}Au, as a rule.

To measure the activity of γ-radiation from irradiated DNA there was used a certified radiometric complex including a coaxial semiconductor detector of extra-purity germanium and a system of spectrometric information processing.

At the reactor operating power equal to 2500 kW the density of maxwellian neutron flux in irradiation positions was as follows:

VEC-25, $F_{th} = 1.98 \cdot 10^{13}$ 1/cm^2 s, L_{Cd} = 5.06 by gold;
VEC-28, $F_{th} = 5.75 \cdot 10^{10}$ 1/cm^2 s, L_{Cd} = 13.5 by gold.

The error of F_{th} measurement constitutes $\approx$4% and is conditioned in the main by uncertainty of γ-radiation registration efficiency by semiconductor detector.

3 Production and calibration of ^{241}Am samples

In the measurements there was used ^{241}Am of high enrichment, the relative content of ^{242m}Am and ^{243}Am atoms in the samples was $< 3 \cdot 10^{-6}$. Prior to application on substrates americium underwent purification of inert and radioactive admixtures using a method of ion-exchange chromatography on cation-exchange and anion-exchange columns. The layers for fission cross section measurement were produced using a method of electrodeposition from alcoholic solution. After application each layer was annealed in a flame of a spirit lamp. The active spot diameter on each backing was 6.0 ± 0.1 mm.

The content of ^{241}Am isotope was determined by two methods independently: alpha-counting and gamma-spectrometry. In both cases a relative method was applied, the certified samples whose activity was determined with the accuracy of 1.4% at p = 0.95 were used as standards. The deviations in the results of "weighing" by two methods did not exceed 2% as a rule.

To measure radiation capture cross sections there were produced several ampoules of quartz glass containing purified nitrate of americium-241. The content of ^{241}Am in the layers constituted from 12.9 to 16.8 μg; in the ampoules – from 32.3 to 35.5 μg.

4 Irradiation of samples, registration of fission fragments and spectra of alpha-particles

Three layers of ^{241}Am containing $3.60 \cdot 10^{16}$, $4.05 \cdot 10^{16}$ and $3.19 \cdot 10^{16}$ nuclei of americium were irradiated in VEC-25 channel within 30 min, 1.0 hour and 1.5 hour, correspondingly. The layers were arranged in aluminum chambers. Immediately after irradiation dielectric track detectors (plates of silicate glass) were established in the chambers, and the measurements were continued using VEC-28.

The chambers were designed in such a way that the detecting plate of glass occurred at a distance of 6.00 ± 0.02 mm from the surface layers, the diameter of the detector registering surface being also 6.00 ± 0.02 mm. At such geometric dimensions the probability of one of fission fragments hit to the area of the detector sensitive surface constitutes (9.37 ± 0.10)%; the error is conditioned by a possible irregularity of americium application on the backing.

The measurement cycle, i.e. time interval from termination of irradiation on VEC-25 to conclusive irradiation on VEC-28 constituted some more than 50 hours.

The chambers were irradiated in turn: without a cadmium jacket and in a jacket; the irradiation duration with no jacket was 2 minutes, in a jacket – 5 minutes.

For additional control and normalization, the chambers with americium layers were irradiated back-to-back jointly with similar chambers with certified layers of ^{235}U or ^{239}Pu. Also for the purpose of additional control, along with the chambers there were several times irradiated activation detectors of golden foil. During the measurement cycle there was measured the yield of spontaneous fission fragments.

After each irradiation event in VEC-28 channel the chambers were kept for 30–40 minutes beyond the radiation screen in order to decrease the radiation dose conditioned by aluminum activation. After that glass detectors with latent tracks caused by fission fragments were taken from the chambers for further etching with hydrofluoric acid, new glasses were established in the chambers and all operations were repeated.

Within the measurement cycle there were registered the α-particle spectra of all the three layers of ^{241}Am. The measurements were performed on a semiconductor alpha-spectrometer with 30-keV resolution. There was thoroughly measured the relative yield of ^{242}Cm α-particles with E_α = 6113 keV and E_α = 6069 keV to the yield of ^{241}Am α-particles with E_α = 5485 keV and E_α = 5443 keV.

Two ampoules with americium-241 nitrate were irradiated inVEC-25 channel within 69.6 hour. The neutron fluence in this case constituted $(0.495 \pm 0.021) \cdot 10^{19}$ 1/cm^2.

5 Data processing, results

5.1 Spectra of α-particles

Figure 1 presents as an example a spectrum of α-particles from the layer of ^{241}Am irradiated in VEC-25 channel within 1.5 hours and measured after the completion of the measurement time, i.e., 66.3 hours after irradiation termination.

Figure 2 illustrates the rise of relative yield of ^{242}Cm α-particles conditioned by β-decay of ^{242g}Am from three irradiated samples of ^{241}Am with different duration of irradiation T_{ir}.

Through the experimental points there are drawn approximating curves $Y_i(t) = A_i(1 - \exp(-\lambda t))$, where λ – constant of ^{242g}Am decay, t – time after termination of the layer irradiation in VEC-25 channel, i – sample number, A_i – parameters determined using a method of least squares.

Basing on the obtained data in the yield of α-irradiation and taking into account the probability of ^{242g}Am β^--decay,

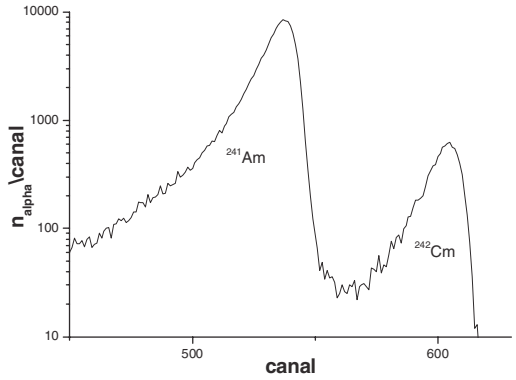

Fig. 1. Spectrum of α-particles from irradiated sample of ^{241}Am.

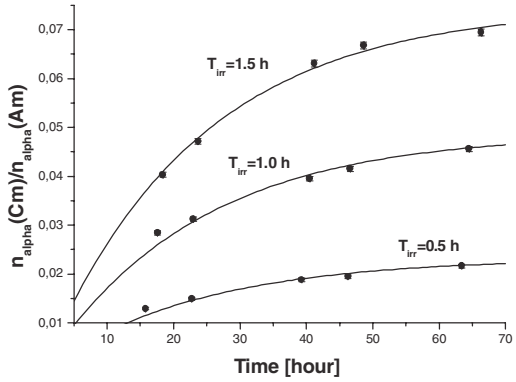

Fig. 2. Time dependencies of ^{242}Cm α-particle yield from irradiated samples of ^{241}Am.

Table 1. Results of ^{241}Am layers irradiation in VEC-25 channel.

	$N_{nucl.}(^{241}Am)$	$N_{nucl.}(^{242g}Am)$
1	$3.60 \cdot 10^{16}$	$(1.03 \pm 0.03) \cdot 10^{12}$
2	$4.05 \cdot 10^{16}$	$(2.44 \pm 0.04) \cdot 10^{12}$
3	$3.19 \cdot 10^{16}$	$(2.94 \pm 0.03) \cdot 10^{12}$

there is calculated the number of ^{242g}Am nuclei accumulated in each layer of ^{241}Am by the end of irradiation by neutrons in VEC-25 channel. The results of calculations are available in table 1.

Basing on the results of measuring α-spectra from americium-241 layers the cross section of ^{241}Am(n,γ)^{242g}Am reaction on the spectrum of neutrons in VEC-25 channel constitutes σ(n,γ) = 836 ± 32 barns.

5.2 Fission fragments

Dielectric track detectors underwent etching with hydrofluoric acid. The observation of detectors and count of tracks caused by fission fragments were implemented visually with the aid of optical microscope.

On each detector registering fission fragments there were fixed ≈30 000 tracks (two-minute irradiation in VEC-28 channel with no cadmium jacket) and ≈10 000 (five-minute irradiation in a cadmium jacket).

Figure 3 presents time distribution of the yield of registered fission fragments from layer 3 (irradiation with no cadmium jacket). There is also presented in the figure the time dependence of the assumed yield of ^{242g}Am fission fragments in a supposition that $\sigma_f(^{242g}Am)$ = 2100 barn. Unfortunately, we did not manage to find out reliably the exponent with $T_{1/2}$ = 16.02 hour in the experimental data on the yield of fission fragments presented in figure 3. In the irradiated layer of americium-241 there are contained the nuclei of ^{241}Am, ^{242m}Am and ^{242g}Am and the accumulation of ^{242}Cm nuclei takes place. Fission of ^{242m}Am nuclei and sub-barrier fission of ^{241}Am nuclei gives a constant contribution to the yield of fission fragments but this contribution turned out to be too large, the scattering of experimental points almost completely "disguised" the yield of ^{242g}Am nuclei fission fragments exponentially dependant on time. The statistical analysis of data made it possible to get the upper evaluation: $\sigma_f(^{242g}Am) \leq 1000$ barn.

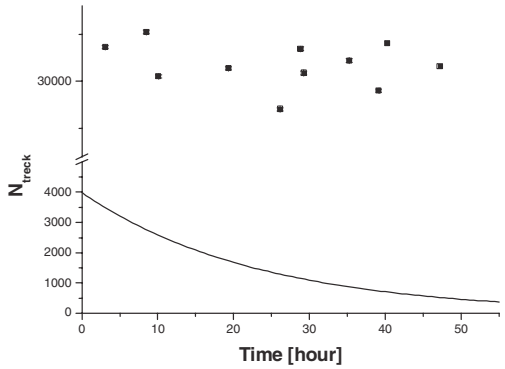

Fig. 3. Yield of fission fragments from layer 3.

The experimental data obtained at measurements with cadmium jacket turned out to be more informative. As in these measurements a calibrated layer of plutonium-239 was used as a standard and basing on the results of the experiment there was obtained the value of the ratio of resonance fission integrals: $I_f(^{242g}Am)/I_f(^{239}Pu) = 0.325 \pm 0.047$. By using the recommended value $I_f(^{239}Pu)$ = 299 barn we get the value $I_f(^{242g}Am) = 97 \pm 18$ barn.

5.3 Mass-spectrometric analysis of irradiated samples of americium-241

After the ampoules with irradiated nitrate of americium-241 had been hold for some time they were crushed by a special appliance, the content of ampoules underwent purification on cation-exchange resin. To refine the results there were performed isolation and purification of americium that did not undergo irradiation. The isotopic analysis of americium samples was realized on mass-spectrometer with resolution power 500.

Basing on the results of measurements there was obtained the following isotopic ratios: $^{243}Am/^{241}Am < 10^{-6}$, $^{242m}Am/^{241m}Am = (7.36 \pm 0.08)10^{-4}$; confidence probability p = 0.95 (2σ).

Most probably, the fluence of $(0.495 \pm 0.011)10^{19}$ 1/cm^2 turned out to be insufficient to produce the required quantity of ^{243}Am.

Basing on the results of mass-analysis the cross section of ^{241}Am(n,γ)^{242m}Am radiation capture on the spectrum of neutrons in VEC-25 channel of IRT MEPhI reactor constitutes σ(n,γ) = (148.6 ± 7.4) barn.

The value of branching ratio obtained in our measurements is as follows: ^{241}Am(n, γ)^{242g}Am/(^{241}Am(n, γ)^{242g}Am + ^{241}Am(n, γ)^{242m}Am) = 0.848 ± 0.034.

6 Conclusion

The results on cross sections of neutron radiation capture with the formation of ^{242g}Am and ^{242m}Am nuclei obtained in the given paper are somewhat higher than the data given in paper of Fioni et al. [1], ^{241}Am(n,γ)^{242}Am = (696 ± 48) barn and also the value given in ENDF/B-VII (620.8 barn). Most probably, this difference is conditioned by the fact that the neutrons in the IRT MEPhI reactor channels are thermalized insufficiently.

The data on neutron cross sections of ^{242g}Am nucleus given in paper [1] and obtained in our measurements give grounds to assume that the recommended cross sections for this nuclide based on theoretical evaluations are essentially overstated. In any case, further experimental researches of characteristics of neutrons-^{242g}Am nucleus interaction are required.

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