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On a problem of assumed ^{232m}Pa isomer characteristics

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Abstract. To study fission characteristics of protactinium-232, the isotope under investigation was built up in the ^{232}Th(p, n) reaction at a proton energy of $E_p \approx 11.5$ MeV. At irradiating the layer by thermal and resonance neutrons the short-lived components with a half-decay period of ∼ 6 hours were found in the yields of fission fragments. The totality of experimental data can be explained by the fact that at the thorium-232 bombardment by accelerated protons the protactinium-232 nuclei are formed in the isomer state with a half-life of $T_{1/2} \approx 2.3$ hour. The isomer of ^{232m}Pa decays approximately with the similar probability by isomer transition to the ground state ^{232}Pa and, as a result of α-decay, to the isotope ^{228}Ac ($T_{1/2} = 6.15$ hour).

1 Introduction

In 1995–1996 several series of cross-section measurements of fission by thermal neutrons and resonance fission integral for odd-odd nuclide ^{232}Pa ($T_{1/2} = 1.31$ day) were performed in VNIIEF. The motivation of this research and the technique of experiments are described in detail in other papers [1,2]. In the course of measurements the layer of protactinium-232 and the layer of uranium-235, used as a benchmark, were irradiated by moderated neutrons. The fission fragments were registered by dielectric track detectors. One of the measurement results obtained in one of the series is available in figure 1.

The experimental results on the fission fragment yield were approximated by a three-parameter curve:

$$Y(t) = p_1 \cdot \exp(-t/8.8726) + p_2 \cdot \exp(-t/45.358) + p_3[1 - \exp(-t/45358)], \quad (1)$$

where $p_2 = (0.02245 \pm 0.00063)$ is a parameter proportional to the yield of ^{232}Pa fission fragments ($T_{1/2} = 31.44$ hour), $p_3 = (0.00200 \pm 0.00030)$ is a parameter proportional to the yield of ^{232}U fission fragments, and $p_1 = (0.0136 \pm 0.0012)$.

The issue of the short-lived component ($T_{1/2} \sim 6$ hour) originating from the time dependence of the fission fragment yield at the irradiation of the protactinium-232 layer by thermal and super-cadmium neutrons has been discussed for quite a long time. Different versions were studied and the additional cycle of measurements was performed. Finally, the assumption was made that a short-lived component is conditioned by fission by neutrons of the odd-odd nucleus Ac-228 ($T_{1/2} = 6.15$ hour).

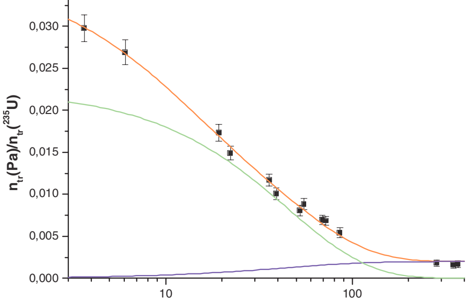

Fig. 1. Time distribution of fission fragments yield from the layer of protactinium under irradiation by thermal neutrons.

It should be mentioned that the Ac-228 isotope is a daughter product of Th-232 decay and it is available in any thorium sample in equilibrium with thorium, i.e., at the protactinium isolation from the irradiated thorium target some number of Ac-228 nuclei can get to the protactinium target. However, because of a large difference in decay periods of Ac-228 and Th-232, the natural content of actinium-228 nuclei in thorium samples is so negligible that one can neglect its effect on the results of the majority of nuclear-physics research.

Thus, the hypothetic mechanism of Ac-228 nuclei generation is as follows: at the irradiation of Th-232 nuclei by protons with the energy of $E_p \approx 11.5$ MeV, some portion or all nuclei of protactinium-232 are formed in the isomer state, then these nuclei undergo an isomeric transition to the ground state of Pa-232 and, partially, α-decay with the formation of Ac-228 nuclei. As a result of β-decay ($T_{1/2} = 1.31$ day) the nuclei of Pa-232 pass to the levels of U-232 nuclei. In this case the 2^- level (E = 1016.8 MeV) of the ^{232}U nucleus is occupied with the probability of 71.7% [3].

This level decays to lower levels with the emission of γ-quanta with an energy of 969.2 keV (41%), 453.6 keV (5.7%), etc. [3].

As a result of β-decay ($T_{1/2} = 6.15$ hour), the nuclei of ^{228}Ac occupy the levels of the ^{228}Th nucleus. The transition of ^{228}Th to its ground state is accompanied by the emission of γ-quanta, among which a highly considerable (16.2%) γ-line with the energy of 968.971 keV [3] is available.

Thus, the newly produced sample of protactinium-232 can emit a group of γ-quanta with the energy close to 969 keV, however, being dependent on the nucleus emitting these γ-quanta, the intensity decrease rate of quanta having this energy will be different ($T_{1/2} = 1.31$ day or 6.15 hour).

The peculiarity of experimental research with samples of sufficiently short-lived nuclides consists in the fact that in the newly produced sample, a variation of elemental composition through natural radioactivity takes place in the course of time.

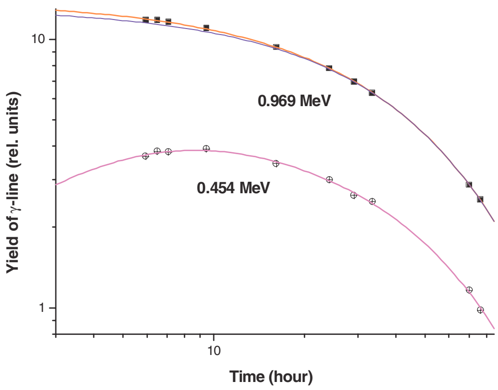

Fig. 2. Time dependencies of the yield of γ-quanta with E_γ = 453.6 keV and $E_\gamma \approx$ 969 keV from the thorium target irradiated by accelerated protons.

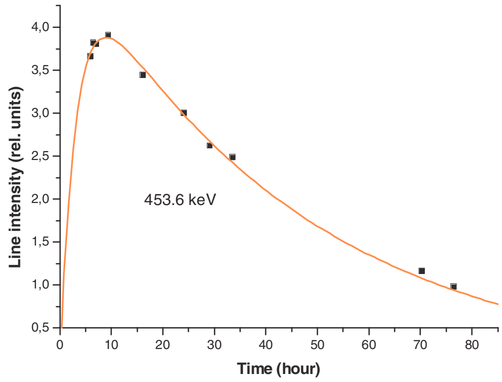

Fig. 3. Time dependence analysis of γ-quanta yield for E_γ = 453.6 keV.

Thus, in the layer of protactinium-232, at different time moments the nuclei of the ^{232m}Pa isomer (on assumption), actinium-228, protactinium-232 and uranium-232 may be available at different amounts. In the practice of the nuclear-physics measurements undertaken with the use of thermal neutrons one can most probably neglect the presence of ^{228}Ac and ^{232}U decay products.

Analysis of time dependence of gamma-quanta yield

During one of the research sessions of ^{232}Pa fission characteristics, the irradiation of a plate of thorium-232 by protons with an energy of $E_p \approx$ 11.5 MeV was performed on a tandem electrostatic accelerator EGP-10. Then, with no isolation of the accumulated protactinium-232 the yield of separate lines of γ-quanta from the irradiated sample of thorium was measured. The main goal of these measurements was to estimate the amount of accumulated protactinium-232 and to confirm the fact that the rate of the decrease of intensity of the given γ-lines corresponds to the half-decay period of protactinium-232.

A HPGe-detector with an energy resolution $\Delta E_\gamma \approx 1.7 \div 1.8$ keV at $E_\gamma \leq 1.5$ MeV was used in a gamma-spectrometer. The duration of the thorium target irradiation constituted 2.5 hours, then, in 5.5 hours and gamma-spectrometric investigations were initiated. Within the 5.95 ÷ 76.43-hour time interval after irradiation termination there were registered 10γ-spectra.

At that time the analysis of separate γ-lines output for short time intervals after irradiation completion was performed not thoroughly enough; it was assumed that, by confirmation of the availability of time decrease with $T_{1/2}$ = 1.31 day, the value of the obtained information was concluded.

At processing the data for the given paper we mainly took advantage of the data on γ-lines with E_γ = 453.6 keV and $E_\gamma \approx$ 969 keV. We have already mentioned that ^{232}Pa and ^{228}Ac contribute to the line with $E_\gamma \approx$ 969 keV.

Figures 2 and 3 give the time dependencies of the yield of γ-quanta with E_γ = 453.6 keV and $E_\gamma \approx$ 969 keV. Even without a thorough computer analysis it is evident that the formation of the source of emitting the 453.6 keV line has at least two stages: build-up (ascending segment of the curve) and decay (descending segment of the curve). In our opinion this fact is already a serious argument in favor of a hypothesis on the generation and subsequent decay of the ^{232m}Pa isomer state at the thorium-232 bombardment by accelerated protons. For the lines with $E_\gamma \approx$ 969 keV the character of time dependence is not so evident.

The following approximating function most precisely describes time dependence of the 453.6 keV γ-line yield:

$$Y_1(t) = p_2[1 - \exp(-t/p_1)]\exp[-t/45.368], \qquad (2)$$

where t stands for time, p_1 = (3.347 ± 0.152) hour for the life-time of isomer state, and p_2 for the parameter defining the yield of protactinium-232 in the isomer state only.

Our conclusion is that at the thorium-232 bombardment by ≈ 11.5 MeV protons the nuclei of protactinium-232 are formed predominantly in the isomer state (^{232m}Pa). According to our measurements the life time of ^{232m}Pa is (3.35 ± 0.15) hour, thus, the half-life is $T_{1/2}$ = (2.32 ± 0.10) hour. This value was used in further calculations.

It has already been mentioned that γ-quanta with $E_\gamma \approx$ 969 keV are emitted both at β-decay of ^{232}Pa and at β-decay of ^{228}Ac. Thus, the time dependence of the yield of these γ-quanta, conditioned by β-decay of ^{232}Pa, should be the same as in case of the 453.6 keV γ-line. For γ-quanta with $E_\gamma \approx$ 969 keV emitted at β-decay of ^{228}Ac the situation is somewhat different. The accumulation of ^{228}Ac occurs through the decay of ^{232m}Pa isomer, while the descending segment of the curve of these γ-quanta yield – through β-decay of ^{228}Ac with a half-life $T_{1/2}$ = 6.15 hours. Thus, for the time dependence analysis of the yield of γ-quanta with $E_\gamma \approx$ 969 keV the following function was applied:

$$Y_2(t) = (P_1 \cdot \exp(-t/45.358) + P_2 \cdot \exp(-t/8.87)[1 - \exp(-t/3.347)], \qquad (3)$$

where P_1 stands for the parameter proportional to the probability of isomer transition ^{232m}Pa → ^{232}Pa and probability of the 969.2 keV γ-line emission (41%), and P_2 is the parameter proportional to the probability of α-decay ^{232m}Pa → ^{228}Ac and the 968.971 keV γ-line emission (16.2%). From the calculated values of P_1 and P_2 parameters follows that the relation between the probability of α-decay of the isomer ^{232m}Pa and the probability of ^{232m}Pa isomer transition (branching ratio) is 1.09 ± 0.11.

Thus, the scheme of ^{232m}Pa isomer decay can be presented in the following form:

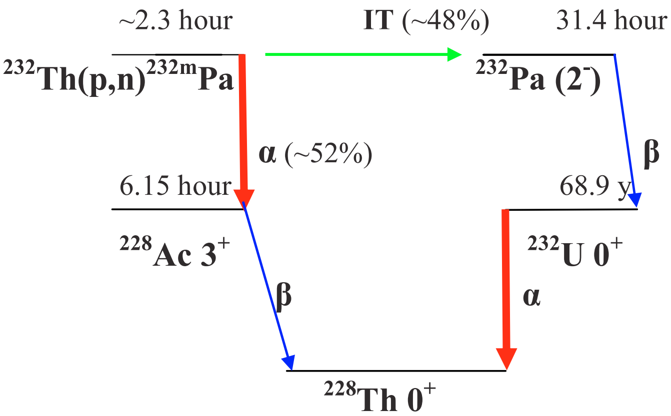

Fig. 4. Scheme of ^{232m}Pa formation and decay.

2 Conclusion

As a result of the analysis of previously obtained experimental data, the hypothesis on the existence of an isomer state of the ^{232m}Pa nucleus got a ponderable confirmation. The lifetime of this state was measured, and the decay channels were determined.

The results of measurements presented in refs. [1,2] were slightly corrected and added. We report here only the final results: $\sigma_f(^{232}\text{Pa}) = (1021 \pm 37)$ b, $I_f(^{232}\text{Pa}) = (1066 \pm 375)$ b, $\sigma_f(^{232}\text{U}) = (83.3 \pm 11.0)$ b, $I_f(^{232}\text{U}) = (1194 \pm 102)$ b, $\sigma_f(^{228}\text{Ac}) = (520 \pm 100)$ b.

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