β -delayed fission of ²²⁸Ac^{*}

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Abstract The basic process of an exotic decay mode namely β -delayed fission is simply introduced. The progress status of the study in the world is essentialized. The observation of β -delayed fission of ²²⁸Ac is reported. The radium was radiochemically separated from natural thorium. Thin Ra sources in which ²²⁸Ac was got through ²²⁸Ra $\xrightarrow{\beta^-}$ ²²⁸Ac were prepared for observing fission fragments from β -delayed fission of ²²⁸Ac. They exposed to the mica fission track detectors, and measured by an HPGe γ -ray detector. The β -delayed fission events of ²²⁸Ac were observed and its β -delayed fission probability was found to be $(5\pm 2)\times 10^{-12}$.

Key words 228 Ac, β -delayed fission, radiochemical separation, mica fission track detector

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Nuclei far from stability are characterized by a number of new phenomena. In heavy neutron-rich region one of these is β -delayed fission (β DF). A β -delayed fission island was predicted to occur in heavy neutron-rich region by E.Ye. Berlovich and Yu.N. Novikov in 1969^[1]. β DF is a nuclear decay process in which a β -decaying nucleus populates exited states in its daughter nucleus which then fission, because these states can be above the fission barrier(s) of the daughter. The β DF process can be represented schematically as follows:

$$(Z,N) \xrightarrow{\beta} (Z+1,N-1)^* \xrightarrow{\theta} \sum_i (Z_i,N_i),$$

where (Z, N) is the fission precursor with proton number Z and neutron number N, (Z_i, N_i) the fission products and an asterisk indicates excited state of an intermediate nucleus (Z+1, N-1). And β DF region can be obtained from the condition of complying with the following inequalities (1) and (2) using mass value from^[2]:

$$T_{1/2}(\beta) \leqslant T_{1/2}(SF), \tag{1}$$

where $T_{1/2}(\beta)$ and $T_{1/2}(SF)$ are half-lives of β -decay

and spontaneous fission of a precursor, respectively.

$$Q_{\beta}(Z, N \to Z+1, N-1) \ge B_{\rm f}(Z+1, N-1),$$
 (2)

where $Q_{\beta}(Z, N \to Z+1, N-1)$ is β -decay energy of a precursor and $B_{\rm f}(Z+1, N-1)$ is fission barrier height of its daughter.

Thielemann and Klapdor et al. have carried out theoretical investigation of β DF probabilities of the nuclei in the island region and shown its role in the production of heavy elements and built up of heavy isotopic abundances in astrophysical r process as well as the influence of the delayed fission on the production of cosmochronometers^[2-8].

In principle, direct tests of β DF theory could be provided by measurements of β DF probabilities $P_{\beta \text{DF}} = \frac{N_{\beta \text{DF}}}{N_{\beta}}$ (the ratio of the number of fission events after β decay to the total number of β -decays of the parent). Unfortunately, the number of isotopes (accessible through practical nuclear reactions) for which detectable $P_{\beta \text{DF}}$ might be expected is very limited. There have been some reports of experimental measurements for β DF probabilities in neutron-rich

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actinide and the nearby region $^{[9-13]}$.

In this paper, we present the search for βDF of ²²⁸Ac and the determination of its βDF probability.

In view of the very low production cross sections in this mass region, efficient reactions with high enough production rate are crucial for the experimental study of β DF. We notice that natural thorium in equilibrium with its decay chain contains long-lived ²²⁸Ra and ²²⁴Ra in amounts corresponding to a large production rate^[14]. There are ~1×10¹² nuclei of ²²⁸Ra in 1 g of natural thorium. Therefore separating radium from natural thorium should be a good way to acquire ²²⁸Ra.

In present work the radium was separated from the mixture of the bulk of thorium and its decay products by the method of the BaCl₂ co-precipitation using Ba carrier. The ThO₂powder (2 g) was dissolved, and radium was separated. Then the thin solid sources ($\sim 0.2 \text{ mg/cm}^2$) containing ²²⁸Ra^{β^-} ²²⁸Ac activities were prepared for the detection of fission tracks. A total number of 22 such sources were made.

The trace radium can be carried by BaCl₂ precipitate quantitatively. The chemical yield of Ba was determined to be $\sim 70\%$ by using ¹³³Ba as tracer. And the decontamination factor for thorium was $\sim 2 \times 10^4$.

At the same time, five thin sources each containing 1 mg thorium were also prepared.

The mica foils, as fission track detectors, were preetched in a solution of 40% HF at 50°C for 4 hours in order to identify the fission tracks that already existed in the detectors (natural background). Then the sources (²²⁸Ra $\stackrel{\beta^-}{\longrightarrow}$ ²²⁸Ac sources and thorium sources) were stuck on the mica foils. They together with an HPGe γ -ray detector were well shielded with lead and paraffin. The sources were exposed to the mica fission track detectors for 720 days (from December 1, 2003 to Nov. 20, 2005). And the γ -spectrum measurements for the sources were performed.

After the process mentioned above, all mica foils were etched again for 4 hours. Then they were scanned by an optical microscope. The same 22 run experiments (including the whole process from preetching to scanning) mentioned above without the sources (blank experiments) were carried out.

As a result, 18 fission fragment tracks were observed in the mica foils exposed by $^{228}\text{Ra} \xrightarrow{\beta^-} ^{228}\text{Ac}$ sources. A sample of them is presented in Fig. 1(a).

And in the blank experiments, one fission event shown in Fig. 1(b) was found. This event should originate from spontaneous fission or induced-fission in the fissile contaminants in the mica track detectors.



¹Ig. 1. (a) A sample of fission fragment tracks observed in the mica foils exposed by ${}^{228}\text{Ra} \xrightarrow{\beta^-} {}^{228}\text{Ac}$ sources; (b) The observed fission fragment track in the blank experiments; (c) A sample of natural background fission fragment tracks.

The observed 17 (18-1) fragment tracks should com the β DF of ²²⁸Ac.

The observed 17 fragment tracks differ clearly in size from those of natural background of fission fragments [(A sample of them is shown in Fig. 1(c)]. Because the total etching time for natural fission fragment tracks was 8 hours whereas it was only 4 hours for fission fragment tracks from the ${}^{228}\text{Ra} \xrightarrow{\beta^-} {}^{228}\text{Ac}$ sources and from the fissile contaminants in blank experiments. Therefore the natural background fission fragment tracks can be excluded as the origin of the 17 fission events. The observed 17 fission events cannot come from the thorium remained in $^{228}\text{Ra} \xrightarrow{\beta^-} ^{228}\text{Ac}$ sources. In the five prepared thorium sources mentioned above, the quantity of thorium was about twice of remained thorium in the 22 228 Ra $\xrightarrow{\beta^-}$ 228 Ac sources because of the $\sim 2 \times 10^4$ decontamination factor for thorium. The same procedure described above (including exposition, etching, and scan) was performed. As a consequence no fission event was found. The observed 17 fission events can not come from cluster emitter nuclei in our sources. As for mica foils, cluster lighter than about mass 30 will not register^[15, 16]</sup>. In our experiments, the mass of clusters originating from the sources is less than 30, because nuclei with $Z \leq 90$ will only emit cluster lighter than mass $30^{[17-19]}$. Only ²²⁴Ra was reported to be the cluster emitter nucleus in our sources $(^{224}\text{Ra}\rightarrow^{14}\text{C}+^{210}\text{Pb})$. The measured γ -ray spectra

indicate that all of the observed γ rays came from daughters of the ²²⁸Ra such as ²²⁸Ac and ²²⁴Ra. The possibility of the β DF of ²²⁴Ra could also be excluded because of its 100% α decay property^[20]. Consequently the fission fragment tracks observed in the mica foils exposed by ²²⁸Ra $\stackrel{\beta^-}{\longrightarrow}$ ²²⁸Ac sources might come from ²²⁸Ac or ²²⁸Ra. Furthermore the fission events should be attributed to ²²⁸Ac rather than ²²⁸Ra by analysis of β -decay energy and fissility systematics. β DF should appear first of all in add-odd nuclei, since they have the greatest β -decay energy

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and the daughter even-even nuclei are characterized by high fissility^[9].

All the arguments mentioned above indicate that the 17 fission events observed in the experiments could be assigned to the β DF of ²²⁸Ac.

A total of $3.3 \times 10^{12} {}^{228}$ Ac β decay accepted by the fission track detectors has been determined through peak areas and branching ratios of the γ rays of 228 Ac in the measured γ spectra^[21]. Thus the β DF probability of 228 Ac was determined to be $(5\pm 2) \times 10^{-12}$.

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