Search for β-delayed fission of ^{228}Ac

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Radium was radiochemically separated from natural thorium. Thin $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ sources were prepared and exposed to mica fission track detectors, and measured by an HPGe γ-ray detector. The β-delayed fission events of ^{228}Ac were observed and its β-delayed fission probability was found to be $(5 \pm 2) \times 10^{-12}$.

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For the first time the existence of the β-delayed fission (βDF) island of nuclei in the heavy neutron-rich region and its role in the production of heavy elements in astrophysical r-process were pointed out by Berlovich and Novikov in 1969 [1]. βDF is a nuclear decay process in which a β-decaying nucleus populates excited states in its daughter nucleus which then fission.

The wide theoretical investigations of the βDF rates in a very large region of nuclei were performed and the influence of this process on the production of cosmochronometers has been shown [2–6].

In principle, direct tests of βDF theory could be provided by measurements of βDF probabilities $\mathrm{P}_{\beta\mathrm{DF}} = \frac{N_{\beta\mathrm{DF}}}{N_\beta}$ (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 $\mathrm{P}_{\beta\mathrm{DF}}$ might be expected is very limited. There have been some reports of experimental measurements for βDF probabilities. Gangrskii *et al.* [7], Baas-May *et al.* [8], Hall *et al.* [9], Mezilev *et al.* [10], and Shuanggui *et al.* [11] successively performed experimental searches and probability determinations for βDF in neutron-rich actinide and the nearby region.

In this Brief Report, we present the search for βDF of ^{228}Ac 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 rates are crucial for the experimental study of βDF. We notice that natural thorium in equilibrium with its decay chain contains long-lived ^{228}Ra and ^{224}Ra in amounts corresponding to a large production rate [12]. There are $\sim 1 \times 10^{12}$ nuclei of ^{228}Ra in 1 g of natural thorium. Therefore separating radium from natural thorium should be a good way to acquire ^{228}Ra.

In the present work the radium was separated from the mixture of the bulk of thorium and its decay products by the method of the $BaCl_2$ co-precipitation using a Ba carrier. The procedure was described as the following:

(i) The ThO_2 powder (2 g) was dissolved into 5 ml of boiling conc. HNO_3-0.05 mol/L HF solution containing two drops of $(NH_4)_2SiF_6$.

(ii) The solution was transferred into a 50 ml centrifuge tube containing 1 ml of conc. hydrochloric acid. Then 4 mg of barium carrier was added and the solution was stirred. The tube was put in an ice-water bath. After 7 min, it was centrifuged and the supernatant was discarded.

(iii) Several drops of hot distilled water were added to the tube to dissolve the $BaCl_2$ precipitate completely.

(iv) The test tube was put into the ice bath. 6 ml of 5:1(V/V) conc. HCl-ether mixture was added to the tube. After 7 min, it was centrifuged and the supernatant was discarded.

(v) Steps 3 and 4 were repeated two times. Finally the $BaCl_2$ precipitate containing radium activities was obtained.

(vi) The $BaCl_2$ precipitate was dissolved with high purifying water and the thin solid sources ($\sim$0.2 mg/cm^2) containing $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ activities were prepared for the detection of fission tracks.

The trace radium can be carried by $BaCl_2$ precipitate quantitatively. The chemical yield of Ba was determined to be $\sim$70% by using ^{133}Ba 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.

Mica foils, as fission track detectors, were pre-etched in a solution of 40% HF at 50°C for 4 h in order to identify the fission tracks that already existed in the detectors (natural background). Then the sources ($^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ 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 mica fission track detectors for 720 d (from 1 December 2003 to 20 November 2005). And the γ-spectrum measurements for the sources were performed.

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

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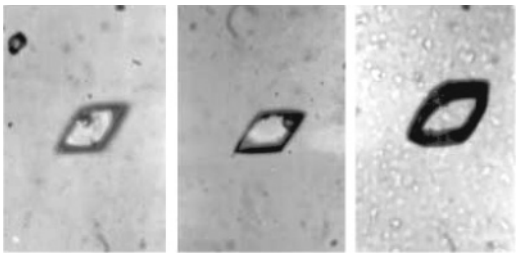

FIG. 1. (a) A sample of fission fragment tracks observed in mica foils exposed by $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ sources. (b) The observed fission fragment track in the blank experiments. (c) A sample of natural background fission fragment tracks.

As a result, 18 fission fragment tracks were observed in the mica foils exposed by $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ 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. The observed 17 (18–1) fragment tracks provide the evidence for the βDF of ^{228}Ac based on the following arguments:

(i) Firstly, the natural background fission fragment tracks [a sample of them is shown in Fig. 1(c)] can be excluded as the origin of the 17 fission events based on the clearly different size comparing to the tracks show in Fig. 1(a) and 1(b). Because the total etching time for natural fission fragment tracks was 8 h whereas it was only 4 h for fission fragment tracks from the $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ sources and from the fissile contaminants in blank experiments.

(ii) The observed 17 fission events cannot come from the thorium remaining in $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ sources. In the five prepared thorium sources mentioned above, the quantity of thorium was about twice the remaining thorium in the 22 $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ 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.

(iii) The observed 17 fission events cannot come from cluster emitter nuclei in our sources. As for mica foils, cluster lighter than mass about 30 will not register [13,14]. In our experiments, the mass of clusters originating from the sources is less than 30, because nuclei with $Z \leqslant 90$ will only emit cluster lighter than mass 30 [15–17]. Only ^{224}Ra was reported to be the cluster emitter nucleus in our sources ($^{224}\mathrm{Ra} \rightarrow {}^{14}\mathrm{C} + {}^{210}\mathrm{Pb}$).

(iv) A sample of the measured γ-ray spectra as shown in Fig. 2 indicates that all of the observed γ rays came from daughters of the ^{228}Ra such as ^{228}Ac and ^{224}Ra. The possibility of the βDF of ^{224}Ra could also be excluded because of its 100% α decay property [18]. Otherwise, ^{224}Ra is more near to the β stability line. As for βDF, decrease of each neutron will result in great decrease of βDF probability. In addition, ^{224}Ra is not an odd-odd nucleus. Based on the systematics [see details in point (5)], it is not favored for the βDF.

(v) Therefore the fission fragment tracks observed in mica foils exposed by $^{228}\mathrm{Ra}\xrightarrow{\beta^-}{}^{228}\mathrm{Ac}$ sources might come from ^{228}Ac or ^{228}Ra. Furthermore the fission events should be attributed to ^{228}Ac rather than ^{228}Ra by analysis of β-decay energy and fissility systematics. βDF should appear first of all in odd-odd nuclei, since they have the greatest β-decay energy and the daughter even-even nuclei are characterized by high fissility [7]. As for ^{228}Ac, the β-decay energy is 2127 keV [19], which feeds to more than 50 excited states of ^{228}Th including some higher excited states with energies up to 2123.1 keV through β^- decay. Whereas the β-decay energy for ^{228}Ra is only 45.9 keV [19]. Only two very low excited states of ^{228}Ac with the energies of 6.67 and 31 keV are populated through β^- decay. At the other hand, even-even nuclei ^{228}Th (β^- decay daughter of ^{228}Ac) have a higher fissility than odd-odd nuclei ^{228}Ac (β^- decay daughter of ^{228}Ra) [1–11]. So from the above arguments, we can exclude ^{228}Ra as the resource of βDF. Up to now, the known nuclei whose βDF probability have

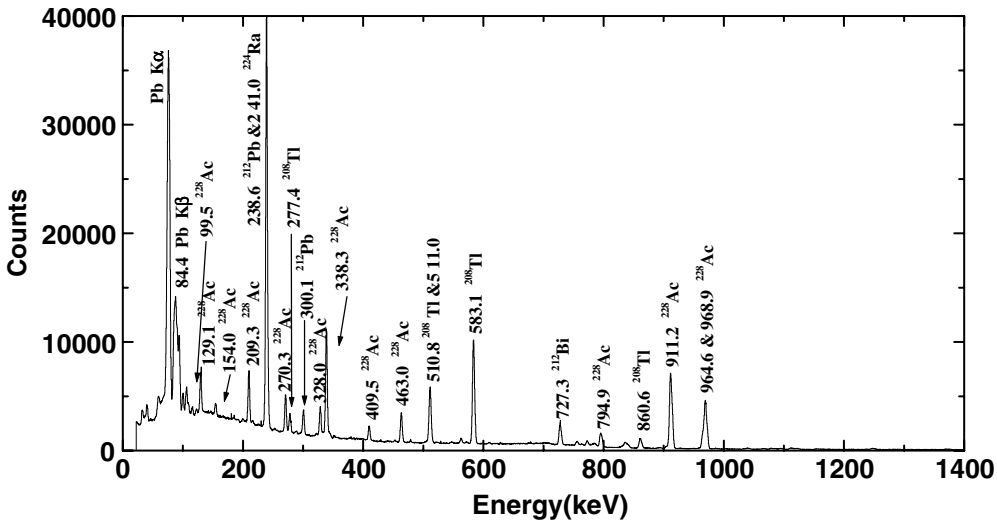

FIG. 2. The measured γ-ray spectrum.

been determined are all odd-odd nuclei, such as ^{234,236,238}Pa, ^{256m}Es, and ^{230}Ac, etc. [7,9,11]. Even in the neutron-deficient region, the events of electron capture delayed fission (εDF) are all happened in odd-odd nuclei, such as ^{228}Np, ^{232,234}Am, ^{238,240}Bk, ^{242,244,246,248}Es, and ^{250}Md, etc. [20–28]. This also supports our conclusion.

All the arguments above indicate that the 17 fission events observed in the experiments could be assigned to the βDF of ^{228}Ac.

A total of 3.3×10^{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 [19]. It is consistent with this number obtained through the used ThO_2 quantity and chemical yield. Consequently the βDF probability of ^{228}Ac was determined to be $(5 \pm 2) \times 10^{-12}$.

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[1] E. Ye. Berlovich and Yu. N. Novikov, Phys. Lett. **B29**, 155 (1969).
[2] H. V. Klapdor *et al*., Z. Phys. A **299**, 213 (1981).
[3] F. K. Thielemann, J. Metzinger, and H. V. Klapdor, Z. Phys. A **309**, 301 (1983).
[4] H. V. Klapdor, Fortschr. Phys. **33**, 1 (1985).
[5] F. K. Thielemann, J. Metzinger, and H. V. Klapdor, Z. Phys. A **309**, 301 (1983).
[6] A. Staudt and H. V. Klapdor-Kleingrothaus, Nucl. Phys. **A549**, 254 (1992).
[7] Yu. P. Gangrskii *et al*., Sov. J. Nucl. Phys. **27(4)**, 475 (1978).
[8] A. Baas-May, J. V. Kratz, and N. Trautmann, Z. Phys. A **322**, 457 (1985).
[9] H. L. Hall *et al*., Phys. Rev. C **39**, 1866 (1989).
[10] K. A. Mezilev *et al*., Z. Phys. A **337**, 109 (1990).
[11] Y. Shuanggui *et al*., Eur. Phys. J. A **10**, 1 (2001).
[12] J. Gilat and S. Katcoff, J. Inorg. Nucl. Chem. **40**, 369 (1978).
[13] J. Hudis and S. Katcoff, Phys. Rev. **180**, 1122 (1969).
[14] P. R. Prevo, R. E. Dahl, and H. H. Yoshikawa, J. Appl. Phys. **35**, 2636 (1964).
[15] R. K. Gupta and W. Greiner, Int. J. Mod. Phys. E **3**, 335 (1994).
[16] S. Misicu and W. Greiner, J. Phys. G: Nucl. Part. Phys. **29**, 67 (2003).
[17] R. Blendowske *et al*., in *Nuclear Decay Modes*, edited by D. N. Poenaru (IOP Publishing, Bristol, 1996).
[18] A. Artna-Cohen, Nucl. Data Sheets **80**, 227 (1997).
[19] A. Artna-Cohen, Nucl. Data Sheets **80**, 723 (1997).
[20] S. A. Kreek *et al*., Phys. Rev. C **50**, 2288 (1994).
[21] H. L. Hall *et al*., Phys. Rev. C **42**, 1480 (1990).
[22] H. L. Hall *et al*., Phys. Rev. C **41**, 618 (1990).
[23] S. A. Kreek *et al*., Phys. Rev. C **49**, 1859 (1994).
[24] D. Galeriu, J. Phys. (London) G **9**, 309 (1983).
[25] D. A. Shaughnessy *et al*., Phys. Rev. C **61**, 044609 (2000).
[26] D. A. Shaughnessy *et al*., Phys. Rev. C **65**, 024612 (2002).
[27] D. A. Shaughnessy *et al*., Phys. Rev. C **63**, 037603 (2001).
[28] Yu. P. Gangrsky *et al*., Sov. J. Nucl. Phys. **31**, 162 (1980).