Observation of exotic nuclear decay of ^{230}U

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Abstract ^{230}Pa was produced through the ^{232}Th(p,3n)^{230}Pa reaction in the irradiated ThO_2 powder targets with 35 MeV proton beam. Pa was radiochemically separated from ThO_2 and other reaction products, and then thin sources of $^{230}Pa \xrightarrow{\beta^-} {}^{230}U$ were prepared. The polyethylene-terephthalate (lavsan) films were selected as solid track detector searching for the cluster activity of ^{230}U. The gamma activity of ^{230}Pa in the sources was measured using a HPGe detector. The lavsan films were etched in NaOH solution and the etched films were scanned under an optical microscope. The exotic nuclear decay of ^{230}U by emission of ^{22}Ne was observed. A branching ratio relative to α decay was deduced, $B = \lambda_{Ne}/\lambda\alpha = (1.3\pm0.8)\times10^{-14}$ for ^{22}Ne emission from ^{230}U.

Keywords ^{230}U, ^{22}Ne emission, ^{232}Th(p,3n)^{230}Pa reaction
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1 Introduction

Sandulescu *et al*[1] predicted in 1980 that some particular parents such as ^{222,224}Ra, ^{230,232}Th and heavier nuclides up to ^{254}No may emit various heavy clusters such as ^{24}Ne, ^{24}Mg, etc. In fact, before the prediction, Prof. LU Xi-Ting[2] from Peking University had made similar prediction that some heavy nuclei, especially ^{223}Ra, may spontaneously emit ^{12}C. Experimentally, cluster radioactivity was discovered for the first time in 1984[3] when Rose and Jones found in an ingeniously simple experiment that rare events of spontaneous decay of ^{223}Ra in a light ^{14}C fragment and a daughter ^{209}Pb nucleus had been observed with a branching ratio to the α -emission of around 6×10^{-10}.

Gales *et al*[4] confirmed soon the exotic nuclear decay of ^{223}Ra by using an intense radioactive ^{227}Ac source and a magnetic spectrometer. Since that pioneering experiment, intense experimental and theoretical research[5-8] has been focused on studying properties of the exotic decay of the nuclei far from the valley of β stability. These studies have involved in measurements (or predictions) of decay models and branching ratios of the rare decay of the cluster emission.

The heavy fragment emission — the two-body decay of a heavy nucleus into a fragment with Z>2 and a nearly magic residual nucleus by a barrier penetration process — can be viewed as an intermediate process between α decay and fission. Theoretical models fall into two categories, i.e. cluster model and unified model. The former regards the fragment as having a certain probability of forming out of a cluster of nucleus inside the parent nucleus and of assaulting the barrier with a characteristic frequency, while the latter views alpha decay, heavy fragment radioactivity and spontaneous fission as similar processes in which the parent nucleus spontaneously deforms through a shape barrier that includes both Coulomb and nuclear forces.

In the region of neutron-deficient nuclides with Z= 56- 64 in chart of the nuclides, it is possible that there is another heavy fragment emission island. Researchers in Dubna[9] and GSI[10] have reported that they observed ^{12}C emission of ^{114}Ba. But, Guglielmetti *et al*[11] claimed through later experiment that no ^{12}C cluster decay of ^{114}Ba was found by means of on-line mass separator. So far more than twenty nuclides have been known to decay by emission of heavy fragments such as ^{14}C, ^{20}O, ^{19}F, ^{24,25,26}Ne, ^{28,30}Mg, ^{32,34}Si etc. and with branching ratios relative to α-decay from 10^{-9} down to 10^{-16} and partial half-life from 10^{11} up to 10^{28} s.[7]

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However, none of the models agrees with the experimental data within an order of magnitude, and none disagrees so badly with the data as to be ruled out for the branching ratios. Obviously, more experimental studies of heavy fragment radioactivity over a wider range of fragment charges are needed.

Recently Bonetti *et al*[12] reported cluster decay of ^{230}U via Ne emission. Their work is similar to that the present paper will report. In their experiment, ^{230}Pa activities were produced via the ^{232}Th(p, 3n) reaction by irradiating metallic thorium targets. ^{230}U sources were prepared by means of electroplating after separation of ^{230}U from ^{230}Pa. The task of detecting Ne cluster emission was carried out using solid-state track detectors. The branching ratio of ^{230}U for emission of Ne clusters was measured to be $B = (4.8 \pm 2.0) \times 10^{-14}$. On the basis of theoretical calculations, they considered the most probable neon cluster coming out to be ^{22}Ne.

In this paper we report experimental details on observation of ^{22}Ne cluster decay events of ^{230}U.

2 Experimental

2.1 Irradiation

The irradiations were performed at the proton liner accelerator of the Institute of High Energy Physics (IHEP), the Chinese Academy of Sciences. The thorium targets of 2.0 g/cm^2 (in the form of ThO_2) were bombarded by a 35 MeV proton beam with an intensity of 3-4 μA. The irradiation had gone on for a total of 100 h. The ^{230}Pa activity was produced via the ^{232}Th(p, 3n) reaction. During the irradiations, thorium targets were cooled by circulating tap water. The irradiated targets were taken out 6-12 h after the irradiation finishing in order to reduce radiation damnification.

2.2 Radiochemical separation

To remove any contaminating spontaneous fission activities the following chemical procedure was performed. The thorium oxide samples were dissolved in 0.05 mol/L HF-concentrated HNO_3 solution, then precipitated with NH_4OH. The precipitate was dissolved in 0.5 mol/L HF-12 mol/L HCl solution. The Pa in the solution was extracted with 5% tri-iso-octylamine(TIOA)- xylene. The organic phase containing Pa was washed twice with 12 mol/L HCl-0.5 mol/L HF solution in order to further remove the other impurities remained in the organic phase. The Pa in the organic phase was back-extracted with 0.1 mol/L HCl solution. A few drops of $FeCl_3$ solution (0.1 mg Fe) was added into the back-extracted aqueous phase and then the aqueous phase was made alkalinity by adding ammonia spirit. The $Fe(OH)_3$ precipitate carried quantitatively the trace protactinium. The thin Pa sources containing $Fe(OH)_3$ precipitate of 40 μg/cm^2 were prepared by filtering the $Fe(OH)_3$ precipitate.

2.3 Exposure of the lavsan films

A total of six thin sources of $^{230}\mathrm{Pa} \xrightarrow{\beta^-} {}^{230}\mathrm{U}$ were prepared in the experiment. Each of the sources was covered with the lavsan films. The source and the lavsan film were fixed by adhesive tapes on glass plates and placed in a dryer to expose the lavsan films to the sources for 248 days.

2.4 Gamma ray measurement and data analysis

After the chemical separation of the target samples, gamma spectroscopy measurements could not be immediately made for the $^{230}\mathrm{Pa} \xrightarrow{\beta^-} {}^{230}\mathrm{U}$ sources, or else the detector system would be blocked because the activity of ^{230}Pa in the sources is extremely intense. Three months after finishing the irradiation, the activity of $^{230}\mathrm{Pa} \xrightarrow{\beta^-} {}^{230}\mathrm{U}$ sources was measured using a HPGE detector with a resolution of 2.3 keV (FWHM) for the 1332 keV ^{60}Co gamma ray. The photoelectric efficiencies of the detector were determined with a standard ^{152}Eu source. The measured data were stored on the magnetic disks with a PC-CAMAC multi-parameter data acquisition system.

The measured γ-ray spectrum data were analyzed employing a set of computer programs. The γ-rays were identified by the half-lives and the gamma ray intensities of the isotopes. The numbers of nucleus ^{230}Pa in $^{230}\mathrm{Pa} \xrightarrow{\beta^-} {}^{230}\mathrm{U}$ sources were obtained from the γ-ray peak area of ^{230}Pa.

2.5 Etching of lavsan films and scanning of tracks

After a total exposure time of 248 days (14.2 half-lives of ^{230}Pa), each of the films was etched for 2 h in 6.25 mol/L NaOH solution at 70°C. The time was chosen on the basis of previous etching experiments with lavsan films.[13] The etched films were then manually

scanned under an optical microscope to locate the developed cluster tracks, and the lengths as well as mouth diameters of these tracks were measured.

3 Results and discussions

Fig.1 shows the measured partial gamma-ray spectrum of a ^{230}Pa $\xrightarrow{\beta^-}$ ^{230}U source. In Fig.1, the energies (443.8, 898.7, 981.5 and 1009.6 keV) and relative intensities of the γ-rays for ^{230}Pa are in agreement with literature values[13] for ^{230}Pa. Because the gamma spectroscopy measurements were carried out three months after the end of irradiation, all of the gamma rays in Fig.1 came from the isotopes with long half-lives. Therefore, all of the γ-rays should be easily identified from their energies and relative intensities. It can be seen from Fig.1 that the γ-rays can be attributed to the decay of the Pa isotopes produced in the experiment and their daughters except for those of Te^{129m} (33.6 d) and Nb^{95g} (35.0 d). It demonstrates that the decontamination effect for the chemical separation is satisfactory. The γ-ray of 324.2 keV (I_γ = 2.83) in Fig.1 can be attributed to ^{230}U. The numbers of nucleus ^{230}Pa in six ^{230}Pa $\xrightarrow{\beta^-}$ ^{230}U sources were determined to be $(3.0\pm0.2)\times10^{15}$ on the basis of the γ-ray peak areas of ^{230}Pa.

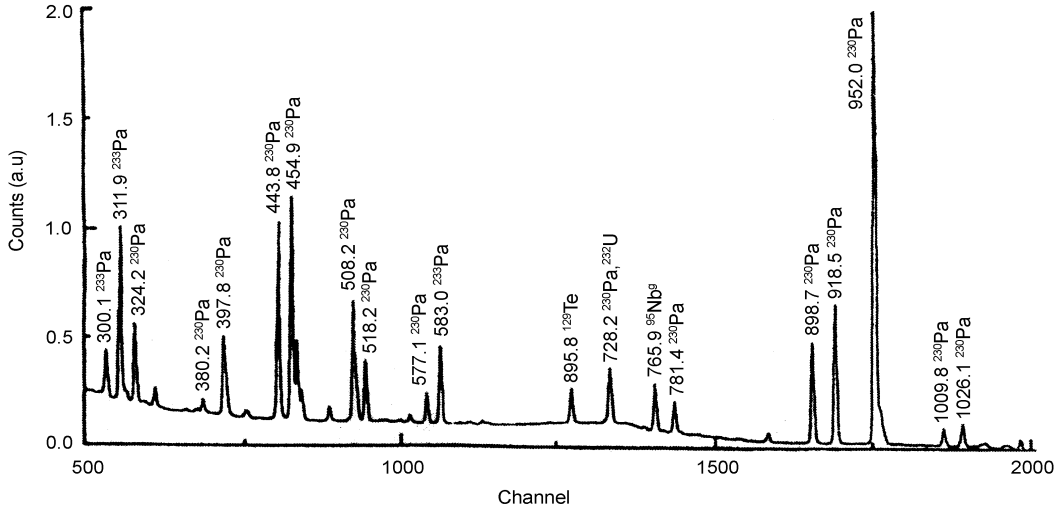

Fig.1 A partial gamma-ray spectrum of ^{230}Pa fraction chemically separated from the 35 MeV protons irradiated thorium dioxide.

As a result of scanning the films, two cluster events have been found in the etched films. Fig.2 illustrates photomicrographs of the two cluster tracks (a) and (b) against the background of α-particles and of track(c) of fission fragments from ^{252}Cf. Both track (a) and track (b) have the same ranges and dimensions, but differ greatly from tracks of α-particles, as well as those of spontaneous fission fragments.

In the series of ^{230}Pa decay chain, only ^{230}U and ^{222}Ra are the candidates for the ^{22}Ne and ^{14}C emission, respectively. It should be indicated that the lightest particles capable of producing an etchable track in the lavsan films are $Z \approx 6$. Controlling the film etching time (2.0 h), the optimum conditions for Ne etching could be selected. Under that condition, therefore, the carbon cluster tracks from ^{222}Ra, having less specific energy loss than Ne, would be etched not fully and were excluded from registration in optical scanning.

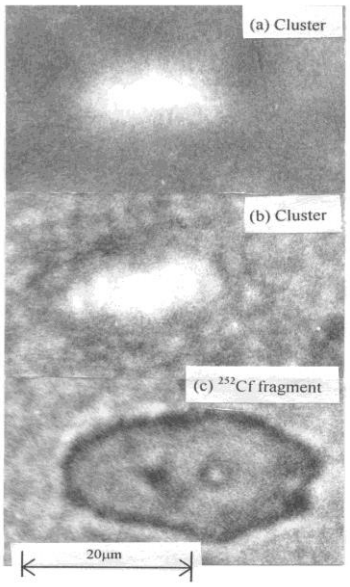

Fig.2 Photomicrographs of two cluster tracks (a) and (b) detected in ^{230}U decay. For comparison, track (c) of spontaneous fission fragment of ^{252}Cf is also shown at the bottom.

Theoretically speaking, ^{230}Pa has the following decay: $^{230}\mathrm{Pa} \xrightarrow{\beta^-} {}^{230}\mathrm{U} \rightarrow {}^{208}\mathrm{Pb} + {}^{22}\mathrm{Ne}$. Therefore, both track (a) and track (b) in Fig.2 should be attributed to ^{22}Ne emission of ^{230}U. From the experimental data, we deduced a branching ratio for ^{22}Ne emission of ^{230}U relative to α decay, $B = \lambda_{\mathrm{Ne}}/\lambda\alpha = (1.3\pm0.8)\times10^{-14}$. Its partial half-life for the emission of ^{22}Ne nuclei corresponds to $(4.4\pm2.7)\times10^{12}$ a.

In summary, we conclude that the exotic nuclear decay of ^{230}U by emission of ^{22}Ne has been confirmed by our experiment described above.

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