Separation of protactinium in the reaction of 60 MeV/nucleon ^{18}O ions with natural uranium

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Abstract The activities of protactinium were produced by the multi-nucleon transfer reactions in bombardment of the natural uranium with 60 MeV/nucleon ^{18}O ions. A simple, relatively fast radiochemical procedure was used for extraction separation of protactinium from the uranium and a variety of reaction products using 1-phenyl-3-methyl-4-benzoyl-5-pyrazolone and tri-iso-octylamine as extractants. The γ ray spectrum of the separated protactinium fractions showed that the protactinium could be separated from all of the main impurity elements. The decontamination factors of the uranium and the main reaction products produced in the reaction are given.

Keywords Pa separation, Extraction with PMBP and TIOA, Heavy ion reaction

CLC numbers O674.233^{+}, O571.42^{+} A

1 INTRODUCTION

For studying the decay properties of short-lived protactinium isotopes produced in the heavy ion induced nuclear reactions with heavy targets, it is often desirable to remove a bulk of target materials and the other reaction products. A number of papers[1-7] have been published on radiochemical separation of the protactinium, but up to now there have been few papers[8] describing the radiochemical isolation of the protactinium from the mixture of many kinds of reaction products. Thus, we studied the separation of the protactinium from the main reaction products in bombardment of ^{232}Th with 14 MeV neutrons by using 1-phenyl-3-methyl-4-benzoyl-5-pyrazolone (PMBP) as an extractant.[9] From the γ ray spectrum of the separated protactinium fraction, we found that the protactinium could be separated from most of the main reaction products except for Zr and Nb. Subsequently, an improvement for the method was carried out. A simple, relatively fast radiochemical procedure was developed, by which the protactinium was extracted into tri-iso-octylamine(TIOA) after extraction and back-extraction of protactinium with

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PMBP as an extractant, in order to remove Zr and Nb from Pa. This paper aims to present the detailed separation method of protactinium produced in the reaction of 60 MeV/nucleon ^{18}O ions with the natural uranium.

2 EXPERIMENTAL

2.1 Heavy ion irradiation

The irradiations were performed at the Heavy Ion Research Facility (HIRFL) in the Institute of Modern Physics (IMP). The natural uranium targets were bombarded with 60 MeV/nucleon ^{18}O beams. The targets had a thickness of 1.5 g/cm^2, in the form of $(NH_4)_2U_2O_7$. The irradiation time was 10–30 min and the average current intensity was about 30–40 enA. The activities of the protactinium isotopes were produced through the exotic multi-nucleon transfer reactions. The beam intensity was monitored with a Faraday cup. After the end of irradiation, the target was rapidly transferred to a radiochemical laboratory by a pneumatic transfer system where radiochemical isolation was carried out.

2.2 Chemical separation

The irradiated uranium target was dissolved in 4.0 mol/L HCl solution. The isolation of the protactinium from the uranium and other reaction products was completed with the following chemical procedure.

The radiochemical separation procedure of Pa is illustrated in Fig.1. In the procedure the protactinium was first extracted from the target solution into 0.05 mol/L PMBP-benzene and then back-extracted into the aqueous phase by 4 mol/L HCl–0.5 mol/L HF solution. In these two steps, the protactinium could be separated from the uranium and most of the other reaction products except for Zr and Nb. Only a tiny portion of I and Po still remained in the protactinium back-extraction aqueous phase. In the following step, protactinium together with Nb was extracted into 5% TIOA - xylene from 12 mol/L HCl–0.05 mol/L HF solution, while Zr remained in the aqueous phase. Nb then remained in the organic phase after protactinium was back-extracted into 4 mol/L HCl–0.5 mol/L HF solution and finally separation of Pa from Nb was achieved.

The samples for counting were prepared by co-precipitation of the Pa fraction with ferric hydroxide. The chemical yield of the radiotracer protactinium was determined to be about 80% with the aid of a ^{233}Pa radiotracer.

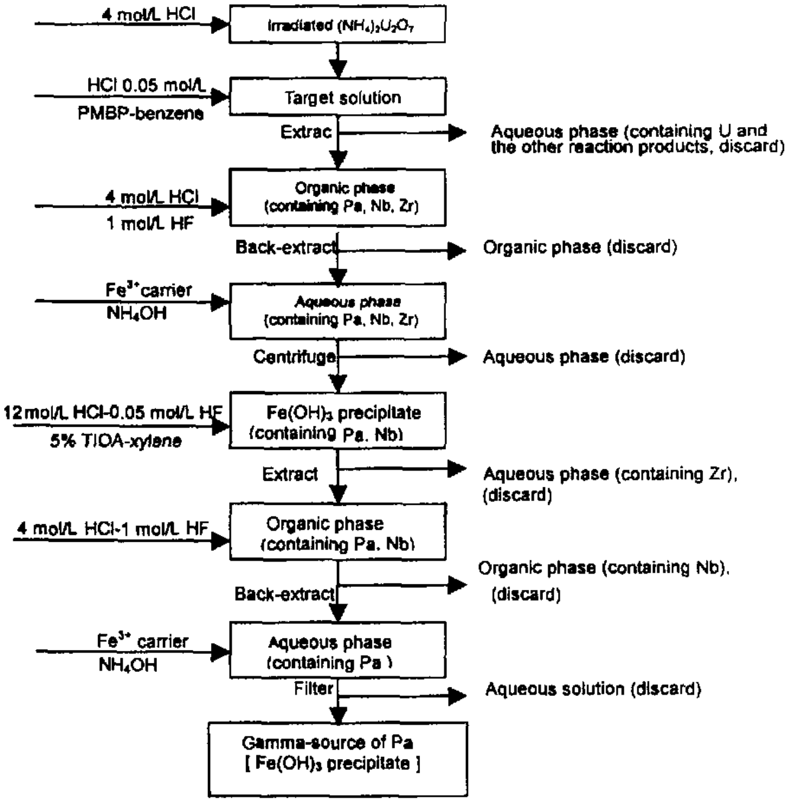

Fig.1 The chemical procedure for separation and purification of the protactinium from the natural uranium irradiated by 60 MeV/nucleon ^{18}O ions

2.3 Radioactivity measurements and data analysis

The time between the end of the irradiation and the start of counting was about 10–15 min. The separated Pa fraction was measured with a HPGe gamma ray spectrometer system in a low-background lead chamber. The HPGe detector with 30% efficiency has an energy resolution of 2.1 keV (FWHM) at the 1332 keV line of ^{60}Co. The energy calibration of the system was performed with a set of standard γ ray sources. Measurements were made in a multi-spectrum mode. The acquired data were stored on the magnetic disks with a PC-CAMAC Multi-Parameter Data Acquisition System[10], and analyzed by a γ spectrum analytical program. Radionuclides were identified according to their gamma-ray energies and relative intensities.

3 RESULTS AND DISCUSSION

The γ ray spectrum of separated protactinium fractions is shown in the Fig.2. It can be seen from Fig.2 that there are no other impurity isotopes except for protactinium isotopes in the Pa fraction.

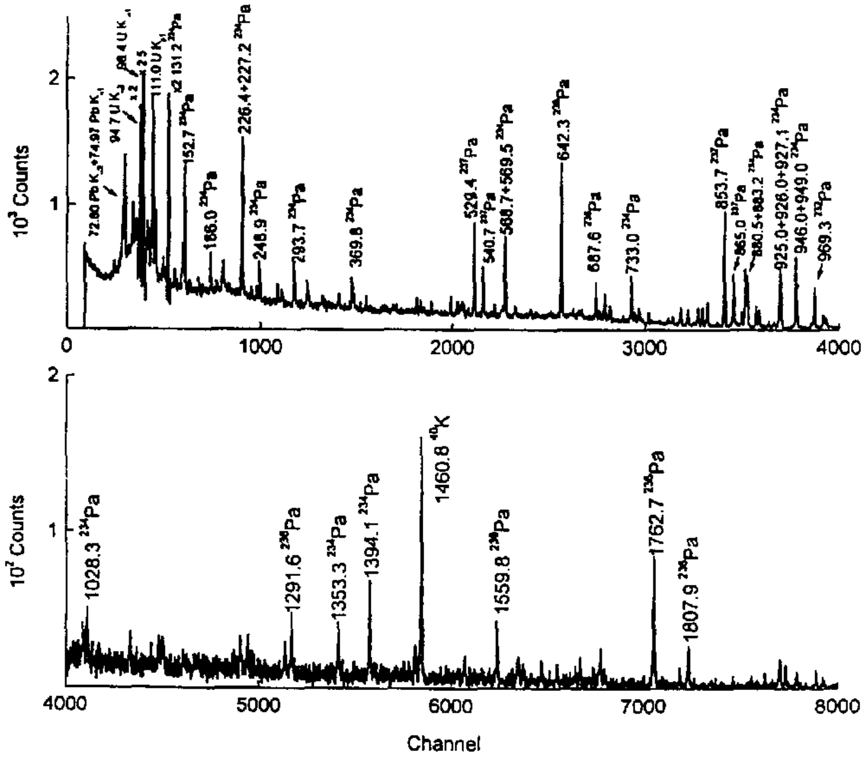

Fig.2 The measured γ ray spectrum of chemically separated Pa fraction in the reaction of 60 MeV/nucleon ^{18}O ions with the natural uranium

The decontamination factors obtained by the experimental measurements are listed in Table 1 for the uranium and the main reaction products. The energies and branching ratios of γ rays for each isotopes listed in the Table 1 were taken from references.[11] The majority of the decontamination factors for the impurity elements are larger than 10^4, indicating that the resulting decontamination for most impurity elements, especially uranium, niobium and zirconium, is satisfactory. The chemical procedure described above is very useful for identification of any unknown neutron-rich isotopes of the protactinium produced by the multi-nucleon transfer reactions in the bombardment of natural uranium with 60 MeV/nucleon ^{18}O beams.[12]

Table 1 Decontamination factors of the main impurity elements in the protactinium fractions separated from the natural uranium irradiated by 60 MeV/nucleon ^{18}O ions

Isotopes	Energy (keV)	Branching ratio (%)	Decontamination factors	Isotopes	Energy (keV)	Branching ratio (%)	Decontamination factors
^{24}Na	1368.5	100	$> 3.1 \times 10^5$	^{111m}Pd	172.2	33.5	$> 6.5 \times 10^5$
^{48}Sc-^{48}V	983.5	100	$> 4.3 \times 10^2$	^{112}Ag	617.4	42.5	$> 8.6 \times 10^5$
^{59}Fe	1099.3	56.5	$> 3.9 \times 10^3$	^{115g}Cd	336.2	49.7	$> 4.2 \times 10^4$
^{65}Ni	1481.8	23.5	$> 5.3 \times 10^3$	^{117g}In	158.6	86.4	$> 4.4 \times 10^4$
^{69}Zn	438.6	94.8	$> 2.8 \times 10^4$	^{118m}Sb	253.7	94.2	$> 6.7 \times 10^5$
^{73}Ga	297.3	80	$> 3.0 \times 10^5$	^{127}Cs	411.9	58.4	$> 5.8 \times 10^4$
^{82g}Br	1474.8	16.6	$> 6.2 \times 10^3$	^{131}I	364.5	81.2	$> 3.1 \times 10^5$
^{84}Rb	881.6	67.8	$> 5.6 \times 10^4$	^{132}Te	228.2	88.1	$> 1.4 \times 10^5$
^{90m}Y	202.5	96.5	$> 3.2 \times 10^5$	^{135g}Xe	249.8	90.4	$> 7.8 \times 10^4$
^{91}Sr	1024.3	33.4	$> 4.6 \times 10^4$	^{140}Ba	1596.5	109.9	$> 3.6 \times 10^4$
^{95}Zr	756.8	55.4	$> 5.5 \times 10^4$	^{141}Ce	145.4	48.4	$> 4.1 \times 10^5$
^{96}Nb	778.2	96.9	$> 3.9 \times 10^4$	^{234}Th	92.8	2.67	$> 1.0 \times 10^5$
^{99}Mo-^{99}Tc	140.5	90.7	$> 7.4 \times 10^5$	^{237}U	208.0	21.7	$> 2.0 \times 10^5$
^{105}Ru	724.2	46.7	$> 6.8 \times 10^4$	^{239}Np	277.6	14.5	$> 6.3 \times 10^4$

The trace protactinium could be quantitatively carried by the $Fe(OH)_3$ precipitate and the extraction efficiency for Pa in each step of the procedure was also high enough. It is reasonable to get a higher chemical yield of Pa by using the procedure.

Table 2 shows the variation of the extraction yields of Pa with the shaking time using PMBP as extractant. The back-extraction yields of Pa from TIOA as a function of the shaking time are given in Table 3. Table 2 and Table 3 show that the extraction or back-extraction equilibrium of Pa can be achieved within 20–30 s. Therefore, the procedure could be preformed in a short time. It was sometimes necessary to wash the resulted organic phases once or twice with 4 mol/L HCl or 12 mol/L HCl–0.05 mol/L solution after each extraction in order to further remove some of the trace impurities.

Table 2 Relation of the extraction yield of Pa and the shaking time*

T(s)	10	20	30	60	120	180	240
E(%)	98.3	98.9	99.1	99.4	99.7	100	100

*The aqueous phase is 4 mol/L HCl solution and the organic phase is 0.05 mol/L PMBP-benzene

Table 3 Relation of the back-extraction yields of Pa and the shaking time*

T(s)	10	20	40	60	120	180
E'(%)	99.2	99.3	100	99.3	99.6	100

*The organic phase is TIOA-xylene and the aqueous phase is 4 mol/L HCl–1 mol/L HF solution

In the experiment, the use of PMBP and TIOA as extractants has the distinct advantages, e.g., no third phase is formed and phase separation is very fast. This is very important for practical application in the nuclear chemical separation. It should be noted that protactinium activities were easily adsorbed on the container wall. Therefore, every attempt should be made to minimize loss of the protactinium in the experiment.

4 CONCLUSION

(1) The high pure protactinium fraction can be separated from large amounts of uranium and the complex reaction products using the present procedure.

(2) The use of PMBP and TIOA as extactants has the distinct advantages of fast phase separation with no third phase forming.

(3) The procedure is simple, relatively fast and has higher chemical yield for protactinium.

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