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Radiochemical separation of thorium from ^{18}O induced reaction with natural uranium

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Abstract A radiochemical procedure used to separate and purify trace concentration thorium produced in heavy ion reaction with uranium targets is presented. The procedure can rapidly yield thorium fraction suitable for gamma-ray spectroscopy studies. The resultant gamma-ray spectra showed that Th was isolated from a large number of elements produced in the reaction, and there were only a few contaminating activities of isotopes of Sc, Cd, In, etc. The decontamination factors for the main reaction products are given.

Keywords Radiochemical separation, Gamma-ray spectra, Decontamination factors.

CLC numbers O674.233^{+}, O571.41^{+}3

1 INTRODUCTION

In an attempt to synthesize and identify new neutron-rich isotopes, it is desirable to completely remove the target materials and all of the other reaction products. In the heavy ion induced reactions with uranium targets the radioisotopes of many elements will be produced[1], including a large amount of near target products and target fragments besides the fission products. In the meanwhile, the half-lives of new neutron-rich isotopes are expected to be very short[2]. In order to identify a new neutron-rich nuclide ^{238}Th, it is essential to find a rapid and high selective chemical separation method used for separation of thorium. In the previous similar studies[3,4], it was found that there were contaminating activities from some isotopes, especially iodine isotope, in the γ ray spectra of thorium fractions. The present paper describes an improved radiochemical procedure applicable to separation and purification of thorium and gives decontamination factors for main reaction products.

2 EXPERIMENTAL

2.1 Heavy ion irradiation

The irradiation was performed at the Heavy Ion Research Facility at Lanzhou

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(HIRFL) in Institute of Modern Physics (IMP) in China. Natural uranium targets (1.5 g/cm^2 of $(NH_4)_2U_2O_7$ powder) were irradiated by 60 MeV/u ^{18}O beams with a current intensity of 40~60 nA. The bombardments lasted about 30 minutes for each target. After the end of irradiation, targets were rapidly transferred to a chemical laboratory by a pneumatic transport system. There the radiochemical separation for irradiated targets was carried out.

2.2 Chemical separation

2.2.1 Preparation of ^{234}Th tracer

The old natural uranium was used to prepare ^{234}Th tracer. The chemical procedure for ^{234}Th preparation has been described elsewhere[3].

2.2.2 Separation procedure

The isolation of thorium from uranium and other reaction products was completed with an improved chemical procedure. The procedure is shown in Fig.1. The irradiated uranium powder was dissolved into 1.8 mol/L HNO_3. The PMBP extraction provides the selective separation of thorium from the other elements. After washing with an equal

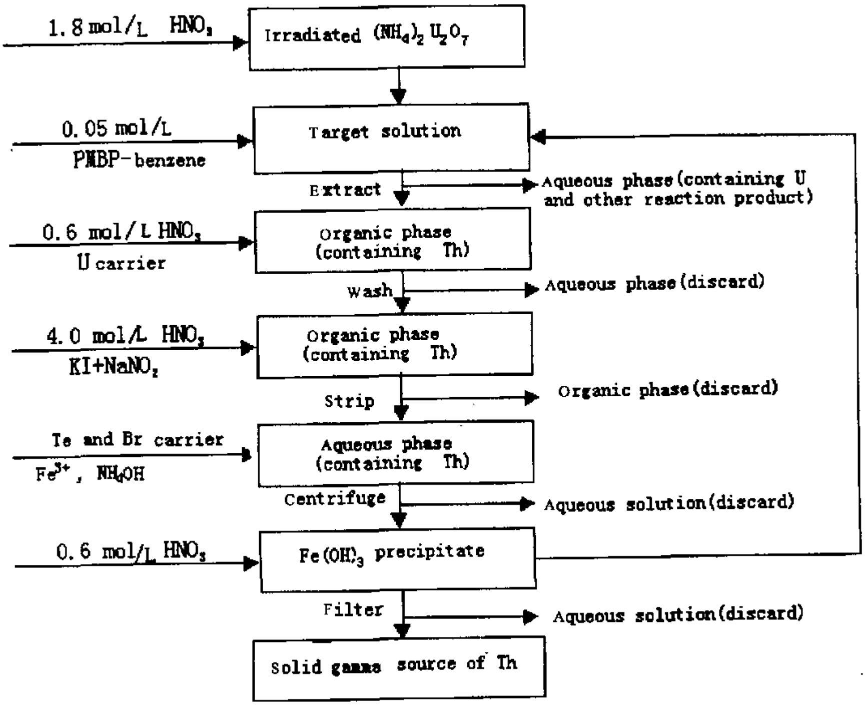

Fig.1 Chemical procedure for separation of thorium from the irradiated natural uranium targets

volume of 0.6 mol/L HNO_3, the organic phase was shaken with 4 mol/L HNO_3 for 1 minute to strip thorium into the aqueous phase. The extraction and back-extraction of Th were repeated once again. The trace thorium was carried quantitatively by the precipitate of $Fe(OH)_3$, here $Fe(OH)_3$ was filtered onto the filter paper. Thorium solid sources were prepared for γ counting. It took about 10 min to complete the procedure described above. By applying ^{234}Th tracer as a yield monitor, the yield of thorium is determined to be about 70%.

Thorium was extracted and stripped only one time in a preceding procedure[3]. However, in the improved chemical procedure the extraction and back-extraction for thorium was carried out two times to remove impurities further. In order to remove the impurities of radioactive Br and I isotopes, we employed a simple method i.e. adding Br-carrier in the step of $Fe(OH)_3$ precipitate and oxidizing I^- into I^0 by $NaNO_2$ during the stripping of thorium. The improved procedure was used in the experiment for identification of ^{238}Th.

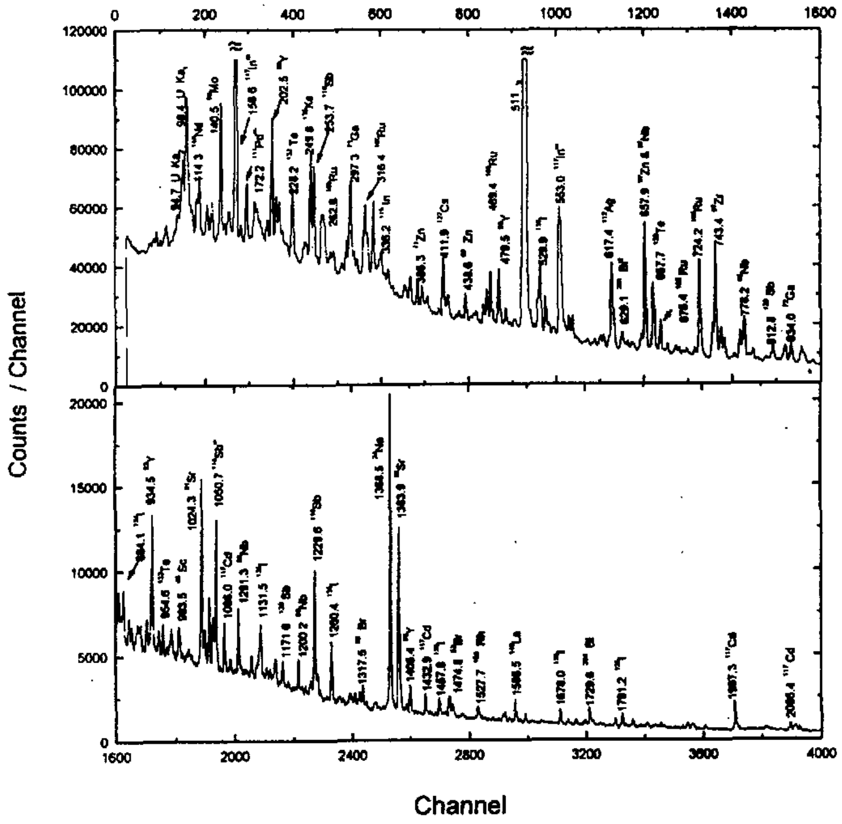

Fig.2 Measured γ ray spectrum of the uranium target sample

2.3 measurement

About 10 minutes after the end of irradiation, measurements of single γ ray spectra started using a calibrated GMX HPGe detector. The chemically separated thorium fractions and another irradiated uranium target sample were measured, respectively. The HPGe detector has 30% efficiency and an energy resolution of 2.0 keV for the 1332 keV line of ^{60}Co. The γ ray spectra were recorded in a multi-spectrum mode in order to obtain half-lives for the interesting nuclides. The time sequence spectra were recorded on magnetic disks by a PC-CAMAC Multi-Parameter Data Acquisition System[5]. The measurements lasted 30 min.

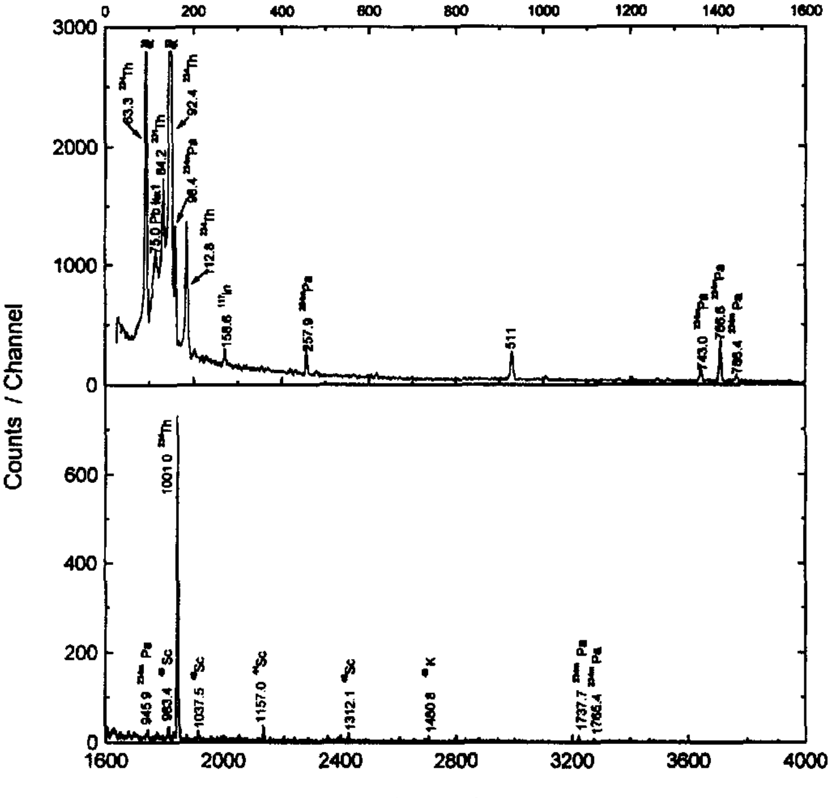

Fig.3 Measured γ ray spectrum of the Th fraction separated from the uranium target

3 RESULTS AND DISCUSSION

Fig.2 and Fig.3 show the single γ ray spectra for the unseparated uranium target sample and for the chemically separated thorium fraction, respectively. Fig.2 shows that there are many γ rays from kinds of reaction products and the intensities for some of γ rays are very strong. By comparing Fig.2 with Fig.3, it is seen that the procedure using decontamination of the impurity elements for the thorium fraction is satisfactory. In the ^{18}O induced reactions of heavy elements Th or U, the fission products are always prominent. A large amount of near target products and the target fragments are also mixed up with the reaction products. Therefore thorium in trace amounts must be isolated from a great amount of uranium and a mixture of reaction products. The separation of Th from Zr, Nb, Mo, Sb, Te and rare earth, etc. is very important as Th are often associated with them in the reaction products. It was found that the thorium can be isolated from most of such elements using the procedure described above. Fig.4 shows clearly that uranium does not interfere with the extraction of trace thorium under the experimental conditions. It should be pointed out that the activities of I and Br isotopes have been completely removed in the thorium fractions obtained by employing the improved procedure. In the previous studies[3,4], iodine activities in the thorium fractions could not always be removed completely by the similar chemical procedures. However, a few amount of impurity for isotopes of Sc, Cd, In, etc. still retained in the thorium fractions in this study. It seems that the procedure needs further improvement so that very pure thorium fractions would be obtained. Table 1 gives the decontamination factors for the main reaction products, in which the decontamination factor is defined as the ratio of the radioactive intensity for some isotope in the unseparated target sample to the radioactive intensity for the same isotope in the separated source.

Table 1 The decontamination factors of the impurity elements in the Th fractions separated from uranium targets irradiated by 60 MeV/u ^{18}O ions

Nuclides	Energy /keV	Branching ratio /%	Decontamination factors	Nuclides	Energy /keV	Ratio /%	Decontamination factors
^{7}Be	477.6	10.4	$> 6.3 \times 10^4$	^{111m}Pd	172.2	33.5	$> 1.6 \times 10^5$
^{24}Na	1368.5	100	$> 1.1 \times 10^5$	^{112}Ag	617.4	42.5	$> 4.8 \times 10^5$
^{48}S-^{48}V	983.5	100	2.1×10^2	^{115}Cd	336.2	49.7	$> 1.1 \times 10^4$
^{56}Mn	1810.7	27.2	2.2×10^2	^{117}In	158.6	86.4	1.5×10^4
^{59}Fe	1099.3	56.5	$> 7.7 \times 10^3$	^{118}Sb	253.7	94.2	$> 1.2 \times 10^5$
^{65}Ni	1481.8	23.5	$> 2.8 \times 10^3$	^{127}Cs	124.7	15.6	$> 3.9 \times 10^4$
^{69}Zn	438.6	94.8	$> 3.7 \times 10^4$	^{131}I	364.5	81.2	$> 1.8 \times 10^5$
^{73}Ga	297.3	80	$> 1.1 \times 10^5$	^{132}Te	228.2	88.1	$> 2.4 \times 10^5$
^{82}Br	1474.8	16.6	$> 4.5 \times 10^3$	^{133}I	529.9	87.0	$> 8.9 \times 10^4$
^{84}Rb	881.6	67.8	3.4×10^4	^{135}Xe	249.8	90.4	6.9×10^3
^{95}Zr	756.8	55.4	$> 6.8 \times 10^4$	^{140}Ba	1596.5	109.9	$> 4.5 \times 10^4$
^{90}Y	202.5	96.5	$> 1.5 \times 10^5$	^{141}Ce	145.4	48.4	$> 2.2 \times 10^5$
^{91}Sr	1024.3	33.4	$> 4.4 \times 10^4$	^{233}Pa	311.9	38.6	$> 1.6 \times 10^4$
^{96}Nb	778.2	96.9	$> 5.7 \times 10^4$	^{237}U	208.0	21.7	$> 4.0 \times 10^5$
^{99}Mo-^{99}Tc	140.5	90.7	$> 6.4 \times 10^5$	^{239}Np	277.6	14.5	$> 5.0 \times 10^4$
^{105}Ru	724.2	46.7	3.4×10^4				

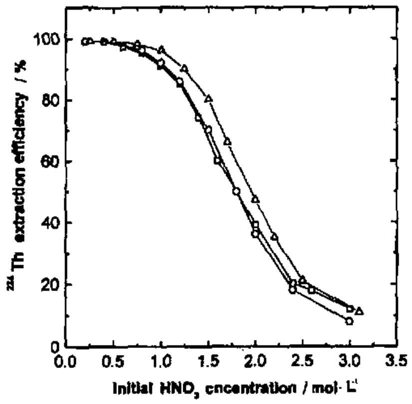

Fig.4 Effect of UO_2^{2+} and $U_2O_7^{2-}$ ions on the percentage extraction of ^{234}Th
□ 1 g $(NH_4)_2U_2O_7$, ○ 1 g $UO_2(NO_3)_2$,
△ No $UO_2(NO_3)_2$ and $(NH_4)_2U_2O_7$

4 CONCLUSION

(1) The solvent extraction of thorium by PMBP in benzene provides a method of rapid and selective separation of thorium from nitric acid aqueous solution.

(2) A large amount of uranium does not interfere with thorium extraction by PMBP in benzene from nitric acid aqueous solution.

(3) Activities of iodine and bromine isotopes can be removed completely in the thorium fractions by using the present procedure.

(4) The single γ-ray spectra of the thorium fraction shows there is a little contamination from the isotopes of Sc, In, etc. So the procedure needs further improvement in order to obtain purer thorium fraction.

(5) The chemical procedure used in the experiment is simple, rapid, highly selective and applicable to separation of trace thorium.

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