1.28 Fission ^{99}Mo Production technology

Miao Zengxing, Luo Zhifu, Ma Huimin
Liang Yufu, Yu Ningwen

Department of Isotope, China Institute of Atomic Energy
P.O. box 275-103
Beijing, China

Abstract:

This paper describes a production technology of fission ^{99}Mo in the Department Isotope, CIAE. The irradiation target is tubular U-Al alloy containing highly enriched uranium. The target is irradiated in the swimming pool reactor core. The neutron flux is about 4×10^{13}/cm^2.see. The production scale is 3.7-7.4 TBq (100-200Ci) of fission ^{99}Mo per batch. Total recovery of ^{99}Mo is more than 70%. The production practice proves that the process and equipment are safe and reliable.

I . Introduction

^{99}Tc^{m} is the most widely used radioisotope in nuclear medical diagnosis. In order to meet the ever increasing demand for ^{99}Tc^{m} generator of high specific activity, the Department of Isotopes, CIAE, began to execute a research project on fission ^{99}Mo production technology using 90% highly enriched uranium (HEU) target in 1990.

The primary objective of the project is to set up a production unit in CIAE, which can produce 3.7-7.4TBq (100 to 200Ci) of fission ^{99}Mo of high purity per batch.

The production of high purity fission ^{99}Mo for medical use is a very sophisticated technique, including the fabrication of HEU target; establishment of irradiation condition, chemical processing and production equipment for separation of ^{99}Mo; radioactive waste disposal of gases, liquids and solids; recycling of HEU target material etc.

Since the first chemical process for separating fission ^{99}Mo had been described by the Brookhaven laboratory[1], the production technology of fission ^{99}Mo for medical use are developed greatly in several countries[2,3,4,5].

In most of the published procedures for large scale production of fission ^{99}Mo, highly enriched uranium was adopted as irradiation target element. The advantages of this target are the smaller processing unit and solution volume as well as the very little plutonium produced.

The irradiation target can be fabricated in the forms of U-Al alloy、UO_2 films or dispersions. The dissolution of the irradiated target can be carried out in alkaline or acidic solution. Chemical separation process of high purity fission ^{99}Mo include several different processing steps. Alumina column chromatography or solvent extraction are the most commonly used method for the extraction of ^{99}Mo from fission products.

This paper describes briefly the outline of the fission ^{99}Mo production technology in CIAE.

It is necessary to point out that over yeas the ANL and the radioisotope production center of BATAN get a great progress in developing ^{99}Mo production technology using a LEU target. We are interested in this project.

II. Production technology

1. irradiation target

The irradiation target used to produce fission ^{99}Mo is a tubular U-Al alloy. The specifications of the target are: highly enriched uranium wrapped in aluminum 0.4mm thick, internal diameter of the target element 16.4mm, external diameter 19.0mm, Al-U weight ration of about 12:1. The target length 180mm(containing ^{235}U 3g) or 270mm(containing ^{235}U 5g).

2. Irradiation conditions:

The irradiation target contained ^{235}U 3g or 5g is placed in the swimming-pool reactor core. The specific power of the reactor is 3.5MW. The thermal neutron flux is 4×10^{13}n/cm^2 • sec. The irradiation time is 4-7 days. The target is cooled directly by the reactor recycling water during irradiation.

3. Chemical process

The flow scheme of fission ^{99}Mo production technology is presented in Fig 1. The main steps of the procedure are described as following:

After 24 hour cooling, the irradiated target is dissolved in a closed stainless steel container with the alkali solution. The released fission rare gases ^{133}Xe、^{135}Xe、^{85}K are collected in the 13X molecular sieve column cooled by liquid N_2.

The solution after cooling is filtered through a 0.4 μ micropore film or a sintered glass filter. The insoluble residue containing uranium and most of the hydroxides of fission products are remained as a black filter cake.

The filtrate is acidified with concentrated nitric acid in a stainless steel container

connected to a closed de-iodine system. Heating the solution, released iodine carried with N_2 is absorbed in sodium hydroxide solution. A small quantity of unabsorbed radioiodine can be cleaned up by two stages of activated charcoal column. The tail gases are collected in pre-evacuated stainless steel tank. After two weeks, these gases are discharged through the ventilation center with activated charcoal filter. The operations for dissolution and acidification are carried out under de-pressure condition to avoid the escape of radioxenon and radioiodine.

The separation of ^{99}Mo from fission products is carried out with an alumina chromatographic column. But the concentration of nitric acid and aluminum ion in the raw solution should be controlled at lower than 1 mol.

The purification of ^{99}Mo is carried out by the steps of anion exchange and activated charcoal chromatography or anion exchange and evaporation method.

III. Tracer study

The tracer studies are carried out in the simulated condition with 1 mol HNO_3 and 1 mol Al $(NO_3)_3$ solution. The experimental results about the separation and purification of ^{99}Mo are obtained as following:

1. Al_2O_3 chromatographic column

The activity of 99% ^{99}Mo can be retained on the Al_2O_3 column, if the flow rate through the Al_2O_3 column is controlled better. The elution yield of ^{99}Mo is about 80-85%. The decontamination factor for ^{131}I is more than 10^2 in the tracer test conditions.

2. Anion exchange:

The ^{99}Mo is adsorbed quantitatively on the anion exchanger. The elution yield of ^{99}Mo is over 98%, and the decontamination factor for ^{131}I is about 10^2. By additional elution of malonic acid, the decontamination factor for ^{131}I can be enhanced to 10^3.

3. Activated charcoal column:

The adsorption of ^{99}Mo on the activated charcoal column is quantitative, but the elution yield is a little less (about 80-85%).

The decontamination factor is 5-10 for ^{131}I.

4. Evaporation

After the ^{99}Mo is eluted with 3 mol. HNO_3 from anion exchange column, the HNO_3 solution evaporated to dryness in quartz Erlenmeyer flask and the dry residue heated at 400℃ a few minute. The recovery of ^{99}Mo is more than 95% and the de-contamination is much better for ^{131}I.

IV. Production performance

The production equipment of this process are installed in 3 hot cells. They are major consist of several stainless steel containers and some grass ware for separation

and purification of ^{99}Mo.

Routine production had been carried out for more than eight years. The whole processing time is about 15 hours. The total yield of ^{99}Mo is more than 70%. Some nuclear purity data of product ^{99}Mo are listed in the tables (1-2).

Conclusion

The production technology developed at CIAC have been used to produce fission ^{99}Mo since 1993. At present time, it is operated two time every month. The quality of product ^{99}Mo is accord with the demands for medical use. It is testified in the production practice of eight years, this process and equipment are safe and reliable.

References

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Table 1. nuclear purity of product ^{99}Mo

Impurity/^{99}Mo radio activity

No / Nuclide	1	2	3	4	5
^{95}Zr	5×10^{-7}	6.7×10^{-7}	1.4×10^{-7}	1.0×10^{-8}	1.8×10^{-7}
^{95}Nb	3.4×10^{-7}	4.1×10^{-7}	7.1×10^{-9}	3.2×10^{-9}	1.3×10^{-7}
^{103}Ru	2.4×10^{-7}	7.4×10^{-7}	1.2×10^{-7}	1.1×10^{-7}	1.5×10^{-6}
^{125}Sb	$<5.9\times10^{-7}$	$<9.1\times10^{-7}$	$<7.1\times10^{-10}$	4.3×10^{-7}	ND
^{131}I	$<3.0\times10^{-5}$	$<7.3\times10^{-5}$	$<1.3\times10^{-5}$	$<1.2\times10^{-5}$	$<1.6\times10^{-5}$
^{132}Te	$<4.2\times10^{-7}$	$<1.0\times10^{-6}$	$<6.5\times10^{-6}$	$<1.1\times10^{-5}$	$<1.9\times10^{-5}$
^{137}Cs	5.0×10^{-8}	7.1×10^{-8}	$<1.2\times10^{-9}$	$<6.2\times10^{-10}$	$<1.3\times10^{-7}$
^{140}Ba	$<6.1\times10^{-6}$	$<9.1\times10^{-6}$	ND	ND	ND
^{141}Ce	$<2.4\times10^{-7}$	$<9.5\times10^{-7}$	ND	ND	ND

Table 2. α 、 β impurity in product ^{99}Mo

Run No.	1	2	3	4	5	6
α (ratio to ^{99}Mo)	4×10^{-11}	8×10^{-12}	5×10^{-11}	1×10^{-11}	2×10^{-11}	3×10^{-11}
β (^{99}Sr÷^{90}Sr) ratio to ^{99}Mo	6×10^{-8}	6×10^{-8}	4×10^{-9}	5×10^{-9}	2×10^{-9}	—

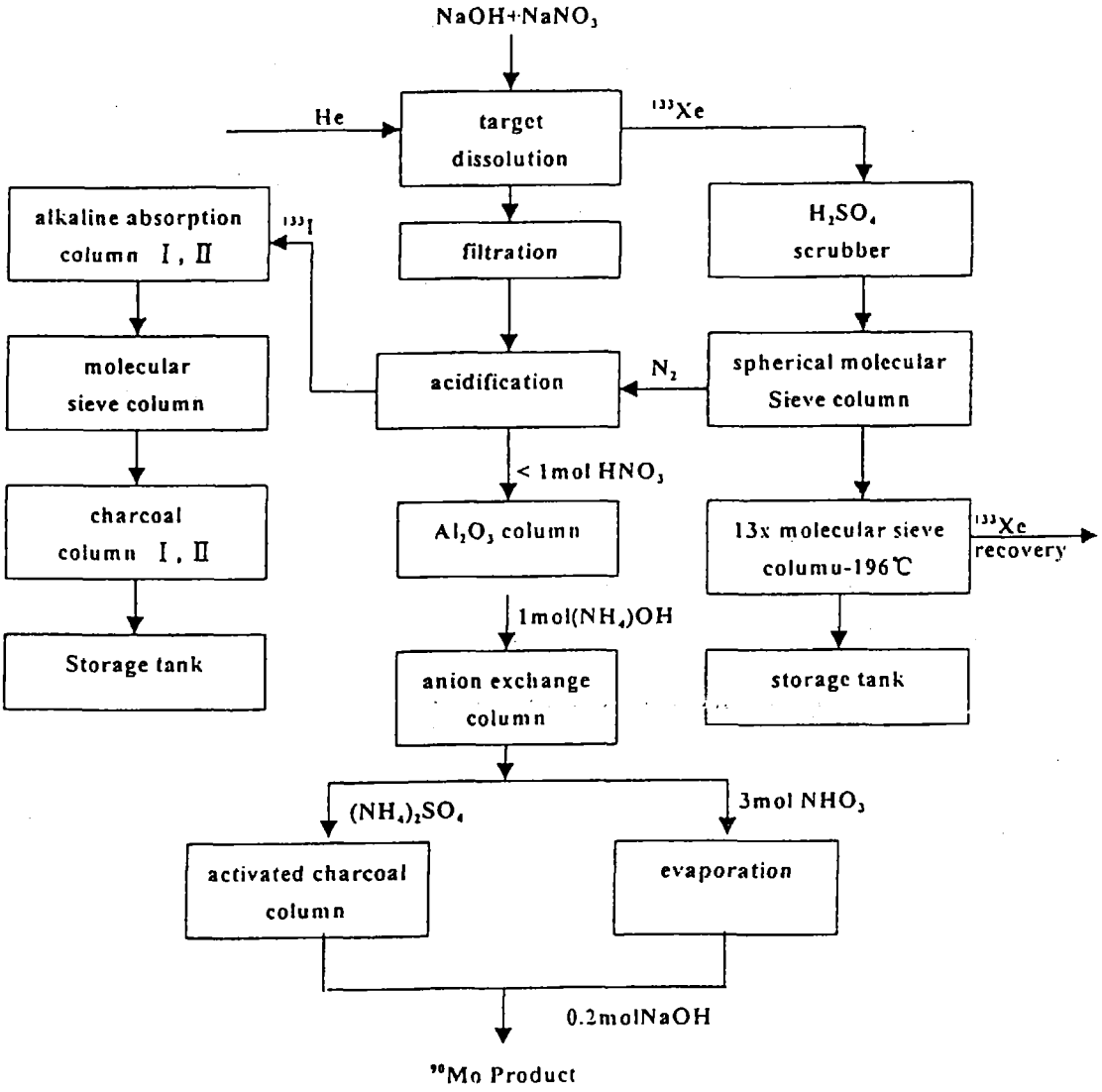

Fig. 1 ^{99}Mo Production technology