PRODUCTION OF MOLYBDENUM-99 FROM LOW ENRICHED URANIUM TARGETS

Pablo R. Cristini, Héctor J. Cols, Ricardo Bavaro, Marcelo Bronca, Roberto Centurión, Daniel Cestau

Comisión Nacional de Energía Atómica, Centro Atómico Ezeiza
República Argentina

Abstract

Fission molybdenum-99 is being produced in Argentina, at the Ezeiza Atomic Centre, since 1985. The procedure involved the irradiation of HEU targets with an Uranium-Aluminum alloy "meat" cladded with aluminum. Chemical processing of the targets is based on the method developed by Dr. Sameh Ali at KfK in Germany.
Some years ago the Atomic Energy National Commission of Argentina (CNEA) started the development of new LEU targets for its Mo-99 production, in order to replace present HEU miniplates. In a previous paper several tests were described, employing uranium silicide and uranium aluminide as target materials. Based on those tests, the method for Mo-99 production from HEU targets was adapted to LEU aluminide targets.

A description of the method, yielding results and quality control data of the final product are shown. At present, this is the method employed in Argentina to obtain high quality fission Mo-99 from LEU targets to be used in nuclear medicine.

Introduction

Technetium-99m (Tc-99m), decay product of molybdenum-99, is the radionuclide most widely employed in nuclear medicine practices. By this reason, production of fission Mo-99 has become a major concern in radioisotope production.

As it was described in several previous papers (1,3), fission Mo-99 is being produced in the CNEA of Argentina since 1985, employing highly enriched uranium/aluminium alloy targets. Chemical processing of the targets is based on the method developed by Dr. Sameh Ali at KfK in Germany (2).

During 1994, the CNEA started a development program in order to reduce the enrichment of uranium in the targets for Mo-99 production from 90% to 20%. Initial tests were performed with uranium silicide as target material, but dissolution of silicide in alkaline medium presented problems and also further purification of Mo-99 was affected (4,5). Next material tested was uranium aluminide. At first, the behavior of this compound during alkaline dissolution was similar to the silicide one. The study of U/Al phase diagram, led to changes in the target production method, yielding a compound suitable for alkaline dissolution (6,7).

Molybdenum-99 production facility

The building is located at Ezeiza Atomic Center, neighbor to RA-3 reactor. Production facility has two groups of four hot cells each. The first group, were dissolution of targets is performed, has a shielding of 30 cm of lead and the other group, employed in the purification stages, has 20 cm of lead walls.

All the cells have α β γ tight stainless steel boxes with epoxi cover

Uranium aluminide targets

Targets were described in previous papers, as well as the tests that led to final dissolving conditions (6,7).

Targets are irradiated in the RA-3 reactor core during 108-120 hours with a neutron flux of $1x10^{14}$ n/cm^2.seg and ten hours of cooling in reactor pool.

Transportation from RA-3 is accomplished through an internal corridor, with a motorized shielding of 23 cm of lead with a capacity for carrying up to four targets.

Dissolution of targets

Irradiated plates are dissolved in hot alkaline solution. Dissolving solution is sodium hydroxide with a concentration around 3.2 M, depending on the number of targets to be dissolved. Hydrogen and fission gases produced are kept in evacuated charcoal filled tanks and, after four weeks, they are released to the atmosphere.

Dissolution is completed in about 1.5 hour and after cooling, solution is filtered through a sintered stainless steel plate, leaving non-fissioned uranium in the precipitate. Mo-99 is present in the filtrate as molybdate anion. Precipitate is washed with fresh sodium hydroxide solution in order to recover remaining Mo-99.

Purification of molybdenum

As first purification stage, alkaline molybdate solution is fed through a column of strong anion exchange resin where molybdenum is retained together with iodine. After washing of the column with dilute sodium hydroxide solution, molybdenum is stripped with an alkaline sodium sulfate solution, leaving iodine in the resin.

Resulting solution goes to a second purification stage. A complex of Mo with thiocyanate is formed in sulphuric acid medium; at this stage, the adding of sodium sulfite, a reducing reagent, is needed in order to form the complex. The solution is passed through a column of chelating resin where Mo-SCN^- complex is retained. Column is washed with H_2SO_4-KSCN solution, and Mo is stripped with NaOH 1 M.

Third purification stage repeats the formation of the complex with SCN^- in acid medium, retention of it in chelating resin and elution with NaOH solution.

The elution of the third column is then boiled for half an hour, cooled down to room temperature and pH is adjusted to 3. Under these conditions, Mo is loaded to an aluminum oxide column. Stripping of Mo from this column is performed with ammonium hydroxide 1 M, and final solution is delivered to other facility for conditioning, quality control and dispatch.

Quality control

Typical values of quality control results for Mo-99 samples produced by this method are shown in table I.

TABLE I

I-131	**$1.46 \cdot 10^{-7}$**
Ru-103	**$1.63 \cdot 10^{-7}$**
Total β	**$< 5 \cdot 10^{-6}$**
Total α	**$< 8 \cdot 10^{-10}$**
Total γ	**$< 5 \cdot 10^{-7}$**

References

1) "Operation of the installation for fission Mo-99 production in Argentina. Fission molybdenum for medical use" MARQUÉS, R. O.; CRISTINI, P. R.; FERNANDEZ, H.; MARZIALE, D. Proc. of Technical Committee Meeting (I.A.E.A.) Karlsruhe, October 13 -16, 1987, IAEA-TECDOC 515, 1989, p.23.

2) "Production techniques of fission ^{99}Mo" A. SAMEH, H. ACHE – Proc. of Technical Committee Meeting (I.A.E.A.) Karlsruhe, October 13 -16, 1987, IAEA-TECDOC – 515 (1989).

3) "Producción rutinaria de Mo-99 a partir de productos de fisión" MARQUÉS, R.; MANZINI, A.; CRISTINI, P.; MONDINO, A.; FERNANDEZ, H.; FURNARI, E.; GONZÁLEZ, N. Actas de la XIX Reunión Científica de la Asociación Argentina de Tecnología Nuclear, 1991

4) "Preliminary Investigations on the Use of Uranium Silicide Targets for Fisson Mo-99 Production" COLS, H. J.; MARQUÉS, R. O.; CRISTINI, P. R. 1994 International Meeting on Reduced Enrichment for Research and Test Reactors. Williamsburg, Virginia. U.S.A.September 18-21, 1994.

5) "Research on Behavior of the Irradiated Uranium Silicide for Mo-99 Production" H. COLS; P. CRISTINI; R. MARQUÉS (CNEA). International Meeting on Reduced Enrichment for Research and Test Reactors. Paris, France. 18-21, Sept. 1995.

6) "Mo-99 from Low Enriched Uranium" H. COLS; P. CRISTINI; A. MANZINI (CNEA) International Meeting on Reduced Enrichment for Research and Test Reactors. Las Vegas, Nevada. U.S.A.1-6, Oct. 2000.

7) "Development of targets of low enrichment for the production of ^{99}Mo for fission." KOHUT, C.; DE LA FUENTE, M.; ECHENIQUE, P.; ADELFANG, P.(CNEA) International Meeting on Reduced Enrichment for Research and Test Reactors. Las Vegas, Nevada. U.S.A.1-6, Oct. 2000.