SMALL SCALE INDIGENOUS MOLYBDENUM-99 PRODUCTION USING LEU FISSION AT CHILEAN NUCLEAR ENERGY COMMISSION

1. INTRODUCTION

This report contains the results of the activities carried out in the Chilean Nuclear Energy Commission (CCHEN) under CRP Nº 13358 “Small Scale Indigenous Molybdenum-99 Production Using LEU Fission” started in October 2005 to November 2011.

The object of the project was to develop the basic infrastructure and to establish the conditions to obtain fission molybdenum-99 (^{99}Mo) by neutron irradiation of uranium-235 (^{235}U) targets in RECH-1 reactor located in Santiago, Chile.

2. WORK DONE AND RESULTS OBTAINED

2.1. CALCULATIONS

Multiple calculations were implemented to determine activities of ^{99}Mo and other fission products, seeking operating conditions for RECH-1 reactor at 5MW power, using ORIGEN-S-SCALE-4.4a and ORIGEN-S-SCALE 5.1 packages.

The main calculations were:

- ^{99}Mo activities obtained by the irradiation of 8 gram LEU foils target in the RECH-1 reactor, during 24 to 48 hours irradiation with a thermal neutron flux from 5.0×10^{13} to 1.0×10^{14} n×cm^{-2}s^{-1}.
- ^{99}Mo activities and total fission products activities obtained by the irradiation of 13 grams LEU foil target in the RECH-1 reactor, during 48 and 72 hours irradiation with a thermal neutron flux of 5.37×10^{13} n×cm^{-2}s^{-1}, 6.6×10^{13} n×cm^{-2}s^{-1}, 7.0×10^{13} n×cm^{-2}s^{-1} and 8.06×10^{13} n×cm^{-2}s^{-1}.
- Fission products activities obtained by the irradiation of 13 grams LEU foil target in the RECH-1 reactor, during 48 hours irradiation with a thermal neutron flux of 7.0×10^{13} n×cm^{-2}s^{-1}, for different decay times.
- Activities of the nuclides produced by the irradiation of 13 grams LEU foil target in the RECH-1 reactor, during 48 hours irradiation with a thermal neutron flux of 8.0×10^{13} n×cm^{-2}s^{-1}.
- Comparison of the activities obtained using the programs ORIGEN-S-SCALE 4.4a and ORIGEN-S-SCALE 5.1 for irradiation of the 13 gram LEU foil target during 48 hours, with a thermal neutron flux of 8.0×10^{13} n×cm^{-2} s^{-1} and different decay times.
- Determination of the most important activities of nuclides produced and the activities of alpha emitters to implement the technique of alpha spectroscopy.
- Activities of activation products from the aluminum and nickel, from gaseous fission products like tritium, krypton, iodine and xenon, and from isotopes identified in IAEA-TECDOC-1051; to identify the main actinides from irradiated target remaining one year after irradiation.

We will work irradiating 13 gram LEU foil target in the RECH-1 during 48 hours with a thermal neutron flux of 8.06×10^{13} n×cm^{-2}s^{-1}, to obtain 155 Ci ^{99}Mo (EOI).

Also neutronic and thermal-hydraulic calculations were done considering a LEU (19.75% of ^{235}U) foil target of 50 mm x 100 mm and 130 µm thick, covered by both sides with a nickel foil of equal dimensions but 15 µm thick. The uranium foil with its nickel covering surrounds an aluminum tube of 152 mm long, 27.99 mm outer diameter and 26.42 mm inner diameter, which has a reduction to take the uranium and nickel foils. This set, as well, is surrounded by an aluminum tube of 28.22 mm inner diameter, 30.15 mm outer diameter and 152 mm long. The neutronic calculations were done using the WIMS-D spectral transport code and the diffusion code CITATION. The thermal-hydraulic calculations were done using COSMOSFLOWORKS code, utilizing as a graphical platform the software SOLIDWORKS to obtain a virtual model of the target.

The neutron analysis has been done for the target irradiation system formed by the element to irradiate targets, the target basket and the target itself. It is noticed that due to the width of the uranium foil (only 50 mm), approximately 43% of the perimeter of this inner tube is not covered by the uranium foil. This zone without covering will lodge air that could affect the heat transfer adversely, reason why the process of target manufacturing will address this issue. Additionally the air gap effect in heat transfer has to be studied.

Calculations to improve the estimation of the power generated by the LEU foil during the 48 hours irradiation at thermal neutron flux of 8.0×10^{13} n×cm^{-2}s^{-}1 and 3-D neutron analysis for a 13 gram LEU foil irradiation in D5 position of the RECH-1 core grid, for the Nº 62 core configuration were done.

To transfer irradiated targets from the RECH-1 reactor to the hot cell for processing, lead shielding thickness and dose calculation were done for the irradiated target transport cask. Dose calculations were done for the lead shielded cask to be used for transport of 100 Ci of fission ^{99}Mo from the high activity hot cell to the ^{99}Mo-^{99m}Tc generators production cell.

Finally, the activities remaining inside the hot cell after four months of continuous ^{99}Mo production, the shielding thickness of the room for the temporary storing of the 3 month decayed liquid and solid wastes from the Modified Cintichem process in the hot cell, and the room with the filter batteries and exhaust fans for the hot cell ventilation system were calculated.

2.2. TARGET FABRICATION

Design and construction of tools and machines used for the LEU target preparation were made, and include:

- the draw die assembly (the expansion tool)
- the welding assembly (machine for rotation of targets in welding process)
- the diameter sizing assembly (the plate-roller for target diameter sizing)
- two cutting machines for irradiated targets: a remote controlled machine and a manual machine that will be operated through manipulators.

The parameters for target assembly were settled down with prototypes using copper foil instead of uranium foil. The tests were done using foils of 130 µm thick, with dimensions of 44 mm x 76mm; 50 mm x 100 mm and 76 mm x 88 mm for simulation of different LEU targets of 8, 13 and 16 grams, respectively.

Targets were assembled, first using stainless steel foils, then Cu foils, and later using Korean NU foils and LEU foils following all assembly procedures and quality controls. Targets were made including nickel foils and Al 3003 tubes.

Quality control of target gas tightness were implemented: first leak test putting the target in liquid N and then in boiling water for gross leaks; then helium leak test for analyzing small leaks. Also metallographic tests were done.

Between 2009 and 2011 were assembled two targets with Cu foils, five with NU foils and five with LEU foils.

Other activities:

- Experiences on hot rolling of uranium encapsulated in low content carbon steel, obtaining uranium foils with thicknesses between 130 and 150 µm.
- Measurement of air gaps in targets.
- Design and construction of a Technyl target holder to protect the target during transport and manipulation.
- Construction of target irradiation device.
- Cutting tests of targets were already completed.
- Cold lamination of the Korean LEU foil to homogenize its heterogeneity, arriving from 70 µm to 22 µm.
- Experiences on thin nickel film electrolytic deposit over copper and uranium foils.
- Construction of definitive irradiation device.

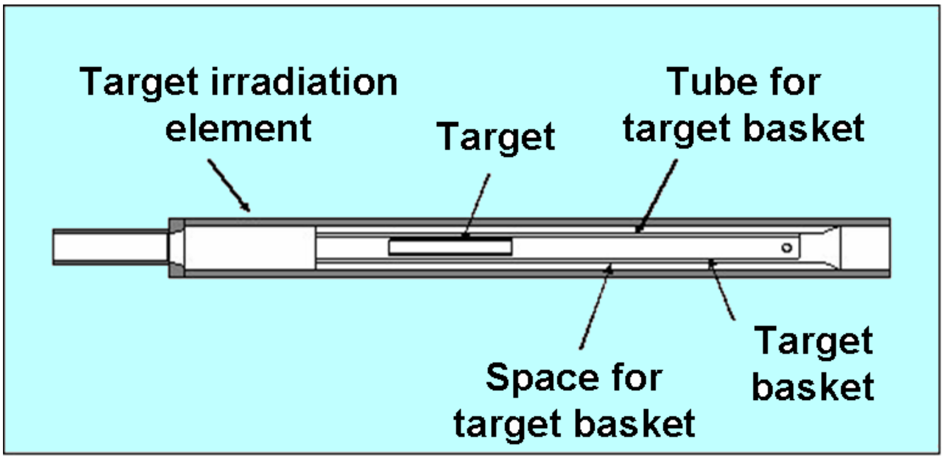

FIG. 1. Target irradiation system.

2.3. CONDITIONING HIGH ACTIVITY HOT CELL

To separate ^{99}Mo it will use a high activity hot cell placed in the reactor building, previously used for the inspection and burn up measurements of fuel elements, so it needed to be decommissioned before commissioning it to its new use.

Adapting the hot cell included design and construction of the new stainless steel lining, installation of new seals and pneumatic system to activate the rear doors, re-design and installation of the electric systems including lighting, fitting of an air injection duct for the ventilation system inside the hot cell and design of the layout of equipments inside the hot cell.

Two news Hans Wälischmiller A100GL master slave manipulators and support elements for replacement of the two old ones were mounted. Also, mechanical belts for feeding supplies were installed.

Finally, a new hot cell ventilation system was designed to be independent from the reactor ventilation system. The construction project is on course.

2.4. MODIFIED CINTICHEM PROCESS

To implement the chemical process a rotating 316L stainless steel dissolver and a second ANL stainless steel dissolver were designed and constructed.

Also it was necessary to design and fabricate glass material of 180 ml capacity for the process. The volume was decreased to get a better handling in the hot cell.

Auxiliary stainless steel devices for the chemical process (bottles holder, columns holder, etc.) were designed and constructed too.

15 experiments were done preparing non-radioactive synthetic solutions, that is, without irradiated uranium. Synthetic solutions were obtained from uranyl nitrate equivalent to 1.8 g and 12 g of uranium shaving dissolved in 40 ml of HNO_3 1.87 M and 8.12 M, and 0.1 g and 1.9 g of nickel powder dissolved in 0.3 ml of concentrated HNO_3, respectively. Carriers were added to the mixture of the solutions: 5.0 mg of Mo (10 mg/ml), 10.0 mg of Ru (5 mg/ml), 12 mg of Rh (8 mg/ml) and 4.0 mg of NaI (1 mg/ml) that corresponds to equal amount of carriers described in the Cintichem procedure. 0.5 ml of $AgNO_3$ to 10 % in HNO_3 0.1 M and 0.3 ml of HCl 1N were added, and the process was continued following the 61 steps described in the procedure.

6 experiments were done with NU foils and mean value of process efficiency obtained was 80.1%.

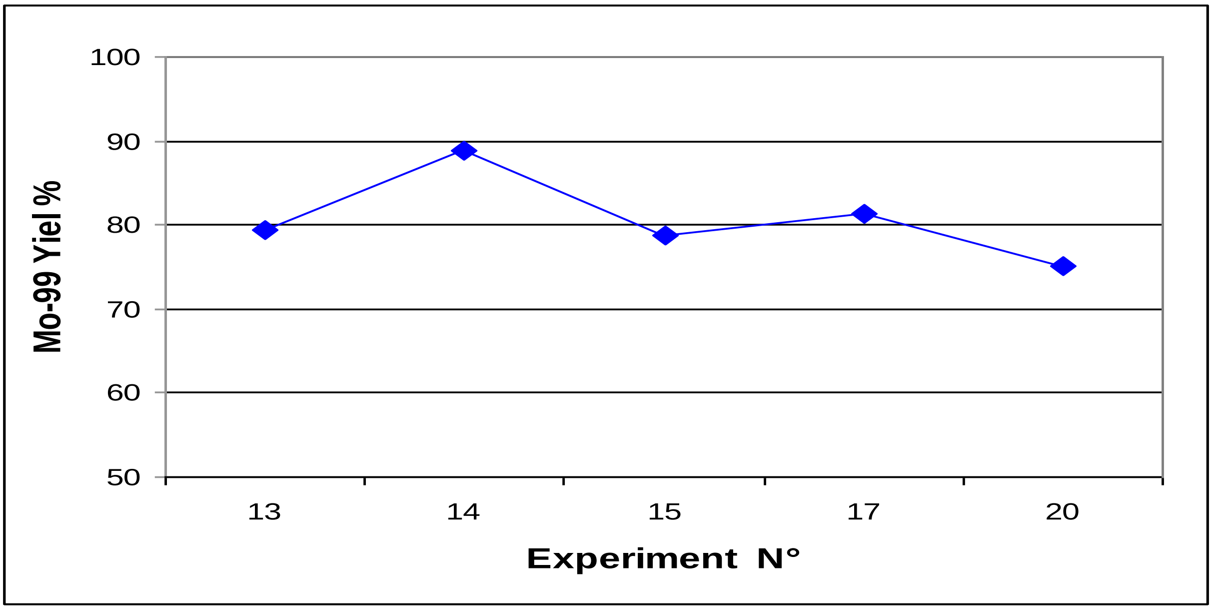

FIG. 2. Experimental yield ^{99}Mo.

2.5. QUALITY CONTROLS

To perform control tests an alpha spectrometry system was assembled including four PIPS detectors and was afterwards calibrated.

Also a control sample for alpha spectrometry was prepared by drying. The procedure resulted in a non-homogeneous deposit, which was considered to be not useful for the alpha spectrometric measurements. Then, it was developed a procedure for sample preparation by electro deposition technique in an electrolytic cell.

86% efficiency was obtained for a sample prepared with 1 Ampere of current during 60 minutes of plating, and a solution adjusted to pH 2.33.

For gamma spectrometry controls and measurement of solution from ^{99}Mo activation, a new gamma spectrometry system with HP Ge detector was purchased.

2.6. RADIOACTIVE WASTES TREATMENT

Design and construction of devices to collect both solid and liquid wastes from the production process and for extraction of wastes from high activity hot cell were done.

Based on calculations of waste activities it was designed a temporary storage room for liquid and solid wastes. This room should be constructed to allow wastes to decay for three years before treatment.

3. CONCLUSIONS

Foil targets of 50 mm x 100 mm, 130 µm thick and 13 grams of LEU (19.75% of ^{235}U) irradiated in RECH-1 by a thermal neutron flux of 8.0×10^{13} $n\times cm^{-2}s^{-1}$ during 48 hours (position D5 of the reactor grid) will produce ^{99}Mo activity of 155 Ci (EOI), useful for CCHEN´s production of Mo-Tc generators.

The irradiation of LEU foil in RECH-1 in the definitive irradiation rig with a thermal neutron flux of 8.0×10^{13} $n\times cm^{-2}s^{-1}$ (position D5 of the reactor grid) during 48 hours generates 7.440 W that is 3.720 W on each face of the uranium foil. The maximum wall temperature on the target surface will be 88.9 ºC in the inner face of it, and the wall temperature to reach ONB is 124 ºC; this is acceptable for reactor safety conditions.

The velocity of the cooling flow inside the target located in the definitive irradiation rig is 3.0 $m\times s^{-1}$, and outside the target is 1.9 $m\times s^{-1}$, both for a pressure drop of 31.159 kPa. The conclusion is that the irradiation rig will fulfill all the thermal-hydraulic requirements.

Cold rolling of the Korean foils was done to reduce heterogeneity in its thickness from an average thickness of 137.2 µm (minimum value of 95 µm to maximum value of 207 µm) to an average thickness of 117 µm.

Own manufacture of thin NU foils was done by a hot rolling process (630 ºC), obtaining foils from 130 to 150 µm thick.

Three machines to assemble the target are constructed and operative, and the methodology of target assembly is known. Targets were assembled using copper foil, and then targets were assembled using Korean NU foils and finally three targets with LEU foils.

The tightness of each target is controlled first submerging it in liquid nitrogen and then in boiling water, for the detection of greater leaks. Then the target is put under a test for small leaks detection using a helium mass spectrometer. Also metallographic tests are done.

The irradiated target cutting machine with remote control to be used in the high activity hot cell was designed, constructed and tested. Better results were obtained in the cutting of irradiated targets using the new manual machine. This is important to prevent damage in the uranium foil.

Multiple experiments were done simulating the conditions and equipment following the modified Cintichem process. In experiments with NU not irradiated shavings and foils, the mean value of molybdenum recovery was 80.7%. The last experiments were done working with NU and Ni foils and the mean value of molybdenum recovery was 80.1%, and the presence of uranium was under the detection limit of the NAA technique used for this kind of sample (< 1 ppm).

At the end of the radiochemical Modified Cintichem process, and considering the efficiency of the process of 80.1%, we would obtain 127 Ci of ^{99}Mo; with half this quantity of ^{99}Mo we would satisfy Chile's need of ^{99}Mo for the production of Mo-Tc generators.

86% efficiency was obtained in sample preparations by an electrodeposition technique for alpha spectrometry quality control. Samples were obtained with 1 A of current during 60 minutes of plating, and in a solution adjusted to pH 2.33.

The calculated remnant activity inside the hot cell after four months of continuous ^{99}Mo production will be 1.08×10^3 Ci.

The calculated remaining activity in the temporary room to be built for radioactive wastes after three years of continuous ^{99}Mo production will be of 3.85×10^2 Ci.

Nowadays, CCHEN is developing a big project to refurbish laboratory for radioisotopes production and include the ventilation system for high activity hot cell. The cell will be completed in 2013. Then we will finish the licensing management and we will irradiate the first LEU targets.