IRRADIATION TECHNOLOGIES AND THE RADIOISOTOPE PRODUCTION AT INR PITESTI

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Abstract

Over the time, Institute for Nuclear Research (RATEN ICN) Pitesti, Romania, developed activities for radioisotopes production. INR is able to supply radioactive sources for industry and performed research work for Mo^{99} production by activation and by fission from Low Enriched Uranium (LEU) targets. In the framework of an IAEA project, INR Pitesti adjusted and developed its infrastructure (hot cells, tools for chemical separation in the hot cell by remote handling and transfer cask) for small scale Mo^{99} production from LEU target. The motivations of our institute to develop technology for Mo^{99} separation using modified CINTICHEM method consist in: ability to manufacture targets using LEU metallic foils; ability to irradiate in TRIGA 14MW reactor; ability to assure handling, disassembling and chemical processing of the target, in the existing hot cell facility; ability of INR Pitesti to handle, treat and condition radioactive waste. This paper will present the existing facilities for radioisotopes irradiation, tools and some result obtained in the implementation of CINTICHEM method for molybdenum separation and available methods for determination of decontamination factor and characterization of the final solution. Types of radioactive sources for industrial use will be also presented.

Keywords: *Radioisotopes, Molybdenum-99, Cintichem method*

Introduction

Over the time, Institute for Nuclear Research (INR) Pitesti, Romania, developed activities for radioisotopes production. INR is able to supply radioactive sources for industry and performed research work for ^{99}Mo production by activation and by fission from Low Enriched Uranium (LEU) targets.

Nuclear medicine diagnostic scans can image and demonstrate the physiology and function of many body parts, including the heart, the skeleton, the thyroid and salivary glands, and the brain, supporting a broad range of medical specialities.

Nuclear medicine scans involve the administration of trace amounts of radioactive pharmaceuticals, referred to as radiopharmaceuticals, into a patient's body. Technetium-99m (Tc-99m) is used as the medical isotope in 85% of nuclear medicine diagnostic scans performed worldwide, or around 30 million scans, every year.

Once internalised by a patient, radiopharmaceuticals are physiologically distributed within the body. Tc-99m-based scans allow diagnoses of a broad range of diseases in many parts of the human body, including cancer, heart disease and neurological disorders.

Substitution of Tc-99m is difficult. No comparable substitutes are available for diagnoses of various cancers, such breast, melanoma and head/neck cancer, and for a range of diagnostics in children, in particular paediatric bone and renal scans.

Supply of Tc-99m to health care providers has been unreliable over the past decade due to unexpected shutdowns and extended maintenance periods at several of the facilities in the supply chain [1].

In this context, in the frame of an AIEA project, INR Pitesti adjusted and developed its infrastructure (hot cells, tools for chemical separation in the hot cell by remote handling and transfer cask) for small scale ^{99}Mo production from LEU target. The goal is to supply the national demand (100 6-day Ci/week) [3].

The motivations of our institute to develop technology for ^{99}Mo separation using modified CINTICHEM method consist in:

- Ability to manufacture targets using LEU metallic foils;
- Ability to irradiate in TRIGA 14MW reactor;
- Ability to assure handling, disassembling and chemical processing of the target, in the existing hot cell facility;
- Ability of INR Pitesti to handle, treat and condition radioactive waste.

The paper includes description of the experimental set-up, some results obtained in the implementation of Cintichem method for Mo separation, discussion on the results, conclusions, and acknowledgements.

Romanian TRIGA SSR 14MW irradiation potential

TRIGA SSR 14MW reactor core is shown in Fig. 1, and the main irradiation parameters are given as follows:

- The maximum thermal flux is 2.9 x 10^{14} n/cm^{2}s at 14 MW rated power in XC-1 (G7);
- Other in-core experimental channels can reach 2x10^{14} n/cm^{2}s at 14 MW;
- Several channels with Ø32 in the Beryllium reflector, thermal flux ranges from 3.0 to 8.0 x 10^{13} n/cm^{2}s at 14 MW rated power;
- Neutron spectrum, water-filled channel (ex. for XC-1):
  - Thermal / epithermal ≈ 30;
  - Fast / integral flux ≈ 0.16 (fast > 1MeV);
  - Cd ratio for Au = 2.45.

Beryllium channels can be loaded and discharged during the reactor operation.

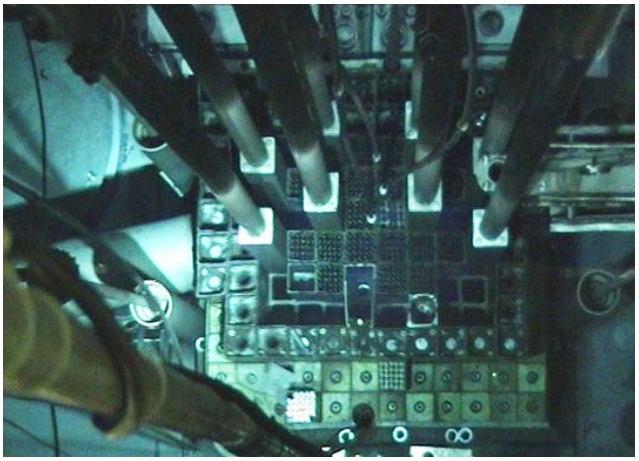


Fig. 1. TRIGA SSR 14MW reactor core

Until now, in the INR TRIGA reactor have been produced the following radioisotopes:

- Ir^{192} sealed sources for industrial defectoscopy
- Co^{60} sealed sources with our own raw material
- Mo^{99} by irradiation of MoO_3 powder targets
- I^{131} by irradiation of TeO_2 powder targets
- I^{125} by irradiation of pressurized gaseous targets of Xe
- Tl^{204} beta emitter by irradiation of solid targets of Tl.

Production of Mo^{99} using LEU targets

The molybdenum production process consist of:

- fabrication and irradiation of the target in the TRIGA SSR 14 MW reactor;
- target dissolution in nitric acid and separation of Mo^{99} using alpha benzoinoxime;
- filtering and washing of the dissolved solution, in order to drain apart the traces of uranium left after irradiation and of fission products;
- purification of Mo^{99} solution in two steps, using a silver coated charcoal column followed by a second resin column of silver coated charcoal, HZO and activated charcoal;
- waste treatment and disposal

Target manufacturing

The goal is to obtain a target geometry that can conduct to a maximum Mo^{99} activity/gU. Thus, it was chosen a target that consists of a LEU metallic foil covered with nickel foil and tight fixed between two aluminium cylinders (see Fig. 2 below).



Fig. 2. Aluminium cylinders for target manufacturing

After mounting the LEU foil between cylinders, they are welded on both ends as is presented in Fig. 3. An important step in preparing the target is the test for leaks. A helium leak test is used in order to ensure there are no leaks in the welds.



Fig. 3. Welding machine for target endcaps welding

After welding, the squeezing device is used to swell the inner cylinder and tightly fix the uranium foil between the two-cylinders (see Fig. 4).

The main characteristics of uranium foil used for target manufacturing are: dimensions 5x7 mm, thickness 140µm, mass 9.2g and U^{235} enrichment around 19.75%. This type of target with a higher density of uranium is expected to produce more Mo^{99} with fewer target irradiations and is useful for small scale production.

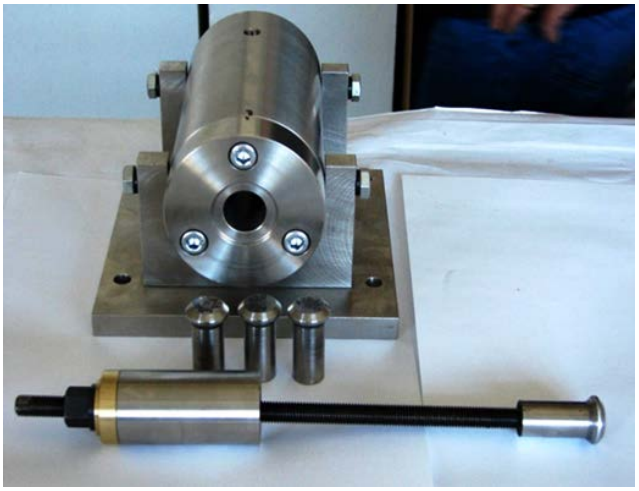


Fig. 4. Squeezing device

The design specifications for the irradiation device for LEU target irradiation were established in order to ensure the safety and efficiency of the irradiation process without modification of reactor core structure or mode of operation. Three targets are mounted on a rod and after this in the irradiation device like in Fig. 5.



Fig. 5. Irradiation device

Hot cell infrastructure and developed devices for irradiated target processing

A transfer container was equipped with an electrical motion engine to make faster and easier the transfer of irradiated target from the reactor to hot cell dedicated for target processing (Fig. 6).

For irradiated target processing, a hot cell was refurbished (Fig. 7). Thus, the window was changed with one with better protection, biological protection improved by adding an additional layer of lead and a dedicated wall penetration for fast transfer of the vial with final sodium molybdate solution to the transport container.

Also, devices for dissolution of uranium foil, precipitation and separation of Mo^{99} and purification were designed, manufactured and mounted inside of the hot cell.

Before processing, we have to cut the target end extract the uranium foil between the aluminum cylinders. For this reason, a cutting machine (Fig. 8) was manufactured so that allows extraction of uranium foil using only remote handlers. The cutting machine and some additional tools for target disassembling were designed, manufactured and tested by INR Pitesti.



Fig. 6. Transfer container



Fig. 7. Hot cell for target processing

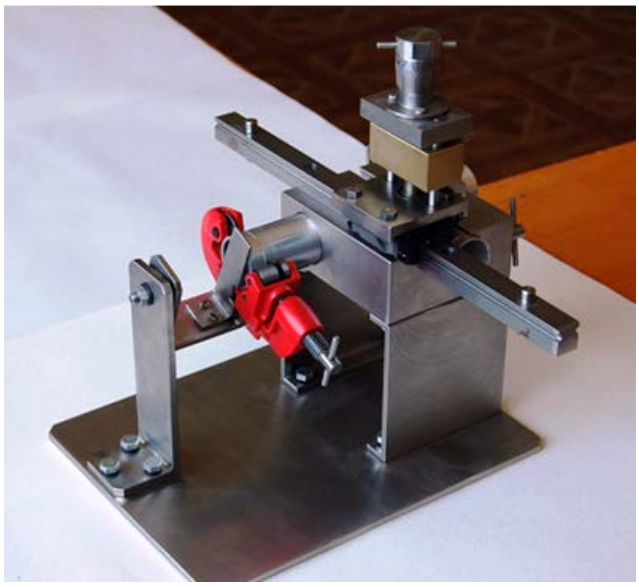


Fig. 8. Cutting machine

Chemical processing of the LEU foil

Chemical processing of the LEU foil consists of dissolution in nitric acid, precipitation, separation of molybdenum and finally purification of the sodium molybdate solution as is presented in Fig. 9.

The dissolution step is carried out using the dedicated tight closed device. For dissolution of LEU foil in nitric acid, the dissolver is heated up to 105 ^{o}C. Because the dissolution reaction takes place with gaseous emission, the dissolver is equipped with a manometer for pressure monitoring. When the reaction is initiated, the pressure rises sharply in the first 15 min and then levels off for 15 min. When the pressure starts to drop the reaction is over. The dissolution reaction lasts for 25 to 30 min.

After dissolution, the molybdenum is precipitated with α-benzoinoxime. The Mo^{99} is retained on a fritted column and the fission products remain in the liquid waste. Then the precipitate is washed and subsequently dissolved using sodium hydroxide in peroxide [4].

The last step is purification. This step is necessary in order to remove all impurities from the molybdenum solution taking into account that the solution is to be used in the pharmaceutical condition. For this purpose, two glass columns are used. The columns are filled with different resins which allow the molybdenum to pass and retain the radioactive iodine and any other impurities. More details about target processing are presented in [4 - 6].

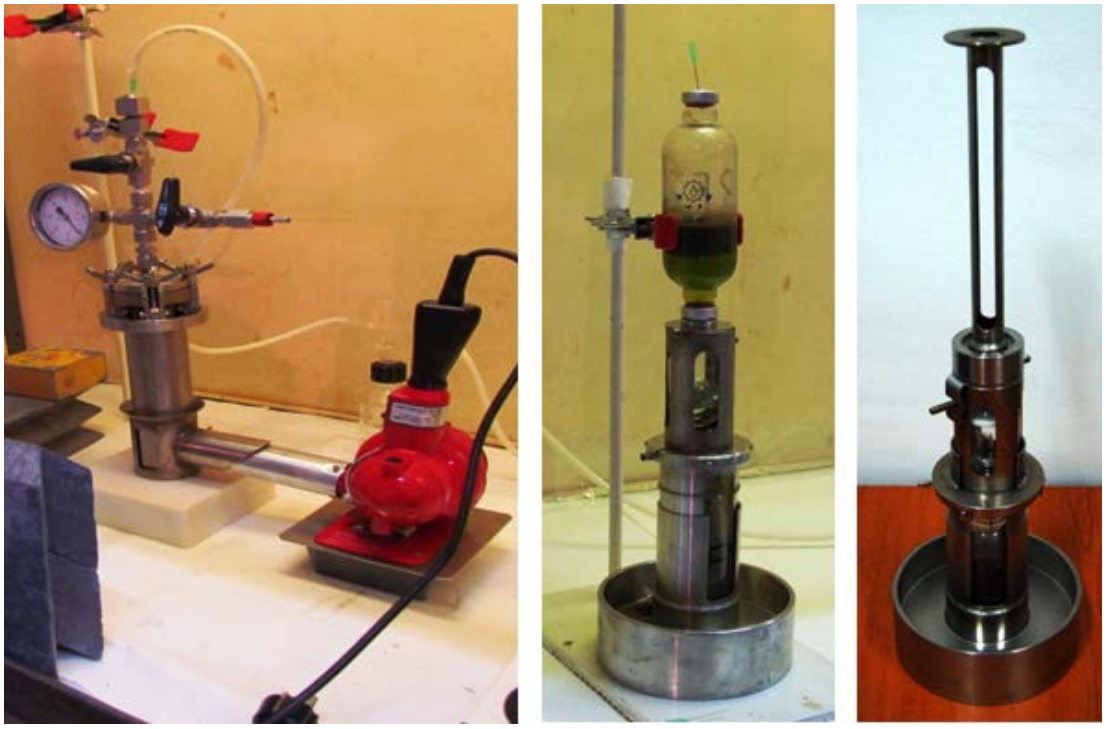


Fig. 9. Devices for LEU foil chemical processing (dissolution, precipitation and separation, purification)

Radioactive waste treatment

Without taking into consideration Uranium recovery, the estimation for average amounts of radioactive waste for a target containing 16g of LEU are as follows: maximum 200 ml high radioactive level of liquid; maximum 200 g of metallic waste; maximum 2000 g of low and medium-level solid waste (plastics, glassware, textures) and maximum 300 ml of low and medium-level liquid waste.

The treatment and elimination of gaseous waste are solved by an efficient off-gas system. Xenon and Iodine isotopes are removed from the dissolver using a cold trap. The cold trap is stored to allow decay and then eventually vented when the gases have decayed to background levels [7].

The solid waste consists of the discarded target tubes after disassembly, and glassware, columns, filters, and syringes used during processing. This material is usually all low-level waste. Volume reduction of solid waste is carried out through cutting and subsequently, they are conditioned in special containers.

Liquid waste contains enriched uranium and almost all fission and activation products from the irradiated target is high-level waste and it will be first stored for one year for decay and then recycled for uranium recovery.

Useful fission products obtained with Mo^{99}

In order to increase the process efficiency, separation of useful radionuclides like I^{131}, Xe^{133}, Sr^{90} and Cs^{137} resulted beside the Mo^{99} may also be carried out for their application in the medical field. Research work for iodine recovery was already done in our laboratory.

Mo^{99} production by neutron activation

For the indigene market, the production of Mo^{99} by the neutron activation of natural molybdenum targets is an attractive alternative to the fission of targets containing U^{235}. The main disadvantage of activation Mo^{99} production is the low specific activity of the Mo^{99} (3.7–74 GBq/g Mo) output.

However, this specific activity can be increased with the use of highly enriched Mo^{98} targets and high neutron fluxes. The advantages of this method are the generation of very low levels of radioactive waste, because the chemical processing necessary is much simpler; this method also reduces the need for hot cells. The target that is commonly used is a high purity MoO_3, either in a powder or compact form.

In order to solve the problem of low specific activity from (n, γ) produced Mo^{99}, the Tc^{99m} gel generator makes activation Mo^{99} into an insoluble zirconium molybdate Mo^{99} gel that contains about 25% of molybdenum. The zirconium molybdate gel is predominantly a cation exchanger with an open structure which allows the free diffusion of pertechnetate anion under simple elution by saline. This enables the gel concept approach to retain the advantages of ease and reliability of (alumina) column chromatography generators.

Mo^{99} production technology based on gel generator was tested at INR Pitesti with good results.

Developing and validation of methods for quality control of produced radioisotopes

The final product must be certified to meet specified purity standards, and so a quality control laboratory must be a significant part of any production operation. For this reason, INR Pitesti developed and validated radiochemical methods for quality control of produced radioisotopes and for radioactive waste characterization. A laboratory was accredited in accordance with international standard ISO/CEI 17025:2018 ″General requirements for the competence of testing and calibration laboratories″ for analysis methods like gamma spectrometry, alpha spectrometry, gross alpha-beta analysis, liquid chromatography, and chemical analysis by Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

Ir^{192} radioactive sealed sources production

Ir^{192} radioactive sealed sources constitute the active part of radiography installations with gamma radiations used in nondestructive testing of welded joints, castings and reinforced concrete.

Conclusions

Since RATEN ICN operates a TRIGA research reactor which can provide thermal neutron fluxes up to 2 x 10^{14} n/cm^2s, it is recommended to develop the technology for obtaining the Mo^{99} radioisotope by fission or by neutron activation using the method of zirconium molybdate gel for making the medical use generator Mo^{99} / Tc^{99m}.

RATEN ICN Pitesti has proved its ability to manufacture the entire equipment necessary for molybdenum production by fission – dissolver, fission gasses trap, precipitating/ dissolving and purification devices – and to fully run the process at a small scale.

The radioactive content of the Ir^{192} sealed sources are the irradiated iridium pellets. The pellets are obtained by a technological process based on pressing and sintering of iridium metallic powder, followed by electroplating with nickel for surface protection.

The pellets are irradiated in the TRIGA 14MW Steady State Reactor, using an irradiation device specially designed for this purpose. At the end of irradiation time (100 days) the pellets are transferred at hot cell facility for processing. At the hot cell facility, the iridium pellets are sorted in accordance with their radioactivity.

In order to produce an Ir^{192} radioactive sealed source, the pellets are encapsulated in a stainless steel capsule by tungsten inert gas welding as is presented in Fig. 10.

INR Pitesti produces and sells Ir^{192} radioactive sealed sources with different focal spots and activities up to 4.44 TBq (120 Ci). For sources having the focal spot smaller than 3x3 mm, the irradiated iridium pellets are inserted in aluminium disks.

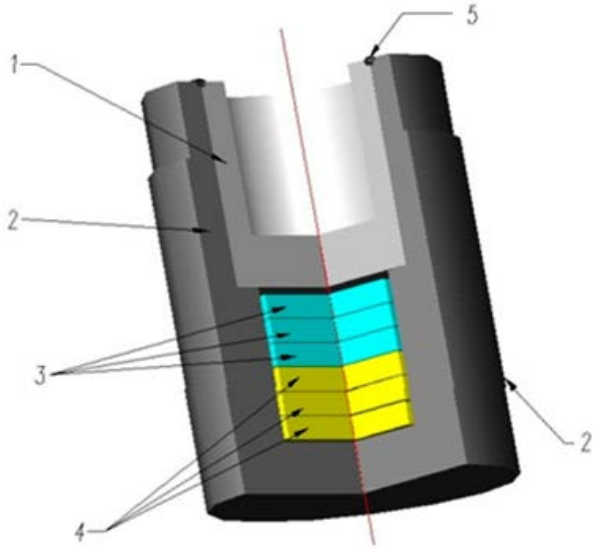


Fig. 10 Ir^{192} radioactive source
1- capsule lid; 2- capsule body; 3- filling pellets;
4- iridium pellets; 5- TIG weld

In the last time, the focus was on approaching the activation method that involves the activation of natural molybdenum targets (24.13% Mo^{98} abundance) or enriched molybdenum in the Mo^{98} isotope (> 80%).

The activation method has been approved by the RATEN ICN because it can be assimilated with the existing infrastructure and can ensure the relatively limited requirements of the indigenous market. The assimilation of the technology to obtain the activation molybdenum is also supported by the fact that the radioactive waste produced is in much smaller quantities and less active. In the work conducted in the last years, irradiation tests at laboratory scale were performed on molybdenum trioxide and the results showed good behaviour.

Studies and activities for optimizing the technological processes for obtaining and sintering Ir irradiation targets in the form of metal disks were addressed.

In order to reduce the material costs, the development of the methods for manufacturing of the targets was undertaken, for obtaining irradiation targets in different forms of metallic microstructures.

With the objective to optimize the manufacturing process for obtaining iridium targets with diameters below 2 mm, RATEN ICN Pitesti demonstrates the capability to produce iridium targets, embedded in aluminium, down to 1.75 mm diameter.

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