

DEVELOPMENT OF TECHNIQUES FOR SMALL SCALE INDIGENOUS ⁹⁹MO PRODUCTION USING LEU TARGETS AT ICN PITESTI-ROMANIA

1. INTRODUCTION

Initiation of the IAEA Coordinated Research Project (CRP) “Development Techniques for Small Scale Indigenous ⁹⁹Mo Production Using LEU Fission or Neutron Activation” during 2005 allowed Member States to participate through their research organization on contractor arrangement to accomplish the CRP objectives. Among these, the participating research organization Institute for Nuclear Research Pitesti Romania (ICN), was the beneficiary of financial support and Argonne National Laboratory assistance provided by US Department of Energy to the CRP for development of techniques for fission ⁹⁹Mo production based on LEU modified CINTICHEM process. The Agency’s role in this field was to assist in the transfer and adaptation of existing technology in order to disseminate a technique, which advances international non-proliferation objectives and promotes sustainable development needs, while also contributing to extend the production capacity for addressing supply shortages from the latest years.

The Institute for Nuclear Research, considering the existing good conditions of infrastructure of the research reactor with suitable irradiation conditions for radioisotopes, a post irradiation laboratory with direct transfer of irradiated targets from the reactor and handling of high radioactive sources, and simultaneously the existence of an expanding internal market, decided to undertake the necessary steps in order to produce fission molybdenum. The Institute intends to develop the capability to respond to the domestic needs in cooperation with the IFINN–HH from Bucharest, which is able to perform the last step consisting in the loading of fission molybdenum on chromatography generators and dispensing to the final client.

The primary scope of the project is the development of the necessary technological steps and chemical processing steps in order to be able to cover the entire process for fission molybdenum production at the required standard of purity.

Molybdenum production process [1][2] can be broken up into the following areas of activity:

- Target fabrication and irradiation of the target in a reactor;
- Target dissolution in nitric acid and separation of ⁹⁹Mo using alpha-benzoin oxime;
- Filtering and washing for decontamination of trace amount of uranium and fission products;
- Purification of ⁹⁹Mo solution through a silver coated charcoal column and once more purification through resin column of silver coated charcoal, HZO and activated charcoal, with development of associated specific measurement method and quality control/assurance;
- Transportation of sodium molybdate from the Institute to the Center for Radioisotopes Production in Bucharest for loading ⁹⁹Mo in their generators and dispensing of the product;
- Waste treatment and disposal.

2. TARGET

The accomplishment of the Institute's medium term plan for development of technology and associated infrastructure was built upon essential information and data received in the framework of activities performed during the CRP as technical visits, workshops and technical information provided by IAEA and ANL experts.

The activities deployed by the institute during the 5 years, 2005-2010, cover a wide range of works starting with analysis and computation of forecasted requirements, design of new devices, laboratory equipment, some redesign of existing ones, manufacturing and testing of devices and equipment by Institute qualified departments, and cold and hot testing of chemical processes developed at the post irradiation laboratory.

The most important analysis concerns the in-core irradiation processes, handling of high radioactivity sources and radioprotection, and radioactive waste management.

The entire activity related to radioisotope production is subject to national regulation for nuclear activities as well as to the provision of international conventions and agreements and for this reason a part of the efforts deployed for technology development were oriented to licensing issues concerning safety and security.

2.1. Target material and size

The modified CINTICHEM target is made [1] of a piece of low enriched uranium foil covered with nickel foil, held and compressed between two aluminium cylinders having welded ends. The annulus target components have the following dimensions:

- concentric aluminium tubes have 165 mm length and diameters of 27.99 mm and 29.77 mm;
- uranium foil has 70 mm x 50 mm x 0.14 mm and weighs approximate 9.25 grams;
- coating Ni foil has 0.015 mm thickness and weighs approximate 1.5 grams.

2.2. Target manufacturing

Dedicated tools and procedures were produced for a qualified stable and reliable process of target manufacturing. Target behaviour during the irradiation process is the most important requirement which depends on material selection, cleanliness of process, achievement of internal gaps and welding of tubes in order to ensure high retention of fission products during irradiation. Target inspection and helium leak testing concludes the manufacturing process. Figure 1 shows the target components assembled and one of welded ends.



FIG. 1. Target components

Figure 2 shows the devices for target assembly and calibration and Figure 3 shows target welding and welded test samples.

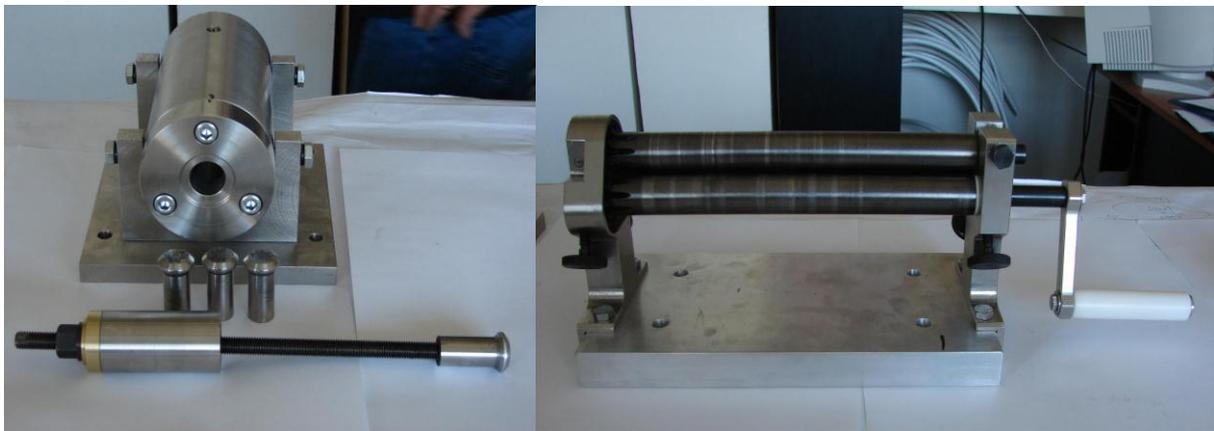


FIG. 2. Devices for target assembly and calibration

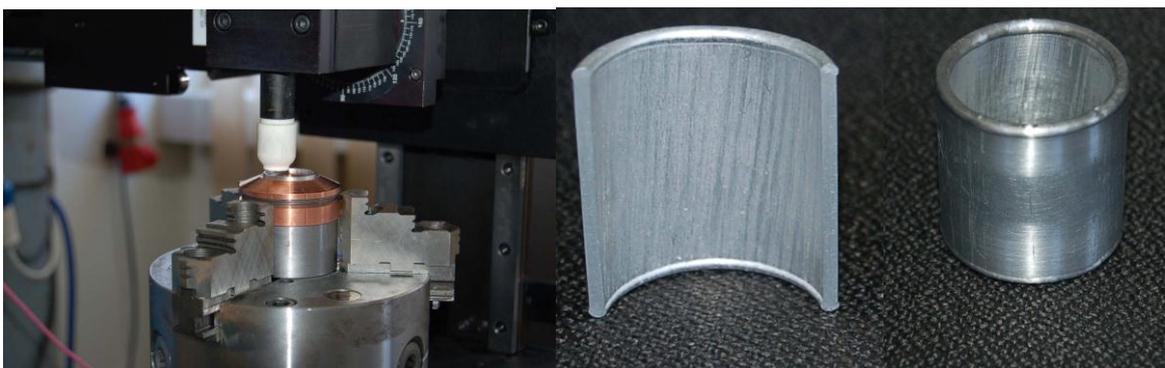


FIG. 3. Target welding and welded test samples

An improvement for target fabrication which leads practically to an absence of the internal gaps between the foil and cladding was obtained by application of internal pressure on the assembly. Figure 4 shows the transversal metallographic cross section of the assembly.

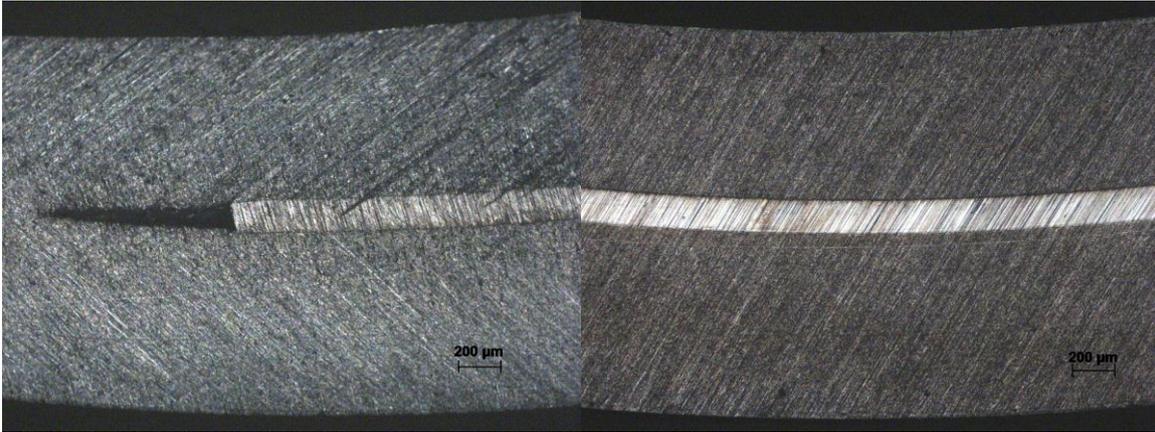


FIG. 4. Transversal metallographic cross section of the assembly

2.3. Irradiation device design and manufacturing

The design specifications for the irradiation device for LEU target irradiation were established in order to ensure safety and efficiency of the irradiation process without modification of reactor core structure or mode of operation. The central channel of a 14MW TRIGA research reactor was proposed for simultaneous irradiation of three annular targets. The main features of this device, Figure 5, are that it could be handled safely with the same tools as the core components and that the targets could be loaded in the existing transfer system and transferred under water without utilization of a cask or container to the transfer cell in the post irradiation examination laboratory. All components of irradiation devices are reusable for every campaign of irradiation reducing practically to zero the amount of radioactive waste generated during the irradiation of targets with positive consequences in the final cost of the radioisotope and safety of irradiation and transfer process.



FIG. 5. Irradiation device and target holder with 3 annular targets

3. NEUTRONIC AND THERMAL HYDRAULIC CALCULATIONS

Neutronic and thermal hydraulic calculations were performed for a given target design and manufacturing conditions and for an in core irradiation device in order to sustain the design and safety analysis of in core assembly operation, and preparation of a safety analysis report for a future utilization license.

3.1. Neutronic calculations of the target

Key parameters for target irradiation are the power developed in the target fission process and temperatures on the target in thermal hydraulic conditions existing in the irradiation device.

The TRIGA neutron calculation system is based on the following codes;

- transport code cell – WIMS permitting to obtain microscopic cross sections of the nuclides, collapsed on 7 energy groups for different burn up steps;
- 3DDT diffusion code supplemented with a burn up loop used for neutron flux distribution calculation and for fuel burn up calculation of the reactor. It is also used to calculate the power and reactivity induced by an experiment in the TRIGA core.

Based on these codes and using the irradiation device model from Figure 6, the effective macroscopic fission cross section of uranium ($\Sigma_f=3.57 \text{ cm}^{-1}$) and the calculated linear power and reactivity change associated with the uranium target inside of the irradiation device introduced in the G7 core location has been determined. This is the most powerful irradiation position in our reactor.

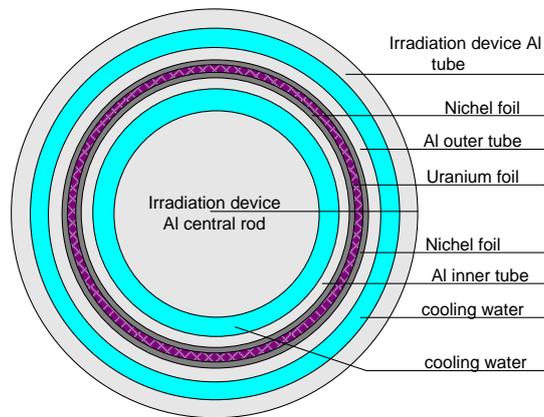


FIG. 6. Cross-section through the irradiation device containing the target

The manufacturing conditions for the target and for irradiation devices which were used in the neutronic calculation model are represented in Figure 7.

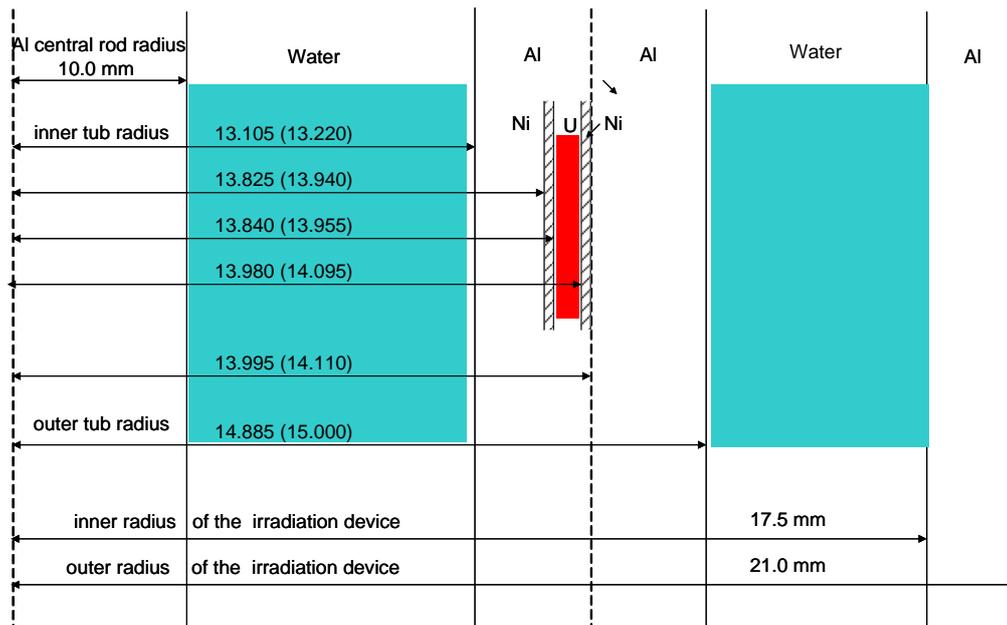


FIG. 7. Vertical section through the irradiation device and target (not to scale) (values in parenthesis are corresponding values obtained after using a sizing draw plug)

Power density in the uranium foil is ($\phi_{\text{thermal}}= 1.6 \times 10^{14} \text{ n/cm}^2 \cdot \text{s}$, $G_f=180 \text{ MeV/fission}$):

$$q''' = \Phi_{\text{th}} \Sigma_f G_f = 1.6 \times 10^{14} \times 3.57 \times 180 = 1.028 \times 10^{17} \text{ MeV/s} \cdot \text{cm}^3 = 16.448 \text{ kW/cm}^3$$
$$P_{\text{target}} = 8060 \text{ W/target};$$

Positive reactivity induced by irradiation device and target is 0.43\$ and this is determined mainly by aluminium water removal and much less by the uranium foil.

3.2. Thermal hydraulic assessment of the target and irradiation device.

Heat transfer computations have been based on the following assumptions [3]:

1. The full power developed by the uranium target is considered 8.0 KW;
2. Uranium foil covered with Ni foil on both sides is tight pressed between the two aluminium cylinders and is considered with no gap. However, because uranium foil thickness proved to be non-uniform [4], calculations were done taking into consideration a hypothetic uniform gap between Ni foil and uranium foil of 0.003 mm and also of 0.005 mm;
3. Heat generated in the metallic uranium foil is propagating through conduction in nickel foils, gaps and aluminium tubes and through forced convection to cooling agent circulating in inner aluminium tube and around outer aluminium tube;
4. The cooling fluid is light water (H_2O) at the temperature of 35°C and gauge pressure of 78000 Pa, which correspond to a water column of approx. 8 m.

The measured water flow through the irradiation device containing the target is equal, with 1.9 l/s, and is circulating through the two channels of the annular target with the same velocity.

The main goal was the evaluation of flow thermal-hydraulic characteristics in the annular flow area around target in the irradiation device, see Figure 6 and 7. The temperature field was calculated using FLUENT code.

A gap must be maintained between foil edges to permit a longitudinal cut of the outer aluminium tube in order to recover the irradiated uranium foil for chemical processing. From a thermal point of view, the unheated gap region provides asymmetry and temperature gradient which were not evaluated. The turbulence model was considered to be suitable for flow conditions.

These conditions are summarized:

- inlet average fluid velocity: 3.88 m/s
- Reynolds numbers for annular flow cooling channels are: $Re_1=10508.71$, $Re_2=8850.335$
- Turbulence intensity for annular flow cooling channels are: $I_1=5.0\%$, $I_2=5.1\%$
- operating gauge pressure: $P=78000 \text{ Pa}$

The following temperatures are very important for target irradiation in the cooling condition for the irradiation device from a safety point of view:

- temperature on the aluminium target inner surface;
- temperature on the aluminium target outer surface;
- uranium foil central temperature.

All these parameters were evaluated starting from the boundary conditions specified above. The analysis shows the influence of gap size on temperatures in uranium foil and near the interface between aluminium and water.

These Figures depict the temperatures in the target as a function of the longitudinal position. The origin corresponds to a central position in the central annular target. Figure 8 shows the temperatures in water, water-Al interface and uranium considering no gap between the different interfaces.

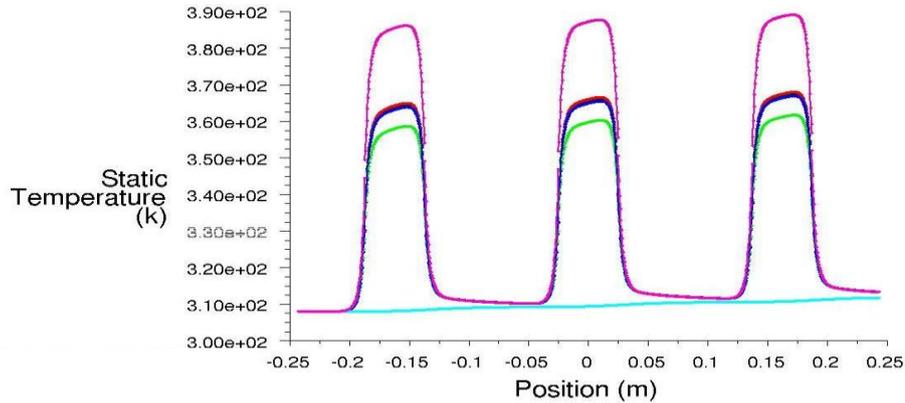


FIG. 8. Temperatures at key locations (water, water-Al interface, uranium) – no gap case

For example, “line-all-0.013125” represents temperature profile across a line situated at a distance of 0.013125 m from central axe of annular target (see Figure 6 and 7). This line is near the water–Al interface situated at 0.013105 m, 20 μm inside Al. The temperature profiles are quite uniform in the cross-section of the annular target containing uranium. However, there is a slight temperature increase in downwards water flow across the section containing uranium. The maximum temperature at aluminium water interface is 365 K for 8 kW of uniform heat dissipation. The temperature profile at a given cross-section of the annular target is quite uniform and does not spread towards the extreme ends of the aluminium tubes. This maximum temperature is well below the saturation temperature of water (388K) at the depth of approximately 8 m where the targets are submerged. When the gap is present, the maximum temperature at the aluminium water interface is practically unchanged - 365K, but the impact on the uranium foil temperature is high. In the no gap case the maximum uranium temperature is 390K (117°C), in the 3 μm gap case (see Figure 9) the maximum temperature increases to 525K (252°C) and to 625 K (352°C) for a 5 μm gap. A uniform gap is not a credible situation because after inserting the foil between two tubes, the outer and inner cylinders are drawn together to obtain good thermal contact.

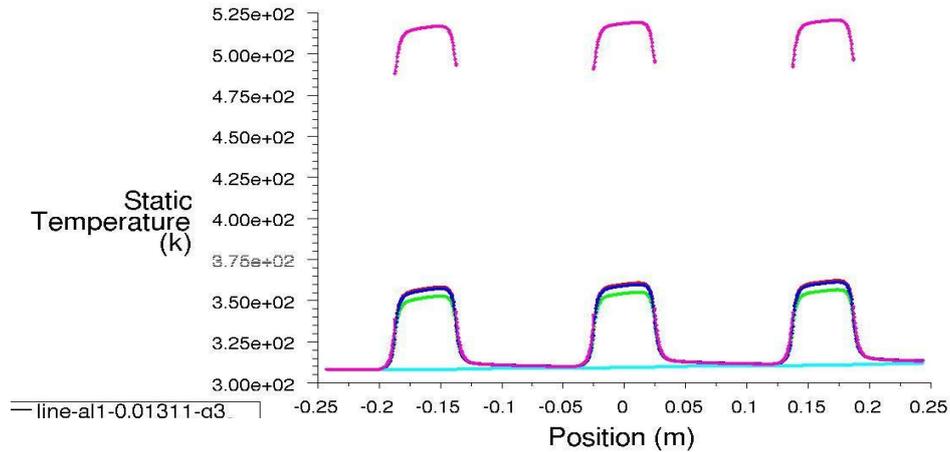


FIG. 9. Temperatures at key locations (water, water-Al interface, uranium) - gap size= 3μ

Heat transfer computations have also been done using RELAP5/MOD3.2, a transient analysis code used for thermal hydraulic design developed at the Idaho National Laboratory. The full power developed by the uranium target was considered in this case to be 9.0 KW.

The hydraulic layout for RELAP5 code of the irradiation device is presented in Figure 10. Irradiation device and target hydrodynamic modelling with specific components of RELAP5/MOD3.2 code are:

- **tmdpvol** 150 component stipulating inside irradiation device pressure;
- **tmdpjun** 145 component stipulating boundary quantity of water flow;
- **pipe** type components (101, 102, 103; 104, 105, 106) stipulating characteristics specific of the tubes;
- **tmdpvol** 140 component stipulating boundary value for pressure;
- **snglvol** 130, 120 components;
- **sngljun** components (124, 123, 122, 121; 128, 127, 126, 125) for consistency of water flow.

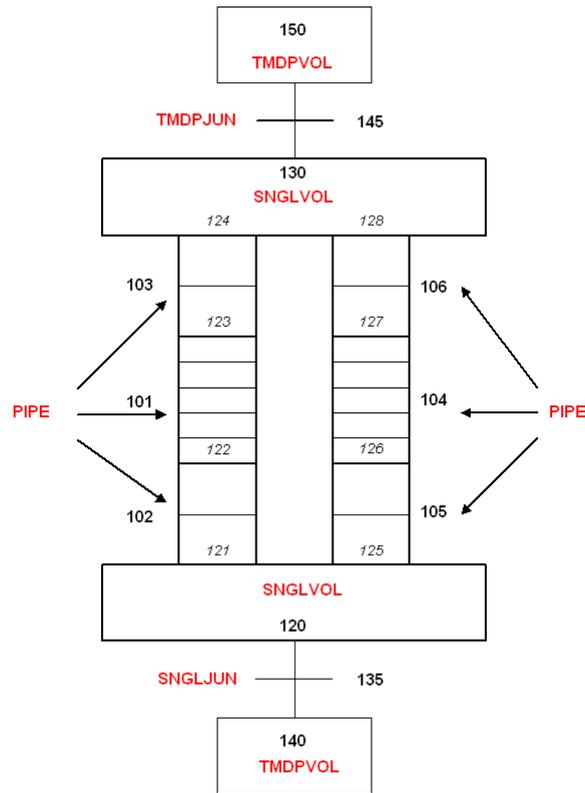


FIG. 10. Irradiation device and target hydrodynamic modelling with specific components of RELAP5/MOD3.2 code

Components involved in heat transfer modelling are 101 and 104. Heat structure is presented in Figure 11.

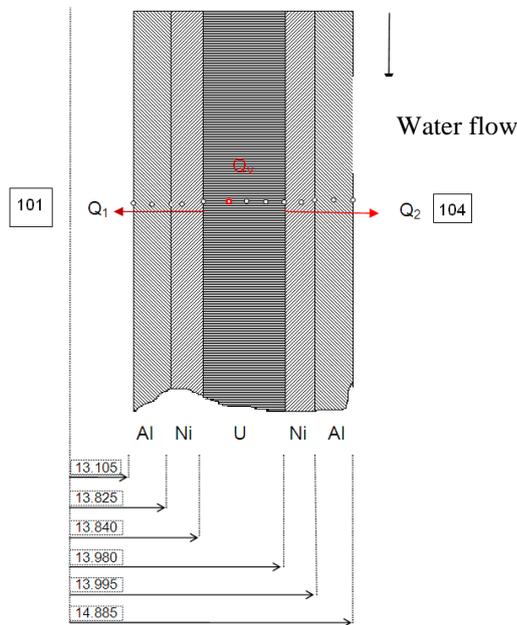


FIG. 11. Heat transfer modelling with specific components of RELAP5/MOD3.2 code

With this data and considering an output power generated during irradiation $P_{\text{target}} = 9$ kW, the temperature distribution into the irradiation target was obtained and is presented in Figure 12. The maximum temperature in the uranium foil is 100.1°C. The temperatures of the outer and inner surfaces of the target are included in intervals (93.5-94.5°C).

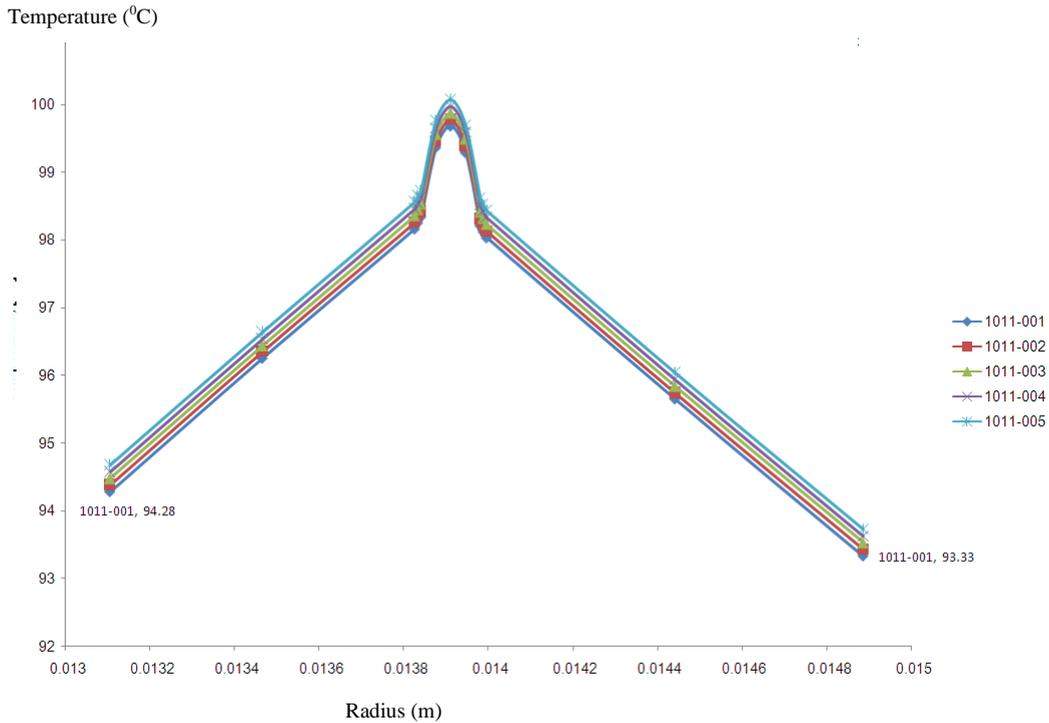


FIG. 12. Temperature distribution inside irradiation target ($P_{target} = 9 \text{ kW}$)

3.3. Activities calculations

Calculation of fission product activities was done with ORIGEN-S code from the SCALE 5 system considering an irradiation at constant neutron flux ($\Phi=1.16 \cdot 10^{14} \text{ n/cm}^2 \cdot \text{s}$). ORIGEN-S code utilizes a Westcott neutron spectrum description in irradiation location. Therefore for calculation of THERM, RES and FAST the TRITON code from the SCALE 5 system is used. To carry out this task, it is needed to work out a 2-D geometric super cell and adequate material description of the core. The result of the 2-D model is presented in Figure 13. The values obtained for the spectral index are: THERM = 0.750, RES = 1.077 FAST = 0.597.

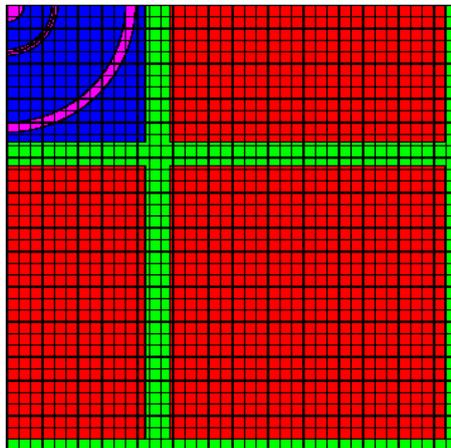


FIG. 13. 2-D geometric super cell description for TRITON code (red: homogenized TRIGA fuel, purple: Al, blue: water, green: shrouds interface)

Different irradiation time can be considered from 5 to 10 days of a target containing 1.0g of uranium, 20% enriched. Calculation of the target irradiated for 5 days after different cooling times (⁹⁹Mo and total activities) produced the results given in Table 1.

TABLE 1. Activities after 5 days of irradiation time and cooling time up to seven days.

Cooling time (days)	0	0.33	0.67	1	2	3	4	5	6	7
⁹⁹ Mo (Ci)	32	30	27	25	20	15	12	9	7	6
Total (Ci)	3685	532	388	311	200	152	124	106	92	81

Irradiation thermal neutron flux is dependent on reactor power and irradiation location and values in our reactor ranges from $1.16 \cdot 10^{14}$ n/cm².s to $1.65 \cdot 10^{14}$ n/cm².s for 10 MW reactor power (perturbed values). Thus, values of specific activities from 32Ci/gU to 48Ci/gU can be obtained at discharge after 5 days of irradiation.

4. SAFETY AND SECURITY OF TARGET IRRADIATION AND HANDLING

Safety and security of target irradiation and handling assessment was a necessary step in the project development performed in order to demonstrate the feasibility of proposed irradiation to sustain the future production of ⁹⁹Mo within the margins of approved limits for reactor operation.

For this reason the typical basic loss of coolant accident (LOCA) and reactivity insertion accident (RIA) considered for the reactor safety case were applied to a dedicated core configuration containing the radioisotope irradiation device loaded with three annular LEU targets.

Suppose a sudden loss of the main coolant loop flow. In 10 seconds flow rate vanishes and the reactor is shut down by scram (this type of accident is considered in TRIGA reactor safety analyses). In such an event, temperature evolution in the fission molybdenum target is presented in Figure 14. The data in this analysis corresponds to a target power of 9000W when the maximum uranium foil temperature is 125°C. Initially, foil temperature decreases to 48°C in two seconds from the reactor scram. In the following 5 seconds the temperature increases to 57°C due to the inversion of the flow rate during natural convection loop formation in the reactor pool. In continuation the temperature decreases constantly without any danger of temperature increasing in the target.

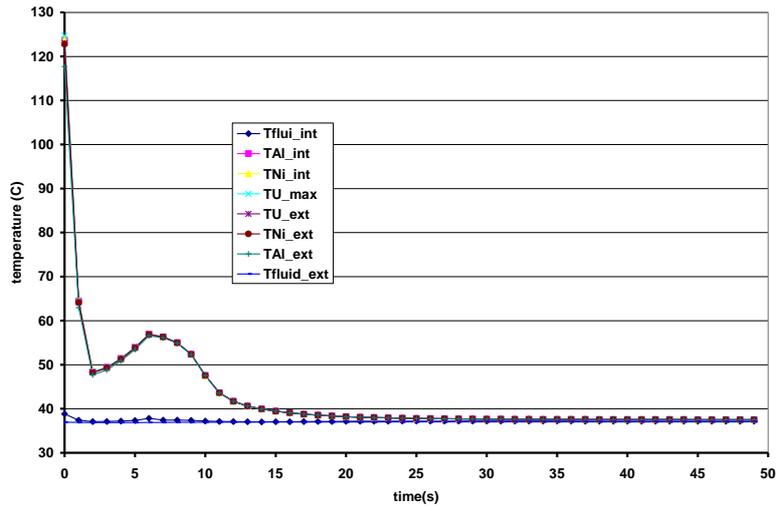


FIG. 14. Temperature evolution in the target during loss of the main coolant flow loop

Insertion reactivity accident is another case taken into consideration. The assumption is made that the reactor is operated at 14 MW and a pulse is produced in 0.1 second which increases the power by a factor of 5. The results obtained in this case are presented in Figure 15. The uranium foil temperature increases in 0.12 seconds from 125°C to 188°C and immediately decreases in a marked manner. The target does not suffer any damage.

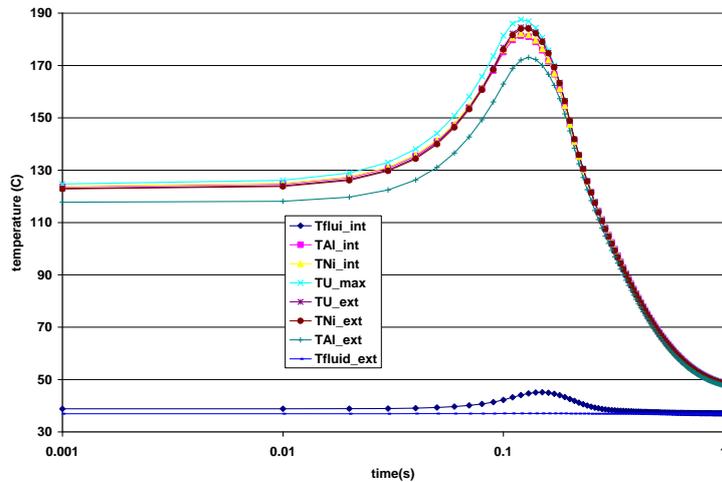


FIG. 15. Temperature evolution in the target during reactivity insertion

Security of fresh target handling, irradiation and post irradiation, in terms of Romanian security regulation concerning LEU storage and handling and other related activities shows that the system implemented for ICN research reactor and post irradiation laboratory do not require any modification.

The above demonstrated steps of small scale radioisotope production by irradiation of LEU foils, which was the subject of IAEA research contract 13364/RO, could be accomplished by the Institute for Nuclear Research without limitation from engineering reasons or/and for safety and security regulation.

5. TARGET IRRADIATION

The target is mounted into the irradiation device and loaded in the TRIGA pool type reactor irradiation location. In the central cavity of the present irradiation device a maximum of three annular targets can be loaded. The amount and the specific activity of ^{99}Mo obtained, depends on the target loading, target power and irradiation time. The un-perturbed neutron thermal flux in the LEU core is $(2.7-2.9)\times 10^{14}\text{ n/cm}^2\text{ s}$. Approximately 40 Ci of ^{99}Mo and 4680 Ci of fission products per gram of U 19.75% enrichment are obtained at the end of 5 days irradiation at 10 MW reactor power.

6. TARGET DISASSEMBLY

Target disassembly is a remote mechanical operation meant to remove the aluminium tubes cladding, having the advantage to be metallic solid waste and to allow a small amount of acid solution for foil dissolution with consequences of relative small dissolver volume and reducing the liquid radioactive waste amount.

The disassembly equipment was designed and manufactured by the Institute department and tested in order to confirm the safety of the target handling and LEU foil recovery. The design, manufacturing and testing of laboratory equipment were performed under the provision of the Integrated Management System of the Institute. Figure 16 shows the as built cutting tool used for target disassembly inside the hot cell.

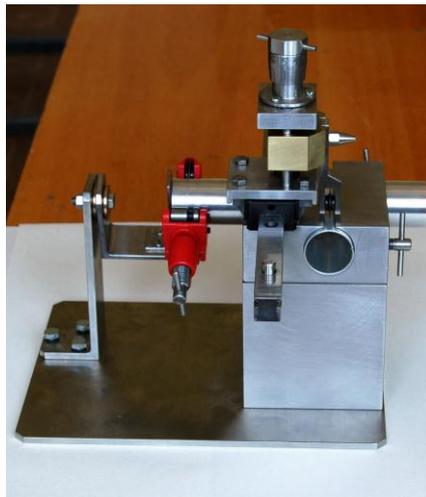


FIG. 16. Cutting tool for target disassembly in hot cell

7. IRRADIATED FOIL PROCESSING

7.1. Uranium foil recovery at the end of disassembly and transfer to the chemistry cell

Following the target disassembly, the irradiated LEU foils are collected into a transfer cylinder in order to use the existing equipment of the post irradiation laboratory. Following the activities calculated after one day cooling the total activity of a foil is 3,000 Ci. In the given conditions the transfer shielding cask and the chemistry cell shielding can't ensure the regular radioprotection condition. For this reason, supplementary analysis led to the conclusion that the existing shielding should be increased or modified. The tight transfer cask mounted on the electrically driven transport vehicle allowed internal supplementary shielding provided by a cylindrical piece of depleted uranium. The final testing of improved shielding

showed the acceptable level of protection for one irradiated, 10 grams LEU foil. This cask travels between the concrete transfer cell and steel shielded chemistry cell. Figure 17 shows the remote actuated transfer cask.



FIG. 17. Transfer cask mounted on transport vehicle

The working side of the chemistry cell was supplemented by a thick lead brick wall and the previous shielding window was replaced with a new window installed in the same frame (see Figure 18).



FIG. 18. New window and additional shielding to radiochemical cell

Following the above design modification of existing equipment, the irradiated foils are safely transferred in a short time between the concrete transfer cell and the chemical cell.

7.2. Testing of dissolution of uranium foil

A small volume dissolver was designed and manufactured for a single foil dissolution connected with an expansion capacity for retaining nitrous vapors and fissile iodine. Figure 19 shows the LEU foil dissolver and top closure.

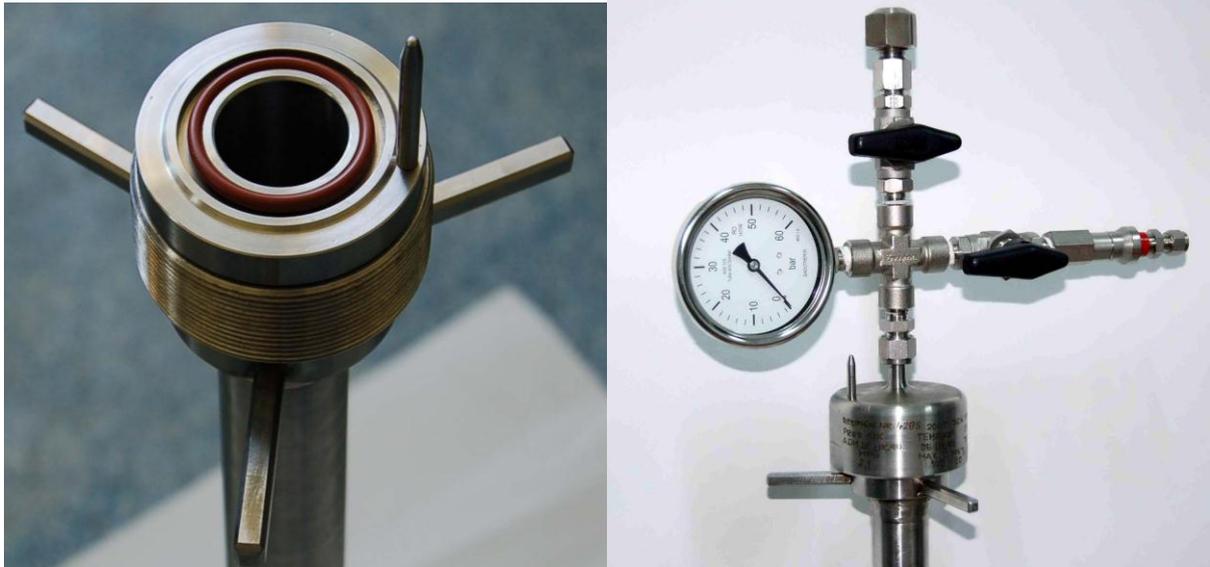


FIG. 19. LEU foil dissolver and top closure

In Figure 20 is the equipped dissolver installed in the working position.



FIG. 20. Uranium dissolution in the dissolver

The gases accumulated during the dissolution process were drained for 20 minutes through an assembly with an iodine trap and cold finger. The cold finger is cooled with liquid nitrogen and is provided with a closure valve. Figure 21 shows the process gases draining.



FIG. 21. Gas draining

The dissolution process evolution, recorded during testing of the dissolver, is presented in Figure 22.

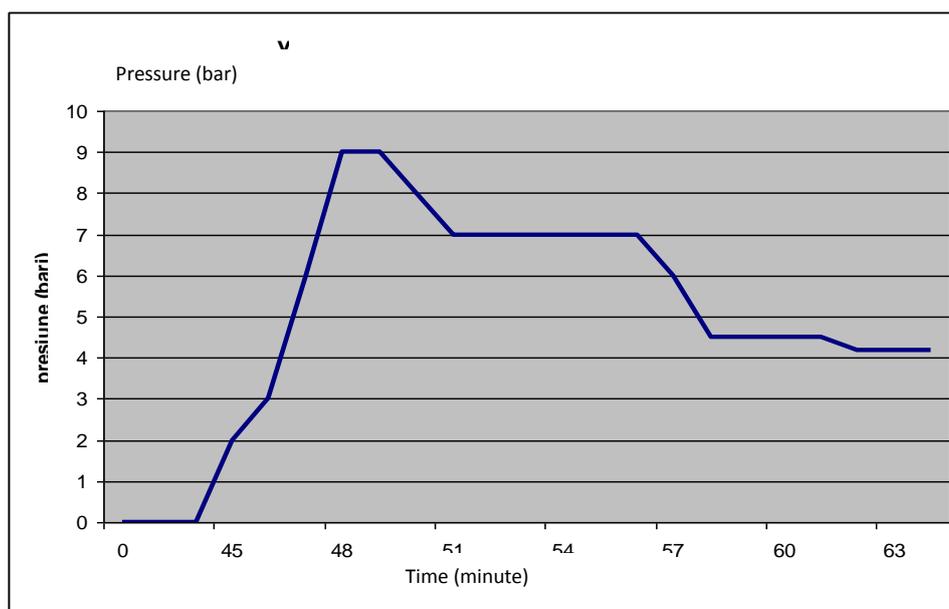


FIG. 22. Dissolution process evolution

The time for complete dissolution of one foil was a maximum of 15 minutes followed by a decompression during 20 minutes through to the expansion capacity.

7.3. ⁹⁹Mo chemical separation and purification test

7.3.1. Molybdenum precipitation and precipitate dissolution

The dissolved LEU solution was drained from the dissolver. The total volume of solution was 66 ml from which a 1-ml sample was taken (QC1).

The following carriers were added:

- 4 mg of iodine carrier (4 ml of KI at 1mg/ml);

- 0.5 mL of 10% AgNO₃ in 0.1M HNO₃
- 0.5 mL of Mo carrier (10 mg/mL).
- 1 mL of 1.0 M HCl
- 2.5% KMnO₄ solution slowly to the raw fission liquor until a deep pink color holds for ~30 seconds. That required 4 mL of solution
- 1.5 mL of Rh carrier (8 mg/mL)
- 2.0 mL of Ru carrier (5 mg/mL)

Molybdenum from the solution is oxidized to Mo⁶⁺ using KMnO₄ and precipitated with alfa benzoin-oxime according to reaction:



The precipitate formed was filtered through a fritted column prefilled with glass beads. Filtered solution was collected in RFW (see Figure 23). 2-mL of sample solution were taken from RFW (QC2)



FIG. 23. Mo Precipitate Filtration

1. Precipitate washing

The precipitate was washed 2 times with 0.1M HNO₃ for each wash. These washes were drained through the glass column in Acid Wash (AW) bottle. The precipitate was washed 5 times more with 10 ml 0.1M HNO₃.

2. Precipitate dissolution

Precipitate dissolution was done injecting 10 mL of 0.4M NaOH with ~1% H₂O₂ directly in the glass column, then the column was heated until the solution began to boil. After 5 minutes of cooling the solution was drained in 1-A bottle.

The dissolution was repeated by injecting 10 mL of 0.2M NaOH with ~1% H₂O₂ directly in the glass column, then the column was heated until the solution began to boil. After 5 minutes of cooling the solution was drained in 1-A bottle.

The fritted column was washed with 10 ml of 0.2 M NaOH and drained in 1-A bottle. In the glass column still remained some brown precipitate.

A 2-ml sample was taken from 1-A (QC3).

7.3.2. Molybdenum purification

- a. **AgC Column:** from bottom to top: glass wool – 5 cm AgC – g.w. – 5 cm C – g.w.
- b. **HZO Column:** from bottom to top: g.w. – 3 cm AgC– g.w. – 3 cm HZrO – g.w.– C g.w.



FIG. 24. First purification



FIG. 25. Second purification

For the first purification, the solution from 1-A was drained through AgC column into the 2-A bottle by gravity. The flow rate was 0.7 ml/min. The 1-A bottle was washed with 10 ml of 0.2 M NaOH and drained in 2-A bottle (see Figure 24). A 2-ml sample was taken from 2-A bottle. (QC4)

Before the second purification we added 4 ml of KI and 0.5 mL of 10% AgNO₃ in 0.1M HNO₃. The solution from 2-A was passed through an HZO column in 1-B bottle (see Figure 25 second purification). Gravity assured a flow rate of 0.45ml/min. The 2-A bottle was washed with 10 ml of 0.2 M NaOH and drained in 1-B bottle. Because the solution appeared cloudy we filtered it through a 0.45µm Millipore filter. The solution was drained in 1-F bottle. A 2-ml sample was taken from 1-F bottle. The approximate volume of 1-F bottle was 50.5 ml.

7.3.3. Results of ⁹⁹Mo chemical and purification tests

Gamma specific activity measurements were performed using an HpGe detector. The detector was calibrated using a standard liquid source containing ⁶⁰Co, ¹³⁷Cs, ²⁴¹Am, and ¹³³Ba. The previously collected samples during process of precipitation and purification (QC1, QC2, QC3, QC4 and QC5 – Quality Control Samples Number) were measured in the same condition. The interest was focused in measurement of molybdenum activity in each sample (see Table 2)

TABLE 2. Fission molybdenum activities in different samples taken during chemical processing

Sample denomination	Total volume from which the sample was taken (ml)	⁹⁹ Mo Activity of sample (Bq)
QC1	66	952380 (Λ_1)
QC2	-	-
QC3	30	509964 (Λ_3)
QC4	40	480616 (Λ_4)
QC5	54,5	431356,6 (Λ_5)

Based on these results we calculated the process yield.

$$\eta = \frac{\Lambda_5}{\Lambda_1} = \frac{431356,6}{952380} \times 100 = 45,3 \%$$

and also partial yield of each step:

- *Before the first purification step:*

$$\eta_{1 \rightarrow 3} = \frac{\Lambda_3}{\Lambda_1} = \frac{509964}{952380} \times 100 = 53,5 \%$$

Note: After the precipitate was dissolved half of molybdenum activity was lost and we assume that most of it remained on the fritted column.

- *After first purification step (before second purification):*

$$\eta_{3 \rightarrow 4} = \frac{\Lambda_4}{\Lambda_3} = \frac{480616}{509964} \times 100 = 94,2 \%$$

Note: AgC column permitted the molybdenum to pass through without considering loss of its activity.

- *After second purification (the final solution I-F)*

$$\eta_{4 \rightarrow 5} = \frac{\Lambda_5}{\Lambda_4} = \frac{431356,6}{480616} \times 100 = 89,75 \%$$

Note: Some of the molybdenum activity was retained on the HZO column, which indicates a poor conditioning of the column.

In Table 3 the measurement results for other than ⁹⁹Mo radioisotope impurities in the samples are presented. In QC5 (the final sample) gamma ray activity of the impurities are lower than the detection limits of the detection system.

TABLE 3. Radionuclide activity in samples taken during chemical processing

Radionuclide	T _{1/2}	Activity in QC1 (Bq/0.1ml)	Activity in QC2 (Bq/0.1ml)	Activity in QC3 (Bq/0.1ml)	Activity in QC4 (Bq/0.1ml)
Ce-143	1,404 days	2923,1	1302,1	-	-
I-131	8,04 days	116,33	55,85	30,58	-
Nd-147	11,06 days	1818,3	-	-	-
Np-239	2,355 days	2425,68	868,9	-	-
Ce-141	32,38 days	124,3	47,7	-	-
Ba-140	12,79 days	483,9	-	-	-
Te-132	3,246 days	492,3	-	97,32	35,88
La-140	40,27 hours	558,9	-	-	-
Ru-103	39,35 days	66,9	-	-	-
I-132	2,38 hours	8.7 · 10 ⁹	420600,7	1,08x10 ⁶	3,1x10 ⁵
I-133	20,3 hours	2362,2	1646,2	148,72	85,7
Sr-91	9,67 hours	14675,3	4856,1	-	-
Nb-97	73,98 min	2.9 · 10 ¹⁶	-	9,6x10 ⁸	1,789x10 ⁹
Zr-97	16,8 hours	5210,6	-	172,8	141,44
Zr-95	64,4 days	57,2	-	14,18	-
I-135	6,61 hours	-	5023,9	-	-

Attempt of measurements of gross α activity for final sample (QC5) were not conclusive.

8. WASTE TREATMENT AND DISPOSAL

The nuclear and radioactive waste management is based on well-established safety standards for the management of radioactive waste.

Romanian legislation and regulations are all developed from internationally agreed standards, guidelines and recommendations in order to ensure the protection of the public and the environment.

8.1. Disposal of radioactive waste in Romania

There is not a geological facility for long lived (half-life >30 years) low and intermediate level or HLW waste disposal in Romania. Short lived low and intermediate level waste may be disposed in a near surface disposal facility (former uranium mine situated 1000m above sea level).

On the Institute site there is a Radioactive Waste Treatment Facility available, which is able to assure the treatment and conditioning of radioactive waste resulting from different on site activities.

The basic treatment procedures are volume reduction, radionuclide removal and change of composition. Operations include: incineration of combustible waste or compaction of dry solid waste, evaporation, filtration or ion exchange of liquid waste streams, and selective precipitation for uranium recovery from liquid radioactive waste produced by fuel fabrication plant.

The most common immobilization method includes solidification of low and intermediate level liquid radioactive waste in cement or bitumen. The waste immobilization method is packaging in 200 liters steel drums containers. The final step is disposal in near surface disposal facility (in uranium mine levels).

8.2. Estimation of radioactive waste amount

Without taking into consideration uranium recovery, the estimation for average amounts of radioactive waste for a target containing 16 g of 19.75% enriched uranium is:

- max. 200 ml high rad. level of liquid;
- max. 200 g of metallic waste;
- max. 2000 g of low and medium level solid waste (plastics, glassware, textile);
- max. 300 ml of low and medium level liquid waste

The treatment and elimination of gaseous waste is solved so as to maintain the exposure of persons in unrestricted areas as low as reasonably achievable.

Xenon, krypton and iodine isotopes are trapped on special materials in absorber vessels (recovery device for the beginning) for decay before elimination in atmosphere (see Figure 26).



FIG. 26. Iodine and noble gases recovery device

Volume reduction of solid waste is carried out through cutting and subsequently, it is conditioned in special containers. Liquids containing enriched uranium will be stored till a decision concerning recycling for uranium recovery will be taken. The preferred option would be the transfer of these wastes to a recycling center in order to avoid long term storage. If not possible after 7 to 10 years of cooling time, uranium and plutonium from liquid waste could be extracted and remaining waste treated and conditioned for storage in National Storage for Radioactive Waste (a former uranium mine). Another option is to convert high radioactive waste solutions to solid form and to use for disposal a similar way as for spent fuel resulted from nuclear power plant.

9. CONCLUSIONS

The Institute's decision to be involved in medical isotope production using low enriched uranium targets has been taken in the context of the need to increase research reactor utilization and in order to have a contribution at the improvement of medical care of the population.

Domestic efforts have been sustained by an existing infrastructure consisting of 14 MW TRIGA reactor, hot cell facility, Radioactive Waste Treatment Facility and the Center for Radioisotopes Production in Bucharest. The Institute considered a medium term plan for development of technology and associated infrastructure complementary to the existing one in order to ensure a stable and licensed process of production considering: target fabrication using imported low enriched uranium foils, target irradiation, target disassembly and dissolution, product recovery and purification, and radioactive waste treatment. The main results obtained in developing technology for ^{99}Mo production show the ability of the Institute to develop the entire proposed program to design, built and test equipment and experiments to assess the progress and nuclear safety and security and to outline the limitation and countered in the performance of the program.

The main issue remains the achievement of a stable process which will meet the good manufacturing practices for ^{99}Mo solution including accredited qualified measurements. The second issue concerns the radioactive waste management interface with existing technologies operated by the Institute and the new one technology which should be developed for safe and economical application of fission Mo technology production.

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