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Feasibility of Conversion to LEU-based Reactor Production of Mo-99

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ABSTRACT

At present JSC "SSC RIAR" employs fission Mo-99 accumulation technology with the use of highly enriched uranium-based targets. In order to implement the conversion of existing nuclear technologies to low enriched uranium (LEU), a calculation data analysis was performed to study feasibility of standard technology conversion. The current stick-type target was used as a basis. A whole scope of scientific research and development study including calculations of neutronic and thermal-hydraulic characteristics was performed. Feasibility of conversion to LEU-based reactor production of Mo-99 was demonstrated. This work was done in cooperation with the National Research Nuclear University Moscow Engineering and Physics Institute (MEPHI) with funding from the Ministry of Science and Education of the Russian Federation within the framework grant #02.G25.31.0069.

Introduction

Nowadays one of high-demand radioisotopes is Mo-99 that is widely used in the nuclear medicine. It is used as a basis for production of technetium -99m generators (Tc-99m), which are used for medical diagnosis of a high number of human diseases including cancerous and cardiovascular diseases above all others.

There are two primary processes for producing Mo-99:

- ✓ Production of Mo-99 as one of the U-235 fission products;
- ✓ Production of Mo-99 via a neutron capture reaction of Mo-99 nuclide.

Major suppliers do not give consideration to the second production method because it does not allow for producing commercial-scale quantities of Mo-99 but the first production method (from fission products) is still the primary production process of Mo-99. Enriched uranium is used for production of this radioisotope. Generally, the enrichment of uranium in U-235 makes up ~90%. Consequently it should be noted that there exists a risk of unauthorized use of such uranium. In order to resolve this challenging task, it was necessary to work out a concept with the focus on conversion of existing nuclear technologies to low enriched uranium (LEU).

Scientific Center “State Scientific Center –Research institute of Atomic Reactors” (Dimitrovgrad) participates in conversion of its nuclear technologies with the use of highly enriched uranium (HEU). Such technologies include the use of 90% enriched nuclear fuel for operation of research reactors as well as HEU target-based production of Mo-99. Specifically, feasibility analysis of conversion of one of the research reactors to low enriched uranium fuel was performed within the framework of the above-mentioned program [1]. As to the conversion of molybdenum-99 radioisotope production, the calculation data analysis was carried out. Presented here are the results of this work.

Mo-99 production and its conversion to LEU

Mo-99 is accumulated in pool-type research reactors RBT-6 and RBT-10 of thermal output of 6MW and 10MW, respectively. They are both located at the JSC “SSC RIAR” site. Research reactor RBT-10 is a water-cooled water-moderated pool-type thermal reactor. The reactor is intended for irradiation tests to investigate properties of materials under irradiation and for production of radionuclides. The fuel assembly slug is 350 mm long. The fuel assembly takes a form of square at the cross-section and the side of the square is equal to 69 mm. Fuel assemblies are spaced for 78 mm. The RBT-10 reactor core is comprised of seventy eight (78) spent fuel assemblies operated in the SM reactor [2] up to a burnup of 20-40 % (but no higher than 53 %), six combined safety-shim rods, one automatic regulating rod and ten experimental channels (Figure 1). Twelve beryllium blocks located at the core corners are also employed as a moderator other than water. The RBT-6 [3] reactor closely resembles the RBT-10 reactor as to the design and neutronic parameters. The main difference is the number of operating fuel assemblies and experimental channels.

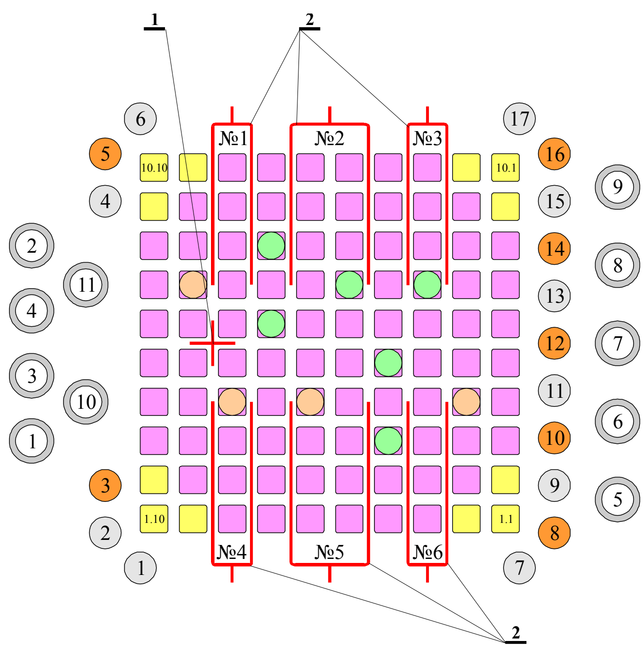

Figure 1. RBT-10 core layout.
1 – automatic regulating rod;
2 – combined safety-shim rods.

The targets irradiated for six days in the reactor and after subsequent cooling period of 24 hours are transported to the radiochemical unit for processing. At present JSC “SSC RIAR” employs the

ROMOL-99 production process to extract Mo-99 from irradiated uranium targets. This process is based on alkaline dissolution, separation of uranium in the form of sodium diuranate precipitate and subsequent extraction of Mo-99 from the resultant solution by sorption onto aluminium oxide. The final purification step of the product is the Dowex-1 ion-exchange resin. The intermetallic compounds i.e. UAl_3 or UAl_4 are used as irradiated fuel to be reprocessed.
The production process used involves a number of restrictions on the Mo-99 production, which are as follows:

- ✓ The maximum mass of aluminium in the targets is 200 g and the target is 200 mm high;
- ✓ Fuel meat of the target comprises uranium intermetallic compounds UAl_3 or UAl_4 enclosed in aluminium matrix;
- ✓ Designed capacity of the molybdenum production facility is 300 Ci at the most that needs to be calibrated on the 6th day that corresponds to activity of 4150 Ci as of the end of irradiation;
- ✓ The limiting high temperature of intermetallic fuel under irradiation is 500 ^{0}C.

The calculation data analysis as to neutronic and thermal-hydraulic parameters was performed with the use of computer codes MCU [4] and RELAP5/MOD3.2 [5] to demonstrate feasibility of Mo-99 conversion in support of new LEU-based targets.

Highly enriched uranium-based targets

Two types of targets are used for the reactor-based production of Mo-99. The first design modification of the target represents itself a rod with a circular cross-section. The target meat incorporates the intermetallic compound (UAl_3+Al). The target cladding is made from aluminium alloy and has a close mechanical contact with the target meat. The cladding is 0.5 mm thick. The target is sealed at both end faces by welding the end plugs to the cladding. The irradiation rig (the first design modification of IR) comprises the flux displacer to attach the grids for fixing targets (Fig. 2). All the components of irradiation rig are made from aluminium alloy. The targets are inserted so that they form one circle around the displacer. Generally, the irradiation rig can incorporate fifteen (15) targets. The irradiation rig is loaded into the flow-through capsule to be irradiated in the core. The irradiation rig output depends on its irradiation position in the core, fuel distribution over the core, position of the safety-shim rods and materials to be irradiated in other channels. According to the above given factors, the IR output, efficiency of fuel utilization (it is estimated as a target power in relation to the mass of fissile isotope) and activity of Mo-99 that is accumulated in the irradiation rig for 6 days are within the ranges given in Table 1.

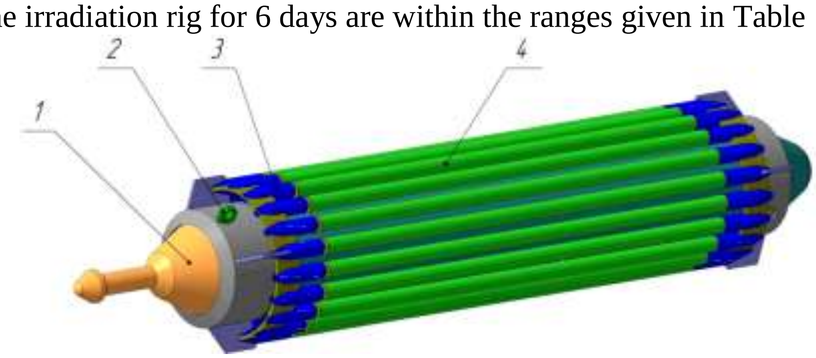

Figure 2. The first design modification of IR
1 – flux displacer, 2 – hook, 3 – holders, 4 – targets.

The second design modification of the target takes a shape of square at the cross-section. The target meat incorporates the intermetallic compound (UAl_4+Al). The target cladding is made from aluminum alloy and has a diffusive bonding with the target meat. The cladding thickness varies within a range of 0.15 to 0.2 mm. The irradiation rig (the second design modification of IR) comprises the flux displacer to attach the spacer components for fixing targets (Fig. 3). All the components of irradiation rig are made from aluminium alloy. Fifty two (52) targets are inserted so that they form two circles around the displacer. Table 1 specifies the parameters of IR and target.

Figure 3. General view of the second design modification of the irradiation rig.

Table 1. Tabulated summary specification of HEU-based targets (IR).

Parameter	target (type 1)	target (type 2)
Total length, mm	192	200
Fuel meat length, mm	168-172	185-195
outer diameter, mm	5.9 (diameter)	2.6 (across flats)
U-235 density in the fuel meat, g/cm^3	0.49	1.47
Volume ratio of intermetallic compound in the meat, %	10.6	40.6
U-235 mass, g	1.5-1.55	1.05-1.1
Enrichment of uranium in U-235, %	90	90
Aluminium content, g	13.1	3.8
IR output, kW	30-40*	60-80*
Efficiency of the fuel used, kW/g_{U-5}	1.55-1.95	1.3-1.65
Activity as of the end of irradiation period, Ci	1400-1800	2800-3800
Note: data with designation "*"specific to irradiation of targets in the RBT-6 reactor.		

Low enriched uranium-based targets

The major challenges of the Mo-99 production conversion to low enriched uranium (uranium enriched less than 20%) are as follows:

- ✓ Decrease in the target radionuclide yield is proportional to the enrichment reduction and subsequently it leads to losses of production process output (production process used);
- ✓ Increase in alpha-emitting transuranic radionuclides yield (primarily, Pu-239) in tens of times and as a result of this the risk of final product quality reduction escalates and subsequently the extraction process\ purification of the target radionuclide needs to be modified.

In order to regain the output of the Mo-99 reactor-based production process, it will be necessary to increase the mass of uranium-235 to be irradiated by increasing the total mass of uranium. Three approaches exist for increasing uranium mass:

- ✓ Increase the number of targets to be irradiated;
- ✓ Increase the target size;
- ✓ Increase the target fuel meat density by increasing uranium.

Once the number of targets and irradiation rigs has been increased, the unit costs of the Mo-99 production will grow considerably and thus up to five times as much volume of waste much be produced as a result. By increasing the target size, it will be possible to reduce the number of targets but the amount of waste will not be reduced.

To obtain higher density of fuel composition in the target by increasing uranium, it will be possible to reduce the number of targets but the volume of waste from the target processing will also increase due to increased mass of uranium. As the stick-type targets of the first design modification have smaller heat pickup surface, it will be possible to increase uranium density by intensifying heat transfer (increase of the coolant rate, extension of the heat pickup surface etc). As to the second design modification of the target, such an increase seems improbable because the density is constrained by the fabrication method of intermetallic compound.

Below are shown the calculated neutronic parameters of irradiation in the RBT-6 (a replica of the RBT-10 reactor) with reference to the first type of targets with due consideration for different uranium density in the fuel meat. Table 2 includes specifications of the LEU-based targets and the irradiation rig of the first design modification, which were used for calculations.

All the calculations provided for the common parameters of the core (fuel distribution over the core, position of the safety-shim rods and materials to be irradiated in other channels) and the same irradiation position of irradiation rig in the core. The reactor power was taken to be equal to 6 MW. Calculations were performed with the use of the MCU code. The obtained calculation results are shown in Fig. 4.

Table 2. Specifications of the LEU-based targets and the irradiation rig of the first design modification used for calculations

Parameter	Calculation variant					
	1	2	3	4	5	6
Total length of target, mm	192					
Length of target meat, mm	170					
Outer diameter of the target, mm	5.9					
Target clad thickness, mm	0.5					
Volume concentration of intermetallic compound in the fuel meat, %	10.6	20	30	40	50	50
Mass of U-235 in the target, g	1.55	0.64	0.96	1.28	1.60	1.60
Enrichment in U-235, %	90	19.7	19.7	19.7	19.7	19.7
Number of targets in IR	15	15	15	15	15	16
Mass of U-235 in IR, g	23.5	9.6	14.4	19.2	24.0	25.6
Total mass of aluminium, g	200.0	196.4	192.7	189.0	185.2	197.6

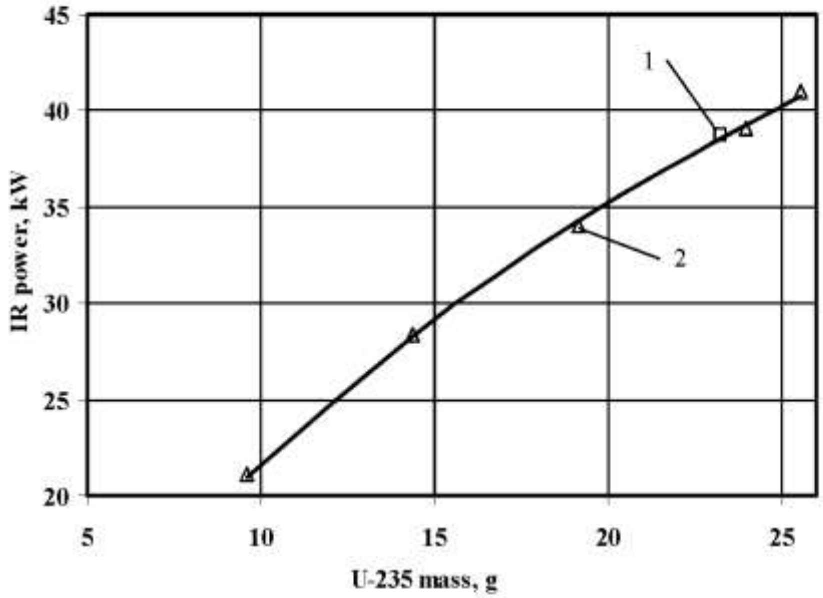

Figure 4. IR power in relation to the U-235 mass in the IR:
1 – HEU-based targets; 2 – LEU-based targets.

The power output of irradiation rig loaded with the LEU targets (see Fig. 4) can be approximated with a satisfactory accuracy:

$$W_{IR} = \frac{2.815 \cdot M_{U5}}{(1 + 0.03 \cdot M_{U5})}$$

where W_{IR} is the IR power output, kW;
M_{U5} is the U-235 mass in the IR, g.

If the volume concentration of intermetallic compound in the target meat is 40% (such a value was achieved in the targets of the second design modification), the U-235 mass in the irradiation rig will be 19.2 g and the IR power will be equal to 34.3 kW based on the above given formula. It is 1.13 times lower compared to the IR power loaded with the HEU targets (38.8 kW). So if the reactor-based production of Mo-99 maintained at the achieved level of production (1400-1800 Ci per week), the number of irradiation rigs will remain basically unchanged.

In the extreme case when the target meat of the first type is made of pure intermetallic compound UAl_3, the irradiation rig can accommodate eighteen (18) targets (aluminium mass is 199.8 g) and thus its power output will be roughly 60 kW. Hypothetically, the LEU-based reactor production output of Mo-99 will be very close to the existing one provided that the second design modification of irradiation rig is used with HEU targets.

Conclusion

The conducted experiments demonstrated a technical feasibility of conversion to LEU-based production of Mo-99 using the RIAR’s proven reactor-based production method of Mo-99 in the targets of with a circular cross-section. However, in order to achieve the same production output of Mo-99 with the use of these targets as in the targets with a square cross-section, it will be necessary to fabricate the target meat out of pure intermetallic uranium and probably it will be necessary to increase the number of irradiation rigs by 20- 30%.

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