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Research Article

Experience in the production of ^{99}Mo from low enriched uranium at the VVR-ts research nuclear facility*

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Abstract

The key industrial method for producing ^{99}Mo is production of the radionuclide as one of the ^{235}U fission fragments. ^{235}U is irradiated with neutrons in a nuclear reactor (both heterogeneous and homogeneous nuclear reactors can be used) and then processed in radiochemical laboratories, where ^{99}Mo is chemically extracted from fission products. Both highly enriched uranium (HEU) and low enriched uranium (LEU) can be used to produce ^{99}Mo by the fragmentation method. To date, almost all world producers, with the exception of Russia, are either in the final stages of transferring production from highly enriched uranium to low enriched uranium, or are already producing ^{99}Mo using LEU. This is due to the problems of non-proliferation of nuclear materials and the prevention of the likelihood of terrorist threats. A number of experimental studies have been carried out on the basis of the VVR-ts research reactor. Experimental studies included the study of the effect of LEU targets on the reactivity reserve of the VVR-ts reactor, irradiation of these targets in experimental channels and separation of ^{99}Mo from them. The paper presents the results of producing and separating ^{99}Mo from targets with LEU material. It is shown that it is necessary to improve the processing technology to increase the production of fragmented ^{99}Mo from LEU.

Keywords

fragmentation ^{99}Mo, research reactor VVR-ts, highly enriched uranium, low enriched uranium, target for ^{99}Mo production

Introduction

Topical nowadays is to increase the production of the ^{99}Mo radionuclide for diagnosis of oncological diseases (World Nuclear Association. Radioisotopes in Medicine). Russia's major producers of fission ^{99}Mo using HEU are the Research Institute of Atomic Reactors (JSC SSC RIAR) in Dimitrovgrad and the L.Ya. Karpov Research Institute of Physical Chemistry (JSC RIPC) in Obninsk covering in full the demand of Russian medicine for this isotope (Molybdenum-99 (2022) Russian Nuclear Community). In connection with threats to nonproliferation of nuclear materials, an issue has arisen regarding the conversion of the fission ^{99}Mo production to LEU targets (Review of Potential Molybdenum-99/Techetium-99m Production Technologies 2010; Non-HEU production technologies for molybdenum-99 and technetium-99m 2013; Muranaka 2022; Program for converting civilian nuclear research

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reactors and molybdenum-99 production targets from HEU to LEU 2012). Achieving this requires undertaking a set of research and technology activities.

Experience of LEU-based ^{99}Mo production in the VVR-ts reactor

HEU-based production of the fission ^{99}Mo radionuclide was started at JSC RIPC in 1985. The technology for manufacturing and irradiating targets has been improved continuously leading to a major increase in production of the desired radionuclide. Currently, the potential output amounts to about 400 Ci (commercial grade) per week.

Activities were undertaken in 2015–2019 to justify the geometry and performance of LEU targets, as well as the possibility for manufacturing such targets (Zevyakin et al. 2022). In 2020–2021, a number of LEU targets were manufactured at JSC RIPC and a range of experimental studies were conducted, including the investigation of the LEU target effect on the VVR-ts reactor reactivity margin, irradiation of these targets in experimental channels, and isolation of ^{99}Mo from them. Targets with direct-flow cooling were used (Fig. 1).

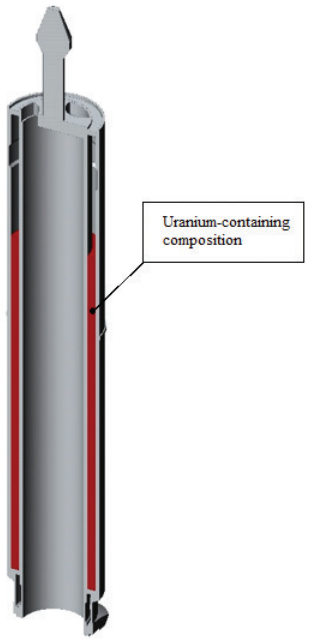

Figure 1. Design of a target with direct-flow cooling for ^{99}Mo production.

A target with direct-flow cooling represents a tubular structure with a water feedthrough for improved removal of heat (Pakholik et al. 2021; Zevyakin et al. 2022). There is a mixture of a uranium-containing material and zinc oxide placed between the internal tube and the external tube. Not more than two targets are installed into the channel at a time.

Standard channels of the MAK and MAK-2 loop devices with standard operating parameters (pressure, flow rate, temperature) were used (Pakholik et al. 2021).

Experimental investigation of the LEU target effect on the VVR-ts reactor reactivity margin

The experimental study was conducted in a depoisoned reactor. The reactivity margin (ρ_{meas}) was determined from the control rod (CR) in-core position (Kochnov et al. 2022) with the reactor brought to the minimum controlled power level (MCPL) prior to and after the LEU target loading into experimental channel (EC) 8–9. The experimental study consisted of the following stages.

- Bringing the reactor to the MCPL, fixing the CR position and the primary circuit water temperature (T_1, °C), and reducing power to zero. The reactor reactivity margin (ρ_1) was calculated based on experimental data.
- Loading LEU targets into EC 8–9.
- Bringing the reactor to the MCPL, fixing the CR position and the primary circuit water temperature (T_1, °C), and reducing power to zero. The reactor reactivity margin (ρ_2) was determined based on experimental data.
- Determining the change in the VVR-ts reactor reactivity margin (ρ) with the LEU targets placed in EC 8–9.

The reactor reactivity margin (ρ_1) with the reactor brought to the MCPL during stage 1 was calculated from the fixed CR position (integral characteristics of the control rods are used):

$$\rho_1 = \rho_{meas1} \quad (1)$$

The reactor reactivity margin (ρ_2) with the reactor brought to the MCPL during stage 3 was calculated taking into account the variation in the primary circuit water temperature (ΔT, °C) relative to stage 1:

$$\rho_2 = \rho_{meas2} - \Delta\rho_t, \quad (2)$$

where $\Delta\rho_t = \alpha_t \cdot \Delta T$ is the temperature reactivity effect, %; $\alpha_t = \pm 0.012\%/°C$ is the VVR-ts reactor temperature reactivity coefficient (Kochnov et al. 2022).

The reactivity margin variation was calculated using the following formula:

$$\Delta\rho = \rho_2 - \rho_1 \quad (3)$$

Table 1 presents the measurement and calculation results.

As it can be seen from Table 1, the positive reactivity increase was 0.042% with the LEU target loaded into the experimental channel of the VVR-ts reactor. In a similar experimental study, a reactivity increase of 0.045% was obtained for a standard HEU target (Table 2). These results show that the neutronic performance of the LEU target is comparable with that of the HEU targets in use. The generation of ^{99}Mo in LEU targets during irradiation in the reactor is comparable with the generation of the same radionuclide in HEU targets.

Table 1. Measurement and calculation results for LEU

Stage No.	$\rho_{measi,}$ %	$T_{i,}$ °C	Δ T, °C	$\Delta\rho_{t,}$ %	$\rho_{i,}$ %	Δρ, %
1	2.552	17.80	+0.30	–	2.552	0.042
3	2.590	18.10		-0.004	2.594	

Table 2. Measurement and calculation results for HEU

Stage No.	$\rho_{measi,}$ %	$T_{i,}$ °C	Δ T, °C	$\Delta\rho_{t,}$ %	$\rho_{i,}$ %	Δρ, %
1	2.920	21.1	+0.10	-	2.920	0.045
3	2.966	21.2		-0.001	2.965	

Description of experiments for LEU-based generation and isolation of ^{99}Mo

Experiment 1

The generation of ^{99}Mo was investigated in experimental channel 8–9 of the VVR-ts nuclear research facility (NRF). Two types of targets were considered for comparison: a LEU target and a HEU target (standard target).

The following schedule was considered for irradiation in the VVR-ts NRF:

- reactor power of 10.36 MW;
- targets are irradiated for 110.90 h.

After the reactor was shut down, the targets were cooled inside the experimental channel for 20 h for the residual heat removal and for short-lived radionuclides to decay. After having been cooled in EC 8–9, the targets were transferred for being processed into a hot cell (a secured compartment in which ^{99}Mo is isolated from ^{235}U fission fragments using remotely controlled manipulators).

Table 3 presents the results of calculating the ^{99}Mo generation for LEU and HEU targets at the reactor shutdown time.

Table 3. Calculation results as of the reactor shutdown time

Calculated yield of ^{99}Mo	LEU targets		HEU targets	
	top	bottom	top	bottom
Number of ^{99}Mo nuclei	$64.89^{-}10^{17}$	$52.25^{-}10^{17}$	$70.56^{-}10^{17}$	$67.89^{-}10^{17}$
Activity of ^{99}Mo, Ci	511.52	411.88	556.22	535.17
Total activity of ^{99}Mo, Ci	923.40		1091.39	

Table 4 presents the radiochemical yield of ^{99}Mo, which is defined as the ratio of the calculated activity and the experimental activity as of the calibration time (41 h after the reactor shutdown).

Table 4. Experimental and calculated yield of ^{99}Mo (bottom target)

Target type	Activity of ^{99}Mo, Ci		Radiochemical yield of ^{99}Mo, %
	Calculation	Experiment	
LEU	267.80	62.40	23.30
HEU (standard reprocessing)	347.96	212.25	61.00

Experiment 2

The LEU and HEU targets were irradiated for two cycles in experimental channel 8–9 (same as in Experiment 1).

VVR-ts irradiation schedule:

1) cycle 1 – reactor power 10.90 MW, irradiation time 115 h.
2) cycle 2 – reactor power 10.76 MW, irradiation time 115 h.

After the reactor was shut down, the targets were cooled inside the experimental channel for more than 20 h for the residual heat removal and for long-lived radionuclides to decay. After having been cooled, the targets were transferred into the hot cell for being processed. The process parameters of the experiment for the LEU targets are presented in Table 5.

Table 6 presents the calculated and experimental activities of ^{99}Mo (as of the calibration time) for the LEU and HEU targets, and the radiochemical yields of the radionuclide are shown for both targets.

Table 5. Process parameters of the experiment

Experiment 2	Reactor operation mode, power (MW) / irradiation time (h)	^{99}Mo obtained, Ci
Cycle 1	10.90 / 115	43,3
Cycle 2	10.76/ 115	

Table 6. Experimental and calculated yield of ^{99}Mo

Target type	Activity of ^{99}Mo, Ci		Radiochemical yield of ^{99}Mo, %
	Calculation	Experiment	
LEU	325.80	43.30	13.30
HEU (standard processing)	418.01	255.00	61.00

Results of the experiments for LEU-based generation and isolation of ^{99}Mo

As the result of testing ^{99}Mo from the LEU-based target, no excessive values of the radionuclide purity (the ratio of gamma impurities to ^{99}Mo with its total activity in a range

of 240 to 407 GBq as of the manufacturing date) have been recorded, but there is a radionuclide pair of ^{95}Zr + ^{95}Nb present in the ^{99}Mo solution. As the result of the ^{95}Zr decay, ^{95}Nb is accumulated ($T_{1/2}$=35 days). As the result of the ^{99m}Tc decay and the ^{95}Nb accumulation, the final spectrum looks as follows (Fig. 2).

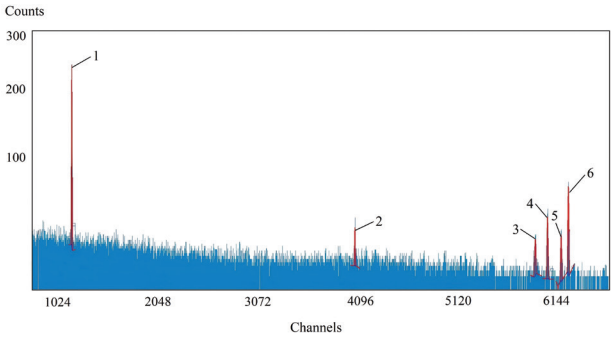

Figure 2. ^{99}Mo solution spectrum: 1 – Tc-^{99}m peak (140.66 keV); 2 – Ru-103 peak (496.96 keV); 3 – Zr-95 peak (723.68 keV); 4 – Mo-99 peak (739.06 keV); 5 – Zr-95 peak (756.22 keV); 6 – Nb-95 peak (765.30 keV).

A number of GT-4K molybdenum-technetium generators (Technetium-99m generator of the GT-4K type) with an activity of 4 to 19 GBq were loaded with the obtained ^{99}Mo. An analysis of eluate has shown a deviation for the 'chemical impurities' parameter, namely an excessive content of manganese. The rest of the parameters are as required by the Manufacturer's Pharmacopoeial Monograph (MPM).

It can be seen from Tables 4, 6 that the yield of the desired ^{99}Mo radionuclides in the two experiments (23.30 and 13.30% respectively) differs greatly from the calculated standard processing values (61%). The radiochemical process is likely to be affected by the presence of a large quantity of ^{238}U, the concentration of the ZnO diluter, and some other factors.

Conclusions

Experimental studies were conducted based on the L.Ya. Karpov Research Institute of Physical Chemistry to investigate the feasibility of ^{99}Mo generation and isolation using a LEU target.

Experiments with a LEU-containing powder target have shown the validity of the neutronic, thermo-hydraulic and strength calculations conducted as part of justifying the safety of the LEU target irradiation in experimental channels of the VVR-ts reactor, including two sequential cycles (no coolant overheating, mechanical changes or target integrity loss).

The possibility has been demonstrated for using the existing ^{99}Mo isolation technology. The radionuclide purity of the molybdate-sodium solution is generally as required by regulatory documentation. The ^{99m}Tc obtained from GT-4K molybdenum-technetium generators largely meets the MPM eluate requirements.

In connection with the fact that there are no currently data on the coefficients of extraction, separation, etc., the effective technology with the existing parameters cannot be conclusively carried over from HEU targets to LEU targets due to multiple uncertainties.

For addressing technological issues, experimental activities are required to define the conditions for the target dissolution (pH of the medium, dissolution time, acid concentration, etc.), and the extraction conditions (ratio of the phase volumes, etc.), that is, full-scale technological development of the process.

There is a need for considering a new design of the target as such, that is, for switching over to an exclusively uranium barrier-enclosed target.

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