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Research Article

Possibility for using a low-enriched target to produce ^{99}Mo in the MAK-2 research channel of the VVR-ts reactor*

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Abstract

Thermal-hydraulic calculations have been conducted with respect to the active part of the MAK-2 loop facility of the VVR-ts research reactor for the ^{99}Mo production. The computational studies were undertaken both for the case of using a highly ^{235}U enriched target and for a low-enriched target. The calculation was performed for the actual technical characteristics of the research channel. The power density for the two simulated cases was obtained in the course of a preliminary neutronic calculation and selected for the most heated channel.

The problem is solved for the steady-state mode of the channel coolant flow and takes into account the dependence of the thermophysical parameters of materials on temperature. The volumetric temperature distribution in the channel was obtained in the process of the calculation.

The calculation results present the maximum temperatures of the target materials for the ^{99}Mo production. An analysis of the obtained results has shown that the maximum temperatures of the aluminum sleeve and the target filling materials do not exceed the critical values. For the analyzed calculation cases, the maximum coolant temperature is localized at a point near the sleeve wall surface and does not reach the boiling temperature for a given pressure. The study has therefore shown that it is possible to reduce the ^{235}U enrichment of the target filling fissile material to 19.7%, provided the average density of the mixture and the amount of ^{235}U in the target remain the same. At the same time, the amount of the medicinally important ^{99}Mo generated will not practically change, which will lead to reduced capital costs for a highly enriched mixture of the target matrix.

Keywords

VVR-ts reactor, MAK-2 research channel, ^{99}Mo production, power density, thermohydraulic calculation, enrichment reduction

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Introduction

Radioisotopes used in nuclear medicine for diagnosis and therapy of cancer are currently in high demand worldwide.

One of such isotopes is ^{99}Mo molybdenum which is a generator of ^{99}Tc technetium. The short-lived isomer of technetium, ^{99m}Tc, is a tracer the movement and accumulation of which in the body can be controlled using tomography of gamma quanta emitted in the course of this nuclide's isomeric transition (Livejournal 2021).

The ^{99m}Tc isotope is extremely sought-after in nuclear medicine for diagnosis of diseases. It is especially suitable for procedures due to being chemically accumulated in ligands and proteins, which concentrate in some of the human organs. Its photons fit ideally the purposes of recording by scintillation detectors (Molybdenum-99).

At the present time, radioisotopes are generated in accelerators and in nuclear research reactors. In Russia, the technetium isotope under consideration is generated predominantly in nuclear research reactors which include dedicated channels for the safe placement of irradiation targets and the isotope production in the reactor core. The efficiency of generating the required isotope depends on many factors, including the efficiency of radiochemical liberation, structural peculiarities of the target, and the target position in the experimental channel.

The paper considers the upgrades to the target for installation in the existing channels (MAK-2) of the VVR-ts reactor facility. The parameters of the facility do not permit target operation modes with coolant boiling in the research reactor and, furthermore, such modes are hazardous for the MAK-2 equipment.

The materials presented in the paper are aimed at analyzing the influence of the initial target material's ^{235}U enrichment on the thermal performance of the VVR-ts reactor's experimental channel.

^{99}MO generation techniques

Initially, two ^{99}MO generation options are considered as the most efficient techniques: those based on fission (generation from ^{235}U-enriched U_3O_8, using the $^{235}U(n, f)^{99}Mo$ reaction), and based on activation (generation by way of ^{99}Mo accumulation, using the $^{98}Mo(n, \gamma)^{99}Mo$ reaction). Both cases require the target irradiation in a nuclear reactor. Each of the above methods has advantages and drawbacks of its own. ^{99}Mo with a high bulk specific activity can be produced from ^{235}U fission products with no or a minor addition of an isotope carrier, which is very important in manufacturing of ^{99m}Tc generators. To be handled, however, fission products require special equipment and a waste disposal solution (Management of Radioactive Waste 1998, Zykov and Kodina 1999, Nuclear Energy Agency 2010). The paper considers the option with generation of ^{99}Mo from uranium fission products.

The yield of ^{99}Mo in a fission reaction is 6.1% (Kolobashkin et al. 1983). The total activity of the irradiated target is 10 times as high as the activity of ^{99}Mo. Apart from handling these isotopes (in gaseous, liquid or solid states), one also needs to clear the ^{99}Mo end product of all such isotopes. Along with clearing ^{99}Mo of fission products, this option requires it to be cleared of alpha-active radionuclides, the requirements to the content of which in ^{99}Mo are highly stringent: the activity of these shall not be more than $1 \cdot 10^{-8}$ to $1 \cdot 10^{-9}$% against the ^{99}Mo activity.

The alpha activity in the irradiated target is defined primarily by the presence of ^{234}U, the daughter isotope of ^{238}U, in initial uranium. The ^{234}U isotope remains in large quantities in enriched ^{235}U as well. The rest of the alpha activity is defined by the accumulation of transuranic elements (TUE) (e.g., ^{239}Pu) in the process of the uranium irradiation in the neutron flux. The accumulation of transuranic elements can be reduced thanks to using enriched ^{235}U (over 90%). However, as noted above, the biggest contributor to the alpha activity of the target is ^{234}U, so using low enriched uranium (LEU) shall be considered impracticable in some cases.

Currently, most of major ^{99}Mo producers use highly enriched uranium (HEU) (90% of ^{235}U). At the same time, many countries do not have HEU reserves and are developing technologies based on using depleted raw materials or alternative technologies (Management of Radioactive Waste 1998, Zykov and Kodina 1999, Nuclear Energy Agency 2010). However, the amounts of HEU used are likely to become smaller due to strengthening the nonproliferation regime. Under the existing agreements, molybdenum shall be produced after 2016 only from less than 20% enriched uranium. This is also the reason for the trend towards the conversion of reactors to low enriched fuel. One of the issues involved in handling of highly ^{235}U-enriched targets is high power density in the target material which leads to the need for heat to be removed from the generator channel.

^{99}MO production in the VVR-ts reactor

The VVR-ts nuclear reactor is the only regionally operating research reactor with a constant neutron flux. An efficient combination of a specific design and the performance of a multipurpose upgraded chemical engineering facility, including an extensive energy spectrum of neutrons with a high flux density, a large number of process channels, advanced process and unique precision measuring systems, and skilled personnel make it possible to undertake investigations in top-priority fields of research, build processing production lines, and develop advanced radiation technologies to obtain new materials, products and medicines (Website of JSC L.Ya. Karpov NRFChI, Scientific and Technical Infrastructure, Chusov et al. 2016).

There are for experimental channels for the ^{99}MO production (Fig. 1) in the VVR-ts reactor based at JSC NRFChI named after L.Ya. Karpov.

The ^{99}MO medical radionuclide is generated in the VVR-ts reactor in a dedicated system, MAK-2, (Kochnov et al. 2019) shown schematically in Fig. 2. The cask unit for the ^{99}Mo production is shown schematically in Fig. 3.

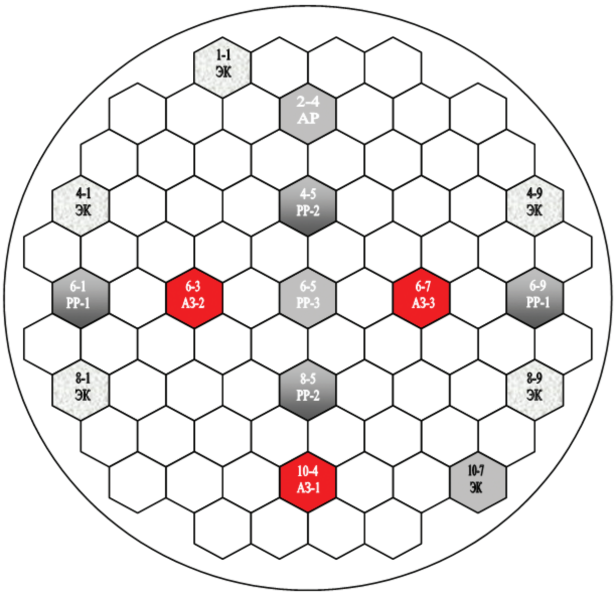

Figure 1. Layout of experimental channels for the ^{99}Mo production (ECs 4-1, 4-9, 8-1, and 8-9) in the VVR-ts reactor core (Website of JSC All-Regional Association Izotop).

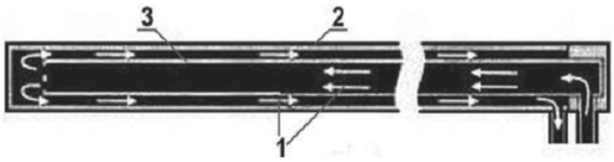

Figure 2. Diagram of the MAK-2 system for the ^{99}Mo generation: 1 – cooling liquid movement direction; 2 – vertical channel with internal tube; 3 – internal tube for container unit installation (90° rotated diagram).

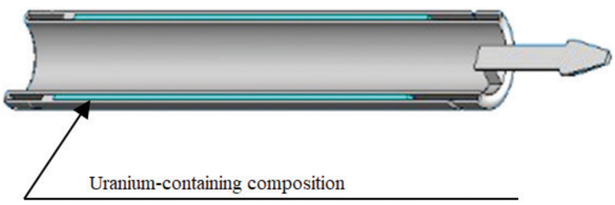

Figure 3. Diagram of a container unit for the ^{99}Mo generation (90° rotated).

The container unit is a metallic double-wall cylindrical target of a tube-in-tube type. The uranium-containing composition is contained between the container unit walls. There is a gap in the upper and lower parts to make up for the swelling of the target's initial material and to discharge gaseous fission products. There is a head attached in the upper part for the container unit to be gripped by the transfer device. The target is made of the CAB-1 aluminum alloy and is sealed at the top and at the bottom.

The target's uranium-containing material is a powder mixture of uranium oxide (U_3O_8) and zinc oxide (ZnO). U_3O_8 (truiranaoctaoxide, uranium oxide concentrate) is an inorganic compound of uranium with oxygen, in which the metal is bivalent. The melting point of this oxide is 1150 °C. In a free state, U_3O_8 is a green-black crystalline material (Boyko et al. 2008).

Analysis of calculation results for container units with a load of different ^{235}U enrichment

In 2012, S.V. Kirienko, Director General of Rosatom State Corporation, approved the program to convert civilian nuclear research reactors and targets for the ^{99}Mo generation from HEU to LEU.

The purpose of the program is to reduce the risk of nuclear material proliferation for building nuclear weapons thanks to minimizing and, in a longer term, excluding the use of HEU in research reactors and targets for the ^{99}Mo generation. At the same time

- the reactor power shall remain the same;
- the neutron flux densities in experimental devices may be just insignificantly smaller;
- the extent of the neutron flux density reduction shall be agreed on with the operators;
- the FA geometries and dimensions shall be the same (the fuel rod dimensions shall be preferably the same as well).

There were two options considered for calculating the ^{99}Mo generation channel's thermohydraulic parameters with a standard (effective) target used:

– a target with a highly (90%) ^{235}U enriched mixture;
– a target with a low (19.7%) ^{235}U enriched mixture.

Fig. 4 shows the layout of the targets in a test channel.

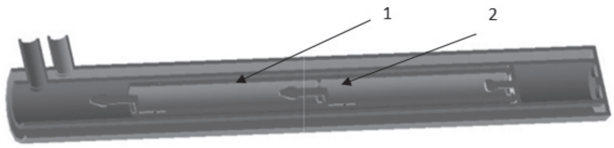

Figure 4. Diagram of the MAK-2 system with installed container units: 1 – upper target; 2 – lower target (90° rotated diagram).

Comparative calculations were undertaken using specialized Monte-Carlo codes to compare the neutronic performance and power densities for molybdenum generation targets made of highly enriched and low enriched fuel (respectively 90% of ^{235}U and 19.7% of ^{235}U) (Kochnov et al. 2012, Pakholik et al. 2021).

The calculations used the condition that the average density of the U_3O_8 and ZnO mixture and the quantity of ^{235}U would be preserved in both calculated cases, the generation of ^{99}Mo being practically the same.

The calculated power density in the targets (for a standard target) was

- $8.27 \cdot 10^8$ W/m^3 for the lower target;
- $8.75 \cdot 10^8$ W/m^3 for the upper target.

The calculated power density for the low enrichment case was

- $8.55 \cdot 10^8$ W/m^3 for the lower target;
- $8.6 \cdot 10_8$ W/m^3 for the upper target.

The thermohydraulic calculations for the two target enrichment options were undertaken using the ANSYS CFX code for the hottest test channel (TC). The calculation was performed for the steady coolant flow in the MAK-2 TC. The preset boundary conditions complied with the rated data for the equipment in the TC circuit (Kochnov et al. 2019). Dependences of the physical parameters of materials on temperature were taken into account (Chirkin 1967, Kirillov and Bogoslovskaya 2000, Alikulov et al. 2014, Volkovich and Smirnov 2014).

Data used for the calculations.

- Aluminum alloy
  - thermal conductivity, 140 W/(m·K);
  - density, 2680 kg/m^3.
- Fissile material powder
  - thermal conductivity, 10.2 W/(m·K);
  - density, 4000 kg/m^3.
- Coolant (water)
  - flow rate, 0.82 kg/s;
  - inlet temperature, 57 °C;
  - static pressure, 171325 Pa (Kochnov et al. 2019).

Table 1 presents the thermohydraulic calculation results for the two target matrix enrichment options.

The errors in the results of the ANSYS CFX thermohydraulic calculations using a k-ε model do not exceed 5% as compared with calculations using analytical formulas and different analytical grid structures (Akhmedzyanov and Kishalov 2009). The computational grid studies are reported in detail in (Akhmedzyanov and Kishalov 2009, Sobolev et al. 2017).

Fig. 5 presents the results of the coolant temperature calculations for the target matrix high ^{235}U enrichment (90%) case.

Fig. 6 presents the results of the coolant temperature calculations for the target matrix low ^{235}U enrichment (19.7%) case.

Table 1. Thermohydraulic calculation results for two target matrix ^{235}U enrichment options

	Target with high U-235 enrichment (90%)	**Target with low U-235 enrichment (19.7%)**
Maximum coolant temperature, °C	96.6	97.3
Maximum sleeve aluminum alloy temperature, °C	228.3	234.1
Target uranium-containing material temperature, °C	316.1	335.8
Average coolant outlet temperature, °C	71.75	71.85
Bulk power density, W/m^3	For lower target $8.27 \cdot 10^8$, For upper target $8.75 \cdot 10^8$	For lower target $8.55 \cdot 10^8$, For upper target $8.62 \cdot 10^8$

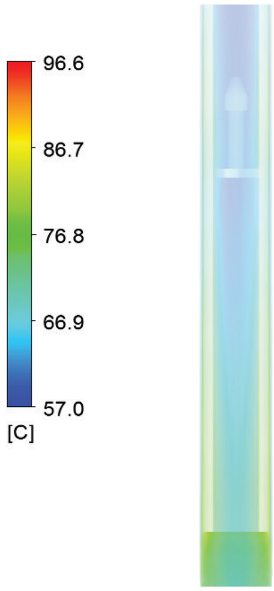

Figure 5. Temperature distribution through the coolant volume in the channel, °C (90° rotated).

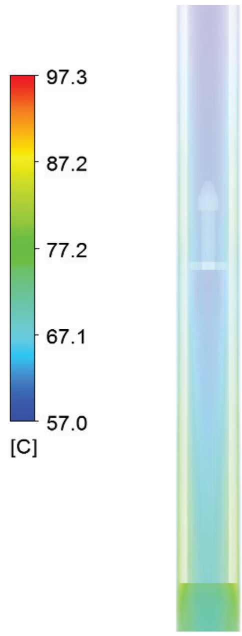

Figure 6. Temperature distribution through the coolant volume in the channel, °C (90° rotated).

As can be seen from the temperature calculation results, the temperature in the channel grows slightly due to neutronic processes in the course of the overall power density increase with a smaller ^{235}U enrichment of the target's uranium-containing mixture. The maximum temperature of the uranium-containing mixture for the low and high ^{235}U enrichment cases does not exceed the critical values for the mixture components.

Provided there is static pressure in the ^{99}Mo generation channel, the maximum value of which is over 168 kPa, the water boiling temperature with such a pressure is approximately 115 °C, which indicates that there is a safety margin to developed boiling (Vargaftik 1972, Termalinfo 2021).

Conclusion

Results were obtained in the course of the study in the form of the temperature field distribution through the liquid volume. The calculations were undertaken for two targets:

- with a high (90%) ^{235}U enrichment;
- with a low (19.7%) ^{235}U enrichment.

As can be seen from the temperature calculation results (Table 1), the temperature in the channel grows insignificantly due to neutronic processes in the course of the overall power density increase with a smaller target matrix ^{235}U enrichment but does not exceed critical parameters. This indicates that it is possible to reduce the target material ^{235}U enrichment provided the average U_3O_8 and ZnO mixture density and the quantity of ^{235}U in the target remain the same.

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