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Research Article

Increasing the production of the Mo-99 isotope by modernizing the design of targets irradiated in the experimental channels of the VVR-c reactor*

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Abstract

There are various ways to obtain Mo-99. Some of them are widely used in industrial production, others are in the research stage with the aim of increasing the product yield. The main industrial method for obtaining Mo-99 using a nuclear reactor is the fragmentation method. This method provides for the presence of a uranium target and a nuclear reactor. The target is placed in the channel of the reactor core and irradiated with neutrons for the required time. After that, the target is removed from the channel to the "hot" chamber for the chemical separation of Mo-99. This is how Mo-99 is obtained practically all over the world.

The paper considers the fragmentation method for producing Mo-99, which is implemented on the basis of the engineering and technological complex of the VVR-c research nuclear reactor. In order to increase the yield of Mo-99, a modernized model of the "tube-in-tube" target is proposed. The assessment of the production of Mo-99 and the cooling efficiency of the modernized target was carried out. The calculations were performed using the VisualBurnOut and Ansys CFX software packages. Computational studies have shown an increase in the energy release and the amount of the produced Mo-99 isotope in the target of the modernized design. In the most stressed zones, the target wall temperature exceeds the water saturation temperature. Surface boiling occurs in these zones. As a result, turbulization and mixing of the near-wall boundary water layer increases. This improves heat dissipation.

Keywords

Molybdenum-99, VVR-c reactor, experimental channel, target for the production of Mo-99

Introduction

The use of radioisotope products for diagnosis and treatment of oncological diseases is an important part of modern medicine. The demand for radiochemical preparations grows steadily as production of medical radionuclides, such as the Tc-99m isomer, also grows (The Supply of Medical Radioisotopes 2014). The half-life of Tc-99m is about six hours which makes it harder to supply to medical centers. To address the problem of the Tc-99m isomer delivery to the end user, the so-called technetium-99m generators are used in the form of lead

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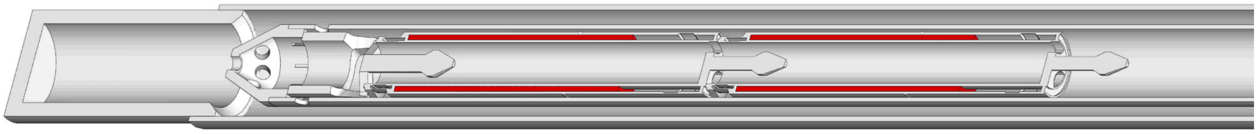

Figure 1. An experimental channel with targets (vertical cut, 90° turn).

containers with columns with Mo-99 inside, which forms the Tc-99m isomer while decaying with a half-life of 66 hours. Uninterruptible generation of Mo-99 is key to production of Tc-99m generators.

Essential to the Mo-99 production using a nuclear reactor is to improve the existing and develop new technologies for producing radionuclides. In this respect, the study is timely as being directly connected with improving the Mo-99 production with the use of the VVR-c research reactor (Chusov et al. 2016) on the basis of JSC L.Ya. Karpov Research and Development Institute for Physical Chemistry.

Production of Mo-99 based on the VVR-c nuclear research reactor

The Mo-99 generation using a nuclear reactor is possible in two ways:

- Mo-99 production as a result of the neutron capture reaction on Mo-98;
- Mo-99 production as one of the U-235 fission fragments.

The Mo-99 production using a capture reaction is not considered at the present time on a commercial scale since it does not make it possible to generate large volumes of the product (the target material cannot be isolated from the generated Mo-99 and the specific activity of the product is not high). Currently, the key commercial method for the Mo-99 production is generation of the radionuclide as one of the U-235 fission fragments. To that end, U-235 is irradiated in a nuclear reactor and Mo-99 is then chemically extracted from the fission products. A fission fragment method is used at JSC L.Ya. Karpov Research and Development Institute for Physical Chemistry for the Mo-99 generation using the VVR-c nuclear research reactor.

VVR-c is a pool-type water-cooled water-moderated reactor of the rated power 15 MW and has a two-loop cooling system. There are six vertical experimental channels (VEC) in the reactor core four of which are used for the Mo-99 generation (Kolesov et al. 2013, Kochnov et al. 2014).

The vertical experimental channels used for the Mo-99 generation are cooled using two loop facilities each of which is a closed water circuit with forced water circulation. One of the loop facilities includes two vertical experimental channels connected in series (one after the other), two circulation pumps (one main pump and one standby pump), a water tank, pipelines, control and shut-off valves, and instrumentation. The VEC outer surface is cooled by the primary circuit water.

The Mo-99 generation channel is designed as a Field tube and represents two coaxially arranged tubes (Fig. 1). The water moves downward in the central tube, cools the targets, and then flows up the tube side to leave the channel.

Targets (irradiated samples containing U-235) of two types are used for in-pile irradiation (Fig. 2).

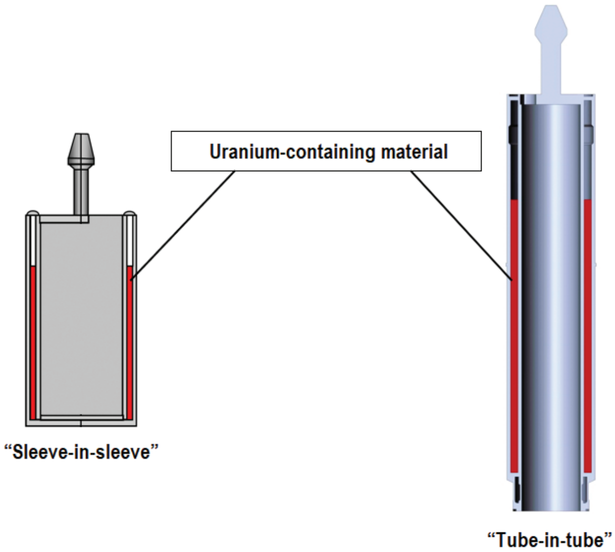

Figure 2. Targets used for the Mo-99 generation in the VVR-c reactor (vertical cut).

The target of type 1 (“sleeve-in-sleeve”) is a structure in the form of sleeves, one inserted into the other. The gap between the sleeves is filled with uranium-containing material (U_3O_8+ZnO). The total height of the target with a head for being grabbed by the transfer mechanism is 140 mm. The target body is made of SAV-1 aluminum alloy. Up to four targets of such design can be irradiated in one channel at a time.

The other target is of the “tube-in-tube” type. The space between two coaxially arranged tubes is filled with uranium-containing material (U_3O_8+ZnO). The tubes are made of SAV-1 alloy. The total height of such target with a head for being grabbed by the transfer mechanism is equal to 250 mm. The target has two-side cooling. Not more than two targets of the type can be inserted into one channel.

Computational study

The purpose of the computational study is to upgrade the design of the “tube-in-tube” target to make it possible to increase the generation of Mo-99 under standard conditions of irradiation while avoiding the VVR-c reactor safe operating limits to be violated.

The major requirements to the target design are as follows (Kochnov et al. 2012):

- the target design shall provide for the best possible use of the neutron flux in the experimental channels of the VVR-c reactor;
- the target shall have such dimensions and design as allow it to be loaded into and unloaded from the VVR-c reactor channel;
- the target shall provide a barrier to prevent the release of radioactive products and, specifically, gases in the course of and after the irradiation;
- the target shall be designed such that to be easy to dismantle in the hot chamber conditions (a hardened box in which Mo-99 is isolated from the U-235 fission fragments using remote manipulators);
- the target design and composition shall allow it to be chemically treated in a hot chamber with the greatest possible product yield for the shortest possible time.

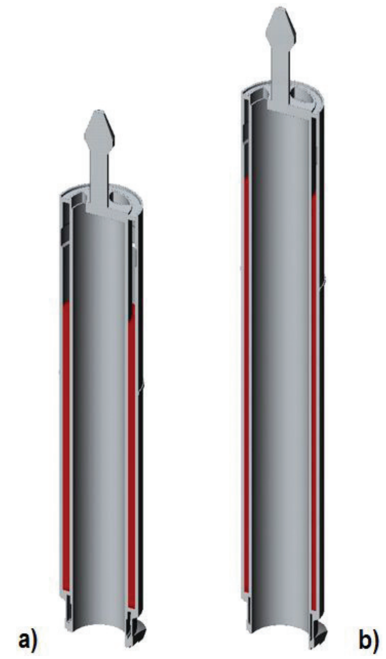

Figure 3. Comparison of targets: a) currently in use; b) upgraded.

An upgraded target has been developed with regard for the above requirements. The target upgrade differs from the targets of the "tube-in-tube" type currently used in the VVR-c reactor in the useful height (increased by 35%), the internal diameter of the cylindrical body (increased by 10%), the size of the space for the uranium-containing material (the space width reduced by 30%) and the total height (increased by 20%). Meanwhile, the volume and the uranium-containing mixture material remain the same (only the target body geometry is changed). It is possible to install not more than two upgraded targets into one channel at a time.

The target upgrade makes it possible to use more advantageously the thermal neutron flux in the experimental channel and provides for a more efficient heat removal, than the current target design, thanks to an increased heat exchange surface. More Mo-99 is accumulated due to the best possible use of the air gap offered by the target for the accumulation of gaseous fission fragments, as well as thanks to the improved use of the neutron flux distribution along the experimental channel's height and a reduced layer of the uranium-containing material (a smaller blocking effect). For comparison, Fig. 3 shows in a similar scale a vertical cut of the target currently in use and the target upgrade.

A precision mathematical model of the VVR-c reactor core was built for the neutronic calculation using the coupled MCNP and VisualBurnOut codes (see (Kamayev et al. 2006, VisualBurnOut 2009) for the code description). The calculations using the coupled codes show a good agreement of the neutron flux densities in the VVR-c reactor core and the change in the reactivity margin in the course of the lifetime with the file data from JSC L.Ya. Karpov Research and Development Institute for Physical Chemistry. The model was used to calculate the power density in the targets and to estimate the Mo-99 generation for the targets currently in use and the upgraded targets of the "tube-in-tube" type for all four vertical experimental channels. Specific to the computational model is that it simulates a structure the upper part of which includes fastening components which cause an angular asymmetry in the liquid flow. This is exactly what necessitates the use of a more complex model which is calculated using the ANSYS CFX code.

The following reactor operating mode was considered as part of the computational studies:

- the targets are irradiated for 120 h (reactor lifetime);
- the reactor power does not change throughout the reactor life and is 15 MW.

The Mo-99 power density and activity calculation results for VEC 8-1 (the largest generated volume of all VECs) are shown in Table 1. The total activity of Mo-99 for all VECs is presented in Table 2. The computational studies show an increase in the power density and the quantity of the generated Mo-99 isotope in the upgraded

Table 1. Mo-99 power density and activity calculated for VEC 8-1

Target type	Power density, kW		Mo-99 activity, Ci		Mo-99 total activity, Ci
	upper	lower	upper	lower	
Target in use	25.47	21.13	993.96	826.06	1820.02
Upgraded target	26.07	25.85	1165.95	1155.93	2321.88

Table 2. Mo-99 activity calculation results for all VECs

Target type	Mo-99 total activity for all VECs, Ci	Mo-99 activity increase, %
Target in use	6711.88	21.41
Upgraded target	8540.20	

target as compared with the targets currently in use. The total activity of Mo-99 for all VECs will be 21.41% as high.

A thermohydraulic calculation, namely numerical simulation of the circulation water flow through a vertical experimental channel of the VVR-c reactor, in which upgraded targets were installed, was undertaken in the ANSYS CFX code. A k-ε turbulence model was used in the calculations (Korkodinov 2013). The code and turbulent model selection is based on multiple calculations (Akhmedzyanov 2009, Kochnov et al. 2014, Kochnov et al. 2019) and a comparison with experimental data. The power density in the targets was defined on the basis of a neutronic calculation. Vertical experimental channel 8-1 was considered in the calculation due to being the highest energy channel.

The values of the circulation water velocity, temperature and pressure components, and the target wall and uranium-containing mixture matrix temperature were obtained as a result of the calculation (Figs 4 through 7).

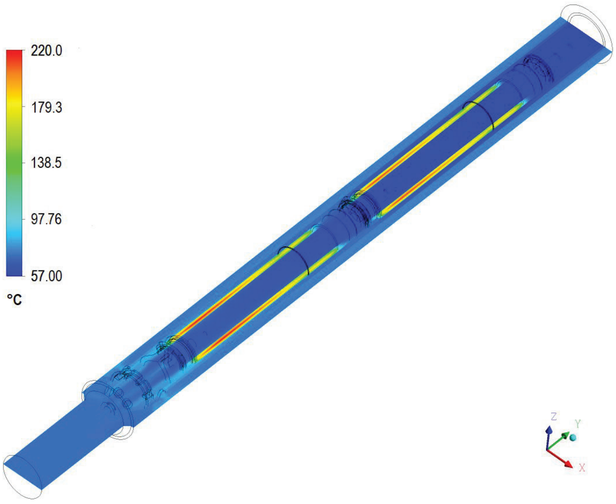

Figure 4. Temperature distribution in the vertical cross-section of the experimental channel.

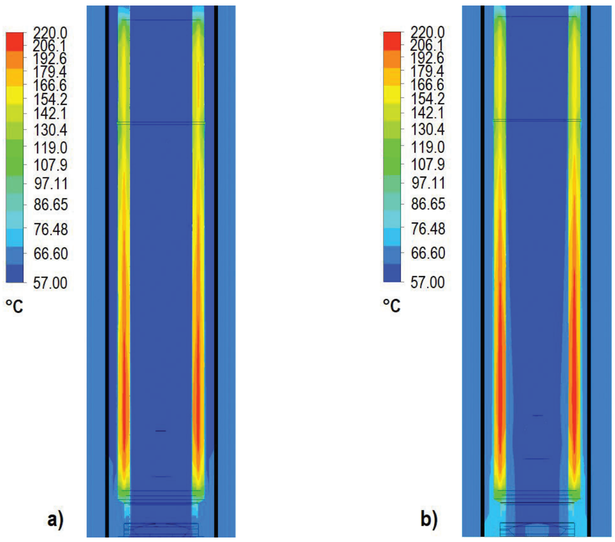

Figure 5. Temperature distribution in the target area: a) upper target; b) lower target.

Fig. 4 shows the temperature distribution in the vertical cross-section of the VVR-c experimental channel with upgraded targets, and Fig. 5 shows the temperature distribution in the target area. It can be seen from the figures that the maximum temperature of the uranium-containing mixture is 220.02 °C. The maximum value of the target body temperature does not exceed 204.95 °C (lower target) which is much below the SAV-1 aluminum alloy melting temperature (T_{melt} ≈ 650 °C). The maximum temperature of the circulation water at the computational region exit is 72.85 °C.

Figs 6 and 7 show the circulation water pressure and velocity distribution in the vertical cross-section of the experimental channel.

It can be concluded from Fig. 6 that the minimum value of the water pressure in the target area is 1.689 kPa which corresponds to the boiling temperature T_s ≈ 115 °C (Rivkin and Aleksandrov 1980).

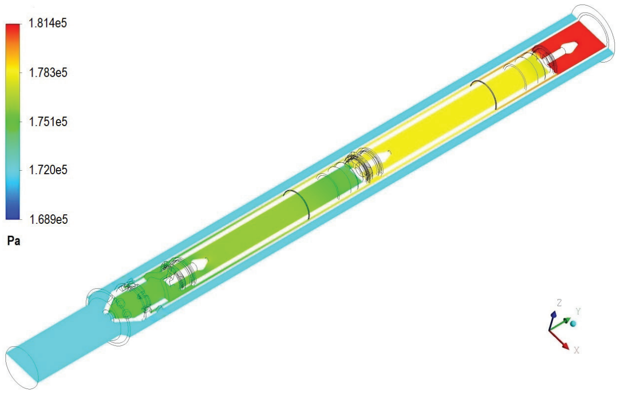

Figure 6. Circulation water pressure distribution in the channel's vertical cross-section.

It can be seen from Fig. 7 that the highest velocity value (3.12 m/s) is achieved in the region of the target head attachment to the target body. This area has a flow of three jets, this affecting the circulation water pressure distribution along the channel radius. The flow velocity has the maximum value and the temperature has the minimum value in the jet movement area.

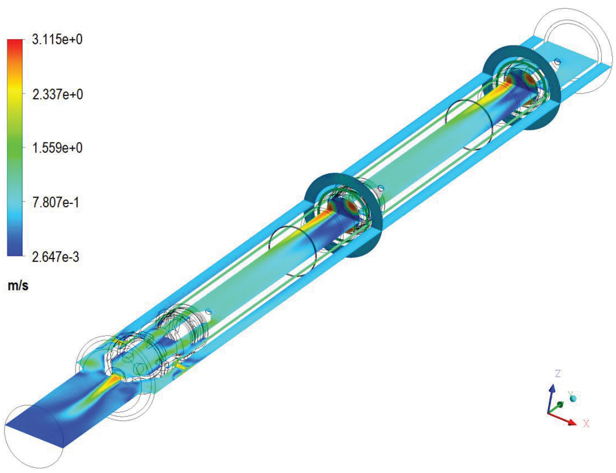

Figure 7. Circulation water velocity distribution in the channel's vertical cross-section.

It needs to be said that there are areas where the target body temperature at the boundary with the circulation water exceeds the water saturation temperature in the channel. This indicates that there is local surface boiling in the near-wall layer. However, since the circulation water flow core temperature is much lower than the channel saturation temperature ($T_s \approx 115$ °C), boiling will be accompanied by the rapid condensation of steam bubbles and will not lead to developed boiling (Kutateladze and Borishansky 1958, Kirillov et al. 1990). This is also confirmed by the maximum water temperature value at the computational region exit (72.85 °C). This type of boiling is called subcooled boiling (Mikheyev and Mikheyeva 1977).

Conclusion

An upgraded target has been developed which fully satisfies the requirements for the Mo-99 production based on the engineering process system of the VVR-c reactor. The target upgrade design makes it possible to integrate the target into the production cycle with no extra manufacturer costs. The upgraded target uses more efficiently the peculiarities of the neutron flux distribution in the reactor's experimental channel thanks to the geometrical dimensions changed as compared with the standard target.

The quantity of radioactive waste grows in proportion to the target radionuclide increase as the Mo-99 generation is increased. This is permissible and there is an extra process capacity provided to that end. The major goal is achieved: it becomes possible to produce an additional quantity of Mo-99, in the event of an increased customer demand, with a limited maximum number of targets (eight).

Computational studies show a 21.41% increase in the quantity of the generated Mo-99 isotope in the upgraded target.

The target body temperature exceeds the water saturation temperature in the highest loaded areas. There is surface boiling taking place in these areas. This leads to increased turbulization and mixing of the near-wall boundary water layer which contributes to an improved heat transfer (Mikheyev and Mikheyeva 1977).

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