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Development of Low Enriched Uranium Targets for Mo-99 Production

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ABSTRACT

JSC "SSC RIAR" conducts the experiments to promote conversion of existing highly enriched uranium-based technologies. These technologies provide for using highly enriched uranium as nuclear fuel in research reactors as well as for Mo-99 production. The present paper presents the results of experiments related to the development of low enriched uranium-based targets with the focus on new design modifications (flat and tubular types). Presented here are the results of neutronic and thermal-hydraulic calculations. The present paper gives an overview of possible target design optimization ways aimed at increasing the Mo-99 production yield. Given here are also the results of comparative analysis of thermal and physical characteristics of targets. This work was done in cooperation with the National Research Nuclear University Moscow Engineering and Physics Institute with funding from the Ministry of Science and Education of the Russian Federation within the framework grant # 02.G25.31.0069.

Introduction

The primary process of Mo-99 production is its separation from fission products of uranium-235. It is the mostly frequently used method that makes it possible to produce over 95% of the global molybdenum production and almost all the molybdenum produced nowadays is produced using highly enriched uranium (HEU). Nowadays the international program "Global Threat Reduction Initiative (GTRI)" has been implemented under the guidance of International Atomic Energy Agency aimed at reducing the risk of unauthorized use of nuclear materials and of nuclear terrorism.

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. 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.

Generation and production of Mo-99

Mo-99 is accumulated in pool-type research reactors RBT-6 and RBT-10 of thermal output of 6MW and 10MW, respectively. [2]. The reason for choosing these reactors is that they have appropriate neutronic parameters, immediately available irradiation positions and suitable reactor operation schedule that provides for long-term operation without outages.

The RBT-type reactors are water-cooled water-moderated pool-type thermal reactors. They are 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. They are spaced for 78 mm in the RBT cores.

The RBT-6 core is comprised of fifty six (56) spent fuel assemblies operated in the SM reactor [3] up to a burnup of 10-30% (but no higher than 47 %), six combined safety-shim rods, one automatic regulating rod and eight experimental channels (Fig. 1). There is one materials test rig (MTR) “KORPUS” in the reflector besides the upright channels. The materials test rig is intended for irradiation of VVER-440 and VVER-1000 vessel steels under the conditions simulating the operating conditions of vessel components in a very wide range as to the neutron density and neutron spectrum, irradiation temperature, gradients as well as simulating changes of all these parameters under operating conditions. A nominal power of the RBT-6 reactor is 6 MW.

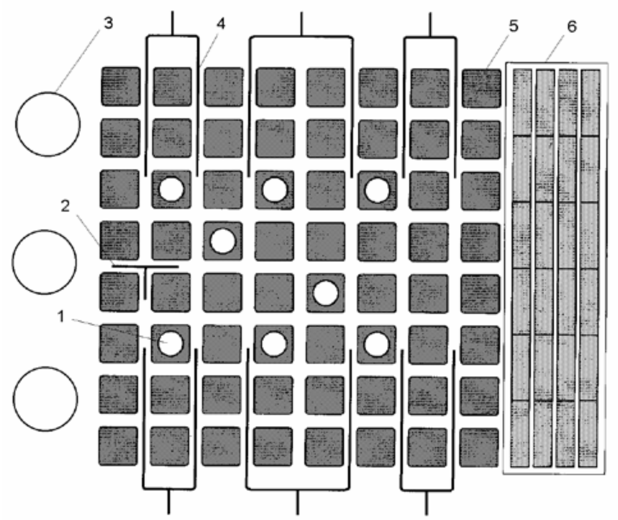

Figure 1. RBT-6 core layout.

1 – test channels; 2 – automatic control rod; 3 –reflector test channels; 4 - shim rod; 5 – operating fuel assembly; 6 – materials test rig “KORPUS”.

The RBT-10/2 reactor core is comprised of seventy eight (78) spent fuel assemblies operated in the SM reactor 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 2). Twelve beryllium blocks located at the core corners are also employed as a moderator other than water. A nominal power of the RBT-10/2 is 10 MW.

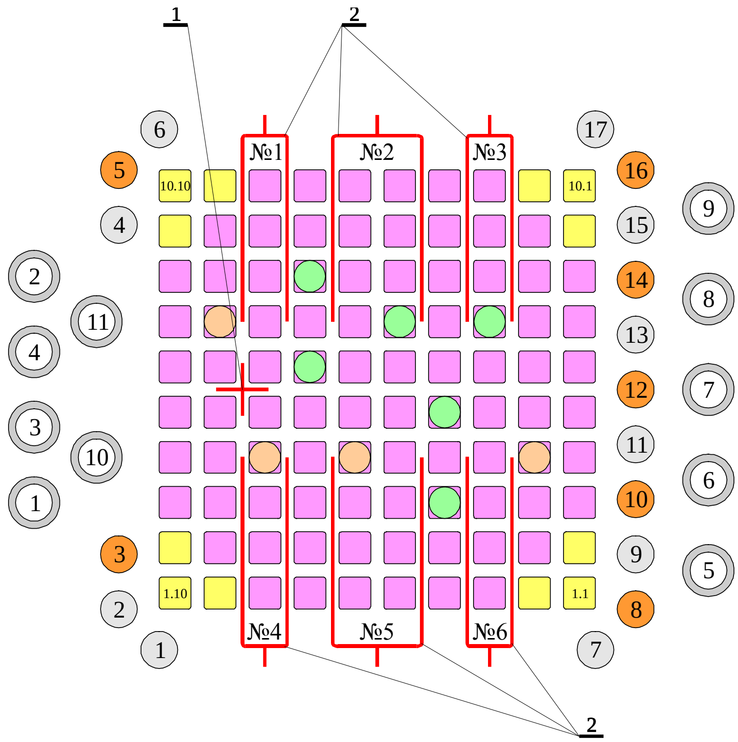

Figure 2. RBT-10 core layout.
1 – automatic regulating rod;
2 – combined safety-shim rods.

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 contained within aluminium matrix;
- Designed capacity of the molybdenum production facility is 300 Ci at the most that needs to be calibrated on the 6^{th} day that corresponds to activity of 4150 Ci as of the end of irradiation;
- The limiting high temperature of uranium intermetallic fuel under irradiation is 500 ^{0}C.

Conversion to low enriched uranium-based production of Mo-99

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.

The principal countervailing measure aimed at reducing the losses of process performance is to increase the total mass of uranium in the targets in order to increase the mass of uranium-235 and/or to increase the number of targets to be irradiated all at the same time. A larger number of targets will impose an additional increase of the target-making costs as well as it will lead to an increasing demand for irradiation capabilities, i.e. appropriate irradiation positions that will increase the cost of irradiation. As the irradiation capabilities are limited, this approach cannot be fully effective.

The best way to handle the above-identified challenging tasks is to obtain higher density of fuel composition in the target by increasing uranium alongside with the increase of the fuel meat volume. Two radically new design modifications of the LEU-based targets were developed in order to implement this approach. 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 analyze their characteristics and operating parameters.

FLAT PLATE TARGET

The target design represents itself a plate comprising three layers. The inside layer contains a LEU-based fissile material (UAl_3, UAl_4 or their mixture), two outside layers serve as the cladding (shell) that is made from aluminium. A calculated value of fuel meat density is 5.7 g/cm^3 given that uranium weight content amounts to 0.702. The target is 200 mm long where the target meat is 190 mm long.

The irradiation rig (IR) represents itself a skeleton-type structure (Fig. 3) that comprises an end cap, frame, supporting ring and pulling fitting for a gripper. The targets to be irradiated are inserted parallel to each other in special slotted holes in order to prevent their displacement during irradiation. They are arranged so that to have a certain gap.

The following parameters were estimated in order to optimize specifications of the targets and irradiation rig:

- The number of targets in the irradiation rig;
- Gap size between the targets;
- Fuel meat thickness;
- Cladding thickness.

The experiments were conducted with the focus on the uranium content limitations in the targets to be loaded in the irradiation rig (200 g) and on the minimum cladding thickness of 0.25 mm (fabrication process specification). The IR power output (Mo-99 generation yielding power).

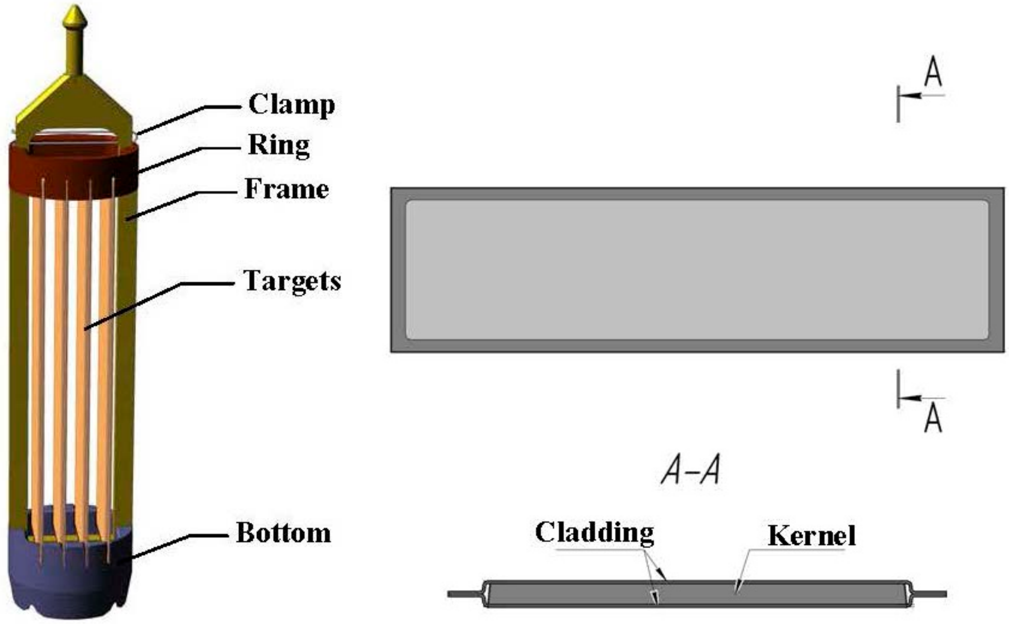

Figure 3. IR design and flat plate target.

First it was necessary to determine the number of targets as may be required to load into the irradiation rig. According to the results of thermal- hydraulic and neutronic calculations, the limiting high temperature (Fig. 4) does not exceed 175°C for the target meat and 145°C for the cladding provided that the number of targets is more than three. In so doing, the DNB value is no less than 6.

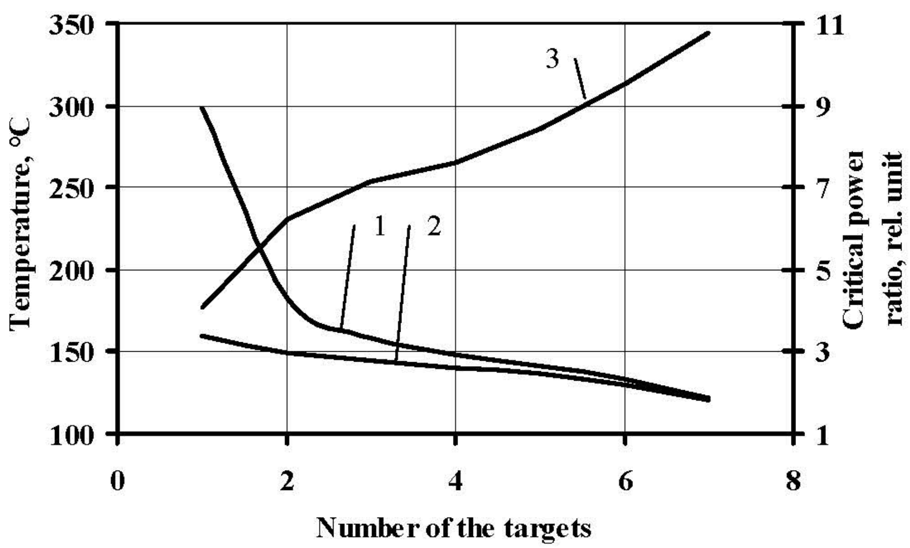

Figure 4. The limiting high temperature values of target meat (1), cladding (2) and DNB values (3) in relation to the number of targets in the irradiation rig (the cladding is 0.25 mm thick; the total aluminium mass in the targets is 200 g).

Variations in the IR power output (thermal heat rate of fuel) were also studied in relation to the number of targets (Fig. 5). As it is shown in the figure, the curve has a flat profile of peak power around 75 kW attributable to uranium enrichment of 20% when the target number amount to 3-4 pieces. This power value makes for activity of generated Mo-99 of 3100 Ci as of the date of its unloading from the reactor Mo-99 that is approximately equivalent to the activity of the product generated with the use of HEU-based targets so far. By way of example, shown in the figure is the IR power output as to fuel enrichment of 70% and 90% that tends to level-off when the number of targets is from 4 to 5 pcs. Later on the irradiation rig loaded with 3-4 targets can be taken as an optimum configuration with regard to irradiation in the RBT-reactors.

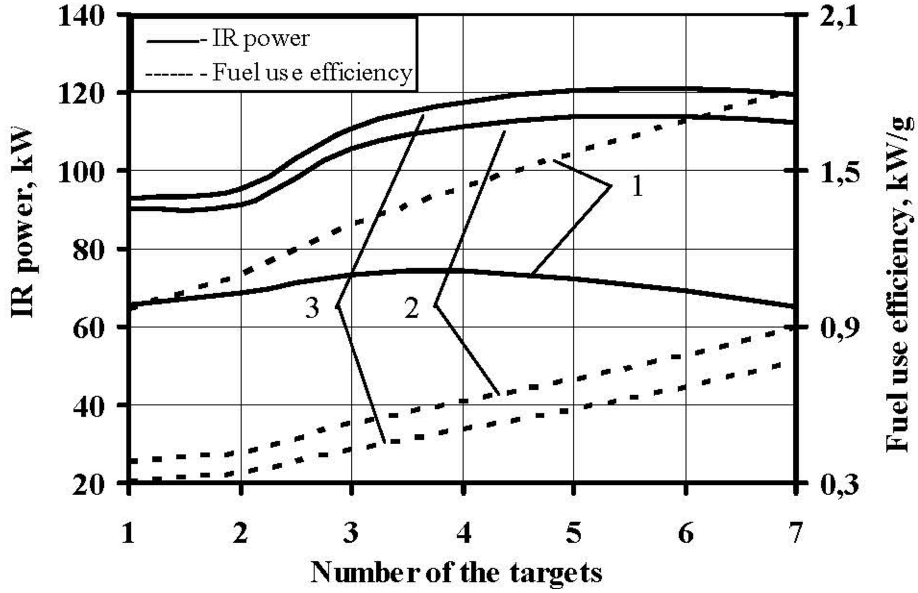

Figure 5. IR power and efficiency of fuel utilizationwith reference to the number of targets:
1 – enrichment of 20%, 2 – enrichment of 70%, 3 – enrichment of 90%
(the cladding is 0.25 mm thick; the total aluminium mass in the targets is 200 g).

Figure 6 demonstrates changes in the IR power output implying that it is loaded with three targets and the cladding becomes thicker. As the following graphs show the IR power tends to decrease by 10% at the most compared to its peak value when the cladding becomes 0.5 mm thick.

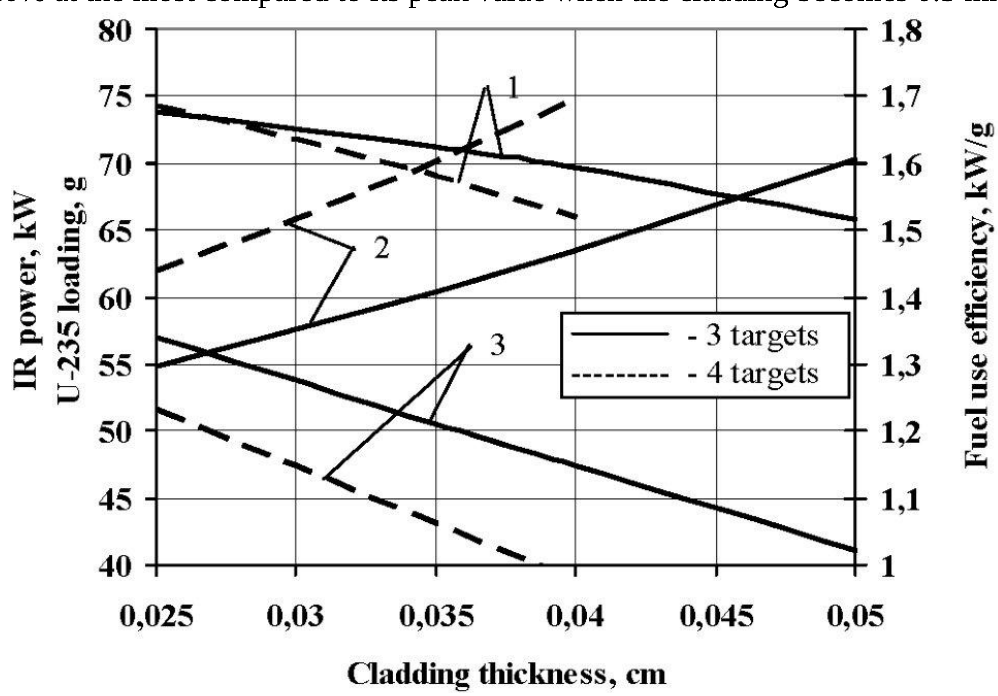

Figure 6. Power output (1), efficiency of fuel utilization (2) and U-235 loading (3) in relation to the cladding thickness (continuous line implies 3 targets; dotted line implies 4 targets; the total aluminium mass in the targets is 200 g).

According to the calculation data analysis:

- Mo-99 accumulation yield increases when the targets are provided with a thin cladding (on the point of fabrication limit value);
- The heating performance can be maintained with the optimum number of targets in the irradiation rig that is 3-4 pieces.

Tube-type target

The second design of the target under development is the tube –type target (Fig. 7). It represents itself two aluminium tubes, which are in axial alignment. There is the LEU-based fuel meat with the bulk of uranium intermetallic powder between the tubes. The displacer is placed inside to create the optimal thermal and hydraulic parameters.

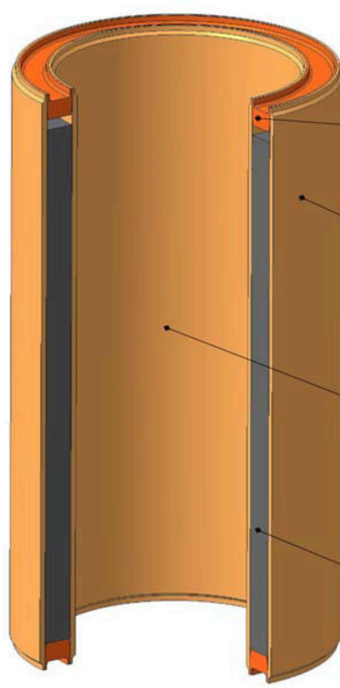

Figure 7. Tube-type target.

Trial calculations of neutronic and thermal-hydraulic parameters were performed with reference to the IR loading with the targets of the tube-in-tube design in order estimate its operating conditions and parameters of the targets to generate Mo-99 assuming uranium enrichment reduction down to 20% U-235. The numerical estimation indicate that the reactor rig loaded with the tube targets in large numbers with reference to uranium enrichment of 20% has a few points in its favor as to the power and fuel heat rate compared with the single-target reactor rig but its disadvantage is a large number of targets and amount of fuel. The present paper focuses on the irradiation rig incorporating one composite tube-type target with the bulk of uranium intermetallic fuel meat enriched up to 20% U-235. To be specific, alloy UAL_4 with a density of 5.7 g/cm^3 and a weight content of uranium of 70% is discussed here.

The use of bulk-type fuel meat has a significant effect on the target performance due to the following reasons:

- Bulky fuel meat makes it possible to use only uranium intermetallic powder without cladding for radiochemical processing that is a “dead weight” due to limitations on the uranium mass in the material to be dissolved or the powder wrapped in a thin aluminum foil during the target assembling. In doing so, the content of fissile material can be increased while preserving to the extent possible the total mass of alumonium in the material to be processed and thus increasing the target capacity and maintaining an appropriate heat pickup;
- Bulky fuel meat makes it possible to remove limitations on the target length (200 mm) that depends on the height of dissolver apparatus and thus the length of target meat can be extended to be equal to height of the reactor core therefore the useful volume of the rig can be occupied more efficiently.

The target incorporating the displacer is expected to be loaded in a cylindrical test channel. In this regard, the trial calculations were focused on optimizing the following design specifications:

- The gap between the cladding and test channel;
- The target-to-displacer gap;
- Fuel meat thickness;
- Fuel meat length.

The choice of the gap value was primarily determined by the heat pickup conditions. The reduction of gap will lead to reduction of DNB. But on the other hand, the increase of the gap size and thus the target meat thickness will lead to fuel temperature increase (Fig. 8). Such an increase manifests itself to a greater extent when the fuel meat porosity is high (that is at lower heat-transfer capacity).

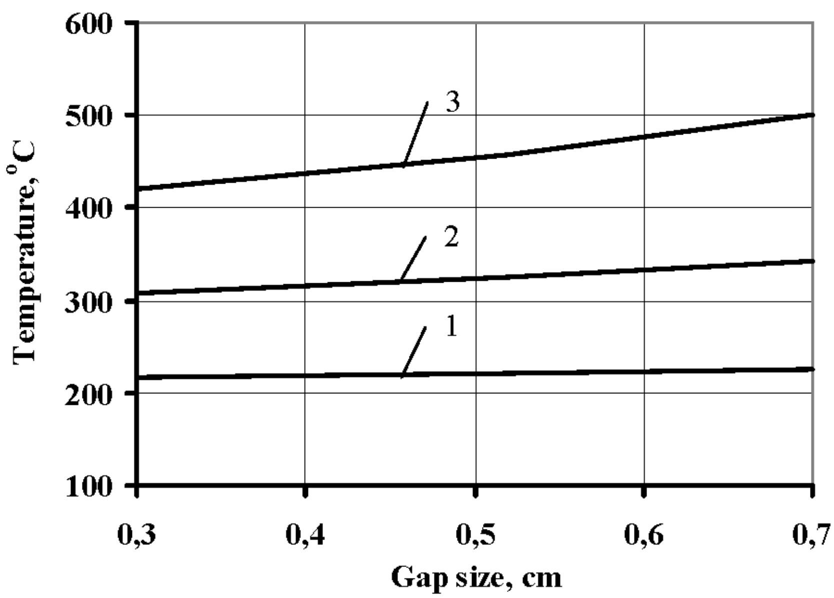

Figure 8. The peak temperature of the target meat in relation to the gap value as to the fuel volume concentration: 1 – 1.0; 2 – 0.6; 3 – 0.5.

When the gap is 5 mm, the peak temperature value of fuel meat does not exceed 450 ^{0}C and the DNB value (Fig. 9) is less than 4 for any porosity of fuel meat and for a length of 190 mm.

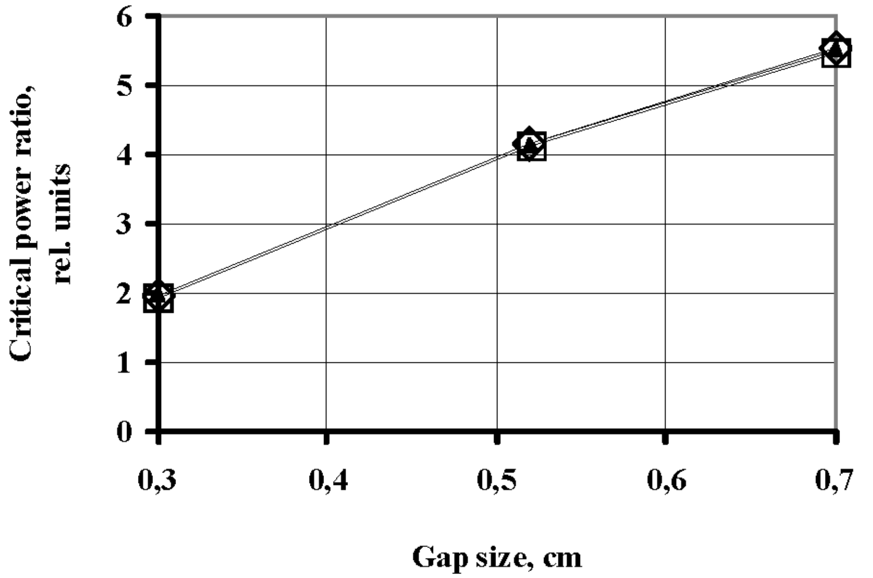

Figure 9. Departure from the nucleate boiling in relation to the gap value (the target meat is 190 mm long).

Due to limitations on the amount of aluminium in the target, the target meat thickness decreases once its length is increased. Thus the fuel heat generation rate and the target power output increase due to reduction of fuel blocking. Shown in Fig. 10 is the target power capacity in relation to the fuel meat length. As shown in the figure, the target power capacity is 110 kW and 114 kW irrespective of the fuel meat porosity as with the length of fuel meat of 300 mm and 350 mm, respectively. This power value makes for activity of generated Mo-99 of 4000 Ci as of the date of its unloading from the reactor after a 6-day irradiation period.

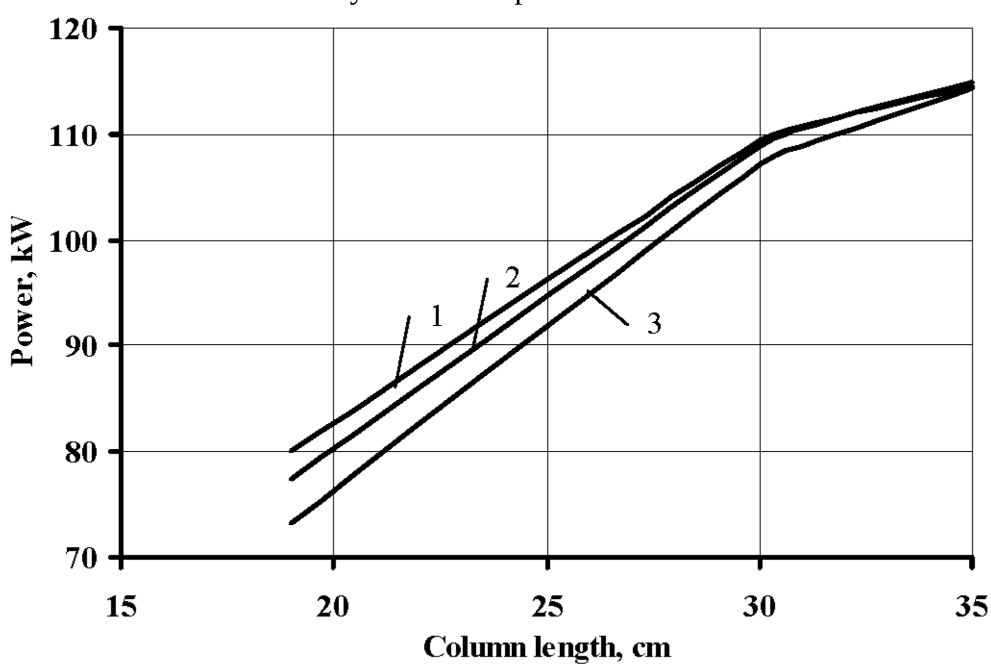

Figure 10. IR power output in relation to the length of target meat as to the fuel volume concentration: 1 – 0.7; 2 – 0.6; 3 – 0.5.

The departure from nucleate boiling ratio (DNB) was calculated in order to estimate thermal and physical reliability and safe operation limits (Fig .11). When the target meat is from 300 to 350 mm long, the peak temperature of the cladding meat does not exceed 320°C (Fig. 12) with any density of the fuel meat and the departure from nucleate boiling ratio is no less than 4.

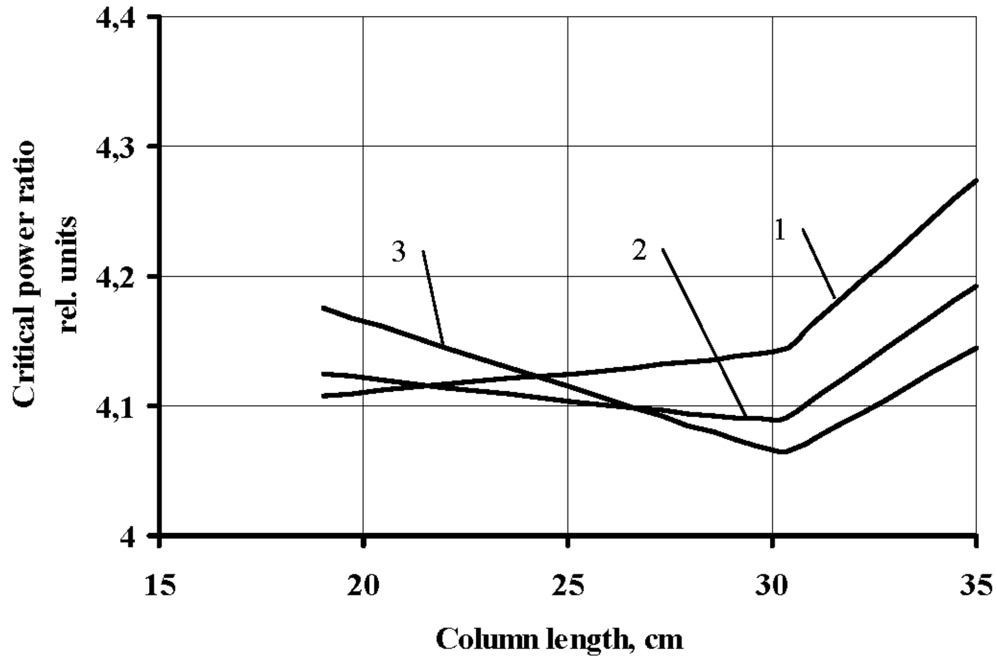

Figure 11. Departure from nucleate boiling in relation to the target meat length as to the fuel volume concentration: 1 – 0.7; 2 – 0.6; 3 – 0.5.

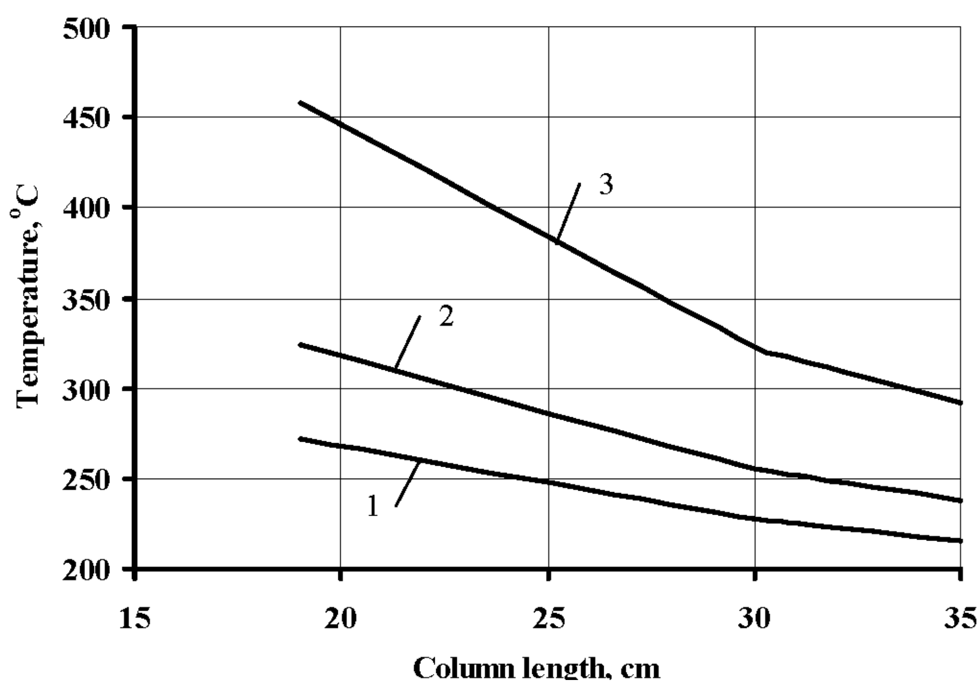

Figure 12. The peak temperature of the target meat in relation to the target meat length as to the fuel volume concentration: 1 – 1.0; 2 – 0.6; 3 – 0.5.

The efficiency of fuel utilization (the target power capacity in relation to the mass of fissile isotope) is no less than 1.1 kW/g-U_5, (Fig. 13) within the given length range that corresponds to a burnup of 1% U-235 after a 7-day irradiation period.

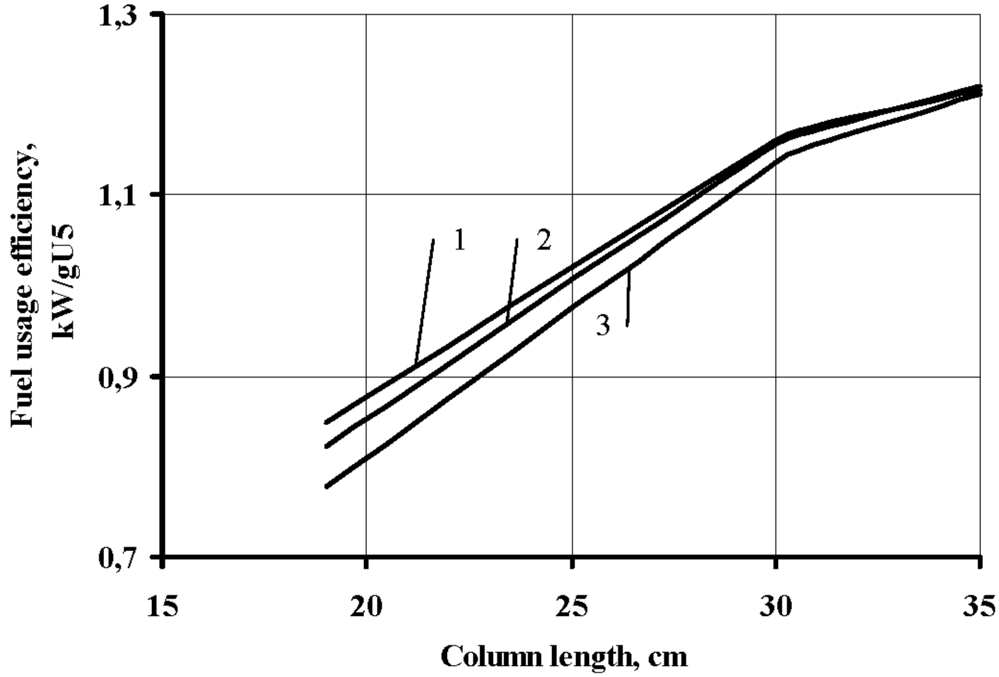

Figure 13. Efficiency of fuel utilization factor in relation to the length of target meat as to the fuel volume concentration: 1 – 1.0; 2 – 0.6; 3 – 0.5.

The major outcome of the accomplished work was feasibility demonstration of target specifications:

- The target-to-cladding gap as well as the target-to-displacer gap are 5 mm;
- The target meat shall be from 300 to 350 mm long.

Table 1 specifies calculated target parameters.

Table 1. Target specifications

Parameter	flat plate target	tube-type target
Peak target meat temperature, ^{0}C	$\leq$ 175	$\leq$ 320
DNB	6	4
Generated Mo-99 activity, Ci	3100	4000
Target meat length, mm	200	300-350
Target power capacity, kW	75	100
Irradiation period, d	6-7	6-7
Target meat composition	UAl_3 or UAl_4 enclosed in Al matrix	UAl_3 or UAl_4
Number of targets in IR	3-4	1

Conclusion

The outcome of calculation data analysis was the development of new design modifications of targets (Table 1), which are capable of the Mo-99 best possible yield capacity with the use of LEU. The next phase of work is to be irradiation test of targets in research reactors.

References

[1] A.L. Izhutov, V.A. Starkov, V.V. Pimenov, V.E. Fedoseev, S.V. Maynskov "Conversion of MIR.M1 Reactor to LEU-Fuel". Proceedings of the 34th International Meeting on Reduced Enrichment for Research and Test Reactors RERTR 2012. Warsaw, Poland, October 14-17, 2012.

[2] Tsykanov V.A., Aver'yanov P.G., Burukin V.P. and others. Issledovatel'skij reactor RBT-6 [RBT-6 research reactor]. Atomnaya energiya. M., 1977. Vol. 43, issue 1. (in Russian).

[3] Starkov V.A., Klinov A.V., Tsykanov V.A., Chertkov Yu.B., Svyatkin M.N., Bestuzheva I.V., Petelin A.L., Gataullin N.G., Fedoseyev V.E., Malkov A.P., Krasnov Yu.A. Kharakteristiki modernizirovannoj aktivnoj zony reaktora SM [Specifications of the SM refurbished core]. Mezhdunarodnaya nauchno-tekhnicheskaya konferentsiya "Issledovatel'skie reaktory v 21 veke" [International Scientific and Technical Conference "Research Reactors in the 21st Century]. June 20- June 23, 2006, Moscow, NIKIET.

[4] Gomin E.A. Status MCU-4. Voprosy atomnoj nauki i tekhniki. Series Phisika yadernyh reaktorov. 2006. Issue 1.pp. 6-32. (in Russian).

[5] Carlson K.E. et al. Code structure, system models and solution metods // RELAP5/MOD3.2: Code manual. NUREG/CR-5535 INEL-95/0174, 1995.