This article is devoted to the development of the production of $^{99}$Mo used in nuclear medicine. The stages in the development of the production of $^{99}$Mo and molybdenum-technetium generators are shown. Production has increased several-fold over the period 2009–2013 and meets the demand of domestic nuclear medicine. The problem of recycling the uranium from an irradiated target after $^{99}$Mo is extracted from the target is important and strongly influences the price of $^{99}$Mo. A technology for reprocessing uranium-containing tailings with the uranium obtained being returned into the $^{99}$Mo production cycle has been developed at the Karpov Research Institute of Physical Chemistry. This has eliminated the need to purchase expensive high-enrichment uranium for targets and has made it possible to use the uranium remaining from the reprocessing of irradiated targets.

The largest supplier of molybdenum-technetium generators for domestic clinics is the Karpov Research Institute of Physical Chemistry (NIFKhI), which has been producing such generators since 1988. $^{99}$Mo is produced from $^{98}$Mo in small quantities for regional medical purposes by the activation method at the Leningradskaya NPP (irradiation of targets) and in the IRT-T reactor at the Physical-Technical Institute at the Tomsk National Research Polytechnic University [1]. At the St. Petersburg Institute of Nuclear Physics, targets containing $^{98}$Mo are irradiated in the VVER-M reactor in order to obtain $^{99}$Mo, and $^{99}$Mo is also separated at the Leningradskaya NPP. In 2012 the production of fragment $^{99}$Mo in the SM-3 and RBT-10 reactors was started at the Research Institute of Nuclear Reactors (NIIAR) with radiochemical separation from an irradiated uranium target using a technology developed by the firm ITD (Germany). This production was developed for domestic nuclear medicine to supplement the existing production at the NIFKhI and is intended mainly for exports.

$^{99}$Mo production runs and the production of molybdenum generators at the NIFKhI were implemented in stages. First, from startup in 1964 the reactor was geared toward scientific research for the Ministry of the Chemical Industry; there were no plans to produce $^{99}$Mo in it. Six vertical experimental channels filled with air or water with cooling by natural circulation were installed in the VVR-Ts core for scientific research.

The problem of mastering the production of $^{99}$Mo was posed at the NIFKhI in 1983. One vertical experimental channel 4-1 was assigned for this purpose (Fig. 1). The first batch of $^{99}$Mo was obtained in 1985, after a channel design with forced cooling and an appropriate target was worked out. The production run consisted of about 3.7 TBq/day (2 days after extraction from the reactor).

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In 2002, the production of fragment $^{99}$Mo was completely concentrated in VVR-Ts after the AM reactor at the Leipunskii Institute for Physics and Power Engineering (FEI) was shut down for decommissioning. The target design was modernized to increase production: the $^{235}$U load was increased from 3.5 to 4 g, and the number of targets in the channel was increased to four. In addition, the processing of the targets was optimized and the radiochemical separation factor of $^{99}$Mo was increased from 60 to 85%. Thus, the capacity of the channel increased to 5.55 TBq/week at the day of release. This was sufficient capacity at that time to meet the needs of clinics for $^{99m}$Tc generators [2].

In connection with the rapid development of methods of diagnostics using radiopharmaceuticals based on $^{99m}$Tc and a shortage of $^{99}$Mo on the world market the production of $^{99}$Mo had to be increased, including for exports. The exports make it possible to compensate some of the losses associated with VVR-Ts operation.

In 2008, another channel with forced circulation was installed in the VVR-Ts reactor instead of the vertical channel in order to increase $^{99}$Mo production. In addition, the target was modernized: the $^{235}$U load was increased to 4.5 g. The system for purifying and catching iodine, radioactive inert gases, and aerosols was also modernized in the ‘hot’ box. A new machine for opening targets was fabricated, and the reprocessing of targets was optimized. The installation of an additional channel in VVR-Ts made it possible to double $^{99}$Mo production since 2009 [3].

In 2009, after the tests performed to evaluate the operational efficiency of the equipment complex were completed, the irradiation time for the uranium targets in VVR-Ts was increased to 72 h, which made it possible to increase $^{99}$Mo production to 14.8 TBq/week on the release date. Thus, since 2009, aside from the required 5.55 TBq/week for domestic clinics, $^{99}$Mo exports up to 9.25 TBq/week became possible; this corresponds to 1.85 TBq/week on the sixth day in terms of the international standards. An improved target of the through type was developed and adopted in 2011; this made it possible to increase $^{99}$Mo production by more than a factor of 2. Two additional experimental channels for the production of $^{99}$Mo were installed in 2012.

The $^{99}$Mo production process with the final product delivered in the form of charge molybdenum-technetium generators to domestic medical centers is shown in Fig. 2. First, the number of targets for loading into a channel and the irradiation parameters for obtaining the required activity in accordance with the delivery schedule are calculated. Next, batches of targets with control of the loaded uranium mass and geometric dimensions and checking of the seal tightness are fabricated. Then, the target is loaded into the irradiation setup, the setup is prepared for operation in the reactor, and the main technological parameters – coolant flow rate and temperature, channel seal, and others – are checked. After the reactor is brought up to power, the targets are irradiated for about 70 h. The radiochemical separation of the $^{99}$Mo is accomplished in hot boxes. After separation, the quality control procedure is performed. Liquid, solid, and gaseous wastes are recycled in accordance with the operative technology. Next, $^{99}$Mo is goes to charging of the pre-fabricated molybdenum-technetium generators. The generators ready for use are delivered weekly to medical treatment institutions. The uranium-bearing residues are separated and sent to a regeneration facility for subsequent recycling.

Irradiation Setup and Target Design for $^{99}$Mo Production in VVR-Ts

Irradiation setup and target specifications. The setup for irradiating the target must meet the following requirements:

1) allow loading into channels with the appropriate thermal neutron flux;
2) reduce the thermal neutron flux density in the materials to a minimum;
3) provide a forced cooling system, adequate for removing the energy release from the target during irradiation;
4) keep fission products inside the target, including gaseous products upon unsealing; and
5) be reusable, i.e., easily disassembled in order to replace targets and maintainable.

The target must contain an adequate amount of $^{235}$U in order to obtain the required quantity of $^{99}$Mo, not allow the emission of radioactive products, especially gases, during and after irradiation, be easy to disassemble in a hot chamber, and allow chemical reprocessing in a chamber in minimal time.

A diagram of the channel setup for producing $^{99}$Mo is displayed in Fig. 3. It is desirable that for radiochemical reprocessing of the target the amount of liquid radioactive wastes be minimal. For this reason, a uranium-zinc target, which after irradiation is dissolved in acid, is used in the VVR-Ts reactor. The reprocessing time for a uranium target strongly influences the $^{99}$Mo yield. About 1.5% of the $^{99}$Mo formed in the target is lost due to decay. For this reason, the operations performed with the target when it is opened in a hot box must be quite simple and easily performed.