Research study of possibility producing radioisotope Mo-99 in the active core of the BN-600 reactor using an irradiated assembly with a high flux of thermal and resonance neutrons

D S Klimenko¹, I A Ignatiev¹, V V Kolesov¹ and V V Korobeynikov²

¹ OINPE NRNU MEPhI, Obninsk, Russia
² SSC RF-IPPE, Obninsk, Russia

E-mail: klimenkods@oiate.ru

Abstract. It is fact that production of secondary fuel isotopes (plutonium) is carried out in fast neutron reactors, and the possibility of production of non-fuel radioactive isotopes for use in medicine and industry is being considered. BN-600 has a high power density in the core. This makes it possible to achieve a high rate of fission of target nuclei, which is especially important in the case of the production of radioisotopes, due to their short half-life. To meet the growing demand for isotopes, it is planned to use irradiation devices in high-power sodium fast reactors. This type of irradiation device assumes a massive production of the necessary isotopes. The paper considers the design of irradiation devices for the production of molybdenum-99 isotope in the BN reactor. The presented design of the irradiation assembly contains a moderator in order to combine the advantages of the high neutron flux inherent in fast reactors and the thermal neutron spectrum inherent in thermal reactors.

1. Introduction

Currently, much attention is paid to the development of methods of medical diagnostics that make it possible to determine the foci of localization of various diseases at the earliest stages of their development. Methods based on the use of radioactive isotopes are among the most informative and accurate diagnostic methods. The bioactive compounds labeled by them allow high-quality imaging of various organs and tissues at the cellular level, which makes it possible to use them in oncology, cardiology, endocrinology, neurology, pulmonology and other fields of medicine. One of the most significant radionuclides for world nuclear medicine is technetium-99m, a daughter product of the β-decay of the radioisotope molybdenum-99.

Molybdenum-99 is produced at nuclear reactors in South Africa, Belgium, Holland, Australia, Russia and other countries, however, the demand for molybdenum-99 in the world is growing every year. The global weekly demand for ⁹⁹Mo is estimated in the range of 9500-1200 Ci [1-2].

It is known that in fast neutron reactors the production of isotopes of secondary fuel (plutonium-239, uranium-233) is carried out [3], a device is known for the accumulation of molybdenum-99 in a nuclear reactor [4], as well as a method for the production of cobalt-60 [5-8] in a fast reactor. The disadvantage of the production of radioactive isotopes in a fast reactor is that the fission cross section of uranium-235 is very small in the neutron spectra characteristic of a fast reactor [9].
2. Construction of a calculation model in the Serpent software

2.1. Serpent
Currently, this software package is being actively developed and, possibly, will replace MCNP.

In the course of the work, the possibilities of neutron-physical calculation of the reactor were studied using the "Serpent" software complex, developed at the State Science and Technology Center in Finland in 2004 [10].

"Serpent" is a software package that implements the Monte Carlo method and allows calculations of the reactor plant and its campaign with the highest possible accuracy:

- changes in the nuclide composition of nuclear fuel during irradiation in the reactor;
- reactor campaign;
- microscopic sections using a multi-group approach;
- infinite and effective neutron multiplication factor;
- distribution of energy release over the zones of the reactor plant;
- activity of spent nuclear fuel;
- temperature effects of reactivity and many others [11].

Also, this program allows you to simulate the loading of the whole reactor with the ability to describe each individual element, channel or cell of the reactor core.

Serpent provides some additional geometry functions specifically for fuel design. These features include simplified definition of cylindrical fuel rods and spherical fuel particles, square and hexagonal arrays for WWER and fast reactors.

2.2. Calculated parameters of the investigated model
Based on the benchmarks published by the IAEA [12-13], a table with the geometric and physical parameters of the investigated model of the BN-600 reactor is presented (see table 1).

| Parameter                                      | Value  |
|-----------------------------------------------|--------|
| Fuel temperature                             | 1500 K |
| Temperature of all other materials            | 600 K  |
| Fuel assembly lattice                         | 99.02 mm|
| Size of fuel assemblies                       | 96 mm  |
| Thickness of fuel assembly case               | 2 mm   |
| Fuel element lattice                          | 7.95 mm|
| Fuel rod diameter                             | 6.9 mm |
| Fuel rod cladding thickness                   | 0.4 mm |
| The number of fuel elements in fuel assemblies| 127    |
| Control rods channel diameter                 | 85 mm  |
| Control rods channel thickness                | 1 mm   |

The core layout of the standard design model of the BN-600 reactor is shown in figure 1. The fuel is uranium dioxide (UO₂). The vertical arrangement of the core elements is shown in figure 2.
Figure 1. Diagram of the elements of the BN-600 core in the model under study. 1) LEZ, 17% U\(^{235}\); 2) MEZ, 21% U\(^{235}\); 3) HEZ, 26% U\(^{235}\); 4, 5) cells for the CPS cluster; 6, 7) steel shielding blocks; 8) radial reflector.

Figure 2. Diagram of the vertical arrangement of the core elements. 1) axial reflector; 2) plugs of absorbing elements; 3) boron shield; 4) sodium plenum; 5) fuel rod plugs; 6) fuel part of fuel rods; 7) first axial blanket; 8) second axial blanket.

3. Calculation of the optimal position of the irradiated assembly

3.1. Target for production Mo-99

In this work, it was decided to use a target similar to the target of the WWR-C reactor [14-15]. In the core of the WWR-C reactor, 2 containers with a target are placed in height. In the core of the BN-600
reactor, 4 containers with a target made of highly enriched uranium are placed in height. The target used in the calculations is shown in figure 3.

![Diagram of reactor setup with target](image)

**Figure 3.** Target for production Mo-99.

3.2. *Assembly position and composition*

For the effective production of Mo-99, it is necessary to select such a position of the irradiated assembly, at which the thermal neutron flux will be maximum. The following models were considered, shown in figure 4.
Metallic beryllium or zirconium hydride was considered as a moderator [16-17]. The neutron spectrum was divided into 4 energy groups: 0 - 1 eV; 1 eV - 150 keV; 150 keV - 0.7 MeV; 0.7 MeV - 20 MeV. Evaluated Nuclear Data Library was used – JEFF-3.1[18].

3.3. Results of calculating neutron fluxes in various targets

The averaged four-group fluxes for targets with different channel locations in the case of using beryllium as a moderator are shown in figure 5.
As can be seen from figure 5, in the case of using beryllium as a moderator, the best variation for the arrangement of the irradiated assemblies is variation 4 (see figure 4). In this case, the maximum heat flux can be $\sim 6E+14$ n/(cm$^2$s).

Now we will give a similar diagram for the same options for arranging the assemblies, but now zirconium hydride is used as a moderator (see figure 6), in this case, the heat flux can be more than $10E+15$ n/(cm$^2$s).

![Figure 6. Average four-group fluxes for different assembly arrangements using zirconium hydride as a moderator.](image)

From the data shown in figures 5 and 6, it can be concluded that the optimal arrangement of assemblies is corresponding to variation 1, and zirconium hydride should be used as a moderator. The use of non-hydrogen-containing moderators, for example, beryllium, due to the limited cross-sectional area inside the assembly, on which the moderator can be placed, as shown by computational studies, does not allow the formation of a neutron spectrum with a significant fraction of neutrons in the thermal energy range and, accordingly, it is not possible to obtain target radioisotopes with economically acceptable specific activity.

3.4. Equalization of energy release from next to target fuel assemblies.

Due to the fact that a moderator is placed in the core, it softens the neutron spectrum in the fuel assemblies adjacent to the irradiated assembly. Such an impact leads to an undesirable increase in energy release. To prevent this effect, it makes sense to surround the assembly with a thermal neutron filter made of absorbing material; in this work, gadolinium oxide is used (see figure 7).
Figure 7. Assembly with a gadolinium filter. 1) case; 2) Gd$_2$O$_3$; 3) ZrH$_2$; 4) container; 5) fuel.

Comparison of the power and effective multiplication factor of the adjacent fuel assembly for different models is shown in table 2.

Table 2. Design model parameters.

| Parameter | Original design | Without Gd$_2$O$_3$ filter | With Gd$_2$O$_3$ filter |
|-----------|----------------|----------------------------|-------------------------|
| N, MW     | 2.889          | 6.331                      | 2.884                   |
| K$_{eff}$ | 1.06936+/-0.00022 | 1.09134+/-0.00021         | 1.02874+/-0.00023       |

From the data presented, it can be concluded that the use of a gadolinium oxide filter makes it possible to prevent an undesirable increase in the energy release of standard fuel assemblies in the core.

4. Results of calculating the operating time of Mo-99
Calculations were made of the Mo-99 production (in Ci) in the core targets at a time interval of 0-300 hours, for 2 models - a model with a moderator and a model without a moderator. The corresponding graphs are shown in figure 8.

We are interested in the operating time of Mo-99 after 120 hours, the corresponding values are shown in table 3.

Table 3. Result of molybdenum-99 production after 120 hours of target irradiation.

| Target | With moderator | Without moderator |
|--------|----------------|-------------------|
| Target 1 | 9900           | 230               |
| Target 2 | 13000          | 300               |
| Target 3 | 13200          | 300               |
| Target 4 | 10300          | 240               |
| Sum     | 46400          | 1070              |
Figure 8. Average four-group fluxes for different assembly arrangements using zirconium hydride as a moderator.

5. Conclusion

Based on the table shown on the slide, an obvious conclusion follows - the production of radioisotopes in a fast reactor without the use of moderating materials does not make sense. The activity of the produced molybdenum differs by more than an order of magnitude in favor of the model using irradiated assemblies with zirconium hydride as a moderator and a gadolinium oxide filter. $^{99}$Mo is sold in units of activity at a given future point in time. The standard calculation procedure is based on the amount of activity that will occur on the sixth day after $^{99}$Mo is removed from the manufacturer's premises.

The total worldwide requirement for $^{99}$Mo is 12000 Ci per week on the sixth day. One assembly of the proposed model can produce about 10000 Ci in 6 days.

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