Radioactive isotope production in the fast neutron nuclear reactors

V Risovanyi

“Science and innovations” JSC of the State Atomic Energy Corporation ”Rosatom”, Moscow

Abstract The present work reviews the primary benefits of radioactive isotope production in the fast neutron reactors the accumulated foreign and local experience in usage of this type of reactors for radioactive isotope production, and also the potential new trends in development of the existing technologies. The main advantage of isotope production in the fast neutron reactors is their high capacity and lower value in general as compared with the accelerating installations. One nuclear reactor may produce dozens of different isotopes including Cj-60, Sr-89 (targets on the base of yttrium), Ni-63 (targets on the base of copper), Eu-152, Eu-154, Sm-145, Sn-117n, Cu-64 and others. It should also be emphasized that a number of radioisotopes are most efficiently produced only in fast and intermediate neutron spectra, which is not possible in nuclear thermal reactors and, above all, in research reactors. Studies have shown that the organization of large-scale production of radioisotopes can significantly increase the economic attractiveness of fast-neutron reactors.

1. Introduction

The reactor isotopes became widely used in the nuclear medicines and different industrial branches. They are used for diagnostics and treatment of oncolgical diseases, in manufacture of diagnostic equipment for compositional analysis and material and structure quality review, for radiation treatment of materials in order to provide them with new application properties, for product disinfection to ensure its long term storage, for better sprouting and germination ability in the agricultural sector. The nomenclature and scope of the reactor isotope supply to the world market are increasing constantly.

The main advantage of isotope production in the nuclear reactors is their high capacity and lower value in general as compared with the accelerating installations. One nuclear reactor may produce dozens of different isotopes.

Now, the reactor isotopes are mainly produced in the research reactors in the world generally by installation of the targets in the idle cells which are not engaged in the reactor experiments and researches. If targets only are irradiated, the production value rises sharply, as the operating expenses of the research reactors are relatively high and may exceed the isotope value in the world market. It requires additional grants or search for alternative reactor technologies for isotope production. Among them, for example, the technology of Co-60 radioactive isotope production in the control rods of CANDU energy nuclear reactors with heavy water coolant and RBMK-1000 shows good results. In these types of nuclear reactors, cobalt is the neutron absorber, and the irradiated absorptive core is used for gamma-source manufacture after control rod operation. Radioactive isotope production in power nuclear reactors enables cost reduction and output increase. According to the investigations and accumulated experience, the fast neutron nuclear reactors are the most effective for this purpose.
Organization of large-scale radioactive isotope production in the fast neutron nuclear reactors will improve greatly their economic attractiveness.

The present work reviews the primary benefits of radioactive isotope production in the FN reactors, the accumulated foreign and local experience in usage of this type of reactors for radioactive isotope production, and also the potential new trends in development of the existing technologies.

2. Benefits of radioactive isotope production in the fast neutron reactors

The fast neutron reactors have the following advantages in radioactive isotope production: the large available capacity for installation of targets in the lateral blanket or in the reactor core directly, high neutron fluxes \(10^{16}\) cm\(^{-2}\)s\(^{-1}\) instead \(10^{13}-10^{15}\) cm\(^{-2}\)s\(^{-1}\) in the thermal neutron nuclear power facilities), ability to form the required neutron energy spectrum in targets using moderating materials. Also, it is necessary to emphasize that some isotopes are produced most effectively only in the fast and intermediate neutron spectrums, that is not possible in the thermal neutron nuclear reactors and, most significantly, in the research reactors. Examples of such isotopes include Sr-89 (targets on the base of yttrium), Ni-63 (targets on the base of copper), Sm-145, Sn-117n, Cu-64 and others.

Figure 1 shows the dependence of neutron fluxes on their energy in different nuclear reactors. It is apparent that introduction of devices with moderating materials in the reactor decreases the neutron energy in the fast neutron reactors while maintaining neutron flux at a relatively high level.

![Figure 1](image)

**Figure 1.** The corner cell of the lateral blanket of BOR-60 reactor core with moderating devices and without them (red line).

3. Irradiation capacity of BN-600 reactor and experience of radioactive isotope production in it

Figure 2 shows BN-600 reactor core. The reactor core contains 6 emergency shutdown rods (AZ), 18 compensating rods (KP-TK) and 2 emergency shutdown rods (AR). During reactor operation, the bottom end of AZ rods with the diameter of ab. 80 mm and the height of 500 mm resides in the reactor core. It is made out of stainless thick shell and designed for handling operations. There is some practice to use the inner volume of the AZ rod end for testing of constructional material samples. It also may contain the targets for radioactive isotope production. Even in one AZ rod, the volume available for target installation exceeds 1500 cm\(^3\), which is equivalent of (5-6) SM-3 reactor channels where radioactive isotopes are produced. But, the moderating elements cannot be placed in the ends, so the ends may contain the targets where the fast neutron radioactive isotopes are produced.

There is a vast successive practice of using trap type constructions in KP-TK and AR rods of BN-600, BN-350, BOR -60 reactors which have zirconium hydride elements moderating neutrons.
Neutron moderation on hydrogen atoms results in increased velocity of neutron capture by boron carbide or europium oxide absorbents, thus increasing physical efficiency of control rods. In the middle of 1990s, these constructions produced Co-60 with the specific activity of about 100Ku/g. The schematic view of the trap rode for Co-60 production is given in figure 3. The trap rods may contain not only cobalt targets but also other targets producing isotopes by absorption of neutrons with the energy of less than 100 eV. There is provided to improve the design of trap rods using homogeneous absorbing and moderating hafnium hydride material (Hf,H). Even with HfH stoichiometry, hafnium hydride has physical efficiency equal to the physical efficiency of boron carbide with 80% enrichment in regard to boron-10 isotopes. The technologies are developed which allow to enter the targets in hafnium hydride directly increasing the neutron capture velocity by the targets by (25-30)%.

Application of two-target trap rods of new design may reduce expenses for radioactive isotope production while utilizing the irradiated items.

The control rods with europium oxide were used in BN-600 reactor for more than 20 years. They produce the radioactive nuclides which have high gamma activity and long half-life surpass their counterparts (Co-60 and Cs-137) by technical and economic features. Specific activity and gamma energy of europium radionuclides are higher than of Cs-137, and their half-life period exceeds by (2-3) times the half-life period of Co-60 (table 1) [2-4]. Application of europium control rods enables production of at least 6 MKu of europium radionuclides per year in BN-600 reactor.

Application of the lateral blanket of the fast neutron reactor provides unique abilities for radioactive isotope production. The volume of only 3 LB cells is equal to the volume of all SM-3 cells. And there are may be more than 40 such cells. The design of devices with targets may be similar to the design of the control rod shown in figure 3, but without absorbing materials. At that, there is more place for targets and moderating materials.

![Figure 2. BN-600 reactor core.](image-url)
Figure 3. The structural diagram of the fast neutron reactor trap rod for Co-60 production: green color - zirconium hydride elements; red - absorbing material (B$_4$C or Eu$_2$O$_3$); yellow - cobalt targets.

Table 1. Comparative characteristics of europium, Co-60 and Cs-137 radioactive nuclides.

| Characteristics                              | Co-60       | Cs-137       | Eu-154       |
|----------------------------------------------|-------------|--------------|--------------|
| Shape of the active part                     | Coated metal| Salt CsCl    | Oxide Eu$_2$O$_3$ |
| Energy (MeV)                                 | 1.17 and 1.33| 0.66 (average)| 0.8 (average) |
| Half-life (years)                            | 5.3         | 30.0         | 13.5         |
| Specific activity (Ci/g)                     | 50-110      | 20-25        | 50-70        |
| Thermal neutron absorption crosssection (barn)| 36 (Co-59)  | -            | 4500 (Eu-153) |
| Time for production of 50Ku/g (eff.days)     | 300-600     | -            | 40-60        |

In the middle of 1990s and at the beginning of 2000s, the lateral blanket of BN-600 reactor produces Co-60 with the specific activity of (90-100)Ku/g. The appearance of the devices without casing manufactured by NIIIAR SSC JSC is given in figure 4. Outside, you can see the absorbing elements with europium oxide along the perimeter. They were used in the design for absorbing of the thermal neurons in order to prevent excessive burn-out of fuel assemblies around the device.

Figure 4. The appearance of the devices with absorbing elements for production of Co-60 in BN-600 reactor.
Since 2016, “Science and innovations” JSC implements the project in cooperation with the Beloyarsk NPP, the LIPPE, OKBM, NRRI, LUCH, BARSRIIM, MSZ for production of the medical grade Co-60 with high specific activity (300 KU/g) in BN-600 and BN-800 reactors[1]. The cluster arrangement diagram and the design diagram of the device with targets is shown in figure 5. The absorbing elements were removed from the design, the content of zirconium hydride moderating material was increased, the targets became smaller in order to eliminate the self-absorption effect on their surface. The devices are surrounded by the steel assemblies which may also contain the targets for production of radioactive isotopes in the fast neutron spectrum. 3 clusters were used at the first stage due to necessity to maintain reactivity margin without additional loading of fuel assemblies.

![Figure 5](image_url)

**Figure 5.** Location of radioactive isotope production clusters in the lateral blanket of BN-600 reactor (left) and the design diagrams of the clusters and the device of Co-60 production with high specific activity (300 KU/g) in BN-600 and BN-800 reactors: green color - elements with zirconium hydride, yellow - targets with cobalt.

Upon performance of the whole test cycle for the devices and development of process operations for their handling and logistics, it is planned to consider loading of the greater number of devices with cobalt targets as well as with the targets for other radioactive isotopes.

4. **American experience in radioactive isotopes production in FFTF**

Figure 6 shows FFTF fast neutron development reactor core (US) with the capacity of 400 MW. It did not generate electric power but had the loopers in the core wherein the targets were loaded and unloaded without the reactor shutdown for production of radioactive isotopes with short half-life.
Figure 6. Location of the targets in FFTF reactor (USA, 1998).

Figure 7. Device design in the FFTF with targets (top photo to the left), device appearances (top photo to the right) and relation between neutron flux change and their energy with moderating elements (red line) and without them.

The appearance of the devices for production of long-live radioactive isotopes is shown in figure 7. The bottom photo shows change of neutron flux with different energy in the presence of moderating elements and without them. Yttrium hydride was used as the moderating material (YHx). At the same
time, Y-90 radioactive isotope was produced in yttrium hydride, its production increased by 25 times within 1991 - 1998. In total, over 30 radioactive isotopes were produced in the FFTF reactor. Table 2 gives the list of some radioactive isotopes produced in the FFTF reactor and shows the advantageous efficiency of this reactor as compared with the HFIR high flow radioisotope research reactor [5]. For some isotopes, the efficiency increases by tens and even hundreds times. The US business structures maintain that production of large quantities of radioactive isotopes in the FFTF fast neutron reactor is cost efficient even without power generation. At the beginning of 2000s, there were great efforts to maintain that production of large quantities of radioactive isotopes in the FFTF fast neutron reactor is some isotopes, the efficiency increases by tens and even hundreds times.. The US business structures resumption evidence that this plant may become the largest radioactive isotope producer in the world. According to K. Holder “There is no reactors in the USA now which can perform this task. FFTF is the perfect choice as the isotope producer”.

Table 2. The comparative characteristics of radioactive isotope production in the FFTF fast neutron nuclear reactor (USA, 400 MW) and HFIR (100 MW, USA, the analogues of SM-3 high flow reactor in the NRRI).

| Isotope  | Target | Activity for 1 g of target Cu | Activity ratio, FTF/ HFIR (rel.un.) | Target volume, cm³ | Ration of target volumes, FTF/ HFIR (rel.un.) | Increase of FTF efficiency |
|----------|--------|-------------------------------|------------------------------------|-------------------|-----------------------------------------------|---------------------------|
| Ac-227   | Ra-226 | 2.53E+00                      | 1.7                                | 38500             | 83.0                                          | 140                       |
| Cd-109   | Cd-108 | 6.19E+00                      | 1.2                                | 77100             | 166.2                                         | 190                       |
| Cu-64    | Zn-64  | 1.56E+00                      | 1.3                                | 72                | 5.6                                           | 9.5                       |
| Cu-67    | Zn-67  | 6.79E-02                      | 1.4                                | 12.9              | 5.6                                           | 7.8                       |
| Ho-166   | Ho-165 | 1.59E+03                      | 1.3                                | 12.9              | 5.6                                           | 7.3                       |
| I-131    | Te-130 | 6.61E+00                      | 0.2                                | 72                | 5.6                                           | 1.1                       |
| Ir-192   | Ir-191 | 1.26E+02                      | 2.4                                | 77100             | 166.2                                         | 400                       |
| Mo-99    | Mo-98  | 7.88E+01                      | 2.9                                | 12.9              | 5.6                                           | 16.2                      |
| Os-194   | Os-192 | 6.80E+00                      | 0.9                                | 77100             | 166.2                                         | 140                       |
| P-32     | S-32   | 5.90E+00                      | 1.4                                | 72                | 5.6                                           | 7.8                       |
| P-33     | S-33   | 1.84E+01                      | 0.4                                | 77100             | 166.2                                         | 62                        |
| Pd-103   | Pd-102 | 1.12E+02                      | 0.4                                | 72                | 5.6                                           | 2.2                       |
| Pt-195m  | Pt-195 | 5.77E+00                      | 1.4                                | 72                | 5.6                                           | 7.8                       |
| Re-186   | Re-185 | 3.51E+03                      | 0.4                                | 72                | 5.6                                           | 2.2                       |
| Sc-47    | Ti-47  | 1.66E+00                      | 1.5                                | 72                | 5.6                                           | 8.4                       |
| Se-75    | Se-74  | 1.43E+03                      | 0.2                                | 38500             | 83.0                                          | 20                        |
| Sm-145   | Sm-144 | 2.00E+01                      | 2.0                                | 38500             | 83.0                                          | 170                       |
| Sm-153   | Sm-152 | 7.33E+03                      | 3.8                                | 72                | 5.6                                           | 21                        |
| Sr-89    | Sr-88  | 6.34E+01                      | 0.4                                | 38500             | 83.0                                          | 30                        |
| Th-228   | Ra-226 | 1.30E+02                      | 1.4                                | 38500             | 83.0                                          | 120                       |
| Th-229   | Ra-226 | 3.70E-02                      | 1.9                                | 38500             | 83.0                                          | 150                       |
| W-188    | W-186  | 5.95E+00                      | 0.1                                | 38500             | 83.0                                          | 9.9                       |
| Xe-127   | Xe-126 | 2.95E+02                      | 0.4                                | 77100             | 166.2                                         | 71                        |

5. Potential areas of radioactive production development in the fast neutron reactors
The main disadvantage in the use of the fast neutron energy reactors including BN-600, BN-800 and BOR-60 is lack of the special serpentine passages which make possible to unload the targets with radioactive isotopes with short half-life without reactor shutdown. In the absence of such loops in fast neutron power reactors, they produce radioactive isotopes with the half-life of more than 50 days. This group of radioisotopes include: C-14 (T½ =5800 years), Ni-63 (T½ =100 years), Ac-227 (T½ =21.8 years), Co-60 (T½ =5.3 years), Eu-152,154 (T½ =8 and 13 years), Gd-153(T½ =242 days), Se-75 (T½ =120 days), Ir-192 (T½ =73.8 days), W-188 (T½ =69.4 days), Y-91(T½ =58.5days), Sr-89(T½
=50.5 days), etc. This restriction is associated with the reactor life time, their shutdown for refueling, control rod loading and repair service. Now, the planned operating periods for BN-600 reactor are 120 and 160 days, and for BOR-60 reactor - about 90 days. The latter may be shut-downed for a short time for target removing and after 40 days of operation during radioisotope production.

All above mentioned radioisotopes are rather popular in the world market or may find application. For example, the sources made out of Ir-192 and Se-75 are used in fault indicators, Co-60 with the specific activity of 100 Cu/g are used in gamma-ray sources. The radionuclides of Europium-152,154 are also considered to be promising in regard to gamma-ray sources. Ac-227, Gd-153, W-188, Y-90, Sr-89, Co-60 (with the specific activity of more than 250 Cu/g) - in nuclear medicine. The self-contained power supplies energy storage units are developed based on the radioactive isotopes on the basis of C-14 and Ni-63. This requires industrial production of large amount of relatively cheap radioisotopes with beta-decay.

There will be provided two-three serpentine passages in the reactor core or in the lateral blanket of the designed nuclear reactors of BN-1200 type like in the FFTF reactor. This decision will sufficiently enlarge the list and the volume of the produced radioisotopes and will reduce their net cost. Three such loops for scientific tasks will be provided in the designed multipurpose research fast neutron reactor (MBIR) with the capacity of 150 MW; these loops may produce short lived radioactive isotopes in the absence of scientific programs.

The control rods and devices based on europium and cobalt have high energy release and may be used as radiant heaters. For example, the TK-KP rod containing about 8 kg of absorbing material 80%Eu₂O₃+20%Mo had the activity of about 400kCu and the thermal power of 6 kW after 420 eff. days of operation in BN-600 reactor, that exceeds the power of irradiated FAs. The devices with (1-2)kg of cobalt with the specific activity of ab. (150-200)кКu/g have the same thermal power. Such radiant heaters with the targets 80%Eu₂O₃+20%Co may be produced, for example, by their installation in the lateral blanket of BN-600 reactor in the areas of the reactor vessel radiation protection.

Preliminary studies testified that organization of extensive radioisotope production may improve significantly the economic attractiveness of the fast neutron nuclear reactors. They may take at least 20% from the cost of power sells in BN-600 and BN-800.

6. Conclusions
There is a positive experience of Со-60, Sr-89, Gd-153, Ni-63, Eu-152,154,155 production in the native FN reactors.

Production of Eu-152,154,155 and Со-60 in BN-600 reactor core requires some little changes in the design and technical documentation of KS and AR rods regarding implementation of europium absorptive cores in the standard dimensions of GIK sources resulting in launch of new GIEK sources production with improved technical and economic features for all operating gamma ray sources.

The advantageous radioisotopes for production in the lateral blanket of BN-600 (800) reactor are Ni-63, Sr-89 ,Y-91, Th-229, Eu-152,154,155, Gd-153, Ir-192, Se-75, W-188, Ac-227 with long half-life (more than 50 days).

The number of target loadings into BN-600 (800) reactors exceeds by 20-30 times the capabilities of all Russian research nuclear reactors.

In the presence of autonomous channels in the FN reactors (they are available in MBIR and it is reasonable to have them in BN-1200), there are no restriction for production of almost all radioisotopes.

The promising area is production of radioisotopes on the basis of europium and cobalt for radiant heaters.

At organization of large scale radioisotope production in BN-600(800) fast neutron nuclear reactors, it is appropriate to build close at hand modern radiochemical plant according to the international standards with potential annual sales of products in the amount of more than 2 bln. RUB beginning from 2022.
Closed Nuclear Fuel Cycle

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