Capability assessment of production of n.c.a. isotope $^{177}$Lu on the average flux reactor IRT-T

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Abstract. The article discusses the possibility of producing $^{177}$Lu nuclide at the IRT-T mid-flow research nuclear reactor. The paper presents possible reactions on thermal neutrons with lutetium and ytterbium nuclei, on the basis of which a theoretical calculation of the activity of $^{177}$Lu, $^{177}$Yb and $^{175}$Yb was carried out taking into account the burnup over the entire energy spectrum of the central channel of TsK-1 of the research nuclear reactor IRT-T. The obtained theoretical calculations were verified experimentally by irradiation of enriched ytterbium targets. It is shown that upon irradiation of an enriched ytterbium target with a mass of 10 mg. within 3 weeks, the obtained activity of $^{177}$Lu was 23.05 GBq, while the activity of $^{175}$Yb was 2.55 GBq. Two Yb2O3 samples with masses of 3.4 and 4.7 mg enriched in $^{176}$Yb to 99.59% were irradiated under different conditions. The first sample (4.7 mg) was irradiated at a power of 100 kW for 60 minutes and the activity of $^{177}$Lu was 23.05 GBq, while the activity of $^{175}$Yb was 2.55 GBq. Two Yb2O3 samples with masses of 3.4 and 4.7 mg enriched in $^{176}$Yb to 99.59% were irradiated under different conditions. The first sample (4.7 mg) was irradiated at a power of 100 kW for 60 minutes and the activity of $^{177}$Lu was 23.05 GBq, while the activity of $^{175}$Yb was 2.55 GBq. Two Yb2O3 samples with masses of 3.4 and 4.7 mg enriched in $^{176}$Yb to 99.59% were irradiated under different conditions. The first sample (4.7 mg) was irradiated at a power of 100 kW for 60 minutes and the activity of $^{177}$Lu was 23.05 GBq, while the activity of $^{175}$Yb was 2.55 GBq. Two Yb2O3 samples with masses of 3.4 and 4.7 mg enriched in $^{176}$Yb to 99.59% were irradiated under different conditions. The first sample (4.7 mg) was irradiated at a power of 100 kW for 60 minutes and the activity of $^{177}$Lu was 23.05 GBq, while the activity of $^{175}$Yb was 2.55 GBq. Two Yb2O3 samples with masses of 3.4 and 4.7 mg enriched in $^{176}$Yb to 99.59% were irradiated under different conditions. The first sample (4.7 mg) was irradiated at a power of 100 kW for 60 minutes and the activity of $^{177}$Lu was 23.05 GBq, while the activity of $^{175}$Yb was 2.55 GBq. Two Yb2O3 samples with masses of 3.4 and 4.7 mg enriched in $^{176}$Yb to 99.59% were irradiated under different conditions. The first sample (4.7 mg) was irradiated at a power of 100 kW for 60 minutes and the activity of $^{177}$Lu was 23.05 GBq, while the activity of $^{175}$Yb was 2.55 GBq.

1. Introduction

In the modern world well known a method of treatment bone tumors and internal organs by irradiation them with radiopharmaceuticals based on nuclide $^{177}$Lu [1]. Significant difference this method from others is enough low toxicological action on patient's organism (maximum energy of beta radiation is 497.1 keV for 79.3%, gamma radiation is 113 keV and 208 keV with intensivity 6.4% and 11% respectively). Lower toxicological action can be got by synthesis of radiopharmaceuticals based on n.c.a. nuclide $^{177}$Lu [2].

Nuclide $^{177}$Lu is producing in active core nuclear reactor by neutrons irradiation ytterbium target by reaction [3]:

$$^{176}Yb(n, \gamma) \rightarrow ^{177}Yb(\beta^{-}) \rightarrow ^{177}Lu(\beta^{-}) \rightarrow ^{177}Hf$$  \hspace{1cm} (1)

In order to significantly increase the activity of the lutetium isotope obtained during the reaction, it is necessary to use an ytterbium target enriched by $^{176}$Yb isotope by at least 90%, for example,
ytterbium can be enriched by electromagnetic or gas centrifuges [4] for the subsequent separation of lutetium from the ytterbium target, both classical separation methods can be used, for example, extraction [5], cementation [6], separation using an ion-exchange resin [7], and non-classical methods, for example, thermal methods [8] or the method or methods operating under the action of an external periodic symmetric electric field [9,10]. By linking the theoretical calculations with those obtained during the experiment, it will be possible to calculate the necessary radiation protective equipment required for the implementation of the above methods for separating the ytterbium target from $^{177}$Lu.

The purpose of the work was theoretical and practical determination activity producing isotope lutetium-177 from ytterbium target on the average flux reactor IRT-T.

2. Reactions

Due to the fact that, there are admixtures in the irradiated target, e.g. $^{174}$Yb, it is necessary to theoretically assess possible radionuclide purity.

Some of the various nuclear reactions when a substance is irradiated with thermal neutrons are given below:

| Target | Reaction | Cross-section (barn) | Activation product | Half-life | Decay Mode | Decay product disintegrating/stable |
|--------|----------|----------------------|--------------------|-----------|------------|-----------------------------------|
| $^{176}$Yb | $(n,\gamma)$ | 2.4±0.2 | $^{177}$Yb | 1.9h | $\beta^-$ | $^{177}$Lu$\rightarrow^{177}$Hf |
| $^{174}$Yb | $(n,\gamma)$ | 69.4±5 | $^{175}$Yb | 4.19d | $\beta^-$ | $^{175}$Lu |
| $^{168}$Yb | $(n,\gamma)$ | 3470±100 | $^{169}$Yb | 32d | EC | $^{169}$Tm |
| $^{168}$Yb | $(n,\alpha)$ | <0.004 | $^{167}$Er | 10.4h | EC | $^{167}$Ho |
| $^{172}$Yb | $(n,\alpha)$ | <0.00003 | $^{169}$Er | 9.4d | $\beta^-$ | $^{169}$Tm |
| $^{174}$Yb | $(n,\alpha)$ | <0.00002 | $^{171}$Er | 7.52h | $\beta^-$ | $^{171}$Tm |
| $^{176}$Lu | $(n,\gamma)$ | 2.1±0.7 | $^{177m}$Lu | 161d | $\beta^-$ | $^{177m}$Hf |
| $^{176}$Lu | $(n,\gamma)$ | 2.1±0.7 | $^{177m}$Lu | 161d | IT | $^{177m}$Lu$\rightarrow^{177}$Hf |
| $^{175}$Lu | $(n,\gamma)$ | 16.2±0.5 | $^{176m}$Lu | 3.68h | $\beta^-$ | $^{176m}$Hf |
| $^{175}$Lu | $(n,\alpha)$ | <0.00006 | $^{172}$Tm | 2.65d | $\beta^-$ | $^{172}$Yb |
| $^{176}$Lu | $(n,\gamma)$ | 1778±75 | $^{177}$Lu | 6.71d | $\beta^-$ | $^{177}$Hf |
| $^{178}$Lu | $(n,\alpha)$ | <0.002 | $^{175}$Tm | 8.24h | $\beta^-$ | $^{175}$Yb |

Some of the given nuclear reactions are significant, some can be neglected due to the small reaction cross section, short decay time, or low content in the initial target. Table 2 shows the approximate content of lutetium isotopes in the ytterbium enriched and natural target.

| Nuclide | Natural Composition (% mass) | Enriched Composition (% mass) |
|---------|-------------------------------|-------------------------------|
| Lu      | 0.003                         | 0.0001                        |
| Yb      | 87.99                         | 87.9998                       |

Table 3 shows the isotopic composition of ytterbium in targets with natural and enriched isotopic composition. The natural isotopic composition in the table is based on the data [11, 12]. The enriched composition of the target is obtained from the passport for the products obtained in FSUE "Integrated Plant "Electrohimpirbor" plant using the electromagnetic method.
Table 3. Isotopic composition of the enriched and natural targets.

| Nuclide | Natural Composition with combined standard uncertainty (% atom.) | Enriched composition (% atom.) |
|---------|---------------------------------------------------------------|-------------------------------|
| $^{176}$Yb | 12.76±0.41                                                   | 99.59±0.04                    |
| $^{174}$Yb | 31.83±0.92                                                   | 0.29                          |
| $^{172}$Yb | 16.13±0.27                                                   | 0.05                          |
| $^{171}$Yb | 21.83±0.67                                                   | 0.04                          |
| $^{170}$Yb | 14.28±0.57                                                   | 0.02                          |
| $^{168}$Yb | 3.04±0.15                                                    | 0.01                          |
|         | 0.13±0.01                                                    | <0.01                         |

In addition to the main elements of interest from the point of view of specific activity, the target may contain elements that affect the radionuclide purity.

3. Theoretical calculations

Figure 1 shows the theoretically calculated spectrum of neutrons for 176 groups according to experimental data in the central channel (TsK-1) of the IRT-T reactor operating at a power of 6 MW.

![Neutron spectrum](image)

**Figure 1.** Neutron spectra in central channel (TsK-1) of nuclear core (IRT-T).

The experimental data were obtained by measuring a number of neutron detectors of the AKN-T type [13]; subsequently, from the obtained data, the spectrum was theoretically reconstructed by the Monte Carlo method.

To calculate the activity of the irradiated isotopes of lutetium-177, ytterbium-177, ytterbium-175 from the calculated neutron spectrum in the central channel of the IRT-T reactor, we used the evaluated nuclear data libraries processed at 300 K.

To search of the nuclear data of several nuclei and to access numerical values and graphical representations was used the JANIS ver. 4.0 [14]. For the calculation, data for cross-sections from the library JEFF-3.3 were used. Figure 2 and Figure 3 show the cross sections for lutetium and ytterbium, respectively. It can be noted that if only the thermal region of the spectrum is taken into account, this will lead to significant discrepancies with the experimental data, since there is a resonance region of ytterbium (the Resonance Integral for $^{176}$Yb is 6.98 barn compared with the cross section in the...
thermal region, which is about 2.82 barn). It is also necessary to take into account the burnup of lutetium over the entire spectrum, both in the thermal region and in the rest of the neutron spectrum.

Figure 2. Cross-section of $^{177}\text{Lu}$

Figure 3. Cross-section of $^{176}\text{Yb}$

For theoretical calculation used classical equation considering growth, decay and burn-up of producing lutetium-177 in time $t$:

$$\frac{dN_{\text{Yb}}}{dt} = \sigma \cdot \varphi \cdot n_0 - \lambda_{\text{Yb}} \cdot N_{\text{Yb}} - \sigma_{\text{Yb}} \cdot \varphi \cdot N_{\text{Yb}}$$  \hspace{1cm} (2)

$$\frac{dN_{\text{Lu}}}{dt} = \lambda_{\text{Yb}} \cdot N_{\text{Yb}} - \lambda_{\text{Lu}} \cdot N_{\text{Lu}} - \sigma_{\text{Lu}} \cdot \varphi \cdot N_{\text{Lu}}$$  \hspace{1cm} (3)

Where $N_{\text{Yb}}, N_{\text{Lu}}$ is the number of $^{177}\text{Yb}$ and $^{177}\text{Lu}$ nuclei, respectively, $n_0$ is the initial quantity of the substance that will irradiation, $\sigma$ is the cross-section of $^{176}\text{Yb}$, $\varphi$ is the neutron flux, $\sigma_{\text{Yb}}, \sigma_{\text{Lu}}$ is the cross-section of $^{177}\text{Yb}$ and $^{177}\text{Lu}$ respectively, $\lambda_{\text{Yb}}, \lambda_{\text{Lu}}$ is the decay constant of $^{177}\text{Yb}$ and $^{177}\text{Lu}$ respectively.

To use library cross-sections with calculated neutron groups, nuclear cross-sections were convolved according to the formula:

$$\sigma = \frac{\int_{E_0}^{E} \sigma(E) \cdot \varphi(E) dE}{\int_{E_0}^{E} \varphi(E) dE}$$  \hspace{1cm} (4)

Where $\sigma(E)$ is cross-section function versus energy (from the library) $\varphi(E)$ is the neutron flux functions versus energy (from the library) $\sigma$ - is the average cross-section between energy $E_0$ and $E$, after convolved
The activity of $^{177}\text{Lu}$ and $^{177}\text{Yb}$ produced from 10mg of enriched $^{176}\text{Yb}$ according to the system of equations (2-4) was calculated using GCC ver. 11.1 which can used for Scientific Computing in C++ [15] and shown in Figure 6. The Research Nuclear Reactor IRT-T operates during a week for about 100 hours recalculating on 6 MW (the increase in the power of the nuclear reactor is taken into account).

4. Experiments

The enriched ytterbium was irradiated in the central channel (TsK-1) of the IRT-T nuclear reactor (see Fig.5). The central channel is "wet", i.e. filled with water. Before irradiation, targets with ytterbium were sealed in a quartz ampoule. The quartz ampoule was irradiated in an aluminum sealed box. Two Yb$_2$O$_3$ samples with masses of 3.4 and 4.7 mg enriched in $^{176}\text{Yb}$ to 99.59% were irradiated under different conditions. The first sample (4.7 mg) was irradiated at a power of 100 kW for 60 minutes. Assuming linearity between the neutron flux and the reactor power, it can be assumed in the first approximation that the irradiation of the first sample was carried out for 1 minute at the power of the IRT-T reactor 6 MW. The second sample was irradiated for 20 hours at 6 MW. After cooldown (for convenience, it was chosen as 2 days since the operating mode of the reactor was taken into account), aluminum capsules and quartz ampoules located in them were opened, the samples were dissolved in 6M HCl to convert them into the chloride form. The acid was evaporated and the sample was dissolved in bidistilled water. Then the selected aliquots were measured on a Canberra GC2018 detector. The detector is calibrated for efficiency using a point source $^{137}\text{Cs}-^{152}\text{Eu}$ (see Fig.7). The activity of aliquots was calculated using the lines 208.4 keV ($^{177}\text{Lu}$, intensity (I) is 11.7%, $\varepsilon = 0.0485$) and 396.3 keV ($^{175}\text{Yb}$, I = 6.55%, $\varepsilon = 0.0281$).

5. Result and discussion

Figure 6 shows the theoretical results of calculating Activity of nuclides $^{177}\text{Yb}$, $^{177}\text{Lu}$ and $^{175}\text{Yb}$ during three cycles of the reactor working (around 500 hours). Since the reactor operates approximately 100 hours per week, after which its power decreases, the rest of the time the nuclides decay. It is also seen that at the end of the third week, the ratio of the activities of $^{177}\text{Lu}$ (23.05GBq) and $^{175}\text{Yb}$ (2.55GBq) is 9, and ytterbium-177 completely decays. During subsequent operations, the main danger for the experimenter is the radionuclide $^{175}\text{Yb}$ with energy of 396.3 keV.
Figure 6. Activity of nuclides during three cycle of the reactor working

For comparison, the data obtained in Section 3 “Theoretical calculations” during theoretical calculations and Section 4 "Experiments", measured on a semiconductor detector made of high purity germanium, are presented in Table 4. The table contains three nuclides: $^{177}$Lu, $^{177}$Yb and $^{175}$Yb. An experimental determination of the $^{177}$Yb nuclide was not carried out due to technical difficulties.

**Table 4.** Theoretical and experimental data obtained by irradiation of an enriched ytterbium target in the central channel (TcK-1) of the nuclear research reactor (IRT-T)

| Nuclide | Target mass (mg) | Theoretical activity (MBq) | Experimental activity (MBq) |
|---------|-----------------|---------------------------|---------------------------|
| $^{177}$Lu | 3.4             | 620.35                    | 497.42                    |
|         | 4.7             | 0.861                     | 0.805                     |
| $^{177}$Yb | 3.4             | 867                       | -                         |
|         | 4.7             | 60.56                     | -                         |
| $^{175}$Yb | 3.4             | 104.21                    | 97.95                     |
|         | 4.7             | 0.126                     | 0.155                     |

Table 4 shows an acceptable convergence of theoretical and experimental data. The error between theoretical and experimental data is less than 20%. The difference between the experimental and theoretical data is explained by many factors that must be taken into account, such as the nonuniformity of the neutron flux, losses due to the sorption of the target on the walls of the irradiation quartz ampoule, evaporation crucible, and the error in calculating the detection efficiency on the detector.

6. Conclusion

The paper presents the results of a theoretical calculation of the activity of $^{177}$Lu, $^{177}$Yb, $^{175}$Yb taking into account the burnup over the entire energy spectrum of the central channel (TsK-1) of the IRT-T research nuclear reactor. These theoretical calculations are verified experimentally by irradiation of enriched ytterbium targets. The comparison between theoretical and experimental results showed that the difference is less than 20%. The calculations and experiments performed will help in a preliminary assessment of the activity, which can be obtained in subsequent experiments, as well as the necessary radiation protection in the separation of lutetium and ytterbium nuclides.
7. References

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