NUCET NUCLEAR ENERGY AND TECHNOLOGY

Nuclear Energy and Technology 9(4): 281–287
DOI 10.3897/nucet.9.116662

Research Article

Study into the dependence of the Co-60 and Lu-177g production efficiency on the energy structure of neutron flux density*

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Academic editor: Yury Korovin ♦ **Received** 4 February 2023 ♦ **Accepted** 6 March 2023 ♦ **Published** 19 December 2023

Citation: Shaginyan RA, Korobeinikov VV, Stogov VYu (2023) Study into the dependence of the Co-60 and Lu-177g production efficiency on the energy structure of neutron flux density. Nuclear Energy and Technology 9(4): 281–287. https://doi.org/10.3897/nucet.9.116662

Abstract

At present, the existing approaches to production of artificial isotopes are mostly based on the development experience from previous years. This work aims to develop an algorithm for selecting the most effective irradiation modes for target materials. The study is based on sequential modeling of irradiation of target isotopes by neutrons of different 'single-group' fluxes at the same neutron flux density within each energy group (BNAB-93). In this study, a flux density equal to 2×10^{15} n/(cm^2×s) was used for each energy group. This approach will help 'designing' and selecting the actual neutron spectrum that has the highest efficiency compared to alternatives. The study modelled Co-60 and Lu-177g production for each energy group. The kinetics was analyzed in the most efficient groups in terms of specific activity. The maximum specific activity for Co-60 is reached in group 17 and is equal to 1 kCi/g. For the scheme of Lu-177g production through Lu-176 the maximum specific activity is reached in group 26 and is equal to 58.5 kCi/g. For the scheme of Lu-177g production through Yb-176, the maximum specific activity is reached in group 17 and is equal to 260 Ci/g, advantageous for production are groups 15–17 and 26.

Keywords

production, isotopes, specific activity, cross-section, group, Co-59, Co-60, Lu-177g, Lu-176, Yb-176

Introduction

Artificial radioisotopes are used extensively in medicine (diagnosis and treatment), industry (sterilization, flaw detection, in-process inspection), scientific research, and space missions (Isotope Production at the Hanford Site in Richland 1999; Kamovich et al. 2019; Risovanyy 2020; Dzugkoeva et al. 2021; Atom for health 2022; Evdokimov et al. 2022; Shaginyan et al. 2022).

The key applications for medical isotopes are diagnosis and therapy.

The purpose of short-lived isotopes is to irradiate or localize the neoplasm lesions and cease to exist. Apart from short-lived isotopes, a range of medium-lived isotopes (with a half-life of up to six months) and even long-lived isotopes are used in medical therapeutic practice (Atom for health 2022).

Radionuclide therapy is one of the malignant neoplasm treatment techniques. This technique suggests that

* Russian text published: Izvestiya vuzov. Yadernaya Energetika (ISSN 0204-3327), 2023, n. 3, pp. 127–138.

therapeutic radiopharmaceuticals (TRP) are introduced into the patient's body to be accumulated selectively in pathologic tissues. TRPs that form a strong bond with the tumor receptors or antigens are used for treatment. Therefore, the preparation is delivered exactly to the pathologic tissue and destroys it while not affecting the healthy tissue. The ionizing radiation from the radioisotope kills cells by damaging their DNA. There are relatively recent preparations based on Lu-177g.

Radionuclide therapy is used not only for cancer treatment but also for treatment of other diseases, e.g. arthritis (Atom for health 2022).

Production of medical isotopes in power and commercial reactors is complicated due to their activation time not exceeding two or three half-lives. In Russia, medical isotopes are produced in reactors at the Karpov Institute of Physical Chemistry, the Research Institute of Atomic Reactors, the Reactor Materials Institute (IRM), Mayak and Rosenergoatom (Atom for health 2022).

The variety of industrial isotopes is not yet such extensive as that of medical isotopes but the demand for these is great and, accordingly, they are produced in large quantities. The applications for these isotopes include sterilization, flaw detection, X-ray fluorescence analysis, etc. Short-lived isotopes are not used here since they normally have a half-life of several years (STRANA ROSATOM. Cobalt-60: points of growth 2017; Risovanyy 2020; Dzugkoeva et al. 2021). Accordingly, they can be produced, e.g. in the fast neutron side screens: in row 1 (isotopes produced in a fast spectrum in the (n, p) and (n, 2n)) reactions, and in row 2 (in irradiation devices with moderator) (Golubev et al. 1991; Zvonarev et al. 1994; Isotope Production at the Hanford Site in Richland 1999; Maltsev et al. 1999; Varivtsev et al. 2014; Kamovich et al. 2019; Risovanyy 2020; Dzugkoeva et al. 2021; Evdokimov et al. 2022; Shaginyan et al. 2022).

The most commonly used industrial isotope is Co-60 with hard gamma radiation which is employed primarily for sterilizing (products in storage, medical appliances, etc.) and, accordingly, the demand for this isotope is not great (tens of MCi per year) (STRANA ROSATOM. Cobalt-60: points of growth 2017). Largely, it is produced in thermal power reactors (CANDU, RBMK), but no high activity (in excess of 80 to 100 Ci/g) can be achieved in these reactors. A fast reactor makes it possible to produce highly active cobalt (more than 300 Ci/g). This isotope is in short supply worldwide and its shortage will only grow.

Description of computational models

Production of any isotope is linked inseparably with the target material cross-sections, the energy structure and the irradiation and cooling times.

Fundamental to investigating the efficiency of the artificial isotope production depending on the energy structure of the neutron flux spectrum is to define the energy structure of the neutron flux spectrum properly. For the efficiency investigation, this paper presents the neutron source spectrum in a 28-group approximation (BNAB-93 (Manturov et al. 1996). The calculations were conducted in the ISKRA code (version 1) for the calculation of the fuel and structural material radiation performance (Tormyshev 2022). It needs to be noted that the calculations were conducted without taking into account the cross-section blocking effect.

As part of the studies, the irradiation of the target neutron isotopes was simulated successively for each energy group (that is, all neutrons were emitted successively by each of the 28 energy groups) with an equal neutron flux density of $2 \cdot 10^{15}$ neutron/($cm^2 \cdot s$) (similarly to the maximum intensity given in Tarasov et al. 2013) within different time intervals: 2 to 12 days under irradiation and further 2.5 days of cooling without irradiation for short-lived isotopes, and 0.5 to 2 years under irradiation and 30 days of cooling without irradiation for long-lived isotopes. A similar approach was used in Kosyakin et al. 2022. The following irradiation times have been selected: 0.5 to 2 half-lives for short-lived isotopes, and proceeding from the fuel life for long-lived isotopes (1 refueling cycle = 0.5 years).

The specific activities presented in the paper are characterized as specific activities of irradiated target materials and are presented in Ci/g of the starting metal.

The microscopic cross-sections as a function of neutron energy are presented in Table 1.

Calculation results for Co-60 production

The most extensively used artificial radioisotope is Co-60. The gamma radiation sources using Co-60 are somewhat of a standard option in all cases which require photons with an energy of over 1 MeV.

The applications for Co-60 are varied.

1. Sterilization of medical products and different foods.
2. Industrial applications for sources with Co-60:
   - thickness and density gages;
   - gamma ray flaw detection (used largely for thick welds);
   - height gages (rarely);
   - irradiation of plastic polymers.
3. Medicine (gamma knife and brachytherapy).
4. Science.

The annual average consumption of Co-60 is estimated at about 2.25 MCi for medicine, and 55 MCi for industry (STRANA ROSATOM. Cobalt-60: points of growth 2017).

Co-60 is produced by irradiating the target consisting of natural Co-59. Fig. 1 presents microscopic cross-sections of Co-59 and Co-60 (logarithmically scaled) as a function of neutron energy.

Table 1. Microscopic cross-sections as a function of neutron energy.

Group No.	E_{low}, eV	E_{up}, eV	Co-59 cross-section, barn	Co-60 cross-section, barn	Lu-176 cross-section, barn	Lu-177g cross-section, barn	Yb-176 cross-section, barn
1	2	3	4	5	6	7	8
-1	1.40E+07	2.00E+07	6.37E-04	6.17E-04	3.31E-03	9.89E-04	9.85E-04
0	1.05E+07	1.40E+07	7.10E-04	7.25E-04	2.66E-03	1.34E-03	1.30E-03
1	6.50E+06	1.05E+07	8.43E-04	4.15E-04	2.57E-03	9.27E-04	7.43E-04
2	4.00E+06	6.50E+06	1.40E-03	2.73E-04	5.87E-03	1.57E-03	9.54E-04
3	2.50E+06	4.00E+06	2.48E-03	3.62E-04	2.27E-02	8.58E-03	4.89E-03
4	1.40E+06	2.50E+06	4.13E-03	5.65E-04	8.60E-02	3.75E-02	1.66E-02
5	8.00E+05	1.40E+06	7.60E-03	7.20E-04	2.28E-01	8.16E-02	1.90E-02
6	4.00E+05	8.00E+05	7.26E-03	1.08E-03	4.14E-01	1.01E-01	1.61E-02
7	2.00E+05	4.00E+05	9.95E-03	2.04E-03	6.36E-01	1.63E-01	1.89E-02
8	1.00E+05	2.00E+05	1.51E-02	3.29E-03	7.98E-01	3.19E-01	2.98E-02
9	4.64E+04	1.00E+05	2.15E-02	5.33E-03	1.02E+00	5.12E-01	7.17E-02
10	2.15E+04	4.64E+04	3.66E-02	9.13E-03	1.42E+00	7.74E-01	1.10E-01
11	1.00E+04	2.15E+04	5.99E-02	1.52E-02	2.19E+00	1.17E+00	1.68E-01
12	4.64E+03	1.00E+04	1.61E-01	1.27E-02	3.56E+00	1.83E+00	2.57E-01
13	2.15E+03	4.64E+03	1.89E-01	3.71E-03	5.87E+00	3.06E+00	3.94E-01
14	1000	2.15E+03	3.40E-02	3.01E+00	9.81E+00	5.44E+00	5.37E-01
15	464.16	1000	4.83E-02	7.96E-02	1.63E+01	9.88E+00	1.93E+00
16	215.44	464.16	2.70E-01	3.29E-02	2.66E+01	1.79E+01	1.61E+00
17	100	215.44	7.38E+01	3.42E-02	4.12E+01	3.19E+01	2.04E+00
18	46.42	100	3.17E+00	4.37E-02	1.06E+02	2.58E+01	3.34E-01
19	21.54	46.42	1.68E+00	6.01E-02	1.17E+02	3.86E-01	5.97E-02
20	10	21.54	1.88E+00	8.59E-02	7.25E+01	3.66E+00	1.02E-01
21	4.64	10	2.48E+00	1.24E-01	1.42E+02	6.30E+01	1.63E-01
22	2.15	4.64	3.48E+00	1.81E-01	6.39E+01	1.11E+04	2.48E-01
23	1	2.15	5.00E+00	2.65E-01	5.54E+02	6.01E+02	3.72E-01
24	0.46	1	7.27E+00	3.88E-01	4.06E+01	3.30E+02	5.50E-01
25	0.22	0.46	1.06E+01	5.70E-01	4.38E+02	3.58E+02	8.11E-01
26	0.0253	0.0253	3.08E+01	1.66E+00	3.65E+03	8.56E+02	2.37E+00

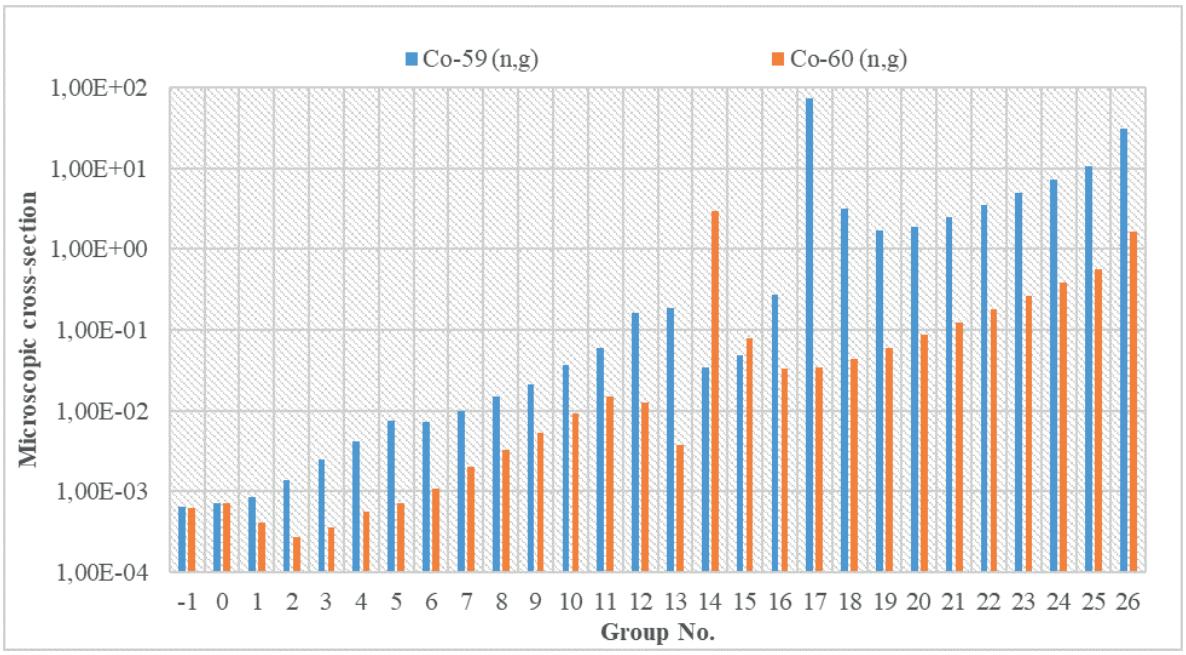

Figure 1. Microscopic cross-sections of Co-59 and Co-60 as a function of neutron energy (Manturov et al. 1996).

It can be seen in the figure that the target isotope cross-section grows noticeably in groups 17 through 26. It is not only high cross-sections of the target isotope are noted but also rather low cross-sections of the desired isotope which differ by an order of magnitude and more.

The following periods of irradiation for production of Co^{60} were assumed for the calculations taking into account its $T_{1/2}$ = 5.27 years: 4 half-life steps and 30 days of cooling (after two years of irradiation) are enough for understanding the overall picture of efficient production.

The Co-59 target material density is equal to 8.9 g/cm^3.

Fig. 2 presents the results of calculating the Co-60 specific activity as a function of the spectrum energy structure and time.

It follows from the results that the most efficient group is group 17 (the peak activity is more than 1 kCi/g). This result is quite evident: it can be seen from Fig. 1 that Co^{59} has one sufficiently broad resonance, and it is exactly the above group that falls within it, which is characterized by high interaction cross-sections and, accordingly, by high reaction and production rates.

Groups 17 and 16 exhibit the specific activity rise that differs from other groups. As judged by the diagram, it can be noted that there is a minor drop in specific activity observed after 30 days of cooling and a conclusion is made therefore that groups 17 and 26 have the maximum specific activity (the concentration of Co-60 becomes so large that even the interaction cross-sections for Co-59 exceeding those for Co-60 cannot maintain such specific activity level) which is achieved in these groups for one year of irradiation and amounts to 1 kCi/g and ~ 800 Ci/g respectively.

If we consider cobalt with an average specific activity of ~ 100 Ci/g as the desired product, groups 17 through 26 can be called the effective production region since the required values are achieved in these for a time interval of six months (groups 17 and 22 through 26) to one year (groups 19 through 21). If cobalt with a high specific activity is considered, values of ~ 200 Ci/g are achieved for a time interval of six months (groups 17 and 24 through 26) to two years (groups 19 and 20).

At the same time, no maximums have been achieved for some groups for two years either due to a low ratio of the target and desired isotope cross-sections, or because of an insufficient irradiation time.

Calculation results for the Lu-177g production

Lu-177g is one of the most promising radionuclides which form the basis for production of modern radiopharmaceuticals. It is highly efficient in target therapy for a range of oncological diseases, including metastatic forms of castration and resistant prostate gland cancer, neuroendocrine tumors, and others. In parallel with its anticancer properties, the radionuclide is explored for applicability in nononcologic therapies, e.g., in local Lu-177g therapy for inflammatory diseases of joints.

The paper considers two desired isotope production patterns (Tarasov et al. 2013):

- Lu-176 (n,γ) Lu-177g reaction;
- Yb-176 (n,γ) Yb-177g Lu-177g reaction.

Figs 3, 4 present microscopic cross-sections (logarithmically scaled) of Lu-176 (Yb-176) и Lu-177g as a function of neutron energy.

As shown by these figures, the neutron capture cross-sections on the Lu-176 target isotope have high values, which is similar to the cross-sections on the Lu-177g desired isotope, this suggesting the fastest possible occurrence of a steady equilibrium state.

The situation is different with the Yb-176 target material. Its cross-sections are an order of magnitude or still smaller than the desired isotope. This limits greatly the maximum possible production due to the desired isotope burnup.

Another factor to be taken into account is a short half-life equal to 6.6 days. In connection with this, the irradiation periods, as compared with the previous case, have been adjusted by 2 to 12 irradiation days and 2.5 days of cooling after irradiation.

The figures below present the Lu-177g production from Lu-176 (Fig. 5) and from Yb-176 (Fig. 6) as a function of the neutron flux energy structure and irradiation time.

Fig. 5 shows that group 26 exhibits the behavior similar to that for production of Co-60 (see Fig. 2), and the occurrence of the maximum specific activity with the subsequent burnup of the desired isotope. The maximum among all groups is observed exactly in the group in question being at a level of about 58.5 kCi/g and produced in the course of two irradiation days.

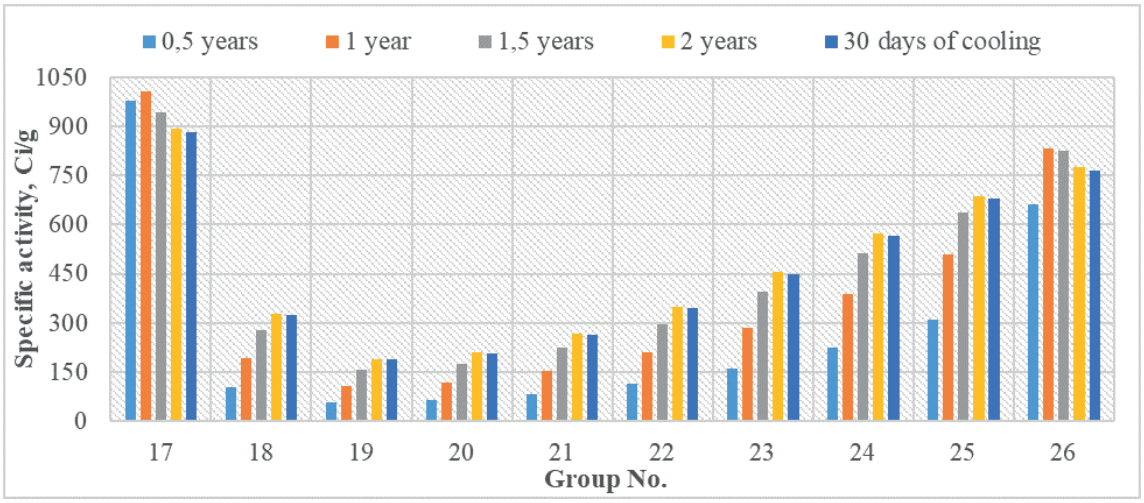

Figure 2. Production of Co-60 as a function of the neutron flux spectrum energy structure and irradiation time.

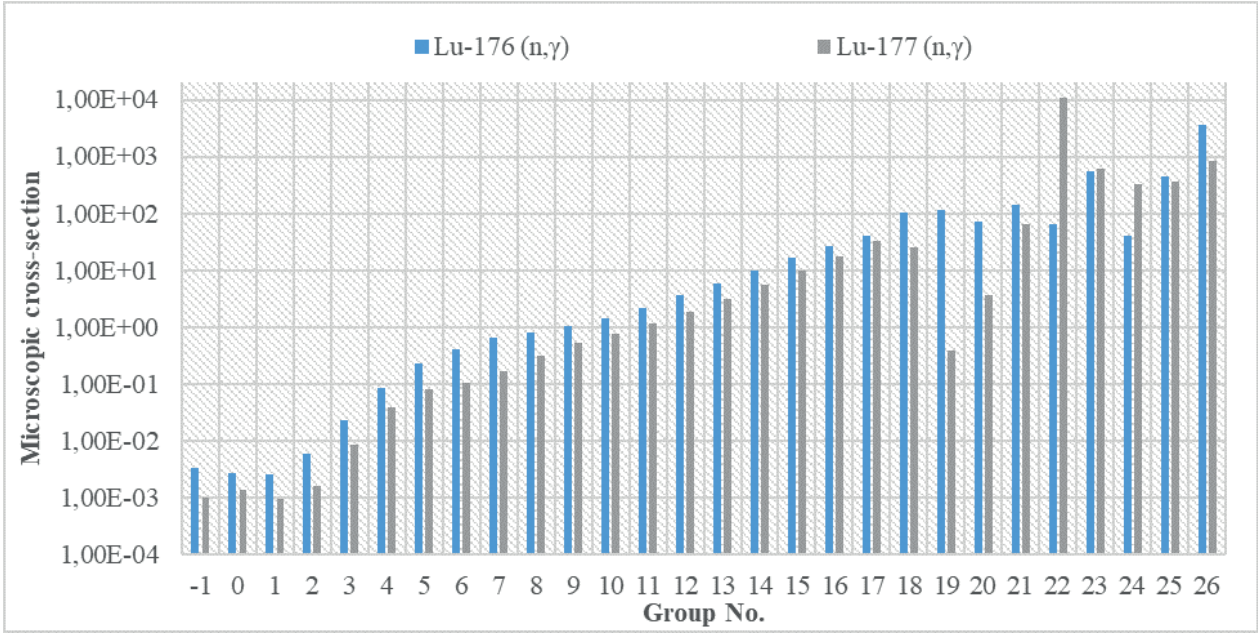

Figure 3. Microscopic cross-sections of Lu-176 and Lu-177g as a function of neutron energy (Manturov et al. 1996).

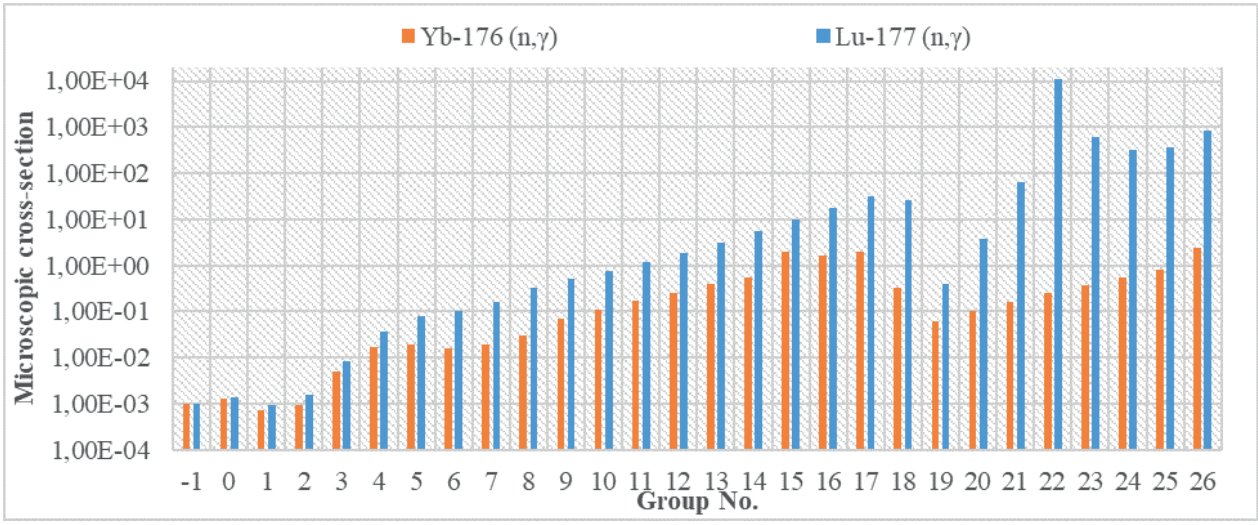

Figure 4. Microscopic cross-sections of Yb-176 and Lu-177g as a function of neutron energy (Manturov et al. 1996).

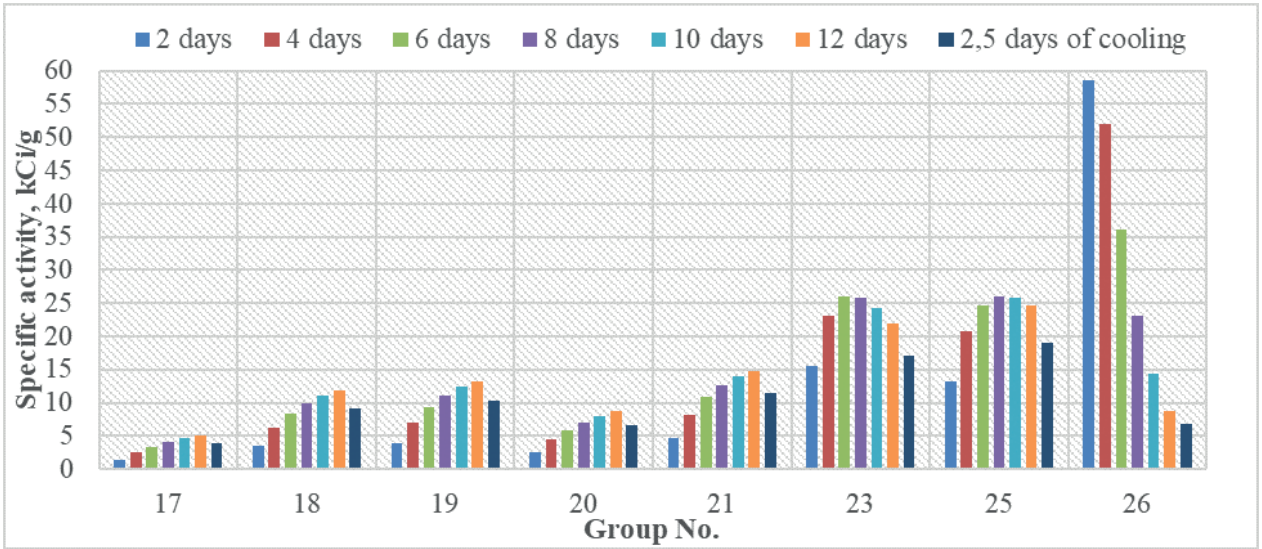

Figure 5. Production of Lu-177g from Lu-176 as a function of the neutron flux spectrum energy structure and irradiation time.

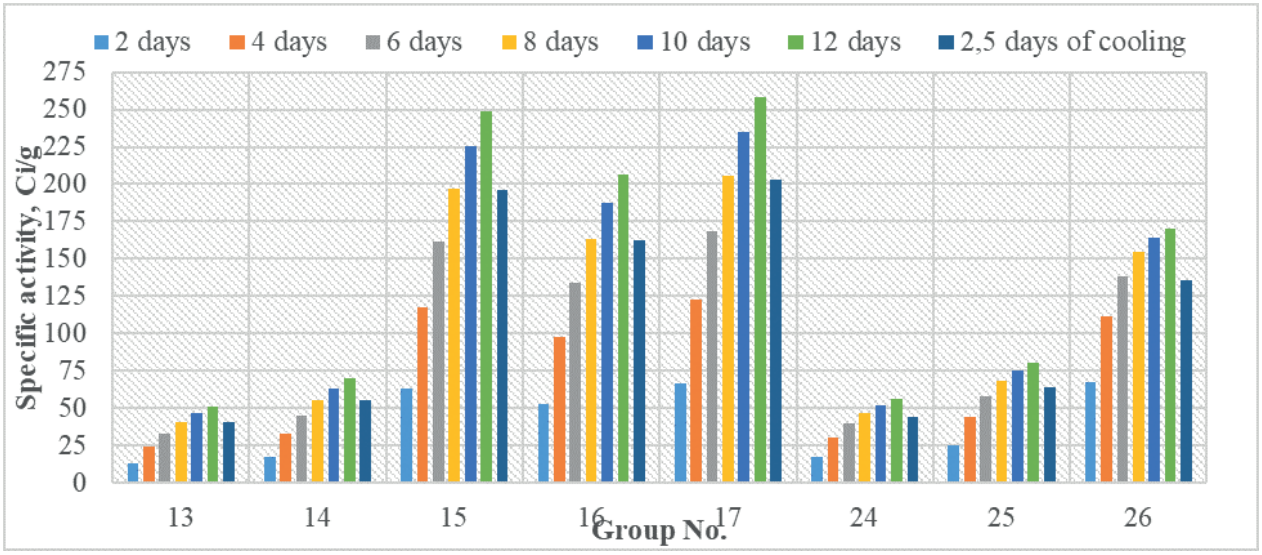

Figure 6. Production of Lu-177g from Yb-176 as a function of the neutron flux spectrum energy structure and irradiation time.

Production is similar for groups 23 and 25. An equilibrium mode is established on the sixth to eighth day of irradiation equal to the half-life of the desired isotope which leads naturally to a reduced specific activity in further irradiation cycles. The maximum specific activity in these cases is at a level of ~ 25.5 kCi/g.

The result is different for groups 17 through 21. In these cases, no equilibrium concentration is achieved and accumulation takes place in all irradiation cycles.

The effects of cooling for 2.5 days cannot be neglected. Calculations show that the specific activity drop is ~ 22%.

Fig. 6 presents the result of lutetium production from ytterbium. Neither maximum specific activity (with further burnup) nor equilibrium state was achieved in any group in the course of irradiation. In this connection, lutetium is produced practically in each group according to one scenario, that is, with the specific activity growth throughout the irradiation cycle.

The maximum specific activity is observed in group 17 and amounts to ~260 Ci/g. Group 15 is also close to these figures with the specific activity equal to 250 Ci/g. Group 16 has somewhat similar scenario and production rates with groups 15 and 17 and is slightly behind in terms of the maximum specific activity value but the maximum is achieved in the same period as in the event of the above-mentioned groups.

The target figures are achieved for group 26 for the entire irradiation period (12 days). After six days of irradiation, however, there is a marked slowdown observed in the specific activity growth rates. The maximum specific activity values for this group are at a level of 170 Ci/g.

As in the case with production of Lu-176, attention needs to be given to the loss of specific activity in the event of cooling for 2.5 days, and the average loss for all groups is about 20%.

The following conclusions can be made based on the calculations and analysis of the results obtained. Groups 18 through 21, 23 and 25 (six days of irradiation) and group 26 (two days of irradiation) are advantageous for the Lu-177g production via Lu-176 taking into account the application peculiarities and its half-life.

The highest production for the other Lu-177g production pattern (via Yb-176) is in groups 15 through 17 and 26.

The results of the studies will help with the design of the irradiation device with the most effective production of the isotopes considered in the paper.

Conclusions

The results of the distributions for the 60-Co and Lu-177g production dependences on the neutron flux spectrum energy structure and irradiation time have been obtained. The groups presented in the results are in accordance with the BNAB-93 group representation (Manturov et al. 1996).

Based on the calculation results for the Co-60 production, two groups (17 and 26) have shown the maximum specific activity which is achieved for one year of irradiation, the specific activity in these groups being 1 kCi/g and ~ 800 Ci/g respectively. The preferred groups have been chosen in the specific activity approximated to the practical application. If cobalt with an average specific activity of ~ 100 Ci/g is considered as the desired product, groups 17 through 26 can be called the efficient production region since they include the required values for the period of six months (groups 17 and 22 through 26) to a year (groups 19 through 21). If cobalt with a high specific activity is considered, figures equal to ~ 200 Ci/g are achieved in a time interval of six months (groups 17 and 24 through 26) to two years (groups 19 and 20).

Based on the calculation results for the Lu-177g production from Lu-176, a specific activity of ~58.5 kCi/g, the maximum one for 2 days of irradiation, is observed in group 26. The occurrence of an equilibrium mode is observed on the sixth to eighth day in groups 23 and 25, the specific activity being ~ 25.5 kCi/g.

Based on the calculation results for the Lu-177g production from Yb-176, the maximum specific activity is

achieved in groups 15 and 17 amounting to 250 and ~ 260 Ci/g respectively. One cannot miss as well the fact that there are groups 16 and 26 with a specific activity of 207 and 170 Ci/g respectively, which is achieved for the entire irradiation period (12 days).

The obtained distributions of production by groups, with regard for time, give a clear notion of the efficiency of accumulation across the energy interval which makes it possible to conclude on the preferred spectral characteristics of particular nuclides and assist with forming the most effective neutron spectrum.

The results include cases of achieving an equilibrium state, both the maximum one with further burnup and insufficiently irradiated.

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