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Research Article

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Computational investigation of 1 up to 3 kCi Co-60 source production at external irradiation boxes of TRR core using MCNPX code

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HIGHLIGHTS

- Kilocurie Co-60 gamma sources are used in low-scale industrial irradiator cells.
- Kilocurie gamma sources could be produced in research reactors.
- Selection of the best irradiation position inside the research reactor helps to obtain the highest possible yield.

ABSTRACT

In many human diseases and health cases, therapy of blood transfusion becomes necessary. In spite of the necessity, there are some risks associated with blood used in the blood transfusion process. The TA-GVHD (transfusion-associated graft-versus-host-disease) is a problem when a blood transfusion occurs. The blood irradiation with gamma rays in blood bags can eliminate this risk. It should be mentioned that Co-60 sources are widely used for such blood irradiators. The present work investigates Co-60 production yield inside the external irradiation boxes of Tehran Research Reactor (TRR) using MCNPX code. 10-rod and 4-rod Co-59 assemblies were modeled at different external irradiation boxes to investigate their negative reactivity impact on TRR core as well Co-60 buildup rate during 3 years operation of the nuclear core at 4 MW power. The obtained results from MCNPX code showed a 4-rod assembly in linear form could obtain the highest specific activity ($Ci.g^{-1}$) inside the external irradiation box faced to the core center. The computational results showed about 8 kCi of Co-60 is produced at the optimized irradiation position after 3 years TRR operation at 4 MW power.

KEYWORDS

Gamma blood irradiator
Co-60 production yield
TRR
MCNPX code

HISTORY

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1 Introduction

One of the most application of kilocurie Co-60 sources is blood irradiators, which are used in hospitals and blood transfer centers. Irradiation of human blood is used to avoid the TA-GVHD (transfusion-associated graft-versus-host-disease), a rare but devastating adverse effect of leukocytes present in blood components for immunocompetent transfusion recipients. Usually, this irradiation practice is performed to a physical elimination of lymphocytes. A proper dose in a range of 15 Gy to 50 Gy is needed usually to be delivered to the blood in the bag collected in a blood tissue bank (Boghi et al., 2008).

Around 10% of all donated blood is irradiated prior to transfusion. Clinically insignificant differences were reported in terms of red cell membrane permeability between X- and gamma-radiation uses, with both being equally effective at reducing TA-GvHD. A recent 2016 survey conducted in the U.S. (but including some international organizations) reported that the most frequent requirement for irradiated blood products is during transfusion from blood relatives in conjunction with histocompatibility testing of the human leukocyte antigen (HLA) gene complex matching. Neonatal exchange transfusions, intrauterine transfusions, and pre-term or low birth-weight babies also have a high requirement for irradiated blood, although patient groups deemed to be less at risk of TA-GvHD might still require irradiated blood products; there are differences in the requirements for the use of irradiated blood between nations and organizations. The British Society for Hematology, during an extensive literature review, also concluded that X- and gamma-irradiation could be considered equivalent in their ability to reduce TA-GvHD incidence (Barnard et al., 2020).

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Figure 1: X-ray blood irradiator cell view (https://bmskgroup.com/x-ray-irradiation-systems).

In overall, blood and its components can be irradiated with gamma rays by Cs-137, Co-60, or X-ray sources (Moroff et al., 1997). Gamma ray irradiators are monoenergetic while x-rays have an energy spectrum. All of these differences imply that there is a periodical need for revision of the protocols of blood product irradiation. Due to problems such as radioactive source age, problems with source replacement, and protection risks, gamma sources are replacing with linear accelerators (Linac) (Aboufazeli et al., 2018).

However, different gamma blood irradiators were used over the world. For example, the Gamma irradiator (Blood Irradiator-2000, developed by Board of Radiation and Isotope Technology (BRIT), India) has a Co-60 source capacity of 675 Ci with photon energies 1.17 MeV and 1.33 MeV (Mhatre et al., 2008; Nagaraj et al., 2013).

Figure 1 shows an X-ray blood irradiator. Its capacity is 4 to 6 blood bags (up to 1800 ml per cycle). Its cycle is up to 6 × 300 ml blood bags in 4 minutes with a dose between 25 Gy and 30 Gy (https://bmskgroup.com/x-ray-irradiation-systems).

X-ray irradiators can cost 250,000 \$ to 300,000 \$ with annual service contract costs running 15,000 \$ to 20,000 \$ (https://www.captodayonline.com/new-rays-blood-safety/). Usually self-contained dry source storage irradiators containing Cs-137 or Co-60 gamma ray sources with activity up to 3 kCi are used as blood irradiator facilities (Bagheri and Ranjbar, 2022).

Co-60 irradiator has multiple fixed sources arranged around a central cavity. These units are stocked with sufficient Co-60 that reloading of the radionuclide is not required for 10 to 15 years. About 31 numbers of Co-60 blood irradiator are reported to be located at U.S (Use, 2008).

A published report denoted that the University of California owned 42 Cs-137 irradiators for a wide range of purposes such as treating blood, exposing cells and small animals. This report has pointed out to the fact that several considerations must be taken into account when replacing a Cs-137 irradiator with a suitable X-irradiator; as discussed, RBE (Relative Biologic Effectiveness, RBE is the ratio of the dose of a reference radiation) values can have a wider range in evaluations using X-rays depending on the energy used in comparison (Barnard et al., 2020).

Another report indicated that using an RS2000 160 kVp X-ray irradiator instead of a Model 68-A Cs-137 irradiator, adds a maximum ∼5% uncertainty for 25 g rodents, and maximum ∼8% uncertainty for ∼50 g rodents. Also, the report showed that using an Precision X-Ray XRAD 160 kVp X-ray irradiator instead of a PNNL (Pacific Northwest National Laboratory) custom Cs-137 irradiator, adds a maximum ∼9% uncertainty for ∼25 g rodents, and maximum ∼13% uncertainty for ∼50 g rodents (Mackenzie and Smith, 2018).

Gamma ray emitters like Co-60 became popular radiation sources for medical and industrial applications. Many gamma ray irradiators have been built. It is estimated that about 200 are currently in operation in Member States of the International Atomic Energy Agency (IAEA). In recent times, the use of electron accelerators as a radiation source (and sometimes equipped with X-ray converter) is increasing. However, gamma irradiators are difficult to replace, especially for non-uniform and high-density products. Co-60 is almost solely used as the gamma radiation source for industrial use now mainly because of its easy production method and its non-solubility in water. However, the use of Cs-137 has been limited to small self-contained, dry-storage irradiators, used primarily for the irradiation of blood and for insect sterilization. Currently, all industrial radiation-processing facilities employ Co-60 as the gamma radiation source (IAEA, 2004).

A gamma cell blood irradiator (Gammacell 3000 Elan; Best Theratronic, Ottawa, Canada) in the Shiraz Blood Transfusion Center was used to irradiate cellular blood products to inactivate T lymphocytes in order to prevent graft-versus-host disease (Fig. 2). The unit has four main components: radiation shield, sample chamber, radiation sources, and control system. The radiation source in the Gammacell irradiator is the Cs-137 in the form of powder embedded in two rod sources with total activity of 1356 Ci (50.2 TBq). Each Cs-137 source is double encapsulated in stainless steel and permanently installed within the radiation shield. The sample chamber has a turntable and removable stainless steel beaker (12 cm diameter and 20 cm height) in which samples or blood products are placed (Mohammadyari et al., 2014).

As Fig. 3 shows, Co-60 sources with less than 3 kCi could be used for some medical and industrial applications such as strawberry, potato, mushroom treatment industry, level-meter nuclear devices, blood products and so on (https://escies.org/webdocument/showArticle?id=251) and (Fernandes et al., 2012; Van Tuyle et al., 2003).

Figure 2: The gamma blood irradiator (a), gel phantom (b), and gel phantom inside the blood container ready to be irradiated (c) (Mohammadyari et al., 2014).

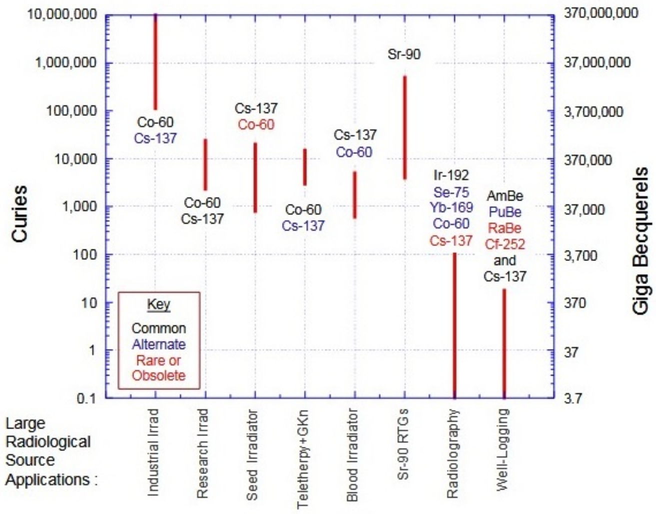

Figure 3: Different application of Co-60 sources with altered activities (Van Tuyle et al., 2003).

Whereas Co-60 irradiators could be effectively used in hospitals and medical centers, as well as the industrial units, the source production possibility with an activity between 1 kCi to 3 kCi in Tehran Research Reactor (TRR) is going to be investigated in the present work using simulation methods.

2 Material and methods

The simulations have been performed by using the MCNPX2.7 code. MCNPX is a general-purpose Monte Carlo radiation transport code with a multi-tasking capability that can be used to reduce the time needed to obtain the computational result (Pelowitz, 2008). The MCNPX code was developed during the last three decades at Los Alamos National Laboratory and can be considered "the state-of-the-art" Monte Carlo code. This code is capable of transporting 36 elementary particles at all energies, and its generalized geometry features and use of continuous-energy cross-sections can generate benchmark-quality results for a variety of nuclear applications (Menarebazari et al., 2023).

TRR core was modeled in details using MCNPX code. The existing TRR is a 5 MW pool-type research reactor with MTR type fuel elements of low enriched Uranium (LEU) with 20% enrichment. The core is built upon an aluminum grid plate, on which there are $9 \times 6 = 54$ holes to accept either fuels or other tools such as irradiation boxes.

Cobalt metal rods with a dimension of 0.7 cm $\times$ 30 cm that were encapsulated inside an aluminum holder were modeled at an external irradiation box of the TRR core (between two graphite reflectors). The rods dimension was chosen according to Ref (Khalafi and Gharib, 2005). The MCNPX model is observed in Fig. 4.

First, two 10-rods and 4-rods configurations were investigated for the production of Co-60 in the irradiation box placed at the core corner. The first irradiation box was selected because of the least possible negative reactivity on the TRR core due to Co-59 neutron absorber loading (Fig. 5). The Co-60 product yield is calculated using the BURN card of MCNPX, which uses CINDER software to solve batman equations as the following.

Buildup of different isotopes inside the core during the reactor core operation is another important concern especially in view of the fissile inventory of the modeled core at any time steps of the core burnup. The values were calculated using the BURN card of the used computational code. CINDER code originally developed for the reactor physics community, while the code today spans applications ranging from reactor burn-up over accelerator driven transmutation to accelerator activation and to astrophysics in the evolution of elements and isotopes since the birth of the universe (Fensin, 2008). Mathematically, the material balance process can be described at any time by the following depletion equation:

$$\begin{aligned}\frac{\mathrm{d}N_i}{\mathrm{d}t} &= \sum_j \gamma_{ij}\sigma_{fj}N_j\varphi + \sum_k \sigma_{ck\to i}N_k\varphi + \sum_l \lambda_{l\to i}N_l \\ &\quad - (\sigma_{fi}N_{ji}\varphi + \sigma_{\alpha i}N_{ji}\varphi + \lambda_i N_l)\end{aligned} \tag{1}$$

where $\frac{\mathrm{d}N_i}{\mathrm{d}t}$ is time rate of change in concentration of isotope i, $\sum_j \gamma_{ij}\sigma_{fj}N_j\varphi$ is production rate per unit volume of isotope i from fission of all fissionable nuclides, $\sum_k \sigma_{ck\to i}N_k\varphi$ is production rate per unit volume of isotope i from neutron transmutation of all isotopes including (n,γ), $(n,2n)$, etc. $\sum_l \lambda_{l\to i}N_l$ is production rate per unit volume of isotope i from decay of all isotopes including β^-, β^+, α, γ, etc. $\sigma_{fi}N_{ji}\varphi$ is removal rate per unit volume of isotope i by fission, $\sigma_{\alpha i}N_{ji}$ is removal rate per unit volume of isotope i by neutron absorption (excluding fission), and $\lambda_i N_l$ is removal rate per unit of isotope i by decay (Gallmeier et al., 2010). Thermal neutron flux (n.s^{-1}.cm^{-2}) as well as deposited heat (W) caused by neutron and gamma were calculated inside any rod using the computational code. Mesh tally card of the code was used

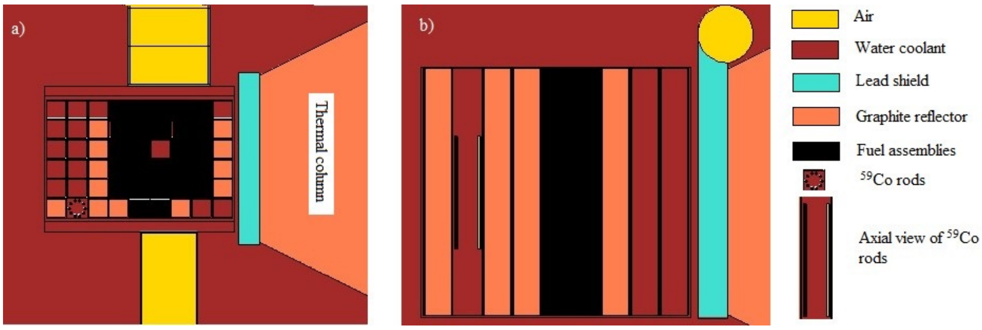

Figure 4: Co-59 rod simulation at the external irradiation box of TRR core, a) cross-sectional view of the core b) axial view of the core.

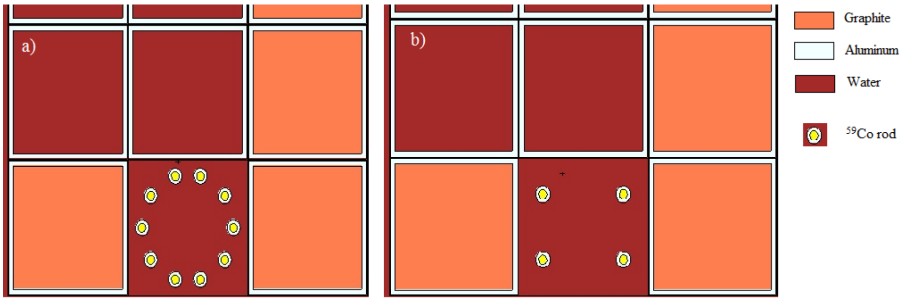

Figure 5: Co-59 rod simulation at the external irradiation box of TRR core, a) 10-rods b) 4-rods.

to model 2D distribution of the neutron deposited heat density (W.cm^{-3}).

In addition, reactivity reduction of TRR core by loading the neutron absorber Co-59 rods was investigated for both 10-rods loading and 4-rods loading. Co-60 yield (Ci) and specific yield (Ci.g^{-1}) were calculated for the modeled rods during their burnup for one, two and three years TRR core operation at 4 MW power (25% less than full power operation of TRR).

Another investigation was done on reduction of the Co-59 rod diameter from 0.7 cm to 0.5 cm and its effect on the product specific yield enhancement. All the above-mentioned calculations were done for the narrower rods too. The Co-59 rods were encapsulated inside an aluminum can with 2 mm thickness.

Another 4-rod assembly arrangement in linear form was selected to investigate the self-shielding effect of rods on each other. Clearly, reduction of cobalt mass loading decreases the Co-60 activity but increases its specific activity because of self-shielding effect reduction. Therefore, to increase the specific activity, the 4-rod assembly was modeled at an irradiation box faced to the core center for the next simulations. To decrease the negative reactivity induction of the cobalt neutron absorbers, the irradiation position was selected after the reflector boxes of TRR core. The new arrangement was modeled in the central external irradiation box to increase Co-60 production rate because of the highest thermal neutron flux, which is available at this box. First, the 4-rod arrangement position was considered at the center of the selected irradiation box. The above-mentioned parameters were calculated for this new arrangement and position. At the next step, the 4-rod arrangement position was modeled in the border of the irradiation box (closer to the TRR core) to investigate the effect of this change on the product yield enhancement after removing the water coolant neutron moderator (about 3 cm) by this displacement than the investigated pervious one. The above-mentioned parameters were calculated for this new position too.

3 Result and discussion

3.1 *Investigation of 10-rod and 4-rod cobalt assembly irradiation at the TRR core corner*

First, 10 Co-59 metal rods (0.5×30 cm^2) were simulated in the corner external irradiation box according to Fig.4. Some parameters such as neutron/gamma deposited heat, average thermal neutron ($E_n < 0.6$ eV) flux and Co-60 production yield (Ci) were calculated for them using MCNPX code. The calculations were repeated for the rods with a thicker diameter (0.7×30 cm^2). Figure 6 shows the Co-60 activity buildup in irradiated metal rods during TRR operation at 4 MW power (the average operational power of the reactor during one year). As the figure shows, after three years irradiation of the thicker rods about 6 kCi of Co-60 is produced in 10-rod assembly. While the product yield inside the thinner rods would be about 5 kCi after the same irradiation condition.

The assembly geometry was changed to 4-rod assembly according to Fig. 5 and all the above-mentioned calculations were repeated for them. Figure 7 shows the Co-60 activity buildup in the irradiated metal rods during TRR operation at 4 MW power. As the figure shows, after three years irradiation of the thicker rods about 4 kCi of Co-60 is produced in 4-rod assembly. While the product yield

Table 1: Comparison of some neutronic parameters by loading 10-rod and 4-rod Co-59 assemblies in irradiation box of TRR.

Co-59 rod specification	Neutron heat (W)	Gamma heat (W)	Average thermal neutron flux ($n.s^{-1}.cm^{-2}$)	Specific activity of the product after 3 years ($Ci.g^{-1}$)	Negative reactivity (pcm)
10-rod assembly					
0.5×30 cm^2	0.26	152	4.14×10^{12}	10.3	31
0.7×30 cm^2	1.19	248	2.74×10^{12}	6.19	45
4-rod assembly					
0.5×30 cm^2	0.33	63.3	5.37×10^{12}	13.6	4
0.7×30 cm^2	0.47	108	4.87×10^{12}	9.59	12

inside the thinner rods would be about 3 kCi after the same irradiation condition.

Table 1 compares the neutron/gamma deposited heat, specific activity, average thermal neutron flux and negative reactivity caused by Co-59 neutron absorber loading in TRR core for any simulated cases. All the simulation calculation errors were less than 3%.

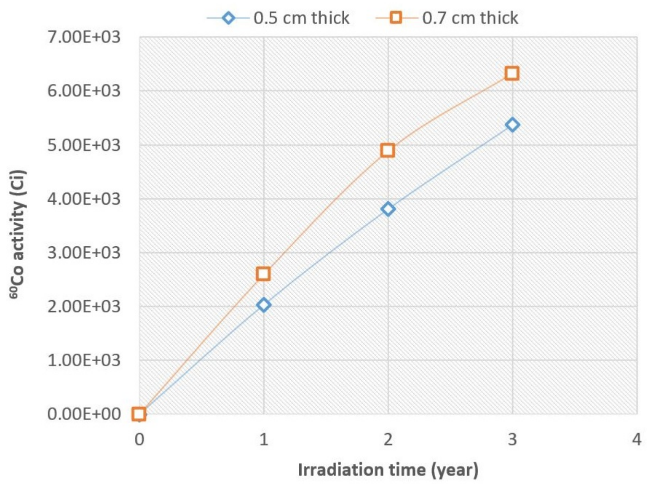

Figure 6: Comparison of Co-60 buildup inside the 10-rod assembly of Co-59 with 0.5 cm and 0.7 cm rod diameter, respectively.

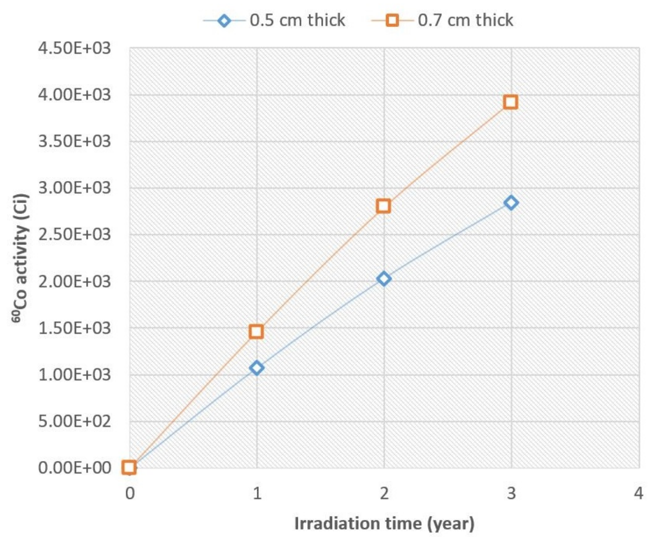

Figure 7: Comparison of Co-60 buildup inside the 4-rod assembly of Co-59 with 0.5 cm and 0.7 cm rod diameter respectively.

The carried out calculations showed there would not be significant negative reactivity in consequence of the Co-59 neutron absorber inside the corner external irradiation box of the TRR core.

Thermal flux distribution inside the corner irradiation box (according to Fig. 4) with loading 4-assembly and 10-assembly Co-59 rods was investigated using a mesh tally card of MCNPX code. The obtained results showed the anterior rods mask the thermal neutron flux of reaching to the posterior rods so that they may be received a third thermal neutron flux or even more in comparison the anterior ones (Fig. 8).

3.2 *Investigation of 4-rod cobalt assembly irradiation at the irradiation box face to the TRR core center*

Hence, another arrangement was considered for the Co-59 rod assembly so that all the rods are in a line according to Fig. 9. Whereas the negative reactivity insertion because of cobalt loading inside the investigated irradiation box was insignificant, in order to the Co-60 product yield enhancement the assembly position was considered in front of the core center.

The carried out calculations for the new 4-rod arrangement showed the Co-59 rods gamma heats are 34.7, 33.2, 32.3, and 32.8 W respectively, which concludes in 172 W total gamma deposited heat inside the assembly; that 133 W belongs to the cobalt rods and the remaining belongs to the aluminum clad. The total neutron deposited heat of the assembly is less than 0.83 W at this position. The calculations showed thermal neutron flux inside the cobalt rods are 6.04×10^{12}, 5.16×10^{12}, 5.05×10^{12} and 5.76×10^{12} $n.s^{-1}.cm^{-2}$ respectively. The Co-60 buildup inside the 4-rod assembly modeled according to Fig.8 was calculated at 4 MW power and is shown in Fig. 10. As the figure shows, after 3 years there is about 6 kCi of Co-60 inside the rods while the specific activity is 14.37 $Ci.g^{-1}$.

In this case, the TRR negative reactivity due to Co-59 4-rod assembly loading is 46 pcm.

Figure 11 shows the thermal neutron flux distribution of this new 4-rod assembly arrangement. As the figure shows, in this new arrangement, all the Co-59 rods receive a thermal neutron flux closer to each other, which makes the Co-60 buildup smoother over the assembly.

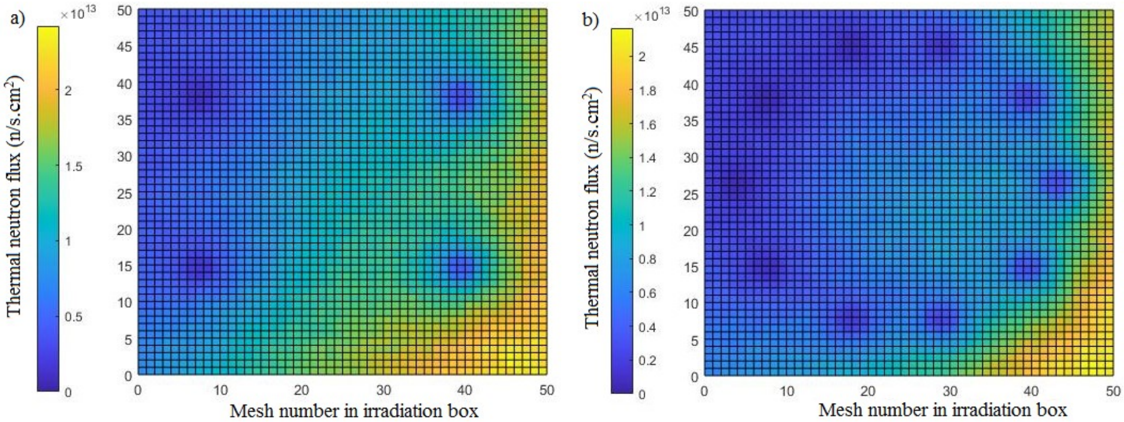

Figure 8: Thermal neutron flux ($E_n < 0.6$ eV) distribution around the Co-59 rods a) 4-rod assembly arrangement b) 10-rod assembly arrangement (the Co-59 rod thickness: 0.7 cm).

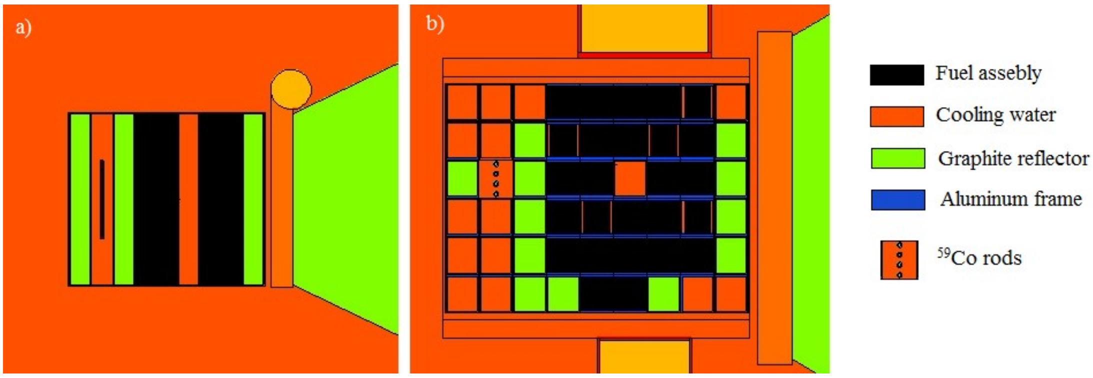

Figure 9: Co-59 rod arrangement in linear form to decrease self-shielding phenomena a) axial view b) cross sectional view.

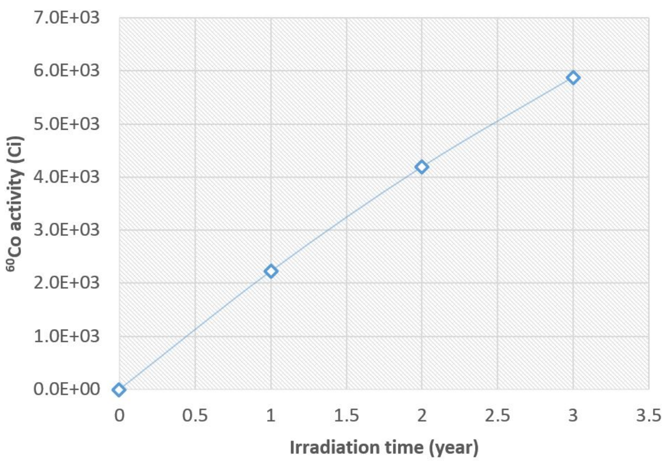

Figure 10: Co-60 buildup inside the 4-rod assembly in linear form of Co-59 rods with 0.7 cm rod diameter.

The carried out calculations for the new 4-rod arrangement showed the Co-59 rods gamma heats are 48.8, 43.1, 43.7, and 43.6 watt respectively, which concludes in 225 W total gamma deposited heat inside the assembly; which 175 W belongs to the cobalt rods and the remaining belongs to the aluminum clad. The total neutron deposited heat of the assembly is less than 1.64 W at this position. The calculations showed thermal neutron flux inside the cobalt rods are 1.24×10^{13}, 1.21×10^{13}, 1.32×10^{13} and 0.81×10^{13} n.s^{-1}.cm^{-2}, respectively. The Co-60 buildup inside the 4-rod assembly modeled according to Fig. 12 was calculated at 4 MW power and is shown in Fig. 13. As the figure shows, after 3 years there is about 8 kCi of Co-60 inside the rods while the specific activity is 19.97 Ci.g^{-1}.

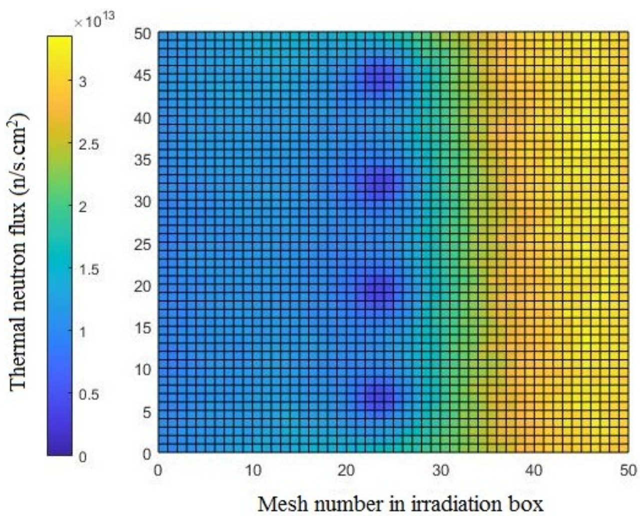

Figure 11: Thermal neutron flux ($E_n < 0.6$ eV) distribution around the 4-rod assembly Co-59 rods (the Co-59 rod thickness: 0.7 cm).

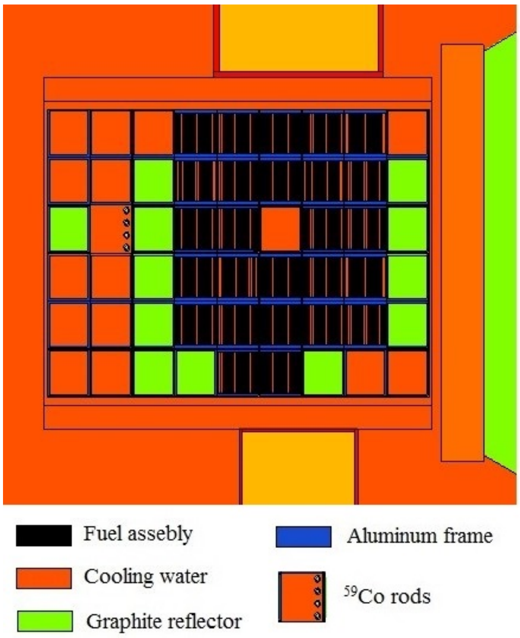

Figure 12: Co-59 rod arrangement in linear form placed close to the irradiation box wall.

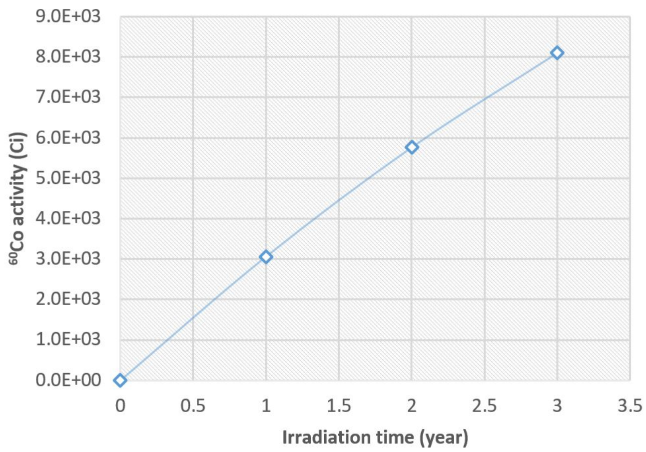

Figure 13: Co-60 buildup inside the 4-rod assembly in linear form of Co-59 rods with 0.7 cm rod diameter, placed close to the irradiation box wall.

In this case, the TRR negative reactivity due to Co-59 4-rod assembly loading is 75 pcm. It should be mentioned TRR operators do not permit a negative reactivity more than 200 pcm. However, internal irradiation boxes have obviously higher thermal neutron fluxes, but they cannot be used for bulk loading of cobalt neutron absorber targets because of a negative reactivity induction more than the mentioned value.

Figure 14 shows thermal neutron flux and gamma deposited heat over the 4-rod Co-59 assembly. As the figure shows, the deposited heat density is less than 0.05 W.cm^{-3}, which indicates there is not any concern for the rods cooling so that the natural convection of the TRR coolant could easily remove the heat. In addition, the figure clearly shows at this position, the rods receive more thermal neutron flux than the previous investigated one.

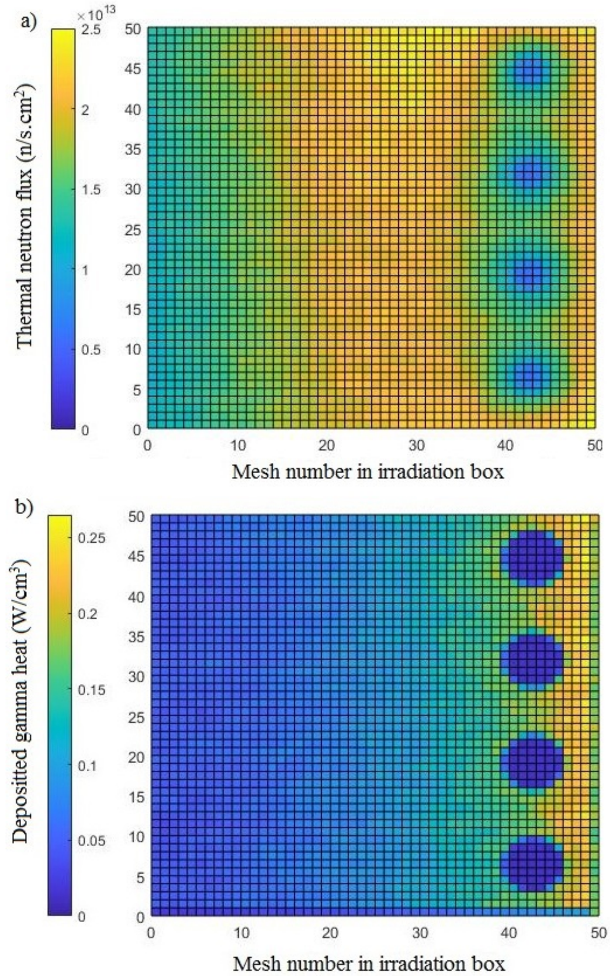

Figure 14: a) Thermal neutron flux (E_n < 0.6 eV), b) Gamma deposited heat, distributions around the 4-rod assembly Co-59 rods placed close to the irradiation box wall (the Co-59 rod thickness: 0.7 cm).

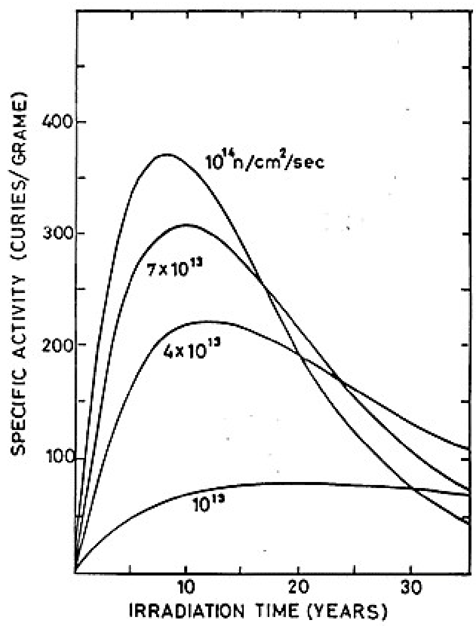

Figure 15: Dependence of Co-60 specific activity to the thermal neutron flux available on the cobalt targets (Villiers, 2003).

3.3 Benchmark study of the calculated values with IAEA reported data

Regarding Fig. 15, a neutron flux in order of 10^{13} n.s^{-1}.cm^{-2} would result in a specific activity about 32 Ci.g^{-1} after 3 years irradiation of Co-59. Clearly, a neutron flux that is half than the mentioned value demonstrates the specific activity of Co-60 product would be definitely less than 16 Ci.g^{-1}. It should be taken in attention moreover the thermal neutron flux at the irradiation position the assembly geometry and the rod diameter play an important role in achieving product specific yield too because of the target self-shielding phenomena. The obtained Co-60 specific activities calculated in this work are in good agreement with the data reported in Fig. 10.

4 Conclusions

New days kilocuries gamma sources are being developed in order to be used in gamma cells to irradiate the small-volume agriculture products such as mushroom, potato etc., level-meter devices, blood gamma cells, as well as research and development centers. The present study investigates the possibility of production of kilocuries Co-60 gamma source in TRR 5 MW research reactor. The carried out investigations using the computational MCNPX code showed that 1 up to 3 kCi Co-60 source could be easily produced in TRR annually using a linear arrangement of the cobalt rods after graphite boxes of TRR (external irradiation boxes of TRR core). Positioning of the rods near to the TRR core (at the border of the irradiation box instead of its center) has a noticeably effect on Co-60 production yield because of removing about 3 cm water moderator. With this change, the product yield increases about 37% because of higher thermal neutron flux, which is available for this position. The carried out calculations showed that after 3 years operation of TRR at this position at 4 MW power, 8.09 kCi of Co-60 is produced. Selection of the best irradiation position helps to increase the Co-60 yield as well as its specific yield without noticeable impact on negative reactivity induced by cobalt neutron absorber loading in TRR core. In addition, linear arrangement of the rods in an assembly helps to decrease the self-shielding effect of the Co-59 rods on each other.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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