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Optimization study to determine the appropriate location for the implementation of silicon doping in Tehran research reactor

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HIGHLIGHTS

- Optimal location for silicon doping in the thermal column of the TRR has been investigated.
- Parameters such as the thermal to fast neutron ratio and heat deposition by gamma and neutron have been obtained.
- The activity level of the silicon ingot and the produced radioisotopes have been obtained.
- The parameters have been determined based on Monte Carlo calculations.
- The P-32 radioactivity level must be lower than the allowed value determined by IEAE.

ABSTRACT

The neutron transmutation doping method is widely used in various fields, such as solar cells, hybrid cars, etc. The Silicon doping process can provide direct commercial income for nuclear research reactors. In this study, we aim to find the optimal location for silicon doping in the thermal column nose of the Tehran research reactor. For this purpose, computational MCNPX and ORIGEN2 codes were used to calculate the neutronic and radioactivity parameters of the silicon ingot. The important parameters such as the thermal to fast neutron ratio, heat deposition by gamma and neutron, and the radioactivity level of the silicon ingot and the produced radioisotopes have been determined to obtain the optimal irradiation channel. The results showed that the irradiation channel placed in the thermal column at a distance of 90 cm from the center of the TRR core has optimal conditions for the implementation of silicon doping. The channel provides a thermal neutron flux in order of 1.721012 $n.cm^{-2}.s^{-1}$ which is the least acceptable value to achieve a proposed neutron fluence during the operation cycles of TRR reactor. Also, the channel has the least possible heat deposition inside the silicon ingot of about 191 W. In addition, the thermal to fast neutron flux ratio of about 311 is enough higher than the determined IAEA limit for NTD.

KEYWORDS
Silicon ingot
Tehran research reactor
Monte Carlo
Neutron transmutation doping
Thermal column

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1 Introduction

Neutron transmutation doping (NTD) is one of the industrys most popular technologies, widely used in various fields such as solar cells, hybrid and electric cars, medical imaging devices, photovoltaic cells, and high-quality semiconductor power devices. The NTD process can provide direct commercial income for nuclear reactors (IAEA, 2011). Doping is the process by which impurity is added to a material to improve its properties for a specific purpose. The most superior materials for doping silicon by radiation method are Ge, Si, GaAs, GaN, GaP, InP, InSe, and HgcdTe (Liesert et al., 1991; Zhao et al., 2004; Meese, 2012; Lark-Horowitz, 1953). Natural silicon consists of three isotopes, Si-28 with an abundance of 92.23%, Si-29 with an abundance of 4.67%, and Si-30 with an abundance of 3.1%. In the case of Si-30, thermal neutron absorption leads to the unstable isotope Si-31, which undergoes beta decay and the final product of this process is a P-31 phosphorus atom. This reaction is as follows, regardless of the amount of antineutrino emission (IAEA, 2011):

$$^{30}\mathrm{Si} + \mathrm{n} \rightarrow \ ^{31}\mathrm{Si} \rightarrow \beta^{-} + ^{31}\mathrm{P} \tag{1}$$

One of the advantages of silicon doping with nuclear reactors compared to chemical methods is the creation of

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uniform impurity inside the ingot (Meese, 2012). The possibility of producing silicon semiconductors with perfectly uniform phosphorus dopant distribution by neutron transmutation was first identified and described by Karl Lark-Horovitz in 1951 (Lark-Horowitz, 1953). Due to the superior features of the NTD method, between 1974 and 1976, this method was offered by several research reactors in the United States, England, and Denmark, and quickly the amount of irradiated silicon reached several tons (IAEA, 2011). The specifications of several examples of NTD facilities in research reactors for silicon doping are summarized in Table 1.

One of the effective factors in silicon doping is the amount of thermal neutron flux. This process must be carried out where the thermal neutron flux parameter is much higher than the fast neutron flux. The minimum thermal neutron flux considered for this process, according to the document of the International Energy Agency, is about 10^{12} n.cm^{-2}.s^{-1} leads to a shorter irradiation time (IAEA, 2011). Another basic parameter in silicon doping is ratio of thermal to fast neutrons flux. This process must be carried out where the thermal neutron flux parameter is much higher than the fast neutron flux. The IAEA recommended that the thermal to fast neutron flux ratio should be at least 7:1 (IAEA, 2011). The gamma rays emission of Si-31 as well as the external gammas received by the crystal from the TRR nuclear core are the major source of heat generation, which may cause the excessive temperature of the ingot and the formation of bubbles on the surface of the ingot. Also, the IAEA recommended that the temperature at the irradiation position must be below 180 ° C to avoid the diffusion of the minority carrier and to minimize lattice defects such as a swirl (IAEA, 2011). The electrical resistivity of irradiated silicon is inversely proportional to the total concentration of the produced dopants such as P-31 atoms and initially existing impurities. In NTD, the added P-31 concentration is proportional to the irradiated neutron flux. In industry, resistance is used instead of a gram for semiconductors (IAEA, 2011). For n-type silicon with phosphorus impurity, the resistance is obtained using Eq. 2 (IAEA, 2011).

$$\rho = \frac{1}{[P]\varepsilon\mu} \tag{2}$$

where $[P]$ is the phosphorus atomic concentration in cm^{-3}, ε is the electron charge (i.e. 1.602×10^{19} C), and is the drift mobility of the electrons in the silicon crystal lattice. Electron mobility depends on the temperature, and it is in the range of 1220 $\sim$ 1500 cm^2.V^{-1}.s^{-1}. In normal conditions at 300 K, it is usually 1350 cm^2.V^{-1}.s^{-1} for silicon.

Another parameter should be taken in attention in NTD process is the amount of radioactivity in silicon ingot. In the process of NTD, side reactions are also carried out, which are as follows according to the aspect of residual activity:

$$^{30}\text{Si}(\text{n},\gamma)^{31}\text{Si} \rightarrow {}^{31}\text{P} \tag{3}$$

$$^{31}\text{Si}(\text{n},\gamma)^{32}\text{Si} \rightarrow {}^{32}\text{P} \tag{4}$$

$$^{31}\text{P}(\text{n},\gamma)^{32}\text{P} \rightarrow {}^{32}\text{P} \tag{5}$$

In addition to the above reactions, there are several threshold reactions involving fast neutrons such as (n, np), (n, α) and (n, 2p), which their contribution to the residual radioactivity is negligible because of their short half-lives together with small cross-sections. Among the interactions, the activity of $(\text{n},\gamma)^{32}$P is important due to its long half-life (172 years). According to regulations, the P-32 radioactivity level at which materials can be classified as exempt is 10^4 Bq.g^{-1} (IAEA, 1996) .

Previously, studies have been carried out to investigate the possibility of the NTD process in the Tehran research reactor (TRR) (Kardan et al., 2022). In this study, the optimal irradiation channelfor silicon doping in the thermal column of the TRR for NTD has been investigated. The proper required paremeters mentioned in this section have been calculated by MCNPX and ORIGEN2 codes.

2 Materials and methods

TRR is a 5 MW pool-type reactor with a graphite reflector and light water moderator. The core of the reactor has fuel elements of the MTR type, which are placed inside the grid plate. The fuel of this reactor is low-enriched uranium oxide (LEU), which includes standard fuel elements (SFE) and control fuel elements (CFE). Both fuel elements have U3O8-Al fuel plates with 20% richness. This reactor includes experimental facilities in the section called stall-end like beam tubes, rabbit system, and thermal column (Pazoki et al., 2022). The thermal column is a stacked graphite and lead assembly for irradiation with highly thermalized neutrons. It is filled with removable graphite blocks which are arranged in four layers. It is about 3 m in length with a wide square shape cross section of 1.2 × 1.2 m^2. The arrangement of the blocks is in such a way that permits us to remove some blocks from the middle of the first layer and put a sample into the thermal column and placed it at the nearest position to the reactor core (Kasesaz et al., 2016).

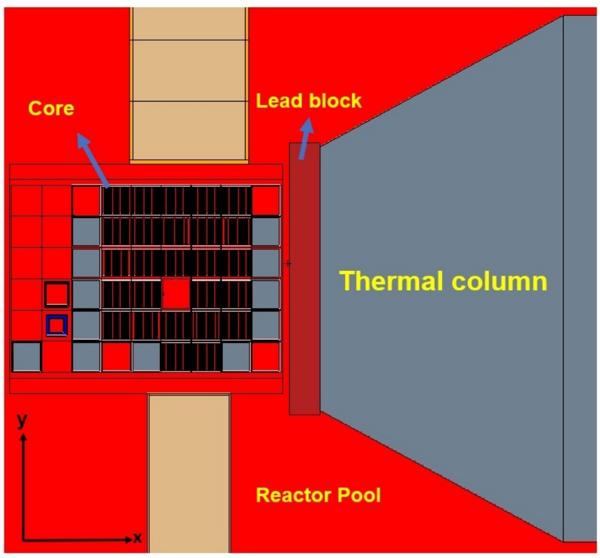

Figure 1: The schematic layout of the TRR and its thermal column.

Table 1: Specifications of several examples of NTD facilities in research reactors for silicon doping.

Country	Reactor name	Power (MW)	Position	Thermal neutron flux ($n.cm^{-2}.s^{-1}$)	Thermal to fast neutron flux ratio	Uniformity approach	Gamma-ray heating or temperature	Reference
Australia	OPAL	20	Channel in Reflector	1.6×10^{12} to 1.7×10^{12}	more than 1000	SS and Al flux screen Rotation ≈ 15 min^{-1}	$\sim 112°C$	(Amos and Kim, 2007)
China	CARR	60	D_2O tank	1×10^{14}	100	Upside down and rotate	- -	(IAEA, 2011)
Brazil	IEA-R1	5	Within graphite reflector	7×10^{12}	- - -	Inversion of 2 ingots and rotation	- - -	(IAEA, 2011) and (Carbonari et al., 1993)
Germany	FRM-II	20	Channel in moderator tank	2×10^{13}	1700	Ni flux screen, Rotation of ingot at 5 min^{-1}	Max.temp: 110°C	(Li et al., 2009)
Belgium	BR2	56 (Nominal :85)	Within Be channel	5.5×10^{13}	27.5	Translation and rotation	$< 200°C$ Si core temp	(Cundy et al., 1992) and (IAEA, 2011)

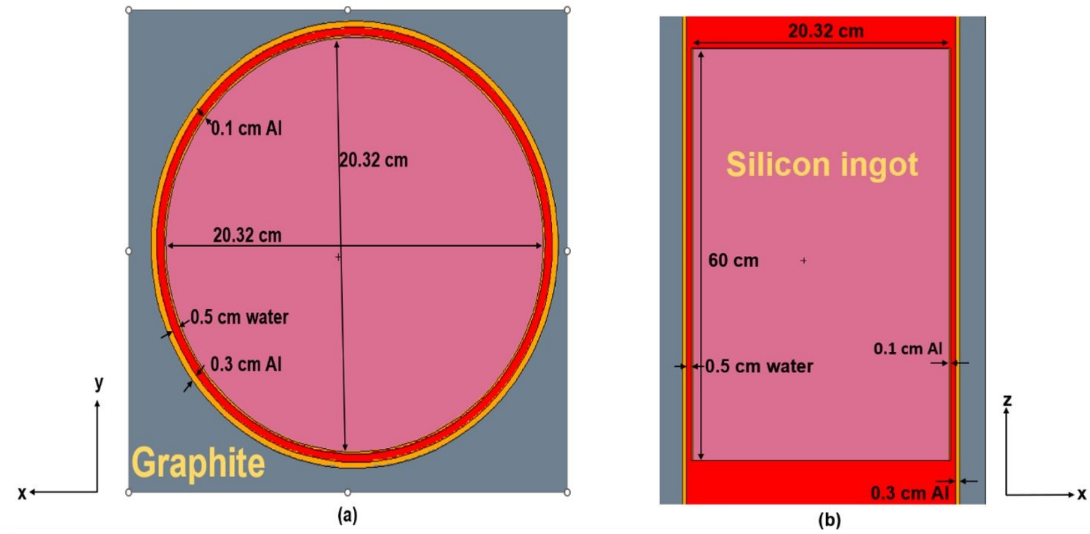

Figure 2: The simulated silicon ingot in irradiation channel a) Crossectional view b) Lateral view.

Therefore, the thermal column is the best location to consider a NTD installation in TRR due to the high thermal to fast neutron flux ratio; the acceptable value according to IAEA document is 7 but achieving higher scores may remove annealing process after NTD process (IAEA, 2011). Figure 1 shows the schematic layout of the TRR and its thermal column which is simulated by MCNPX.

The MCNPX is a general-purpose Monte Carlo particle transport code that began in 1994 as an extension of MCNP4B and LAHET 2.8 in support of the accelerator production of the tritium project (APT). The work envisioned a formal extension of MCNP to all particles and all energies; improvement of physics simulation models; extension of the neutron, proton, and photonuclear libraries to 150 MeV; and the formulation of new variance-reduction and data-analysis techniques. The program also included cross-section measurements, bench-mark experiments, deterministic code development, and improvements in transmutation code and library tools through the CINDER90 project (Fensin, 2008; Pelowitz et al., 2005).

The silicon ingot has been considered as a cylinder with 20.32 cm (8 inches) in diameter and 60 cm in height. Moreover, the ingot has an aluminum holder with a thickness of 0.1 cm. This has been placed inside an aluminum irradiation channel, which has about 0.5 cm of water between the channel and the ingot. Figure 2 shows the two views of the simulated silicon ingot inside the irradiation channel.

In the first stage of determining the optimal location of irradiation, channels at different distances from the reactor core were considered inside the thermal column. This channel was filled with water and neutronic parameters were calculated without silicon ingots. The appropriate location of the silicon ingot was determined to increase the thermal to fast neutron flux ratio, reduce residual heat caused by neutron and gamma particles, and maintain the minimum flux of 1×10^{12} $n.cm^{-2}.s^{-1}$ (Kardan et al., 2022). In the second stage, the silicon ingot was placed in the optimal place, and all these parameters were calculated in addition to the amount of radioactivity and the mass of P-31 produced. In addition, flux and energy deposition calculations in optimal channel locations have been investigated for core configuration variation (change of the number of the loaded fuel assemblies inside the core) since they can alter many of these important parameters in NTD.

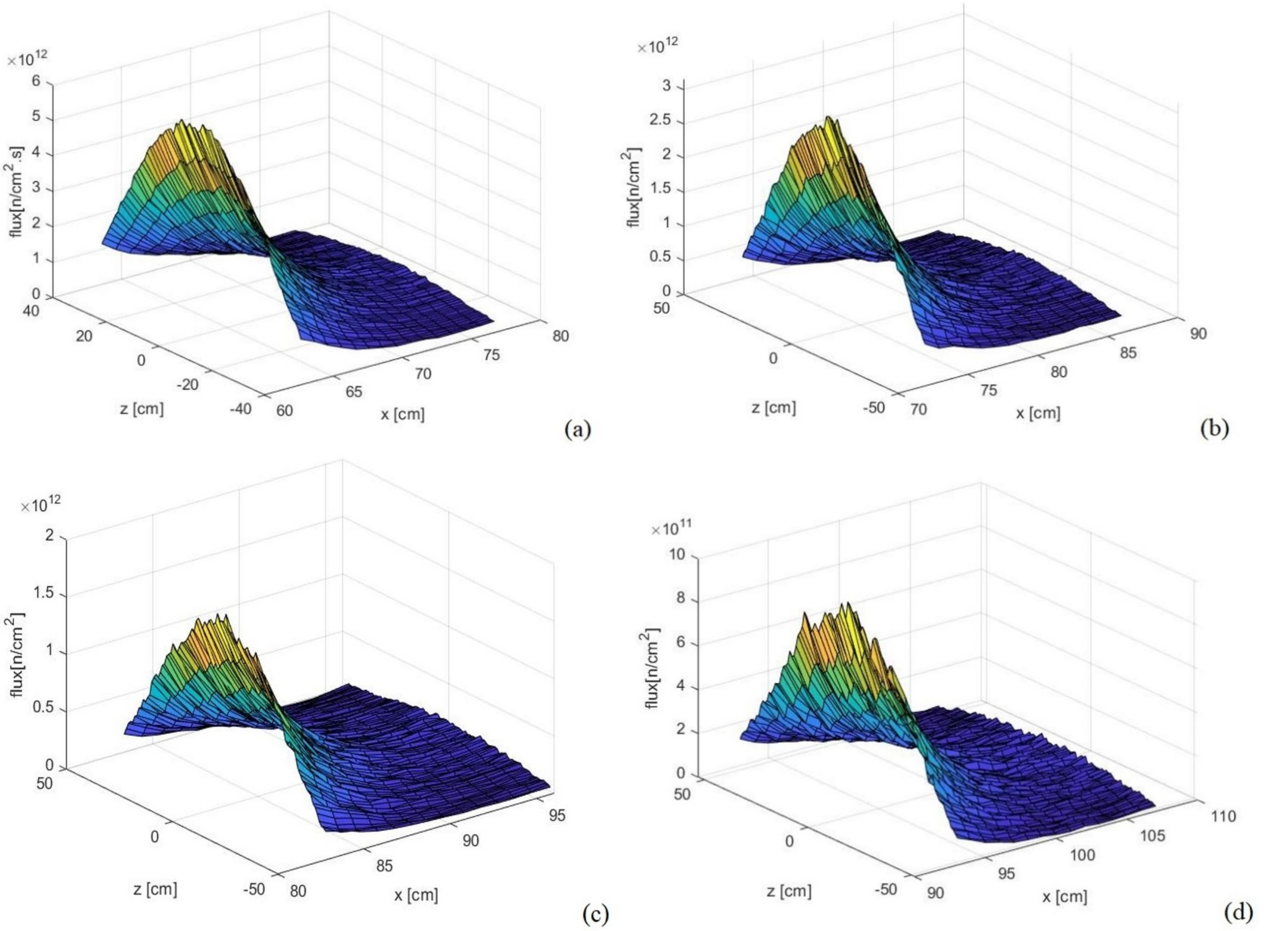

Figure 3: Longitudinal and axial neutron flux distributions for channels in the four distances from center of reactor core in thermal column a) 70 cm b) 80 cm c) 90 cm d) 100 cm.

The radial and axial neutron flux were obtained by the mesh tally feature of the MCNPX code. Furthermore, the track length estimator tally (F4) is used to calculate the neutron flux in the ingot volume. The tally energy card was used to obtain flux in energy intervals. The energy range of thermal neutron was considered as < 0.4 eV and for fast neutron as > 1 MeV. It was necessary to normalize the results to account for the reactors working power intensity of 3.75×10^{17} n.s^{-1} using tally multiplier cards (FM). The energy depositions from the gamma ray and neutron were computed by the F6 tally.

In addition, the activity level of the silicon ingot and the produced phosphorus gram were calculated by the ORIGEN2 code. This code solves the Bateman equation and the exponential matrix using the single-group neutron energy and the 18-group photon energy to enable the user to calculate any changes in activity, power, mass, decay heat, etc. for various reactor operation periods (Croff, 1980). This code has 28 output cards; cards 7, 5, and 9 can be used to calculate the accumulated activity in terms of Curie, the amount of phosphorus produced in grams in the material, and the residual heat due to the delayed gamma in watts respectively.

3 Results and discussion

As illustrated in previous section, irradiation channels have been considered with different distances from the center of the reactor core in thermal column nose (70, 80, 90, and 100 cm). Figure 3 shows the three-dimensional distribution of the thermal neutron flux at the longitudinal (X-axis) and axial (Z-axis) directions without silicon ingot loading inside the channels. It can be seen that the longitudinal thermal neutron flux decreases exponentially. The axial thermal neutron flux also has a maximum in the central part of the channel. The average thermal neutron flux value of the channel 70 cm from the core center in the radial direction and at the middle of the thermal column nose ($z = 0, y = 0$) is 5.9812×10^{12} n.cm^{-2}.s^{-1} at the channel beginning and the value is reached to 5.8655×10^{11} (90.19% reduction) at the end of the channel. The thermal neutron flux value of the channel 70 cm at the bottom of the channel is about 2.6435×10^{11} n.cm^{-2}.s^{-1}, in the center of the channel is 1.2536×10^{12} n.cm^{-2}.s^{-1} (79% increase) and at the top of the channel it reached 2.7845×10^{11} n.cm^{-2}.s^{-1} (77% decrease). It is due to the cosine-shaped distribution of the thermal neutron flux in the axial direction of the TRR core, which propagates the

Table 2: The parameter values without silicon ingot at different distances from the reactor core center.

Channel distance from the center of the reactor core (cm)	Average thermal neutron flux ($n.cm^{-2}.s^{-1}$)	Ratio of thermal to fast neutron flux	Deposited heat by gamma (W)	Deposited heat by neutrons (W)	Average neutron energy (eV)
70	2.208×10^{12}	77	146	40	10
80	1.271×10^{12}	174	84	10	8.3
90	7.583×10^{11}	397	50	2.7	2.29
100	4.657×10^{11}	611	31	0.96	1.31

Table 3: Parameter values with silicon ingots at the two chosen distances from the reactor core center.

Channel distance from the center of the reactor core (cm)	Average thermal neutron flux ($n.cm^{-2}.s^{-1}$)	Ratio of thermal to fast neutron flux	Deposited heat by gamma (W)	Deposited heat by neutrons (W)	Average neutron energy (eV)
90	1.723×10^{12}	311	190.58	0.52	5.25
100	1.056×10^{12}	634	140.15	0.37	1.83

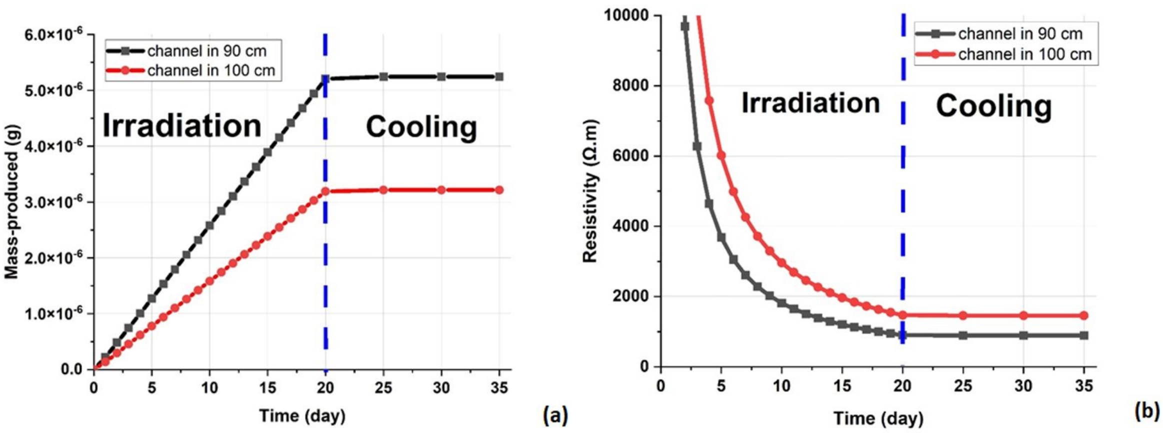

Figure 4: The amount of a) P-31 impurity produced by neutron transmutation in silicon and b) obtained silicon resistance versus irradiation and cooling periods for different channel distances.

neutrons inside the nose of the thermal column in approximately the same axial distribution.

The average amount of thermal neutron flux along with other parameters such as thermal to fast neutron flux ratio, deposited heat by neutron and gamma, and the average neutron energy for these four channels are summarized in Table 2.

The amount of thermal neutron flux decreases from 2.208×10^{12} $n.cm^{-2}.s^{-1}$ to 4.657×10^{11} $n.cm^{-2}.s^{-1}$ as the channel distance increases from 70 to 100 cm in the thermal column. However, the ratio of thermal to fast neutron flux, which is an important parameter in the rate of neutron damage caused by fast neutrons, increases with this distance. The amount of heat caused by gamma and neutrons in the end of channels is less than in the beginning channels of the thermal column, while the average energy of neutrons is closer to the energy of thermal neutrons.

The obtained results show that according to the criteria determined for these parameters in the previous section, the end channels of the thermal column have a better performance for the intended silicon doping work. Therefore, the channels with a distance of 90 and 100 cm from the core of the reactor were chosen to place silicon ingots. The continuation of the results has been carried out by obtaining these parameters on silicon ingots in these channels (Table 3).

It can be seen that the thermal neutron flux in the silicon ingot at 90 cm channel distance is 1.723×10^{12} which is higher than the 100 cm distance. However, the ratio of thermal to fast neutron flux for the channel at a distance of 100 cm is almost twice that of a distance of 90 cm. In addition, the heat deposited by neutrons and gamma inside the silicon ingot in the 100 cm distance channel is less than the 90 cm distance channel (approximately 0.73 times).

Therefore, to find the optimal distance of the irradiation channel, other characteristics such as phosphorus impurity production and radioactivity of the samples should also be investigated. In this regard, Card No. 5 of the ORIGEN2 code was used to calculate the amount of P-31 , which is one of the important impurities resulting from neutron transmutation in silicon. The calculations were carried out for 20 days of irradiation and 10 days of cooling. Since the irradiation time is dependent on the required resistance that should be induced inside the ingot during the neutron transmutation, irradiation can continue until the desired resistance is reached. Of course, the neutron flux plays an important role in this field.

Table 4: The calculated neutron transmutation doping parameters in two different configurations of TRR core.

Reactor core configuration	Average thermal neutron flux (n.cm^{-2}.s^{-1})	The ratio of thermal to fast neutron flux	Deposited heat by neutron (W)	Deposited heat by gamma (W)
26-fuel assembly	1.723×10^{12}	311	0.52	190.58
28-fuel assembly	1.57×10^{12}	300	0.49	169

Figure 4-a shows the amount of P-31 produced impurities in grams as a function of irradiation and cooling during periods for two chosen irradiation channels of 90 and 100 cm from the core center. As seen, the amount of phosphorus produced in the 90 cm channel distance of the core during irradiation is greater than this amount in the 100 cm channel distance from the core, which causes the desired resistance to be reached in a shorter irradiation time. In addition, Fig. 4-b shows the final resistance after irradiation according to Eq. 2 for these two chosen irradiation channels. It can be seen that in the 90 cm channel distance, on the 20th day of irradiation, the resistance reaches from 230000 to 892.53 Ω.m, while for the 100 cm channel distance, it reaches 1456.26 Ω.m during the same period.

Figure 5 shows the amount of produced radioisotope activities during neutron irradiation and cooling time. These calculations have been carried out to show the importance of P-32 as a radioactive element produced with a long half-life. It can be seen that the other radioisotopes produced completely decrease after 5 days of cooling. The P-32 activity changes during irradiation and cooling time have been calculated in Bq.g^{-1} for the chosen channels as shown in Fig. 6. Considering the low probability of this reaction, it can be seen that the activity of P-32 in two channels during irradiation was lower than the allowed value determined by IEAE.

The calculations were all done for the 26-fuel assemblies core configuration of TRR, where the calculations related to the flux and energy deposition for another configuration, i.e. changing the arrangement to the 28-fuel assemblies core, were also performed. Figure 7 shows the difference in the arrangement of the core configuration of TRR in two cases. A comparison of the parameters obtained from both arrangements for the silicon irradiation channel at a 90 cm distance from the core center is listed in Table 4.

It can be seen that the thermal neutron flux decreases by about 9% by changing the configuration of the reactor core to a 28-fuel assemblies. For definition of material in the code input, real burnup of the fuel assemblies were used. The value of the thermal to the fast neutron flux has decreased from 311 to 300, which has increased by about 3.5%. The Deposited heat due to gamma from the photon has been reduced from 190 to 169, a reduction of about 12%. The heat deposited by the neutron has changed a little. According to the mentioned cases, it can be concluded that the parameters of silicon doping in the 28-fuel assemblies core configuration have improved than the previous 26-fuel assembly core. Overall, the most important parameter, which is thermal neutron flux, has a variation of less than 10% then may change the irradiation correspondingly.

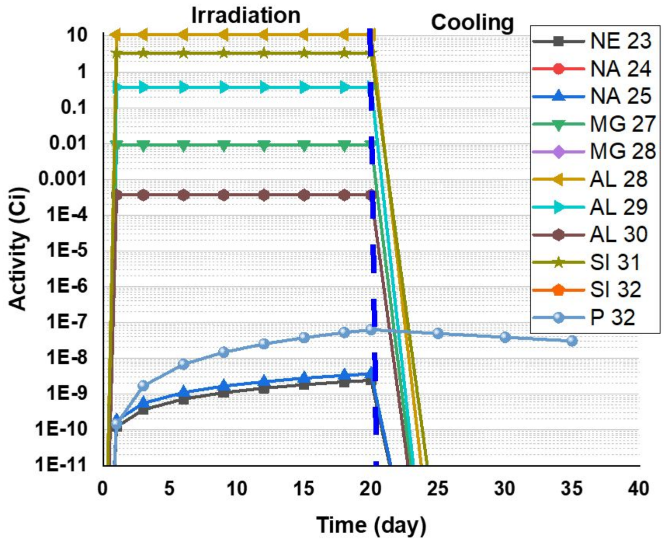

Figure 5: The activity of produced radioisotopes in the silicon ingot for irradiation channel at 100 cm distance from the core.

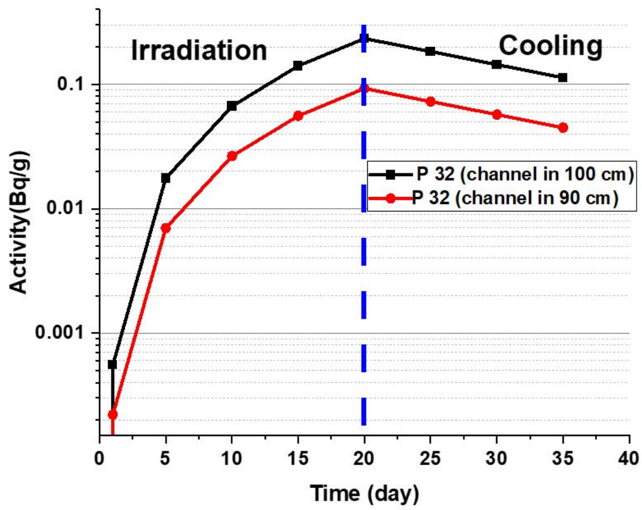

Figure 6: The activity of P-32 during irradiation and cooling time for channels at 90 and 100 cm distance from the core

4 Conclusions

The process of neutron transmutation doping has been used in research reactors around the world for more than several decades and has been able to generate good income for these reactors. Considering the various uses of doped silicon in the industry, the existence of this process inside the country is necessary and saves the country's costs. Investigation of silicon doping process neutronic parameters in TRR showed that this method could be implemented well inside an irradiation channel in the thermal column.

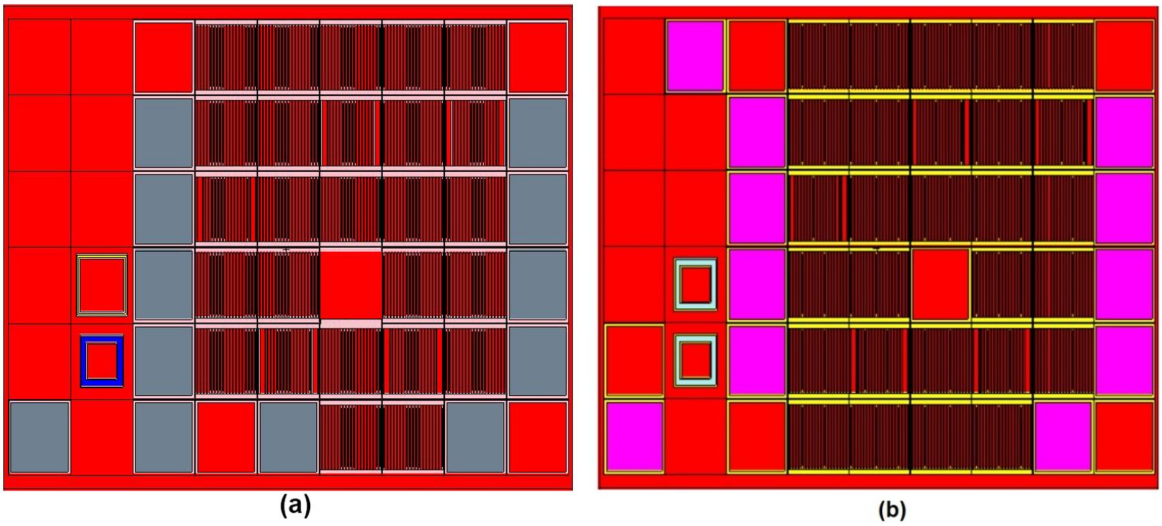

Figure 7: The TRR core arrangement for cases a) 26-fuel assemblies b) 28-fuel assemblies configurations.

The thermal neutron flux and the ratio of thermal to fast neutron flux were 2.11×10^{12} n.cm^{-2}.s^{-1} and 185 in the previous study (Kardan et al., 2022). In this work, we aim to find the appropriate irradiation channel based on optimal distance from core by calculation of the neutronic and radioactivity parameters in the silicon ingot. According to the obtained results, the irradiation channel at 100 cm from the core has superiority over the channel at 90 cm in several parameters, including the ratio of thermal to fast neutrons, the depositions from gamma rays and neutrons, and the amount of energy of neutrons. However, in the irradiation channel at 90 cm from the core, the average thermal neutron flux is higher than the 100 cm channel, which makes it reach the desired resistance in a shorter time. The thermal neutron flux and the ratio of thermal to fast neutron flux were obtained as 1.723×10^{12} n.cm^{-2}.s^{-1} and 311, respectively at 90 cm from the core. This channel has the least possible heat deposition inside the silicon ingot of about 191 W. In terms of similar activities, it can be seen that both channels, after irradiation and cooling, their activity level is lower than the exemption level. According to the above, the irradiation channel at 90 cm from the core center has been chosen as the optimal channel for the NTD facility in TRR.

Of course, it should be noted that changes in the reactor core configuration can also have changes in the effective parameters of the NTD process, some of which were mentioned. Furthermore, the non-uniformity of the radial and axial neutron fluxes was observed on the silicon ingot, which requires another study for a design for their uniformity and is the subject of our future research.

Conflict of Interest

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

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