Study on Concrete Activation Reduction in a PET Cyclotron Vault

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ABSTRACT

Background: Concrete activation in cyclotron vaults is a major concern associated with their decommissioning because a considerable amount of activated concrete is generated by secondary neutrons during the operation of cyclotrons. Reducing the amount of activated concrete is important because of the high cost associated with radioactive waste management. This study aims to investigate the capability of the neutron absorbing materials to reduce concrete activation.

Materials and Methods: The Particle and Heavy Ion Transport code System (PHITS) code was used to simulate a cyclotron target and room. The dimensions of the room were 457 cm (length), 470 cm (width), and 320 cm (height). Gd₂O₃, B₄C, polyethylene (PE), and borated (5 wt% ^{nat}B) PE with thicknesses of 5, 10, and 15 cm and their different combinations were selected as neutron absorbing materials. They were placed on the concrete walls to determine their effects on thermal neutrons. Thin B₄C and Gd₂O₃ were placed between the concrete wall and additional PE shield separately to decrease the required thickness of the additional shield, and the thermal neutron flux at certain depths inside the concrete was calculated for each condition. Subsequently, the optimum combination was determined with respect to radioactive waste reduction, price, and availability, and the total reduced radioactive concrete waste was estimated.

Results and Discussion: In the specific conditions considered in this study, the front wall with respect to the proton beam contained radioactive waste with a depth of up to 64 cm without any additional shield. A single layer of additional shield was inefficient because a thick shield was required. Two-layer combinations comprising 0.1- or 0.4-cm-thick B₄C or Gd₂O₃ behind 10 cm-thick PE were studied to verify whether the appropriate thickness of the additional shield could be maintained. The number of transmitted thermal neutrons reduced to 30% in case of 0.1 cm-thick Gd₂O₃+10 cm-thick PE or 0.1 cm-thick B₄C+10 cm-thick PE. Thus, the thickness of the radioactive waste in the front wall was reduced from 64 to 48 cm.

Conclusion: Based on price and availability, the combination of the 10 cm-thick PE+0.1 cm-thick B₄C was reasonable and could effectively reduce the number of thermal neutrons. The amount of radioactive concrete waste was reduced by factor of two when considering whole concrete walls of the PET cyclotron vault.

Keywords: Concrete Activation Reduction, PHITS Code, PE, B₄C, Gd₂O₃, PETtrace Cyclotron

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Introduction

Medical proton cyclotrons operating below 20 MeV are used to produce radioisotopes for medical diagnostic imaging. Typical short-lived radioisotopes used in posi-

tron emission tomography (PET) are ^{11}C , ^{13}N , ^{15}O , and ^{18}F among which ^{18}F is in particular interest. More than 40 cyclotrons are installed in Korea including 20 non-self-shielded cyclotrons. These cyclotron facilities are expected to be dismantled before the designed lifespan due to upgrade, changes of purpose and place of use, etc. Activated materials inside the cyclotron vault and in concrete walls result in radiological hazards during the maintenance and decommissioning stage, respectively. Additionally, a huge amount of radioactive waste will be generated, after a long period of cyclotron operation, which is one of the main issues during the decommissioning stage because of the high cost of radioactive waste management. The primary radionuclides that cause the concrete activation are ^{152}Eu ($T_{1/2} = 13.52$ y) and ^{60}Co ($T_{1/2} = 5.27$ y). Although ^{151}Eu and ^{59}Co are trace element in the concrete constitutes, they have significant thermal neutron absorption cross sections (Table 1). After a long operation, the specific activities will exceed the order of clearance level [1–3]. The clearance level of the radionuclide in this work is based on the international standards [4]. A material is disregarded as the radioactive waste based on the following clearance criterion:

$$\sum_i \frac{C_i}{C_{Li}} \leq 1 \quad (1)$$

where C_i is the specific activity of the isotope i ($\text{Bq} \cdot \text{g}^{-1}$) and C_{Li} is the clearance level for the isotope i ($\text{Bq} \cdot \text{g}^{-1}$).

The goal of this work is to reduce the total volume of radioactive concrete waste by a factor of two by considering the capability, availability, and the cost of the shielding materials. To achieve this goal, the thermal neutrons, which are responsible for the induced radioactivity in the concrete, were reduced by using the absorbing materials and the reduced amount of activated concrete was estimated. Activation reduction methods are important to decrease the activation level of the concrete in the depth for non-self-shielded cyclotrons that are already installed and are in operation.

Table 1. Major Radionuclides in Concrete and Corresponding Characteristics

Radionuclide	Reaction	Half-life	Thermal neutron absorption cross section (barn) [17, 23]	Clearance level ($\text{Bq} \cdot \text{g}^{-1}$)
^{152}Eu	$^{151}\text{Eu}(n, \gamma)^{152}\text{Eu}$	13.52 yr	5,900	0.1
^{134}Cs	$^{133}\text{Cs}(n, \gamma)^{134}\text{Cs}$	2.06 yr	29	0.1
^{60}Co	$^{59}\text{Co}(n, \gamma)^{60}\text{Co}$	5.27 yr	37	0.1
^{46}Sc	$^{45}\text{Sc}(n, \gamma)^{46}\text{Sc}$	83.79 day	26.5	0.1

Several researchers have proposed different ways to reduce the amount of radioactive waste such as using low activation concrete [5, 6], or reusing the low activated materials [7]. One effective method is to use neutron absorbing materials with high neutron capture cross sections. In addition to the conventional shielding materials as polyethylene (PE) and borated polyethylene (BPE), other novel materials such as boron carbide, and gadolinium are gaining interest [1, 8]. In this work, the effects of conventional and novel materials are considered and discussed. These materials can be used around the target or on the walls [9] in order to absorb thermal neutrons and consequently to reduce the activated concrete. A non-self-shielded PETtrace cyclotron (GE Healthcare, Chicago, IL, USA) has been selected to investigate the neutron reduction method by Monte Carlo code, Particle and Heavy Ion Transport code System (PHITS) version 3.02 [10]. The PETtrace is one of the popular medical cyclotrons which are used to generate ^{18}F by impinging 16.5-MeV protons on a water target with enriched ^{18}O via $^{18}\text{O}(p, n)^{18}\text{F}$ reaction. In our previous work, the neutron production yields from the H_2^{18}O target have been investigated [11]. The secondary neutrons that are produced along with ^{18}F activate the surrounding materials and the bulk concrete, especially, in case of non-self-shielded PET cyclotrons [2, 12–14].

Materials and Methods

In order to consider the activity reduction in the concrete, a geometry of an H_2^{18}O target and a cyclotron vault was simulated in PHITS-3.02 code [10] as shown in Fig. 1. In the PHITS code, by default, proton-induced reactions at energies up to 3 GeV are simulated by the intra-nuclear cascade of Liège model version 4.6 (INCL-4.6) [15]. The INCL-4.6 model is followed by the generalized evaporation model (GEM) [16]. In this simulation, JENDL-4.0 [17] cross sections library was applied for neutron interactions. INCL-4.6 was used for proton-induced reactions on ^{18}O .

The dimension of the cyclotron vault was 457 cm in length, 470 cm in width and 320 cm in height. The neutron spectrum in the forward direction was scored before reaching the concrete (position 1 in Fig. 1). Position 1 was selected in this study as it is in the forward direction with respect to the proton beam and the maximum neutron flux is expected. The spherical detector radius is 10 cm and distance to the concrete wall is 40 cm. Thermal neutrons in the concrete at different depths were scored as well (positions 2, 3, and 4 in Fig. 1). Other tal-

Fig. 1. PETtrace room geometry simulated using the PHITS code (A) top view, (B) vertical view, (C) 16.5-MeV proton beam striking the H_2^{18}O target. Neutron spectra were scored inside the room and in the concrete at positions 1, 2, 3, and 4, respectively. The size of the room is 457 cm in length, 470 cm in width and 320 cm in height. PHITS, Particle and Heavy Ion Transport code System.

lies were also used to score the neutrons at different directions as shown in Fig. 1. However, the results from positions

Fig. 2. Neutron absorption cross sections for natGd , natB , and natH taken from JENDL-4.0 [17]. The vertical dashed line indicates the thermal neutron energy of 0.4 eV.

1, 2, 3, and 4 are shown in this work. In this simulation, the concrete wall was divided into layers with the dimensions of $30 \times 30 \text{ cm}^2$ and the thickness of 3 cm. Neutrons were scored at each layer using the [T-Track] tally in the PHITS code. The scored neutron fluence was folded with the cross sections taken from JENDL-4.0 to consider the impact of the thermal, epithermal and fast neutrons on the radionuclides production. The thickness of the H_2^{18}O target was 1.04 cm with a diameter of 1.4 cm. The target is bombarded with 16.5-MeV protons. The radioactivity is due to trace element in the concrete such as Eu and Co. The magnitude of activation will highly depend on the concrete composition. In this calculations, concrete compositions are taken from PAL-XFEL-2017 (Pohang Accelerator Laboratory X-ray Free Electron Laser) [18] with the density of $2.337 \text{ g} \cdot \text{cm}^{-3}$.

The operation scheme was selected based on the survey that was conducted about the cyclotrons in Korea [19] so that the maximum operation frequency and daily operation was selected to be 5 days per week and 4 hours a day. Additionally, it is expected that many of the cyclotrons would stop the operation before 10 years because of upgrade, changes of purpose, and place of use [20]. Therefore, the operation scheme in this work is quite conservative.

Boron, gadolinium, and hydrogen have high neutron absorption cross sections (Fig. 2) so that they can reduce thermal neutrons significantly and in case of hydrogen, it is relatively an effective neutron moderator. The whole walls of the cyclotron vault were then covered by neutron absorbing ma-

materials so as the effects of each material could be investigated separately. In the calculations, Gd_2O_3 ($7.407 \text{ g} \cdot \text{cm}^{-3}$), B_4C ($2.52 \text{ g} \cdot \text{cm}^{-3}$), BPE ($1.01 \text{ g} \cdot \text{cm}^{-3}$), and PE ($0.94 \text{ g} \cdot \text{cm}^{-3}$), with thicknesses of 5, 10, and 15 cm were considered for each material. In addition, a combination of PE+ Gd_2O_3 , and PE+ B_4C were studied as well. Optimum material was determined by considering the thermal neutron absorption capability, the material cost and availability. Finally, the concrete thickness to be the radioactive waste were determined.

Fig. 3. Neutron spectrum in the forward direction respect to the proton beam indicated as position 1 in Fig. 1 at 40 cm before the concrete. The spectrum contains 20% thermal neutrons and 80% epithermal and fast neutrons. The vertical dashed line indicates the thermal neutron energy of 0.4 eV.

Results and Discussion

1. Thermal Neutron Depth Profile

The neutron spectrum in the forward direction was scored before reaching the concrete (position 1 in Fig. 1) and is shown in Fig. 3. The spectrum contains 20% thermal neutrons and 80% epithermal and fast neutrons that can be down-scattered to thermal neutrons. The neutron spectra without any absorbing material are shown in Fig. 4A at several concrete depths. It is seen that fast neutrons are higher at the very beginning of the concrete depth and decrease as the concrete thickness increases. On the other hand, the number of thermal neutrons increases in the higher depths of concrete. Suppressing these thermal neutrons is extremely important to reduce the radioactivity in the concrete. The impact of the epithermal and fast neutrons on the ^{152}Eu and ^{60}Co production were also considered. Even though considering the wall position where those flux are maximum, the contribution of the epithermal and fast neutrons to the production of ^{152}Eu was less than 10%, while they contribute to the ^{60}Co production by 15% at the concrete surface and in the deeper positions it was less than 10%. Therefore, the thermal neutrons were the major concern in the ^{152}Eu and ^{60}Co production.

Fig. 4B shows the integrated thermal neutron fluence for energies less than 0.4 eV as a function of concrete depth, and illustrates that thermal neutron fluence reaches a maximum around 10 cm. The reason of this increase is that neutrons coming into the concrete interact with the hydrogens in a

Fig. 4. (A) Neutron fluence at different concrete depths without neutron absorbing materials, (B) integrated neutron fluence ($E_n < 0.4 \text{ eV}$) as a function of concrete thickness in the front wall indicated as position 2 in Fig. 1. The proton beam current is 60 μA . Horizontal and vertical dashed lines correspond to the thermal neutron flux of $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ and 64 cm concrete to be radioactive waste, respectively.

concrete and are thermalized, and then reach the maximum around the depth of 10 cm. Thermalized neutrons are captured by the elements of concrete and the number of thermal neutrons decreases after 10 cm depth. The level of concrete activity mainly depends on the amount of Eu and Co, and the thermal neutron flux. We measured the average values of Eu and Co as 1 and 15 ppm, respectively [18]. By considering the operation scheme of 4 hours per day, 5 days per week over 520 weeks (10 years), and the thermal neutron flux of $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$, the specific activities of ^{152}Eu and ^{60}Co were matched the clearance level. In other words, where the thermal neutron flux exceeded $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$, the concrete was considered as the radioactive waste in this work. The horizontal and vertical lines in Fig. 4B correspond to the thermal neutron flux of $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ and the concrete depth which it is radioactive waste, respectively. It is seen that around 64 cm of the concrete becomes radioactive waste in the front wall.

In the simulations, the walls were covered by the absorbing materials with different thicknesses. Table 2 shows the thickness of each material and the corresponding transmitted thermal neutrons ratios obtained as $\Phi_{\text{with shield}}/\Phi_{\text{no shield}}$ at depth of 64 cm in the concrete wall. The thermal neutron fluence and the $\Phi_{\text{with shield}}/\Phi_{\text{no shield}}$ versus the depth in concrete

Table 2. Neutron Absorbing Materials and Thicknesses Used in the Calculations and Corresponding Transmitted Thermal Neutrons at Depth of 64 cm in Concrete

Material	Thickness (cm)	Transmitted thermal neutrons (%)
Gd ₂ O ₃	5	65
	10	40
	15	25
B ₄ C	5	45
	10	20
	15	10
PE	5	58
	10	31
	15	19
BPE	5	59
	10	35
	15	20
PE+Gd ₂ O ₃	10+10	20
	10+5	23
	10+0.4	30
	10+0.1	30
	5+10	23
PE+B ₄ C	10+10	7
	10+0.4	30
	10+0.1	30

PE, polyethylene; BPE, borated polyethylene.

for single materials are shown in Fig. 5 and for different combinations are shown in Fig. 6.

Fig. 5A and 5B indicate the thermal neutron fluence against depth in concrete and corresponding ratios to case with no shield, respectively. The radioactive concrete waste in the front wall decreases to 59, 58, and 49 cm in depth by using 5, 10, and 15 cm-thick Gd₂O₃ shield and transmitted thermal neutrons are 65%, 40%, and 25%, respectively. It is seen from Fig. 5C that B₄C can reduce the concrete thickness as the radioactive waste to 55, 43, and 30 cm. According to Fig. 5D, the thermal neutrons decrease dramatically so that for 5, 10, and 15 cm-thick B₄C, the transmitted thermal neutrons are 45%, 20%, and 10%, respectively. Although boron carbide is quite effective in thermal neutron abortion, it is one of the costly materials, in particular, for covering whole wall of cyclotron walls with thick layers.

In case of PE, thermal neutrons increase at the surface of the concrete due to thermalization process in PE. At inner positions, however, thermal neutrons are attenuated by concrete (Fig. 5E, 5F). Placing 5, 10, and 15 cm of PE shield, the concrete thickness as the waste is 56, 48, and 43 cm, and the transmitted thermal neutrons are 58%, 31%, and 19%, respectively. On the other hand, for BPE, neutrons are elastically scattered by the hydrogen atoms in PE and lose energy and are absorbed by boron. Thus, the thermal neutron fluence is lower than that of only PE at shallow positions in the concrete. The activated thickness of the concrete after using 5, 10, and 15 cm-thick BPE shield is 56, 48, and 43 cm and the transmitted neutrons are 59%, 35%, and 20%, respectively (Fig. 5G, 5H).

10 cm-thick PE+10 cm-thick B₄C can reduce dramatically the radioactive concrete waste to 20 cm in depth and the transmitted thermal neutrons to 7% (Fig. 6A, 6B). As indicated in Fig. 6C and 6D, the activated concrete thickness is reduced to 40, 45, and 38 cm and the transmitted thermal neutrons reach 20% by using 10 cm-thick PE+10 cm-thick Gd₂O₃, and 23% for 10 cm-thick PE+5 cm-thick Gd₂O₃, and 5 cm-thick PE+10 cm-thick Gd₂O₃.

It should be mentioned that in economical view, using a large amount of these absorbing materials are not cost effective. Therefore, thin layer shields of B₄C and Gd₂O₃ were placed between 10 cm-thick PE and the concrete wall one by one and the results were compared together. As shown in Fig. 6A and 6B, using 0.1 or 0.4 cm-thick B₄C layer, the thermal neutron fluence was reduced absolutely in the concrete depth up to 24 cm. Because the fast neutrons are thermal-

Fig. 5. Thermal neutron fluences for different neutron absorbing materials and corresponding ratios, ($\Phi_{with\ shield}/\Phi_{no\ shield}$) as function of concrete thickness in the front wall indicated as position 2 in Fig. 1. (A, B) for Gd_2O_3 , (C, D) for B_4C , (E, F) for PE, and (G, H) for BPE. Horizontal dashed lines correspond to thermal neutron flux of $10^5 n \cdot cm^{-2} \cdot s^{-1}$. PE, polyethylene; BPE, borated polyethylene.

Fig. 6. Thermal neutron fluences for different neutron absorbing materials and corresponding ratios, $(\Phi_{with\ shield}/\Phi_{no\ shield})$ as function of concrete thickness in the front wall indicated as position 2 in Fig. 1. (A, B) for PE+B₄C and (C, D) for PE+Gd₂O₃. The layers of B₄C and Gd₂O₃ are placed between the wall and PE. Horizontal dashed lines correspond to thermal neutron flux of $10^5\ n\cdot cm^{-2}\cdot s^{-1}$. PE, polyethylene.

ized in the PE shield and then are absorbed by boron before the concrete wall. On the other hand, the results for 0.1 cm and 0.4 cm-thick B₄C with 10 cm-thick PE are almost similar so as the transmitted thermal neutrons was reduced to 30%. The thickness of concrete wall to be considered radioactive was also reduced to 48 cm in the front wall. According to Fig. 6C and 6D, the layered shields of 10 cm-thick PE+0.4 cm-thick Gd₂O₃ and 10 cm-thick PE+0.1 cm-thick Gd₂O₃ reduce the radioactive concrete thickness to 48 cm and the transmitted thermal neutrons were 30%.

Fig. 6 illustrates the results for the front wall where the effect of 10 cm-thick PE and 10 cm-thick PE+0.1 cm-thick B₄C on the waste reduction seems similar. However, it is obvious

in Fig. 7 to figure out the effect of B₄C on the radioactive waste reduction compared with only PE layer. The depth profile of thermal neutrons at backward and left side walls, which are indicated as position 3 and position 4 in Fig. 1, are also shown in Fig. 7. It can be seen that in backward (Fig. 7A) and left side (Fig. 7B) walls, no radioactive waste is generated by using 10 cm-thick PE+0.1 cm-thick B₄C, while the concrete waste thickness is around 20 cm for 10 cm-thick PE in both positions.

By placing 0.1 cm-thick B₄C+10 cm-thick PE or 0.1 cm-thick Gd₂O₃+10 cm-thick PE on the concrete wall, the depth of concrete wall treated as the radioactive waste was reduced similarly, from 64 cm of no shield case to around 48 cm. Al-

Fig. 7. Thermal neutron fluences for PE and PE+B₄C combinations as function of concrete thickness. (A) In the backward wall indicated as position 3 in Fig. 1 and (B) in the left side wall indicated as position 4 in Fig. 1. Horizontal dashed lines correspond to thermal neutron flux of $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$. PE, polyethylene.

though the price of 10 cm-thick PE+0.1 cm-thick Gd₂O₃ is around 20% lower than that of 10 cm-thick PE+0.1 cm-thick B₄C. The Gd₂O₃ is in the form of powder and needs to be fabricated in a correct way to be used after the PE layer. On the other hand, B₄C is commercially available in different forms and it is easy to be shaped and used after the PE layer. Therefore, 10 cm-thick PE+0.1 cm-thick B₄C is considered to be the best combination as additional shielding materials.

2. Estimation of Radioactive Concrete Reduction

The thermal neutron versus concrete thickness shown in Fig. 6 are only for the forward direction with respect to the proton beam (front wall). In order to estimate the total reduction of radioactive concrete waste, the thermal neutron distribution inside the cyclotron room as well as all concrete walls are shown in Fig. 8 and discussed. The figures were obtained for the cases without any additional shielding material (Fig. 8A, 8B), with 10 cm-thick PE (Fig. 8C, 8D), and with 10 cm-thick PE+0.1 cm-thick B₄C shield (Fig. 8E, 8F). Fig. 8 illustrates the thermal neutron distributions ($E_n < 0.4 \text{ eV}$) calculated by the PHITS code in the vertical (left column) and top views (right column). The concrete depths at which the thermal neutron exceeds $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ are indicated for each case. Without neutron absorbing materials, more than 60 cm of the concrete was considered to be radioactive waste in the front wall regarding the proton beam as well as in the floor and more than 40 cm in all other walls. By using 10 cm-thick PE, the radioactive waste thickness in the front wall and floor is 48 cm and that of ceiling is 30 cm (Fig. 8C). The radio-

active waste thickness is 20 cm for the left side and backward walls and 32 cm for the right side wall respecting to the proton beam (Fig. 8D). On the other hand, 10 cm-thick PE+0.1 cm-thick B₄C can reduce the concrete waste thickness to 48 cm in the front wall as well as in the floor and 24 cm in the ceiling (Fig. 8E). Thickness of 28 cm of the concrete is radioactive waste in the right side wall regarding to the proton beam. No activated concrete was generated in the backward and left side walls (Fig. 8F).

At forward direction, the effect of 10 cm-thick PE and 10 cm-thick PE+0.1 cm-thick B₄C seems to be similar. For the side walls, ceiling, floor, and back wall, however, 0.1 cm-thick B₄C can absorb the scattered neutrons dramatically as can be seen from Figs. 7 and 8. The reason is that the neutrons are scattered and become thermal and can be captured by B₄C. As a result, the total radioactive waste volume decreases. In the forward direction, most neutrons are epithermal and fast neutrons that cannot be captured by B₄C so that the difference between the 10 cm-thick PE and 10 cm-thick PE+0.1 cm-thick B₄C is small. It is found that the amount of radioactive concrete waste can be reduced by a factor of 2 using the 10 cm-thick PE+0.1 cm-thick B₄C shield. It can be seen from Fig. 8 that the front wall respecting to the proton beam and the floor are the hot spots where the amount of radioactive waste is quite high. Thus, 10 cm-thick PE+0.1 cm-thick B₄C can be used for these areas, and for other side walls, thinner shielding materials is enough to reduce thermal neutron fluence inside the walls and results in lowering the price of the shielding materials. In a real cyclotron vault,

Fig. 8. Distributions of thermal neutron flux ($E_n < 0.4$ eV) inside the cyclotron room and the concrete walls in vertical (left column) and top views (right column) (A, B) without neutron absorbing materials, (C, D) with 10 cm-thick PE, and (E, F) with 10 cm-thick PE+0.1 cm-thick B₄C. The thin layer of B₄C is placed between the wall and PE. The numbers indicate the depths at which thermal neutron flux exceeds $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ and concrete is radioactive waste. PE, polyethylene.

however, neutrons can be scattered by the cyclotron body remarkably and reach to the thermal energy which can be captured by boron in the B_4C . Therefore, 10 cm-thick PE+0.1 cm-thick B_4C is expected to be more effective to reduce the radioactive waste in concrete than the simple geometry considered in this work. It is emphasized that the operation scheme in this work was conservative so that in the case of the operations corresponding to around 20% of our operation scheme and using 10 cm-thick PE+0.1 cm-thick B_4C , one should not consider the concrete as the radioactive waste. By considering the installation cost of the shielding material, it seems to be very beneficial to reduce the radioactive waste storage expenses.

It should be pointed out that the B_4C is available in rubber form that can be shaped easily and can be attached to the walls. The amount of B_4C in the rubber material is 80% and 20% is the glue to form the rubber. Therefore, the required thickness of the B_4C rubber is 0.12 cm to make equivalent to 0.1 cm-thick B_4C .

3. Production of Other Radionuclides

Together with the ^{152}Eu and ^{60}Co radionuclides, the ^{54}Mn , ^{22}Na , and 3H radionuclides can be produced through the $^{54}Fe(n,p)^{54}Mn$, $^{23}Na(n,2n)^{22}Na$, $^2H(n,\gamma)^3H$, and $^6Li(n,\alpha)^3H$ reactions. ^{54}Mn and ^{22}Na are generated by fast neutrons so as their production were approximately in the order of ^{152}Eu and ^{60}Co at the concrete surface. The ^{54}Mn and ^{22}Na production decreased exponentially with the concrete depth and

became one order lower than ^{152}Eu and ^{60}Co at 30 cm. The production of 3H in the concrete was about one order higher than other radionuclides due to the high reaction cross section of the $^6Li(n,\alpha)^3H$ reaction. These results are in agreement with the results reported in the literature [2, 13]. However, the clearance level of 3H is $100 \text{ Bq} \cdot \text{g}^{-1}$ which is three orders higher than that of ^{152}Eu and ^{60}Co . It was also confirmed that 3H is produced mainly by the thermal neutrons and the transmitted thermal neutrons decrease to 30% by using the 10 cm-thick PE+0.1 cm-thick B_4C shielding material and the production of the 3H could reduce correspondingly. The number of fast neutrons at different concrete depths without and with the 10 cm-thick PE+0.1 cm-thick B_4C shielding are shown in Fig. 9A and corresponding ratio is indicated in Fig. 9B. It is seen that the transmitted fast neutrons also decrease to 40% by using the shielding material. It means that the 3H , ^{54}Mn , and ^{22}Na production could also decrease considerably and they could not be a major issue for the radioactive waste management of concrete.

Structural concrete is often reinforced with steel bars that become activated via $^{54}Fe(n,\gamma)^{55}Fe$, $^{54}Fe(n,p)^{54}Mn$, and $^{59}Co(n,\gamma)^{60}Co$ reactions. The reinforcing bars contribution to the activation can be significant [21]. However, the number of fast and thermal neutrons significantly decrease by using 10 cm-thick PE+0.1 cm-thick B_4C as the shielding material and the activation of the reinforcing bars are also decreased proportionally. It is noticed that the activated bars are localized and could be separated from the concrete during the

Fig. 9. (A) Fast neutron fluence ($E_n > 1 \text{ MeV}$) against the concrete depth without and with the 10 cm-thick PE+0.1 cm-thick B_4C shielding, (B) transmitted fast neutrons, $(\Phi_{with\ shield}/\Phi_{no\ shield})$ as a function of concrete thickness in the front wall indicated as position 2 in Fig. 1. PE, polyethylene.

dismantling. They are not as voluminous as the concrete to become a major concern for the radioactive waste storage. For the case of ^{54}Mn , its activation level remains constant after several years of operation [21, 22]. By considering the typical cooling time before the decommissioning the activity level of ^{54}Mn could reduce as well.

Conclusion

The use of non-self-shielded cyclotrons for the PET isotopes production results in serious concrete activation after a long operation. In this study, intensive Monte Carlo calculations were performed to mitigate the concrete activation by using additional neutron absorbing materials. A non-self-shielded PET trace cyclotron vault and an H_2^{18}O target was simulated by the PHITS code to study the effects of neutron absorbers on thermal neutrons reduction. The conservative cyclotron operation scheme of 4 hours per day, 5 days per week over 520 weeks (10 years) and 60 μA proton beams was selected in this work. The amounts of Eu and Co were 1 and 15 ppm, respectively. The thermal neutron flux of $10^5 \text{ n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ was obtained at above specific condition, which the concrete was considered as the radioactive waste. Results showed that Gd_2O_3 , B_4C , PE, and BPE would be effective to decrease the activation level of concrete. However, a combination of these materials is more effective than using single shield material to decrease the price of the shielding. It was concluded that placing 0.1 cm-thick B_4C between 10 cm-thick PE and the concrete wall could reduce the total radioactive concrete waste by a factor of 2 in the whole cyclotron room. However, this combination is expected to reduce more thermal neutrons in a real cyclotron vault. Additionally, the amount of radioactive waste decreases dramatically at cyclotron with lower operating parameters. By taking the price of 0.1 cm-thick B_4C and 10 cm-thick PE into account, it is still cost-effective to use it as a shielding material to achieve a significant reduction of radioactive concrete waste volumes.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

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Author Contribution

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