A Study of Targetry Activation and Dose Analysis of PET Cyclotron Using Monte Carlo Simulation

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ABSTRACT

Cyclotron for medical purposes generates nuclear reaction by accelerating protons in high speed, in order to produce radiopharmaceuticals, and unnecessary neutrons are generated through such nuclear reaction. Neutrons cause activation in the parts of cyclotron which then cause exposure to radiation for people working in the field. This study, in that regard, aims to analyze exposure level by finding out the degree of activation of aluminum body, silver body, and havar foil which are the parts of Targetry where the nuclear reaction takes place.

The results of the experiment showed that aluminum body and silver body had no problems re-using them as the energy and half-life of activated nuclides were small and short, making the affect on the people working in the field extremely low. However for havar foil, its activated nuclides had a high level of energy which resulted in high level of affect to the people working in the field.

The activation factors of the cyclotron were analyzed, and the results showed that the Havar foil was activated the most among the targetry parts, and greatly exposed workers due to regular replacement, and needed special management as radioactive waste.

Keyword: Cyclotron, Activation, Targetry

I. INTRODUCTION

Cyclotron is a device that accelerates particles and installed in industrial facilities, hospitals, and laboratories. Among them, cyclotron used in medical facilities is used as a device to produce radioactive isotope with short half-life which is used in a medical device called Positron Emission Tomography (PET) Prominent radioactive isotopes with short half-life are $^{18}\mathrm{F}$, $^{11}\mathrm{C}$, $^{13}\mathrm{N}$, and $^{15}\mathrm{O}$. About 98% of total domestically produced cyclotrons is $^{18}\mathrm{F}$. In order to produce $^{18}\mathrm{F}$, protons with high energy need to be accelerated [$^{18}\mathrm{O}$] to generate nuclear reaction with $\mathrm{H}_2\mathrm{O}$ target ($\mathrm{H}_2^{18}\mathrm{O}$ + p \rightarrow $\mathrm{H}_2^{18}\mathrm{F}$ + n). From such

nuclear reaction process, unnecessary secondary neutrons are generated. The neutrons then have nuclear reaction with the parts of cyclotron to cause activation so that the parts can indicate radioactivity. [2-8]

According to the results of previous research, it is shown that nucleated radioactive species of ⁴⁸V, ⁵¹Cr, ⁵²Mn, ⁵⁴Mn, ⁵⁶Co, ⁵⁷Co, ⁵⁸Co, ⁶⁰Co, ^{95m}Tc, ⁹⁶Tc, ¹⁸³Re, and ¹⁸⁴Re are generated. These radionuclides are also known to be the main cause of radiation exposure for cyclotron operators. ^[9,10]

Most studies did not know the exact dose because they estimated the exposure rate after a cooling time due to the high exposure of the radioactive component. [11,12] Therefore, in this study, Monte Carlo simulation was used to accurately evaluate the activation of the components of the targetry.

I MATERIAL AND METHOD

1. Monte Carlo Simulation

In this experiment, two types of Monte Carlo simulation codes FLUKA code^[15] and MCNPX code^[16] were used. Fig. 1 shows the geometric structure, targetry was manufactured by referencing PETtrace 800 series (GE Healthcare, USA) model.^[17] Fig. 2 shows the targetry's properties were consisted of basic elements with impurities removed. The impurities were not included in order to maximize the generation of neutrons as a cause of deterioration of ¹⁸F production.

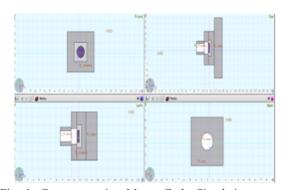


Fig. 1. Geometry using Monte Carlo Simulation.

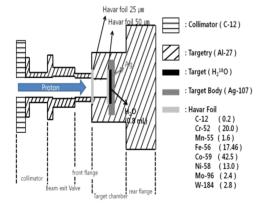


Fig. 2. Elements and their weight percents in Targetry.

2. Experimental Method

2.1 Verification of Reproducibility

Based on the data provided in the manufacturer's manual as shown in Table $1^{[18]}$, protons of 16.5 MeV, 60 μ A were studied for two hours in order to find out reproducibility of the targetry manufactured in this experiment.

Table 1. Manufacturer's ¹⁸F-FDG production volume

S9120NA, PETtrace 840, 60 μA 6500 mCi / 240 GBq ¹⁸F-fluride after 2h of irradiation in single mode

2.2 Radioactivation analysis

conducted The experiment was by setting cyclotron's operating time at four hours a day, five days a week in weekly basis. Radioactivated nuclide's radiation level was measured by FLUKA's RESNUCLEi, and aluminum body, silver body, and havar foil were set as measuring areas. Based on the measured radioactivated nuclide's radiation, equivalent dose was measured using MIRD Phantom which was developed by MCNPX. Fig. 3 shows the MIRD Phantom. The dose conversion factor (ICRP 74) of the de/df card installed in MCNPX was used to measure the equivalent dose, and the measured equivalent dose was comparatively analyzed after converting to valid dose using the organizational weight ratio of ICRP 103.

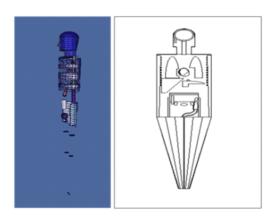


Fig. 3. ORNL MIRD Phantom.

2.3 Radioactive Material Wastes

Based on the radiation level of the measured radioactivated nuclide, according to the IAEA Safety standards series No. RS-G-1.7.^[19] discarding standards of the radioactivated nuclides were comparatively analyzed, and the shielding width that is necessary for storing the radioactive waste materials were calculated through MCNPX. Table 2 shows the self-disposal tolerance level.

Table 2. Self-Disposal tolerance level for each radioactive nuclide

Radioactive nuclide	Tolerance level (Bq/g)
⁵⁴ Mn, ⁵⁶ Co, ⁶⁰ Co, ^{110m} Ag, et cetera	0.1
²⁴ Na, ⁵⁹ Fe, ⁵⁷ Co, et cetera	1
⁵⁶ Mn, ⁹⁹ Mo, et cetera	10
⁵¹ Cr, ⁶⁴ Cu, ^{99m} Tc, et cetera	100

III. RESULT

1. Verification of Reproducibility

When protons 16.5 MeV, 60 μA were studied for two hours in the manufactured targetry, in $H_2^{18}O$ target, about 257 GBq of ^{18}F was generated at 2.5711E+11. Fig. 4 shows the result of FLUKA

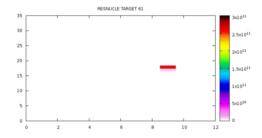


Fig. 4. Radioactivated nuclides generated in the target.

2. Radioactivation analysis

2.1 Aluminum body

As a result ofstudying protons 16.5 MeV, 60 µA

on the aluminum body surrounding the targetry for 20 hours a week and measuring its level of radioactivation, ²⁴Na, ²⁶Al were generated. ²⁴Na attained a near equilibrium at 7.8E+6 Bq at more than 5 weeks, and ²⁶Al was very insignificant with below 1 mBq. As a result of measuring radiation exposure level in human body based on radiation of ²⁴Na, ²⁶Al, ²⁴Na showed about 2.79E-02 mSv/hr, and ²⁶Al showed about 6.45E-12 mSv/hr. Fig. 5 and Table 3 shows the results of the activation in the aluminum body. Table 4 shows the exposure dose by aluminum body.

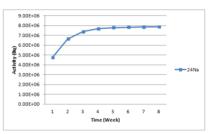


Fig. 5. ²⁴Na that was generated by activation in the aluminum body.

2.2 Silver body

As a result of measuring activation on the silver body that surrounds the Target, ^{110m}Ag was generated. As a result of measuring on a weekly basis, it was confirmed that activation attains equilibrium at 1.21E+8 Bq after about 1500 weeks (30 years), and as a result of testing radiation exposure level through radiation level at the state of equilibrium, it showed about 2.01E-1 mSv/hr. Fig. 6 shows the results of the activation in the silver body. Table 5 shows the exposure dose by silver body.

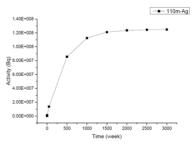


Fig. 6. ^{110m}Ag that generated by activation in the silver body.

Table 3. ²⁶Al that was generated by activation

Table 3. ²⁶ Al that was generated by activation										
	1 week	2 week	3 week	4 week	5 week	6 week	7 week	8 week		
Activity	2.95E-5	5.91E-5	8.86E-5	1.18E-4	1.48E-4	1.77E-4	2.07E-4	2.36E-4		

Table 4. Radiation exposure level per organ that will be received from aluminum body

	²⁴ Na	²⁶ Al
mSv/hr	2.79E-02	6.45E-12

Table 5. Radiation exposure level per organ that will be received from Silver body

	^{110m} Ag
mSv/hr	2.01E-01

2.3 Havar foil

In order to measure havar foil, the study period of protons was set as 5000 µA (4 weeks) which is the replacement period of havar foil based on the recommendation by the equipment vendor, and

activations of Vacuum foil (25 µm) and Window foil (50 µm) were measured.

Among the activated nuclides generated from havar foil, a total of 14 emitted gamma, and the result of evaluating dose based on the measured level of radiation showed a radiation exposure level of 4.13 mSv/hr total. Table 6 shows the amount of nuclides generated in the Havar foil. Table 7 and 8 shows the exposure dose by Havar foil. Among the activated nuclides, the nuclide that affected the most to the radiation exposure level was ⁵²Mn(52.39%), and three nuclides including 58Co and 56Co consisted 97.5% of the total dose. Table 9 shows percentage of dose per niclide.

Table 6. Nuclides generated by acyivation in havar foil

	25 μm	50 μm
¹⁸⁶ Re	2.65E+05	3.97E+05
⁹⁹ Mo	6.11E+04	1.23E+05
⁶⁴ Cu	4.97E+04	1.34E+05
⁶⁰ Co	1.98E+03	6.21E+03
⁵⁹ Fe	4.02E+03	4.00E+04
⁵⁸ Co	2.58E+08	4.19E+08
⁵⁷ Co	2.01E+07	3.13E+07
⁵⁶ Mn	5.84E-04	3.49E-03
⁵⁶ Co	1.31E+08	3.18E+08
⁵⁵ Fe	1.68E+07	2.35E+07
⁵⁴ Mn	3.24E+06	5.76E+06
⁵² Mn	3.35E+08	7.34E+08
⁵¹ Cr	1.67E+08	1.76E+08
^{99m} Tc	6.88E+04	1.39E+05

2.4 Radioactivation analysis

The activated radioactive waste materials were evaluated according to the IAEA Safety standards series No. RS-G-1.7.^[17] As a result, ²⁴Na that was

generated in the aluminum body was lowered below the self-disposal tolerance level after 14 half-lifes (8.75 days) have passed, and ²⁶Al was exempted from consideration since its generated radiation dose was extremely low. 110mAg that was generated in the silver body was lowered down to the self-disposal tolerance level after 25 half-lifes (17 years). Activated nuclides of the aluminum body and the silver body were not considered for shielding location since the effect of their radiation exposure levels on human body was extremely low. ⁶⁰Co was set as the self-disposal basis for havar foil considering its half-life and tolerance base level, and it was lowered to below the tolerance level after about 85 years. Since havar foil generates high level of radiation exposure in its surrounding

areas due to activation, it was determined that a shielding container is needed to safely store it. It showed that such shielding container needs to have shielding dose of at least more than 15 cmPb for the width of the shielding structure based on below 1 μ Sv/hr. However, as the number of stored havar foils increases, it was determined that a shielding structure of maximum 22 cmPb is needed. Table 10 shows the Havar foil dose according to lead thickness.

Table	7. Radi	ation e	xposure	level p	er organ	that	will be 1	received	from 1	havar fo	il (25 µ	m)		Unit: 1	ıSv/hr
	¹⁸⁶ Re	⁹⁹ Mo	⁶⁴ Cu	⁶⁰ Co	⁵⁹ Fe	⁵⁸ Co	⁵⁷ Co	⁵⁶ Mn	⁵⁶ Co	⁵⁵ Fe	⁵⁴ Mn	⁵² Mn	⁵¹ Cr	^{99m} Tc	total
totlal	6.10E -05	2.89E -05	5.47E -05	4.83E -06		3.97E -01	4.72E -03	1.30E -12	2.40E -01	1.58E -05	4.42E -03	6.78E -01	3.81E -02	1.86E -05	1.36E +00

Table	Table 8. Radiation exposure level per organ that will be received from havar foil (50 μm)										Unit : µ	Sv/hr			
	¹⁸⁶ Re	⁹⁹ Mo	⁶⁴ Cu	⁶⁰ Co	⁵⁹ Fe	⁵⁸ Co	⁵⁷ Co	⁵⁶ Mn	⁵⁶ Co	⁵⁵ Fe	⁵⁴ Mn	⁵² Mn	⁵¹ Cr	^{99m} Tc	total
totlal	9.13E -05	5.81E -05	1.47E -04	1.51E -05	9.07E -05	6.44E -01	7.36E -03	7.78E -12	5.83E -01	2.21E -05		1.49E +00	4.01E -02	3.73E -05	2.77E +00

Table 9. Percentage of dose per nuclide

Radioisotope	Percentage	Radioisotope	Percentage
⁵² Mn	52.39%	¹⁸⁶ Re	.%
⁵⁸ Co	25.20%	⁹⁹ Mo	.%
⁵⁶ Co	19.91%	⁵⁹ Fe	.%
⁵¹ Cr	1.89%	^{99m} Tc	.%
⁵⁴ Mn	0.30%	⁵⁵ Fe	.%
⁵⁷ Co	0.29%	⁶⁰ Co	.%
⁶⁴ Cu	.%	$^{56}\mathrm{Mn}$.%

Table	10.	Havar's	shielding	dose	according	to th	e width							unit :]	µSv/hr
	cm	1 1	1:	5	16	17	18	19	20	21	22	23	24	25	26

Hava	ir's	1	15	16	17	18	19	20	21	22	23	24	25	26	27
4 week	1 EA	1.5E+04	 5.3E-01	2.8E-01	1.5E-01	7.9E-02	4.2E-02	2.2E-02	1.2E-02	6.3E-03	3.3E-03	1.8E-03	9.9E-04	5.2E-04	2.6E-04
1 year	12 EA	1.8E+05	 6.4E+00	3.4E+00	1.8E+00	9.4E-01	5.0E-01	2.7E-01	1.4E-01	7.6E-02	4.0E-02	2.1E-02	1.1E-02	6.3E-03	3.1E-03
2 year	24 EA	3.7E+05	 1.2E+01	6.8E+00	3.5E+00	1.9E+00	1.0E+00	5.4E-01	2.8E-01	1.5E-01	8.0E-02	4.3E-02	2.3E-02	1.2E-02	6.3E-03
3 year	36 EA	5.6E+05	 1.9E+01	1.0E+01	5.3E+00	2.8E+00	1.5E+00	8.1E-01	4.3E-01	2.2E-01	1.2E-01	6.4E-02	3.5E-02	1.8E-02	9.5E-03
4 year	48 EA	7.4E+05	 2.5E+01	1.3E+01	7.1E+00	3.8E+00	2.0E+00	1.0E+00	5.7E-01	3.0E-01	1.6E-01	8.6E-02	4.7E-02	2.5E-02	1.2E-02
5 year	60 EA	9.3E+05	 3.2E+01	1.7E+01	8.9E+00	4.7E+00	2.5E+00	1.3E+00	7.2E-01	3.8E-01	2.0E-01	1.0E-01	5.9E-02	3.1E-02	1.5E-02
6 year	72 EA	1.1E+06	 3.8E+01	2.0E+01	1.0E+01	5.6E+00	3.0E+00	1.6E+00	8.6E-01	4.5E-01	2.4E-01	1.3E-01	7.1E-02	3.7E-02	1.9E-02
7 year	84 EA	1.3E+06	 4.5E+01	2.3E+01	1.2E+01	6.6E+00	3.5E+00	1.9E+00	1.0E+00	5.3E-01	2.8E-01	1.5E-01	8.3E-02	4.4E-02	2.2E-02
8 year	96 EA	1.4E+06	 5.1E+01	2.7E+01	1.4E+01	7.5E+00	4.0E+00	2.1E+00	1.1E+00	6.0E-01	3.2E-01	1.7E-01	9.5E-02	5.0E-02	2.5E-02
9 year	108 EA	1.6E+06	 5.8+01	3.0E+01	1.6E+01	8.5E+00	4.5E+00	2.4E+00	1.3E+00	6.8E-01	3.6E-01	1.9E-01	1.0E-01	5.6E-02	2.8E-02
10 year	120 EA	1.8E+06	 6.4E+01	3.4E+01	1.8E+01	9.4E+00	5.0E+00	2.7E+00	1.4E+00	7.6E-01	4.0E-01	2.1E-01	1.1E-01	6.3E-02	3.1E-02

IV. DISCUSSION

In this study, due to the neutrons generated when a cyclotron for medical purposes is operated, the experiment was conducted after analyzing radiation of the Targetry parts. Firstly, when the production level of ¹⁸F was compared by studying the protons of 16.5 MeV, 60 µA based on the recommendation by the equipment vendor, about 257 GBq of ¹⁸F was generated which was a little higher than the base value of the equipment vendor. For production of ¹⁸F, because of the contamination and impurities of other nuclides, it has been known that the production level of ¹⁸F may decrease^[20,21], and it is believed that in this study, basic elements without impurities were used to produce ¹⁸F. Through such process, reproducibility of the targetry manufactured by FLUKA was obtained. Secondly, as a result of testing activation using FLUKA, in the Aluminum body, ²⁴Na, ²⁶Al were generated, and in the Silver body, 110mAg was generated with Havar foil generating ¹⁸⁸Re, ⁹⁹Mo, ⁶⁴Cu, ⁶⁰Co, ⁵⁹Fe, ⁵⁸Co, ⁵⁷Co, ⁵⁶Mn, ⁵⁶Co, ⁵⁵Fe, ⁵⁴Mn, ⁵²Mn, ⁵¹Cr and ^{99m}Tc. Based on the radiation of the above nuclides, valid dose for each part that affect human body was measured using MCNPX and ORNL MIRD Phantom. As a result, Aluminum body's ²⁴Na attained equilibrium at more than 5 weeks, and the result of measuring valid dose using the equilibrium radiation value showed about 2.79E-02 mSv/hr. ²⁴Na was lowered to below 1 Bq/g after about 14 half-lifes (8.75 days) due to it short half-life of 15 hours. ²⁶Al was generated below 1 mBq, and its valid dose of 6.45E-12 mSv/hr was revealed to be barely affecting human body. 110mAg was generated in the silver body, and its radiation attained equilibrium after about 30 years. Based on the radiation at the equilibrium of 110mAg, and the affect on human body was shown to he 2.01E-01mSv/hr. For havar foil, it was confirmed that there were a total of 14 types of radioactivated nuclides. The effect on human body was shown to be about 4.13 mSv/hr, and 3 nuclides of ⁵²Mn, ⁵⁸Co, and ⁵⁶Co consisted more than 97% of the dose. Havar foil is replaced periodically, and it has been known that the time to replace is six minutes (five minutes for replacement and one minute for moving to the storage 20). It was shown that the doses received from the Targetry for 6 minutes were Havar foil 4.13E-01 mSv/6min, Silver body 2.01E-02 mSv/6min, and Aluminum body 2.79E-03 mSv/6min, for a total of 4.36E-01 mSv/6min dose received.

In the past, The research focused on estimating the exposure dose at the time of replacing the air foil after a certain period of cooling time due to the risk of high radiation exposure^[11-13].

Now, we can calculate the dose rate that the worker receives at the time of replacement of the havar foil using the same method as the Monte Carlo simulation^[14].

Currently, Number of studies been published on Havar foil as shown in Table 11 and the difference in the results caused by the location measurement, cooling time, etc. depending on the researchers. However, Havar foil is a very high risk of radiation exposure to the workers was demonstrated through a variety studies. The dose of the aluminum body in equilibrium state consisted 0.64% of the total dose. It is believed that it can be used without replacing it as it has a short half-life and low dose. The maximum dose for the silver body consisted 4.6% of the total dose after 30 years. It is believed that it can be used without replacing it as it has a low dose. For havar foil, it was determined that extra caution is required when changing it as it has high level of radiation exposure due to the activated nuclides. For the radioactive waste materials that appeared due to activation, it is believed that the aluminum body and the silver can be used again based on the radiation dose of the nuclides. For havar foil, as it has a high dose, it needs an independent storage facility, and in order to lower the shielding dose to below 1 µSv/hr, the width of the shielding structure needs to be at

least more than 15 cmPb. However, as the number of stored havar foils increases, it was determined that a

shielding structure of 22 cmPb is needed.

Table 11. Comparison research on radioactive Havar foil

D 4		Spectrometry		Monte Carlo s	simulation
Report -	O'Donnell ^[11]	Manickam ^[13]	Bowden ^[12]	M. Serrano ^[14]	this study
Cyclotron			PETtrace		
Proton energy			16.5 MeV		
Cooling period [d]	18	264	18		
^{48}V	$\sqrt{}$		\checkmark		
⁵¹ Cr	\checkmark		\checkmark		\checkmark
⁵² Mn	\checkmark		\checkmark	\checkmark	\checkmark
⁵⁴ Mn	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
⁵⁵ Fe					\checkmark
⁵⁶ Mn					\checkmark
⁵⁶ Co	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
⁵⁷ Co	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
⁵⁸ Co	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
⁶⁰ Co				\checkmark	\checkmark
⁶⁴ Cu					\checkmark
⁵⁹ Fe				\checkmark	\checkmark
95mTc/95Tc			\checkmark		
⁹⁶ Tc			\checkmark		
⁹⁹ Mo					\checkmark
^{99m} Tc					\checkmark
¹⁸³ Re	\checkmark	\checkmark	\checkmark		
¹⁸⁴ Re	\checkmark		\checkmark		
¹⁸⁶ Re					\checkmark
Dose(mSv/6min)	0.24			0.34	0.41

V. CONCLUSION

When producing radioactive isotope in a cyclotron, not only radioactive isotope that we do not want, but unnecessary neutrons are also generated. Such neutrons become the main cause of exposure to radiation by the people working in the field as they activate surrounding substances. With regards to that, as a result of testing the degree of activation in the parts of Targetry in this study, it was revealed that the main cause of radiation exposure received when replacing the parts was due to havar foil, and the aluminum body and the silver body were less than 5% of the total dose. Additionally, it was confirmed that the replaced havar foil needs to be replaced as it has a high dose of radiation, and the aluminum body and the silver body have low dose of radiation which can be re-used without replacing them as they have short half-lifes.

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몬테카를로 모의 모사를 이용한 의료용 사이클로트론의 Targetry 방사화 및 피폭선량 분석

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요 약

의료용 사이크로트론은 방사성의약품을 생산하기 위해 양성자를 고속으로 가속시켜 핵반응을 일으키게 되며, 핵반응을 통해 불필요한 중성자가 발생하게 된다. 중성자는 사이클로트론의 부품에 방사화를 일으키는 원인으로 종사자들의 피폭의 원인이 된다. 이에 본 연구에서는 핵반응이 일어나는 Targetry 부품들인 Al uminum body, Silver body, Havar foil의 방사화 정도를 분석하여 피폭선량을 알아보고자 하였다. 실험결과 Aluminum body와 Silver body는 방사화된 핵종들의 에너지가 작고, 반감기가 짧아 종사들에게 미치는 선량이 미미하였으며, 재사용하는데 문제가 없었다. 하지만 Havar foil의 경우 방사화된 핵종들의 에너지가 높고 반감기가 길어 종사자들에게 미치는 영향이 매우 높았으며, 방사성폐기물로써 특별한 관리가 필용한 것으로 나타났다.

중심단어: 사이클로트론, 방사화, 타겟

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