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A Study on Neutron Activation in a Cyclotron Vault

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1. Introduction

2023 data showed that diagnosed people with cancer was 136 per 100,000 people in Indonesia [1]. To diagnose cancer, positron emission tomography (PET) is used. Radionuclide and radiopharmaceuticals for PET are produced by another facility, which is a cyclotron. As of 2024, in Indonesia, there are only 4 hospitals that have a cyclotron facility, which are Gading Pluit Hospital, MRCCC Siloam Hospital, Dhamais Cancer Hospital and Abdul Wahab Sjahraine Hospital [2]. Cyclotron produced many types of radiopharmaceuticals, one of them is ^{18}F. Cyclotron accelerates the beam of charged particle in a spiral path. The particle then focused onto a target, in this case, ^{18}O in a form of water H_2O^{18}, and the bombardment cause the production of the desired radioisotopes. Production of ^{18}F is shown in following formula:

$$^{18}_{8}O + \, ^{1}_{1}p \rightarrow \, ^{18}_{9}F + \, ^{1}_{0}n + \gamma$$

During the production of PET radionuclides, substantial amounts of neutron are generated around cyclotrons. These neutrons, produced during operations, can lead to both direct and indirect exposure within the vault. Various elements in the concrete and cyclotron components can become activated by neutrons, leading to formation of long half-life radionuclides such as ^{60}Co and ^{134}Cs which can pose a radioprotection challenge for the public safety [3].

National Research and Innovation Agency of Indonesia is currently planning to build a cyclotron facility at Pasar Jumat Nuclear Area. With the high demand of radiopharmaceutical, it will be a great deal to build a cyclotron facility. While the construction of the cyclotron begins, the calculation of activation caused by neutron at cyclotron facility have to be calculated. Activated matter can be consider as a nuclear waste when the facility is decommissioned. The clearance level has been established to remove the radioactive materials from any regulation control. Any materials which contain radioactive nuclides below the levels can be declared as clearance materials resulting in reuse or disposal of as non-radioactive wastes [4]. Monte Carlo simulation software called Particle and Heavy Ion Transport code System (PHITS) was used in this study.

2. Method and Result

This study will focus on the material comparison and neutron activation inside a cyclotron vault. Data of cyclotron is from a hypothetical study of a constructed cyclotron in Pasar Jumat Nuclear Area, Indonesia. ^{18}F will be the product of the cyclotron operation. To produce ^{18}F radiopharmaceutical, 11~18 MeV energy of cyclotron is needed [5]. In this study, 18 MeV of energy was used. Cyclotron vault will have the height of 3 meters, width of 3 meters and length of 4 meters with the vault wall thickness of 10 cm. Geometry of the cyclotron vault is shown in the Fig. 1.

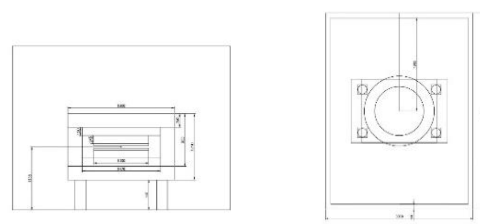


Fig. 1. Front View and Top View.

Composition of cyclotron wall in this study will use 3 different materials which are Portland Cement (Material 1), Barite Cement (Material 2), and Material 3. Material 3 is the material composition of a cyclotron vault wall in S. Orsola Malpighi Hospital, Italy. Material 3 is used as a comparison for other 2 materials. Density of each material are 2.3 g/cm^3, 2.35 g/cm^3 and 3.35 g/cm^3, respectively. Composition of each material is shown in Tabel I [6,7].

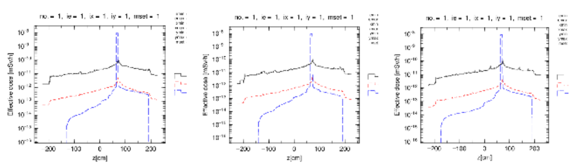
Tabel I: Material Composition (%)

	Material 1	Material 2	Material 3
H	0.168753	0.109599	0.01
C	0.001416	-	0.001
O	0.562526	0.600196	0.667
Na	0.011838	-	0.0013
Mg	0.001400	0.001515	0.0035
Al	0.021354	0.004777	0.018
Si	0.204199	0.011474	0.24

K	0.005656	-	0.0038
Ca	0.018674	0.038594	0.037
Fe	0.004264	0.026213	0.016
Ba	-	0.103985	0.0025
S	-	0.103647	-
Mn	-	-	0.0011
Ti	-	-	0.00095
Ni	-	-	0.000031
Co	-	-	0.0000019
Nb	-	-	0.0000023
Eu	-	-	0.000000066
Cs	-	-	0.0000012

2.1 Material Comparison

Each of the wall material needs to reduce the effective dose from the cyclotron operation. In this study, source is located at the middle of the vault geometry and calculation of the effective dose is performed using PHITS. The effective dose of each material used the conversion coefficient based on ICRP 103. Each material ability to reduce the effective dose is shown in Fig. 2. Material 2 (Barite Concrete) showed to be a better shielding composition for the cyclotron vault. It can reduce the effective dose due to cyclotron operation compared with two other materials.



Material 1 Material 2 Material 3

Fig. 2. Material Comparison for Effective Dose.

2.2 Neutron Activation

To calculate the neutron activation, time of cyclotron operation and shut-down time is needed. The cyclotron is assumed to have 10 years operation. Time after cyclotron decommissioning will be 6 months where there will be a dismantling of the cyclotron wall and its components. Assumption of cyclotron work hour per day is 4 hours in 300 days in 1 year. Using DCHAIN of PHITS code, the calculation of neutron activation that occurs for 10 years can be done. Calculation for activated materials after 10 years of operation is shown in Fig. 3.

```
dominant nuclides (top 10)
--- -------- Activity ----------------------------
no. nuclide     [Bq/cc]       [Bq]  rel. err.    [%]
 1   K  40   1.9473E+01 1.7526E+06 3.1310E-22 100.00
 2   Ca 48   1.2027E-07 1.0825E-02 0.0000E+00   0.00
 3   Sc 48   3.0069E-08 2.7062E-03 2.4232E-06   0.00
 4   Al 28   2.3981E-09 2.1583E-04 7.5169E-02   0.00
 5   Eu151   8.1595E-10 7.3435E-05 2.6413E-33   0.00
 6   Ar 37   2.7811E-10 2.5030E-05 6.4575E-02   0.00
 7   Na 24   1.1664E-10 1.0498E-05 8.7660E-02   0.00
 8   Mn 56   1.1386E-10 1.0248E-05 1.2978E-01   0.00
 9   Mg 27   1.1008E-10 9.9075E-06 8.4785E-02   0.00
10   Na 24m  6.6370E-11 5.9733E-06 9.0574E-02   0.00
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Material 1

```
dominant nuclides (top 10)
--- -------- Activity ----------------------------
no. nuclide     [Bq/cc]       [Bq]  rel. err.    [%]
 1   K  40   1.5900E+01 4.5075E+05 5.1812E-22 100.00
 2   Ca 48   1.1350E-07 3.2178E-03 0.0000E+00   0.00
 3   Sc 48   2.8375E-08 8.0444E-04 6.2249E-07   0.00
 4   Eu151   3.3310E-10 9.4434E-06 5.3272E-34   0.00
 5   Al 28   2.8379E-10 8.0454E-06 2.7804E-01   0.00
 6   Ar 37   1.3619E-10 3.8609E-06 2.1570E-01   0.00
 7   P  32   1.0833E-10 3.0711E-06 2.1658E-01   0.00
 8   Mg 27   1.7881E-11 5.0693E-07 4.0863E-01   0.00
 9   Mn 56   1.4601E-11 4.1394E-07 2.9759E-01   0.00
10   Pm147   1.1833E-11 3.3546E-07 1.3816E-04   0.00
```

Material 2

```
dominant nuclides (top 10)
--- -------- Activity ----------------------------
no. nuclide     [Bq/cc]       [Bq]  rel. err.    [%]
 1   K  40   1.0946E+01 3.1033E+05 4.0271E-22 100.00
 2   Ca 48   6.2910E-08 1.7835E-03 0.0000E+00   0.00
 3   Sc 48   1.5728E-08 4.4588E-04 2.0103E-07   0.00
 4   Al 28   1.5547E-10 4.4075E-06 2.6393E-01   0.00
 5   P  32   1.0910E-10 3.0930E-06 2.1569E-01   0.00
 6   Ar 37   7.6001E-11 2.1546E-06 2.1249E-01   0.00
 7   Eu151   7.4879E-11 2.1228E-06 6.8051E-34   0.00
 8   Mg 27   1.1623E-11 3.2953E-07 2.3719E-01   0.00
 9   N  16   6.0473E-12 1.7144E-07 2.0778E-01   0.00
10   Na 24   4.4831E-12 1.2710E-07 2.6260E-01   0.00
```

Material 3

Fig. 3. Activation Material after 10 Years Operation.

In Fig. 3 above, most of the generated dominant radionuclides are solid form except ^{16}N and ^{37}Ar. The radionuclides came from the neutron activation with the composition of air inside the cyclotron (N, O and Ar). The rest of the dominant radionuclides came from the wall composition. Most of the dominant radionuclides that generated at Material 1 and Material 2 have short half-life except for ^{40}K (1.25×10^{12} years) and ^{151}Eu (4.62×10^{18} years). Both of the radionuclides have low specific activity with ^{151}Eu is 8.1595×10^{-10} Bq/cc for Material 1 and 3.3310×10^{-10} Bq/cc for Material 2, and ^{40}K is 19.473 Bq/cc for Material 1 and 15.9 Bq/cc for Material 2. Both radionuclides are considered as Very Low-Level Waste (VLLW) because of the long half-life and low specific activity. In real life, Eu can be found in form of ^{152}Eu inside of concrete with the concentration of 0.707 ppm [8].

For Material 3, Eu is generated because of the composition of Eu is written for input data. ^{151}Eu specific activity after cyclotron operation is 7.4879×10^{-11} Bq/cc, which is also considered as Very Low-Level Waste (VLLW). Before dismantling, activation of materials is calculated with DCHAIN where time is set to 6 months after shut down. Fig. 4 shows the radionuclides that are generated. Some radionuclides have increased amount of specific activity. A new radionuclide, ^{147}Pm, is also generated.

^{147}Pm has half-life of 2.6234 years and emits beta particle. ^{147}Pm that generated at the wall is considered as Very Low-Level Waste (VLLW).

dominant nuclides (top 10)

no.	nuclide	Activity [Bq/cc]	[Bq]	rel. err.	[%]
1	K 40	1.9473E+01	1.7526E+06	3.1491E-21	100.00
2	Ca 48	1.2027E-07	1.0825E-02	0.0000E+00	0.00
3	Sc 48	3.0068E-08	2.7061E-03	0.0000E+00	0.00
4	Eu151	8.1595E-10	7.3435E-05	4.5599E-31	0.00
5	Pm147	3.1703E-10	2.8533E-05	3.5566E-04	0.00
6	H 3	2.6580E-11	2.3922E-06	9.3263E-02	0.00
7	Ar 37	1.2684E-11	1.1416E-06	6.4361E-02	0.00
8	Mn 54	8.9520E-12	8.0568E-07	8.9552E-02	0.00
9	Fe 55	4.1398E-12	3.7258E-07	8.9576E-02	0.00
10	Ca 45	2.5500E-12	2.2950E-07	1.2809E-01	0.00

Material 1

dominant nuclides (top 10)

no.	nuclide	Activity [Bq/cc]	[Bq]	rel. err.	[%]
1	K 40	1.5900E+01	4.5075E+05	2.4343E-21	100.00
2	Ca 48	1.1350E-07	3.2178E-03	0.0000E+00	0.00
3	Sc 48	2.8375E-08	8.0444E-04	1.0701E-06	0.00
4	Eu151	3.3310E-10	9.4434E-06	4.5819E-32	0.00
5	Al 28	2.8045E-10	7.9508E-06	3.0798E-01	0.00
6	Ar 37	1.5489E-10	4.3912E-06	1.9022E-01	0.00
7	P 32	8.4624E-11	2.3991E-06	1.9487E-01	0.00
8	Pm147	7.4507E-11	2.1123E-06	4.6070E-04	0.00
9	Mn 56	2.1037E-11	5.9640E-07	4.0656E-01	0.00
10	Mg 27	1.5117E-11	4.2856E-07	4.1464E-01	0.00

Material 2

dominant nuclides (top 10)

no.	nuclide	Activity [Bq/cc]	[Bq]	rel. err.	[%]
1	K 40	1.0946E+01	3.1033E+05	3.2542E-21	100.00
2	Ca 48	6.2910E-08	1.7835E-03	0.0000E+00	0.00
3	Sc 48	1.5728E-08	4.4588E-04	0.0000E+00	0.00
4	Eu151	7.4879E-11	2.1228E-06	1.2057E-31	0.00
5	Pm147	2.9093E-11	8.2480E-07	7.5433E-06	0.00
6	H 3	3.7156E-12	1.0534E-07	2.0138E-01	0.00
7	Ar 37	2.9516E-12	8.3677E-08	1.9708E-01	0.00
8	Mn 54	2.0702E-12	5.8689E-08	2.8869E-01	0.00
9	Fe 55	3.2554E-13	9.2290E-09	2.8556E-01	0.00
10	C 14	2.5857E-13	7.3306E-09	2.0136E-01	0.00

Material 3

Fig. 4. Activation Material after 6 Months.

3. Conclusion

Simulation with PHITS code showed that Material 2 (Barite Concrete) was considered as a good choice for a wall composition for a cyclotron vault. It can reduce proton, photon and neutron that are generated by the cyclotron operation. Neutron activation at the wall vault during the cyclotron operation generated some radionuclide with long half-life and low specific activity which are ^{40}K and ^{151}Eu. Because of this, cyclotron wall vault is considered as Very Low-Level Waste (VLLW).

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