Identification of activation products and radiation dose estimates in Havar® foils for a GE PETtrace800 medical cyclotron via high resolution gamma ray spectroscopy and MCNPX simulation

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Abstract. During production of F-18 in a medical cyclotron, high energy protons and secondary neutrons hit the highly reactive components of the target assembly resulting to activation products. In determining the presence of activation products and quantifying their respective activities, high resolution gamma spectroscopy using high purity germanium (HPGe) was used. From the results of the spectroscopy analysis, the doses were estimated using two methods: calculation method and MCNPX simulations. For the gamma spectroscopy analysis, the sample was prepared using aqua regia and was sent to PNRI. The analysis was done using GEM Series HPGe Coaxial Detector System. The results of the gamma spectroscopy showed the presence of ⁵⁶Co, ⁵⁷Co, ⁵⁸Co, and ⁵⁴Mn after a cooling period of eighty (80) days. For the MCNP simulations, the Havar® window foil was simulated as an isotropic disc source and a soft–tissue mimicking box detector was used and positioned at 100 cm from the center of the simulated window foil. *F8 tally was used to estimate the effective dose arising from a single activation product homogeneously distributed in the window foil and was done under the assumption that no other radiation sources were present in the simulation. Using calculation method, the effective dose estimates of ⁵⁶Co, ⁵⁷Co, ⁵⁸Co, and ⁵⁴Mn were found to be of minimal values during replacement of ¹⁸O vial at one meter. Moreover, it was found out that the highest contributors of dose during replacement of an ¹⁸O vial and maintenance of cyclotron are ⁵⁶Co and ⁵⁸Co. After a period of one year, the guiding activation products become ⁵⁷Co and ⁵⁴Mn due to their long half–lives. Furthermore, the annual dose estimates of these activation products were found to not exceed the annual dose limits for the whole body and for the extremities. As for disposal and storage, a period of four years will allow the Havar® window foil to reach the exemption levels set by PNRI.

1. Introduction
A cyclotron is a type of particle accelerator used to produce short – lived radionuclides by bombarding a specific isotope with high – energy charged particles (usually protons, deuterons, or α particles) with the resulting radionuclides being used for PET imaging [1]. During this process, highly reactive components of the target assembly become “radioactivated” because of the high energy protons and
secondary neutrons hitting the target assembly [2]. There are currently 2 medical cyclotron facilities in the Philippines. One is located at Khealth Corporation and the other one is located at St. Luke’s Medical Center – Quezon City. After a certain number of productions, the cyclotron operator goes inside the bunker to replace the empty $^18$O vial located near the target assembly. This replacement process usually takes 2.5 minutes. As part of the preventive maintenance of the cyclotron, the service engineer and cyclotron operator go inside the bunker to perform annual check on the cyclotron. The exposure arising from the activated components of the cyclotron becomes more prominent during annual maintenance since the service engineer and operator spend more time inside the cyclotron bunker. The purpose of this study is to identify and quantify the activation products present in the Havar® foil to determine if the exposure is within the limits set by PNRI.

2. Materials and Methods

Two methods were done to acquire the necessary data: simulations using MCNP to acquire an estimate of the individual dose contribution of each activation products from the Havar® window foil and a high-resolution gamma spectroscopy analysis using HPGe detector in PNRI to identify the presence of these activation products and their count rates as well.

2.1. High Resolution Gamma Ray Spectroscopy and Sample Preparation

The Havar® window foil (figure 1) that was used in this study was removed from the target assembly last July 07, 2017 and was analyzed eighty (80) days post – removal from the target assembly of the GE PETtrace800 medical cyclotron. For the sample preparation, the procedure from the study of O’Donnell et al and Bowden et al was adopted. The window foil, due to its high activity, was disintegrated using aqua regia. Aqua regia can be used to dissolve many metals and alloys including gold, palladium, and platinum. Aqua regia was made by mixing 50 mL of HNO$_3$ and 200 mL of HCl. Due to presence of chromium in the window foil composition, there is a possibility that the spectra might be affected. Aqua regia removes the presence of paramagnetic chromium that might interfere with the spectrum acquisition [3,4].

The disintegrated window foil was placed inside a 250 – mL HDPE wide – mouth Nalgene bottle. A 10 µL sample was taken from this and was volumetrically diluted using distilled water. This sample was sent to PNRI for the gamma spectrum analysis. The gamma spectrum analysis was done using a GEM Series HPGe Coaxial detector system, a high purity germanium detector. A 250 - mL of standard mixed radionuclides like $^{60}$Co, $^{113}$Sn, $^{123}$I, $^{55}$Te, $^{85}$Sr, $^{109}$Cd, $^{57}$Co, $^{139}$Ce, $^{51}$Cr, $^{137}$Cs, $^{241}$Am, and $^{88}$Y were used to calibrate the HPGe detector. The sample was placed on top of the detector with a 0.4 mm distance and the analysis lasted for 6 hours. Equation (1) where $A_a$ = activity per unit area (MBq/m$^2$), $\Gamma$ = specific
gamma ray constant (mSv/hr @ 1 meter/MBq), $L = \text{diameter of source surface in meters (m)}$, $R = \text{distance from source surface in meters (m)}$, and $t = \text{time spent replacing O-18 vial (hr)}$ was used to compute for the dose estimates arising from a disc source [5].

$$D(mSv) = \pi A_a \Gamma \ln \left( \frac{L^2 + R^2}{R^2} \right) t$$

(1)

2.2. MCNP Simulation

For the MCNP simulation, four simulations were done under the assumption that the window foil was made up of a single activation product only, was distributed homogeneously throughout the foil, and was an isotropic source. A box detector was placed 1 m away from the center of the source. The *F8 tally, which calculates the total dose deposition for a given cell, was used for the detector composed of thirteen (13) elements that mimic soft – tissue composition. Each simulation consisted of $10^8$ histories per activation product. MCNP version 5.2 and Visual Editor were used in this study.

3. Results and Discussion of the Study

The results of the analysis showed that $^{56}\text{Co}$, $^{57}\text{Co}$, $^{58}\text{Co}$, and $^{54}\text{Mn}$ were present in the sample. After a cooling time of 80 d, most of the short – lived radionuclides were no longer present during the time of the analysis. The results of the dose estimates using equation (1) and MCNP simulations when treating the Havar® window foil as an isotropic source showed discrepancies. For the MCNP simulations, the four simulations were able to pass the 10 statistical checks. Figure 2 shows the comparison of effective dose for a single replacement of $^{18}\text{O}$ vial while figure 3 shows comparison of the annual effective dose for replacing the $^{18}\text{O}$ vials in one year.

Figure 2. Comparison of effective dose estimates via calculation and via MCNPX simulation for $t = 2.5$ min
Figure 3. Comparison of effective dose estimates via calculation and via MCNPX simulation for \( t = 1 \) hr

The discrepancies seen in figure 2 and figure 3 may be attributed to the stochastic way of solving problems of MCNP. It is also worth noting that the MCNP transport code considers different factors that may affect the effective dose estimates such as the different energies and the respective probabilities of the activation products for each simulation, treating the Havar\textsuperscript{®} window foil as an isotropic source, and oversimplification of the representation of the whole body, among others. The activity decay, shown in figure 4, for each activation product was calculated for a 4-year period. After a period of 1 year, the major contributors, \(^{56}\text{Co}\) and \(^{58}\text{Co}\), would have decayed significantly due to their short half–lives. The guiding activation products would become \(^{57}\text{Co}\) and \(^{54}\text{Mn}\) due to their long half–lives.

Figure 4. Decay of activation products
4. Conclusion
Given that the results of the dose estimates via calculation method do not exceed the annual dose limit of 20 mSv for the whole body and the annual dose limit to the extremities of 500 mSv, it is safe to assume that the contributions of these activation products are insignificant and pose no threat to the radiation worker provided that they maintain the maximum distance possible when replacing the $^{18}$O vial and minimize the time spent inside the bunker. The results of the analysis showed that $^{56}$Co and $^{58}$Co are the highest contributors of dose during replacement of $^{18}$O vial and during maintenance which is consistent with previous studies. After a period of one year, the guiding activation products would become $^{57}$Co and $^{54}$Mn due to their relatively long half-lives compared to the half-lives of $^{56}$Co and $^{58}$Co. For storage and disposal purposes, a period of 4 years will allow the Havar® window foil to reach the exemptions levels set by PNRI.

5. References
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Acknowledgment
The authors would like to thank Khealth Corporation and the Nuclear Analytical Techniques Applications Section (NATAS) of PNRI for the assistance given during this study.