Qualitative Analysis of Long-Lived Residual Radioisotopes in 18 MeV Proton Bombarded Enriched Water Target

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Abstract. Residual radioisotope analysis as a result of cyclotron-based 18F production is of paramount importance since it relates to the radiation safety of patients as well as radiation workers. In this investigation, 18-MeV proton beams were employed to irradiate enriched water (H218O) target for 18F production while Talys Evaluated Nuclear Data Library (TENDL) 2017 were used to study the origins of the radionuclide impurities. Gamma rays emitted by the residual radionuclides were detected using a gamma ray spectroscopic system following a month of decay while their origins were analyzed from the TENDL 2017 nuclear cross-section calculations. Experimental results indicated that several long-lived radionuclides such as 109Cd, 57Ni, 58Co and 56Co were recorded by the gamma ray spectroscopic system. The long-lived residual radionuclides were presumably due to proton interactions with Havar window and Silver body. Using the TENDL 2017-calculated nuclear cross-sections, it was discovered that several nuclear reactions responsible for the residual radioisotopes include 109Ag(p,n)109Cd which corresponded to the generation of 109Cd, 60Ni(p,α)57Co and 58Ni(p,2p)57Co reactions for the formation of 57Co, 58Fe(p,n)58Co reaction for the generation of 58Co, and 56Fe(p,n)56Co reaction for the formation of 56Co. This experimental result can be used as a reference for future production of 18F and other radioisotopes should Havar window and silver body are used in the target system.

1. Introduction

Recent progress on radioisotopes production has been concerning various types of radioisotope production applicable for diagnosis such as 68Ga [1-5], 99mTc [6-7] and therapy such as 153Sm, 186Re [8-17] and many others. Due to emmission of positron particle, 18F radionuclide has been used in Positron Emission Tomography (PET) in nuclear medicine. Fluorine-18 (18F) is currently produced only by cyclotron with proton beam energy ranging from 7 MeV to 18 MeV which has been widely reported elsewhere, in which enriched water (H218O) is used as the target [18-20]. Previous research suggested that the intensity of radionuclidic impurities depended strongly on the proton dose [21]. The target system in 18F production usually consists of a window made of havar foil or Nb foil [22-23], a target body made of silver or titanium [22-23] and the cooling fluid. Interaction between proton beam and havar window and target body could result in radioactivity impurities. In addition the cyclotron
vicinity could be activated by secondary neutrons as a result of radionuclide production which has been reported elsewhere [24].

Using 11-MeV proton beam bombarded to 95% enriched water (H\textsubscript{2}O\textsuperscript{18}) target, Dlugosz-Lisiecka [25] reported formation of \(^{109}\)Cd and \(^{107}\)Cd via \(^{109}\)Ag(p,n)\(^{109}\)Cd and \(^{107}\)Ag(p,n)\(^{107}\)Cd in the target body respectively. Previous investigation by Kamali et al observed other impurities such as \(^{56}\)Co, \(^{52}\)Mn, \(^{54}\)Mn and \(^{110m}\)Ag in 11-MeV proton irradiated enriched water target following 1 hour cooling (after End-of-Bombardment) [23]. Kohler et al [26] investigated radionuclide impurities in 18-MeV proton irradiated enriched water and detected various impurities such as \(^7\)Be, \(^{48}\)V, \(^{31}\)Cr, \(^{52}\)Mn, \(^{54}\)Mn, \(^{55}\)Co, \(^{56}\)Co, \(^{57}\)Co, \(^{58}\)Co, \(^{57}\)Ni, \(^{89}\)Zr, \(^{92m}\)Nb, \(^{93m}\)Mo, \(^{95}\)Tc, and \(^{96}\)Tc. Furthermore, they identified that the impurities came from proton interaction with Nb window and Nb window impurities.

Since the maximum impurities allowed in F-18 is not more than 0.1% according to the European Pharmacopeia [27], the window separating high vacuum cyclotron and the target as well as the target body should be chosen to minimize the impurities. In this work we report the use of Ti and Havar foils as the windows while Niobium is used as the target body. A 97% pure enriched water (H\textsubscript{2}O\textsuperscript{18}) target is bombarded with 18-MeV proton beam while radionuclidic impurities are detected using a gamma ray spectroscopic system. The radioisotope impurities are identified from their gamma ray emissions. In addition, the TENDL 2017 nuclear cross-section data are used to study the origins of nuclear reactions responsible for the experimentally observed radionuclide impurities.

2. Materials and Methods

In this work, the cyclotron employed to generate 18 MeV protons was located in Mochtar Riady Comprehensive Cancer Center (MRCCC) Siloam Hospital, Jakarta, Indonesia. The protons were bombarded to 2 ml enriched water (H\textsubscript{2}O\textsuperscript{18}) target with beam current of 30 µA and irradiation time of 1 hour, which yielded 2.2 Ci \(^{18}\)F at the end of bombardment.

The beam windows were made of a combination 12.5 µm thick natural Ti foil and 50 µm thick Havar foil which consists of cobalt (42.5 %), chromium (20 %), iron (18.1 %), nickel (13 %), tungsten (2.8 %), molybdenum (2 %), manganese (1.6 %), carbon (0.2 %), beryllium (0.04 %), and some other trace elements. The Ti window was placed in front of Havar window so that the Ti window was directly in contact with vacuum chamber. While irradiation of Ti could result in radioactive isotopes, it was expected that they would not contaminate the enriched water target since Havar window was put between the Ti window and enriched water target. The target vessel was composed of a niobium (Nb) cavity which was housed in a stainless steel body. The cooling water flew in the cavity to cool down the Nb material. The whole target system is illustrated in Fig. 1.

![Fig. 1. Target system for \(^{18}\)F production at MRCCC Siloam Hospital, Jakarta, Indonesia.](image-url)

The gamma rays emitted by residual radioisotopes were detected using a gamma ray spectroscopic system consisting of HPGe detector and analytical software and devices following 1 month decay. The
spectroscopic system was calibrated using 3 radioactive sources, e.g. $^{60}$Co, $^{137}$Cs and $^{241}$Am. The radioactive isotopes left over in the enriched water target were then identified from the gamma ray energies and their half lives. In order to study the origins of the residual radioisotopes, the TENDL 2017 data were employed to study nuclear cross-sections and nuclear reactions responsible for the radioisotope formations. In addition, the TENDL 2017 data have been previously used to study cyclotron-based radioisotope production elsewhere [28-36].

3. Results and Discussion

Gamma ray spectrum recorded by the spectroscopic system following 1 month decay is given in Fig. 2, which indicates several gamma rays emitted by residual radioisotopes. The identified radioisotopes include $^{109}$Cd, $^{57}$Co, $^{59}$Ni, $^{57}$Ni, $^{58}$Co, $^{56}$Mn, $^{56}$Co and $^{52}$Mn. The half lives of the residual radioisotopes are between 35.6 hours and 461.9 days. The X-ray recorded at 74.97 keV is presumably due to KL$_3$ transition of Pb [37] as the shielding material. A complete list of nuclear data for the identified long-lived residual radioisotopes is shown in Table 1, which include their nuclear reaction possibilities.

![Gamma rays recorded by gamma spectroscopic system following 1 month decay. The identified peaks belong to (1) X-ray (2) $^{109}$Cd, (3,4) $^{57}$Co, (5) $^{57}$Ni, (6) $^{58}$Co, (7) $^{56}$Co and (8) $^{57}$Ni.](image)

Table 1. Identified long-lived residual radioisotopes and their nuclear reaction possibilities.

| E (keV) | Radioisotope | $T_{1/2}$ | Possible nuclear reaction/occurrence | Threshold Energy (MeV) | Decay mode | Source |
|---------|--------------|-----------|------------------------------------|------------------------|------------|--------|
| 74.97   | X-Ray $^{109}$Cd | -         | KL$_3$ transition of Pb $^{109}$Ag(p,n)$^{109}$Cd | -                      | -          | Shielding |
| 88.03   | $^{109}$Cd    | 461.9 d   | $^{60}$Ni(p,$\alpha$$^{57}$Co $^{58}$Ni(p,2p)$^{57}$Co | 0.27, 8.31 | $\beta^+$ | Havar |
| 122.06  | $^{57}$Co     | 271.7 d   | $^{60}$Ni(p,$\alpha$$^{57}$Co $^{58}$Ni(p,2p)$^{57}$Co | 0.27, 8.31 | $\beta^+$ | Havar |
| 136.47  | $^{57}$Co     | 271.7 d   | $^{60}$Ni(p,$\alpha$$^{57}$Co $^{58}$Ni(p,2p)$^{57}$Co | 0.27, 8.31 | $\beta^+$ | Havar |
| 511     | $^{57}$Ni     | 35.6 h    | $^{58}$Fe(p,$\gamma$$^{57}$Ni $^{58}$Ni(p,n)$^{57}$Ni $^{58}$Ni(p,d)$^{57}$Ni | 0.00, 12.40, 10.20 | $\beta^+$ | Havar |
| 813.32  | $^{58}$Co     | 70.86 d   | $^{59}$Fe(p,n)$^{59}$Co | 3.17 | $\beta^+$ | Havar |
| 848.44  | $^{56}$Co     | 77.24 d   | $^{59}$Fe(p,n)$^{59}$Co | 5.61 | $\beta^+$ | Havar |
| 1365.1  | $^{59}$Ni     | 7.6x10$^4$ y | $^{59}$Co(p,n)$^{59}$Ni | 2.23 | $\beta^+$ | Havar |
Based on the TENDL 2017 nuclear cross-sections, $^{109}\text{Cd}$ is produced from $^{109}\text{Ag(p,n)}^{109}\text{Cd}$ nuclear reaction as seen in Fig. 3 (blue circles), whereas $^{57}\text{Co}$ may be generated from both $^{60}\text{Ni(p,}\alpha)^{57}\text{Co}$ and $^{58}\text{Ni(p,2p)}^{57}\text{Co}$ nuclear reactions, though $^{58}\text{Ni(p,2p)}^{57}\text{Co}$ nuclear reaction may contribute to higher intensity of $^{57}\text{Co}$ than that of $^{60}\text{Ni(p,}\alpha)^{57}\text{Co}$ nuclear reaction since it has much higher cross-section at 18-MeV protons. The presence of $^{57}\text{Co}$ was previously discussed by Kohler et al [26], while $^{109}\text{Cd}$ was earlier reported by Długosz-Lisiecka [25]. Strong gamma ray emission is also seen at 511 keV which presumably due to positrons ($\beta^+$) emitted by $^{56}\text{Co}$, $^{57}\text{Co}$, $^{58}\text{Co}$ and $^{57}\text{Ni}$ that directly interact with electrons and eventually emit gamma ray at 511 keV from the interaction.

Again, based on the TENDL 2017 cross-section sections, three nuclear reactions, i.e. (p,γ), (p,np) and (p,d) may contribute to the gamma rays captured by the spectroscopic system. Of the three reactions, $^{58}\text{Ni(p,np)}^{57}\text{Ni}$ has the highest cross-section while the two others, i.e. $^{56}\text{Fe(p,g)}^{57}\text{Ni}$ and $^{58}\text{Fe(p,d)}^{57}\text{Ni}$ have very low and insignificant cross-section; thus it rules out any possibility of the two reactions responsible for the production of $^{57}\text{Ni}$. To sum up, it is clear that $^{57}\text{Ni}$ is as a result of $^{58}\text{Ni(p,np)}^{57}\text{Ni}$ nuclear reaction. Previous research by Kohler et al [26] also detected $^{57}\text{Ni}$ in the $^{18}\text{F}$ production.

Fig. 3. TENDL 2017 nuclear cross-sections of $^{109}\text{Ag(p,n)}^{109}\text{Cd}$ (blue circles), $^{60}\text{Ni(p,}\alpha)^{57}\text{Co}$ (black squares) and $^{58}\text{Ni(p,2p)}^{57}\text{Co}$ (red filled circles).

Fig. 4. TENDL 2017 nuclear cross-sections of $^{58}\text{Ni(p,n)}^{57}\text{Ni}$ (black circles), $^{58}\text{Ni(p,d)}^{57}\text{Ni}$ (red squares) and $^{56}\text{Fe(p,g)}^{57}\text{Ni}$ (blue filled circles).
The detected $^{58}\text{Co}$ is clearly due to $^{58}\text{Fe}(p,n)^{58}\text{Co}$ reaction, which is proven by the very high nuclear cross-section according to the the TENDL 2017 data. Quite similar result also occurs to $^{56}\text{Co}$ which is generated by $^{56}\text{Fe}(p,n)^{56}\text{Co}$ nuclear reaction. Both $^{58}\text{Fe}(p,n)^{58}\text{Co}$ and $^{56}\text{Fe}(p,n)^{56}\text{Co}$ nuclear cross-sections can be seen in Fig. 5. Furthermore, the present of $^{58}\text{Co}$ and $^{56}\text{Co}$ was previously reported by Kohler et al [26].

Fig. 5. TENDL 2017 nuclear cross-sections of $^{58}\text{Fe}(p,n)^{58}\text{Co}$, $^{56}\text{Fe}(p,n)^{56}\text{Co}$.

After 2.5 years of the $^{18}\text{F}$ production, the residual radioisotopes were measured again, and it was found that only $^{57}\text{Co}$ radioisotope was recoded while the others were not detected. This result agrees with the data captured 1 month after irradiation in which gamma rays emitted by $^{57}\text{Co}$ were the strongest among others. Comparison between the two data are shown in Fig. 6. It should be noted that in this work, radionuclide impurities resulted from proton bombardment of Nb cavity was not observed. Kohler et al [26] reported $^{89}\text{Zr}$ radionuclide impurity which was produced from $^{93}\text{Nb}(p,x)^{89}\text{Zr}$. However, since the half life of $^{89}\text{Zr}$ is only 3.27 days, it could no longer be detected in this investigation.

Fig. 6. Comparison of gamma ray spectrum observed after 1 month decay (red line) and 2 years decay (blue line). Note that the y-axis is in arbitrary unit.
4. Conclusion

Long-lived residual radioisotopes in $^{18}$F production have been detected, identified and analyzed from their gamma ray spectrum following 1 month decay. Several radioisotopes were identified, such as $^{109}$Cd, $^{57}$Co, $^{57}$Ni, $^{58}$Co and $^{56}$Co. The TENDL 2017 nuclear cross-sections have been employed to study the origins of the impurities. It was concluded that $^{109}$Cd was produced from $^{109}$Ag(p,n)$^{109}$Cd reaction, while $^{57}$Co formation was presumably due to $^{60}$Ni(p,$\alpha$)$^{57}$Co and/or $^{58}$Ni(p,2p)$^{57}$Co reactions. In addition, $^{57}$Ni, $^{58}$Co and $^{56}$Co radioisotopes were a result of $^{58}$Ni(p,d)$^{57}$Ni, $^{58}$Fe(p,n)$^{58}$Co and $^{56}$Fe(p,n)$^{56}$Co nuclear reaction respectively. This experimental report agrees with previous reports elsewhere.

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