Neutronic analysis of DECY-13 cyclotron target system as a neutron source for SAMOP

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Abstract. The Centre for Accelerator Science and Technology (PSTA) has developed a subcritical reactor for the molybdenum-99 (\(^{99}\)Mo) production (SAMOP). This device requires an external neutron source. For experimental purposes, SAMOP is still driven by an external neutron source from a critical reactor, i.e., from the radial beam-port of the Kartini reactor. PSTA is also developing a 13 MeV cyclotron (DECY-13) for the production of fluorine-18 (\(^{18}\)F) which can generate neutron as by-products. This cyclotron has an opportunity to be used as an external neutron source for the SAMOP. In this work, particle transport simulations have been carried out to determine the characteristics of the neutron produced by the DECY-13. The simulation results show that DECY-13 produces an average neutron flux of \(2.6347 \times 10^{9}\) particles/cm\(^2\)s and around 97% of them are the fast neutron. Based on the analysis, it concluded that the DECY 13 could be used as an external neutron source for the SAMOP by the addition of a neutron collimator.

1. Introduction
Technetium-99m (\(^{99m}\)Tc) is the most used isotope in medical radiography. Almost all of them are obtained from the molybdenum-99 (\(^{99}\)Mo) decay \([1]\). \(^{99m}\)Tc has a short half-life, so it is necessary to build a production facility that is close to the user \([2]\). \(^{99}\)Mo is generally produced using critical reactors with difficult regulations and expensive construction costs \([3]\). The Centre for Accelerator Science and Technology (PSTA) is developing a device for the production of \(^{99}\)Mo called SAMOP (Subcritical Assembly for Molybdenum-99 Production) \([4]\). SAMOP uses targets in the form of uranyl nitrate [\(\text{UO}_2(\text{NO}_3)_2\)] solution which is placed in the SAMOP core and surrounded by the TRIGA fuel elements. To make it easier to get a license, SAMOP is designed to operate in subcritical conditions \([5]\). However, subcritical devices require an external neutron source \([6]\). For experimental purposes, SAMOP is still driven by an external neutron source from a critical reactor, i.e., from the radial beam-port of the Kartini reactor \([7]\). Furthermore, this device is expected to be operated using an external neutron source from a compact neutron generator \([8]\).

Besides SAMOP, PSTA is also developing the DECY-13 cyclotron. This cyclotron is planned to produce a 50 \(\mu\)A proton beam with 13 MeV energy for fluorine-18 (\(^{18}\)F) production \([9]\). The DECY-13 uses 99% oxygen-18 (\(^{18}\)O) enriched water target. The interaction of protons with the target will produce \(^{18}\)F isotopes and neutrons through the \(^{18}\)O(p,n)\(^{18}\)F reaction \([10]\). These neutrons by-product has an opportunity to be used by SAMOP. The neutron beam that can be used by SAMOP must meet the characteristics such as those from the radial beam-port of the Kartini reactor, which is shown in Table 1.
Table 1. Characteristics of the neutron beam from the radial beam-port of the Kartini reactor [11].

| Neutron energy | Neutron flux (particles/cm²s) |
|----------------|-------------------------------|
| Thermal        | $1.8532 \times 10^8$         |
| Epithermal     | $1.4425 \times 10^8$         |
| Fast           | $2.3076 \times 10^8$         |
| Total          | $5.6033 \times 10^8$         |

To find out the characteristics of the neutron beam produced by the DECY-13 cyclotron, the particle transport phenomena that occur must be resolved. The mathematical method that can be used is the Monte Carlo method [12]. However, due to the complexity of a large number of particles, it is necessary to use a computer program. The software that can be used to simulate the particle transport phenomena based on this method is Monte Carlo N-Particle eXtended (MCNPX) [13]. Thus, the purpose of this work is to carry out particle transport simulations of the DECY-13 cyclotron as an external neutron source for SAMOP, using the MCNPX software.

2. Basic Theory

When particles cross a material, it has a probability to interact or just pass-through without interacting. The probability of specific interaction between particle and matter is called microscopic cross-section and denoted by $\sigma$. It doesn't only depend on the type of material involved, but also by the energy of the incoming particle. The particle may lose energy when interacting with matter. If the particle energy changes, the microscopic cross-section also changes. The microscopic cross-section can be considered as an effective area of a material that is present to the incoming particle [14]. The larger the effective area, the more likely the interaction will occur. The microscopic cross-section is expressed in the units of area. It is described in terms of barns which is equivalent to $10^{-24}$ cm$^2$. If there are particles with flux $\Phi$ that cross one cubic centimeter of material with a microscopic cross-section $\sigma$ and a mass density $\rho$, then the reaction rate $R$ can be calculated using Equation 1.

$$R = \frac{\sigma \Phi \rho N_A}{M_R}$$

$N_A$ is the Avogadro constant and $M_R$ is the molecular mass of the material. An analytical method can be used to determine the condition of the system by assuming the microscopic cross-section value is fixed for a range of energy. The assumption also ignores the beam direction. For a complete picture of the system condition, it is necessary to solve the particle transport phenomena.

There are two types of methods that are generally used to solve particle transport phenomena, that are deterministic and probabilistic. The deterministic method solves the transport equation of the average particles, while the probabilistic simulates individual particles. The Monte Carlo method uses probabilistic analogies and random sampling techniques to obtain numerical solutions. This method has been widely used to solve the particle transport phenomena [15-19].

MCNPX is a software to simulate particle transport phenomena based on the Monte Carlo method [13]. MCNPX performs simulations based on a user input file. The user input file consists of three main parts, namely: cell cards, surface cards, and data cards. Cell cards contain cells definition. Cells are spaces that are bounded by the MCNPX virtual surfaces and are the parts of the MCNPX virtual universe. The user must define every part of the MCNPX virtual universe into specific cells to prevent particles from getting lost. Cells are defined using Boolean algebra based on their position from the MCNPX virtual surfaces. The other part of the input file is the surface cards that describe the surface
specifications that exist in the MCNPX virtual universe. The last part is the data cards that contain data about: the type of the simulated particle, the particle source parameters, the type of material, the output parameters (tally), and the other data required for simulation [12]. In addition to the data in the form of user input files, MCNPX also uses data derived from the libraries, namely cross-section data. If there is no cross-section data of a particular isotope or reaction in the library, then the model-based data will be used [13].

MCNPX is a derivative of MCNP, so the algorithms of both software are similar [13]. MCNP simulates the journey of a particle from its beginning to its end (termination). This process begins by determining the initial energy, position, and the motion direction of the particle. Then MCNP calculates the path length and conducts sampling the type of interaction that the particle will experience based on the cross-section data. Particle is terminated if it experienced an absorption. MCNP will determine the new energy and motion direction of the particle if the interaction is scattering. MCNP will continue to follow the particle until it enters into an unimportant region (zero importance cell), or the particle's energy is below the threshold (energy cutoff). Some interaction produces more than one particle. If it happens, then one particle will be tracked while the others will be stored. One stored particle will be followed after the previous particle is terminated. MCNP repeats this process until no more stored particle left.

Particle history is a series of particle simulations from the creation to the end of its life and its descendants. After completing a particle history, MCNP will start the history of other particles and so on until the program is off or the history cutoff is reached. The MCNP will count the tally that the user requested, after all of the histories is over [20].

3. Method
To find out the characteristics of the neutron beam produced by the DECY-13 cyclotron, the particle transport phenomena that occurred on the target system was simulated using the MCNPX software version 2.7.0 build Win64 MPI. The parameters studied were the flux and energy distribution of the neutrons produced by the DECY-13 cyclotron. In this work, the user input file was created using MCNPX Visual Editor and Microsoft WordPad software. This simulation uses the geometry and material data from the design document of the DECY-13 target system. This system consists of a gas cooling module, the target module, and the liquid cooling module, such shown in Figure 1. The target system is integrated with the DECY-13, using a mounting module made of aluminum 6016. There is a vacuum channel inside the mounting module. The Havar window is used to close this channel. The next part is a gas cooling module that uses argon as a cooling gas and Teflon as a cooling wall. DECY-13 uses 99% oxygen-18 (18O) enriched water target inside a chamber made of aluminum 6061. The Havar window is used again to close this chamber. The last component is a liquid cooling module that uses water as a cooling liquid with aluminum 6061 walls [21].

The simulated particles in this study are neutrons and protons with the source particles in the form of a 13 MeV proton beam with a current of 50 µA which is equivalent to $3.1211 \times 10^{14}$ particles/s [9]. MCNPX uses the cross-section data of neutron interactions from the Evaluated Nuclear Data Files (ENDF/B-V and ENDF/B-VI) and the Evaluated Nuclear Data Library (ENDL 1992), except for $18$O isotopes that use the Intra-Nuclear Cascade Model. This is due to the absence of the evaluated data for these isotopes. MCNPX also uses the cross-section data of the proton interaction from the model, except for the isotopes of hydrogen-1 (H), nitrogen-14 (14N), oxygen-16 (16O) and aluminum-27 (27Al) which uses the ENDF/B-VII data.
4. Result and Discussions

The particle transport simulation of the DECY-13 target system has been carried out using MCNPX software. This software generates the proton source particles on the entry of the vacuum channel. The proton beam then hits the Havar window and enters the cooling gas chamber. After passing through the cooling gas chamber, the proton beam hits the next Havar window and enters the target chamber. On its way, the proton beam interacts with the Havar window, the cooling gas and the target. Most of the interactions that occur are electronic interactions with the water target. This interaction causes many protons to lose energy below the energy limit for a nuclear reaction.

MCNPX will terminate the protons with energy below the threshold (energy cutoff). From the source particles of $3.1211 \times 10^{14}$ protons/s, there are only $1.1891 \times 10^{12}$ protons/s that undergo nuclear interactions. Not all that nuclear reactions are the (p,n) reaction. The DECY-13 only produces a neutron beam of $8.2348 \times 10^{11}$ neutrons/s. The neutrons produced have a diverse angle and energy distribution. On its way out, the neutron interacts with the material of DECY-13. There are $2.4067 \times 10^{10}$ neutrons/s that experience the absorption interactions. There is a neutron flux of $2.6347 \times 10^{9}$ neutrons/cm$^2$s that comes out from DECY-13. Figure 2 shows the energy distribution of the neutron beam produced by DECY-13.

Figure 1. The DECY-13 target system.

Figure 2. Energy distribution of the neutron beam produced by DECY-13
Based on Table 2, the neutrons that come out from the DECY-13 are mostly fast. It must be slowed to match the characteristic of the neutron from the Kartini reactor. We also need to guide neutrons in the desired direction. The addition of a neutron collimator may achieve that condition [22-25]. Overall, DECY-13 produces sufficient neutron flux for SAMOP. It is four times larger than neutron flux from the radial beam-port of the Kartini reactor.

| Neutron energy | Produced by DECY-13 | Radial beam-port of the Kartini reactor |
|----------------|---------------------|----------------------------------------|
| Thermal        | $1.1464 \times 10^6$| $1.8532 \times 10^6$                   |
| Epithermal     | $7.6538 \times 10^7$| $1.4425 \times 10^8$                   |
| Fast           | $2.5570 \times 10^9$| $2.3076 \times 10^8$                   |
| Total          | $2.6347 \times 10^9$| $5.6033 \times 10^8$                   |

5. Conclusion
The DECY-13 produces as much as $8.2348 \times 10^{11}$ neutrons/s through $^18$O($p,n)^{18}$F reaction. There is a neutron flux of $2.6347 \times 10^9$ neutrons/cm$^2$s that comes out from the system, and around 97% of them are the fast neutron. DECY-13 requires a collimator to create a proper angle and energy distribution of the neutron. It concluded that the DECY 13 could be used as an external neutron source for the SAMOP by the addition of a neutron collimator.

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References
[1] Barbosa L et al 2013 Non-HEU Production Technologies for Molybdenum-99 and Technetium-99m (Vienna : International Atomic Energy Agency) p 1
[2] National Academies of Sciences, Engineering, and Medicine 2009 Medical Isotope Production Without Highly Enriched Uranium (Washington DC: National Academies Press) p 17 https://doi.org/10.17226/12569
[3] National Academies of Sciences, Engineering, and Medicine 2016 Molybdenum-99 for Medical Imaging (Washington DC: National Academies Press) p 29 https://doi.org/10.17226/23563
[4] L Wahid et al 2018 J. Phys.: Conf. Ser. 1090 012031 https://doi.org/10.1088/1742-6596/1090/1/012031
[5] S Syarip et al 2018 IOP Conf. Ser.: Mater. Sci. Eng. 434 012012 https://doi.org/10.1088/1757-899X/434/1/012012
[6] Syarip and Tri N H S 2017 Jurnal Pengembangan Energi Nuklir 19 25-31 http://dx.doi.org/10.17146/jpen.2017.19.1.3354
[7] M. Iqbal Farezza W and Syarip 2018 J. Phys.: Conf. Ser. 1090 012013 https://doi.org/10.1088/1742-6596/1090/1/012013
[8] Syarip et al 2018 Proc. of the Pakistan Acad. of Sci.: A. Phys. and Comp. Sci. vol 55 (Pakistan Academy of Sciences) p 21-26
[9] Silakhuddin 2016 Prosiding Pertemuan dan Presentasi Ilmiah – Penelitian Dasar Ilmu Pengetahuan dan Teknologi Nuklir 2016 (Surakarta: Pusat Sains dan Teknologi Akselerator, BATAN – Fakultas Matematika dan Ilmu Pengetahuan Alam, UNS) p 151-157
[10] Sunardi et al 2017 Prosiding Pertemuan dan Presentasi Ilmiah Penelitian Dasar - Ilmu Pengetahuan dan Teknologi Nuklir (Yogyakarta: Pusat Sains dan Teknologi Akselerator) p 41-44
[11] Tegas S and Syarip 2014 Ganendra J. of Nucl. Sci. and Tech. 17 83-90
[12] S A Dupree and S K Fraley 2002 A Monte Carlo Primer: A Practical Approach to Radiation Transport (New York: Springer Science+Business Media) p 1 https://doi.org/10.1007/978-1-4419-9036-5
[13] D B Pelowitz 2008 MCNPX User’s Manual: Version 2.6.0 (New Mexico: Los Alamos National Laboratory) p 1
[14] U.S. Departement of Energy 1993 DOE Fundamental Handbook: Nuclear Physics and Reactor Theory Volume 1 of 2 (Washington DC: U.S. Departement of Energy) mod 2 p 7
[15] Aniti P et al 2017 Indonesian J. of Phys. and Nucl. App. 2 128-136 https://doi.org/10.24246/ipna.v2i3.128-136
[16] Hedarh M. Al-Salahy et al 2019 J. Phys.: Conf. Ser. 1253 012028 https://doi.org/10.1088/1742-6596/1253/1/012028
[17] Farshad Mostafaie et al 2015 Physiol. Meas. 36 2057 https://doi.org/10.1088/0967-3334/36/10/2057
[18] J Inamarga et al 2019 J. Phys.: Conf. Ser. 1248 012002 https://doi.org/10.1088/1742-6596/1248/1/012002
[19] H R Khaleghi and M Hassanzadeh 2018 J. Phys. Commun. 2 035030 https://doi.org/10.1088/2399-6528/aab51d
[20] X-5 Monte Carlo Team 2003 MCNP – A General Monte Carlo N-Particle Transport Code, Version 5: Volume I: Overview and Theory (New Mexico: Los Alamos National Laboratory) ch 1 p 3
[21] Ichwan S 2015 DokumenTeknis:TGRG/PTRR-BATAN/08/2015 (Serpong: Badan Tenaga Nuklir Nasional)
[22] S Wonglee et al 2018 J. Phys.: Conf. Ser. 1144 012095 https://doi.org/10.1088/1742-6596/1144/1/012095
[23] Muhammad Aliff Ashraff Rosdi et al 2019 IOP Conf. Ser.: Mater. Sci. Eng. 555 012016 https://doi.org/10.1088/1757-899X/555/1/012016
[24] D Wahyuningsih et al 2018 IOP Conf. Ser.: Mater. Sci. Eng. 288 012007 https://doi.org/10.1088/1757-899X/288/1/012007
[25] Safwan Shalbi et al 2019 IOP Conf. Ser.: Mater. Sci. Eng. 555 012017 https://doi.org/10.1088/1757-899X/555/1/012017