Moderator material efficiency of neutron energy slowing down on D-T reaction neutron generator for SAMOP

S Santosa¹, A H Anggraini²

¹²Center for Accelerator Science and Technology, CAST- BATAN, Yogyakarta, Indonesia

anjar.ah@batan.go.id

Abstract. We have studied a collimator design setup for the Subcritical Assembly for Mo-99 Production (SAMOP) at the Center for Accelerator Sciences and Technology (CAST), Nuclear Energy Agency of Indonesia (BATAN). The main purpose of the project is for providing sufficient neutrons to produce Mo-99, to be used to produce Tc-99m for nuclear medical applications. The modelling of the collimator is based on Monte Carlo simulation using MCNPX version 5 software to determine the variation of the suitable materials. The collimator consists of moderator, filter, and reflector, where we expected that the output of neutron will be thermal neutron with a sufficient neutron flux of about $10^8$ neutron/cm².s. The neutron source used is a D-T reaction neutron generator with a mono-energetic neutron energy of 14.1 MeV. The simulation has been conducted in nine variations, with each experiment uses five types of materials. The best result in terms of neutron flux is seen from the material combination of Al, C_{25}H_{52}, LiF, and PbF₂, which produces a thermal neutron flux of $1.24\times10^7$ neutron/cm².s and average energy deposition of $1.88\times10^6$ MeV/g. This research needs to be developed further for subsequent experiments to optimize the combination of materials and geometry systems to be used.

1. Introduction

The project has the main purpose of producing Mo-99, which thus is used for producing Tc-99m for nuclear medical applications [1]. The Subcritical Assembly for Mo-99 Production (SAMOP) is a new method for radioisotope production. SAMOP utilizes a solvent of uranyl nitrate salt as irradiation target for a neutron beam output. The neutrons are usually produced by nuclear reactor [2], but it also can be produced by using a neutron generator (NG), in which a D-D (deuterium-deuterium) or D-T (deuterium-tritium) reaction occurs [3]. This paper concentrates on the D-T reaction that produces monoenergetic neutrons at 14.1 MeV. In this collimator modelling study, we used an NG as a neutron source, which is considered as good for use on a SAMOP system because of it has a single energy. In a SAMOP system, NG is one of the alternative replacements of neutron sources as a neutron source. Some countries have been using D-T-/D-D-reaction NG as a compact experimental accelerator driven subcritical facility, because of its superiority in, for instance, safety and waste transmutation [3]. We use the D-T reaction NG with an energy of 14.1 MeV in our simulation study.

In Figure 1, shown is the initial design of collimator using a compact neutron generator, which consists of moderator, filter and reflector. The research was conducted to design a better moderator that produces thermal neutrons for SAMOP, for a NG neutron source. The method used is simulation using
a Monte Carlo N-Particle software package, MCNPX version 5. The simulation was done by optimizing the collimator part to produce the output in accordance with the requirements in Mo-99 production on SAMOP system. Therefore, the variation of material and geometry of the collimator is required to qualify SAMOP system in accordance with the standards of the International Atomic Energy Agency (IAEA) [4]. Part of the materials is varied in order to adjust the degradation of fast neutrons energy into the energy required in SAMOP system using D-T reaction.

![Figure 1: The initial design setup of collimator using CNG.](image)

2. Experimental method
The design of neutron collimator using Monte Carlo N-Particle (MCNPX) method is performed by using cell cards to determine the variation of the materials. It uses a D-T reaction NG with neutron energy of 14.1 MeV. The material on the collimator positioned between the SAMOP system and the NG is divided into five main parts: cell 18, cell 15, cell 12, cell 20, and cell 6, which are moderator, neutron filter, reflector, collimator, and reflector, respectively.

2.1. Moderator
Moderator is a part of collimator that used to reduce the energy of fast neutrons (± 2 MeV) into normal reactor components (± 0.02 to 0.04 eV). As the D-T reaction of NG produces neutron energy of 14.1 MeV, the moderator material must be chosen in accordance with the requirement that it can eliminate most of the fast neutron energy in each collision and small ability to absorb neutrons, as well as a great ability to dissipate neutrons. Materials in this experiment are aluminum (Al), aluminum oxide (Al₂O₃) [4], and paraffin (C₂₅H₅₂) [5].

2.2. Filter
Filters are expected to reduce contamination of gamma rays and fast neutron current. The gamma rays could pose high hazard on the human body, so the presence of filters in such a collimator to reduce the radiation of gamma rays is vital.

Fast neutrons can also cause harm to the human body. In general, since the source of neutron has high energy neutron, materials that can reduce the energy is required. The fast neutrons must not be captured because of the intensity of irradiation will be lowered, but the fast neutrons must be moderated
in order to keep the high intensity. The filter is normally a material with a small atomic mass number. However, the material must also have a small neutron absorption cross-section, a large scattering cross-section, and a good heat conductivity. Materials used in this experimental setup of simulation are aluminium fluoride (AlF₃), lithium fluoride (LiF), and paraffin (C₂₅H₅₂) [6].

2.3. Reflector
The reflector has a function to keep neutrons in the collimator. The reflector material has a scattering cross-sectional requirement of a relatively large atomic mass. The suitable materials that are used in this section of simulation are Pb and PbF₂.

The energy output generated from each material is given in the average flux passing through each cell variation. There are three main parts in the observation of the average flux of neutrons in cell 20 and surface 47, so that the average flux for thermal, epithermal and fast neutrons is known. In addition, the cell 20 is also known deposition energy of the protons and neutrons that have units of MeV/g. In the next section of surface 47, the average flux for protons is obtained.

3. Modelling of collimator geometry
Our collimator design utilized the MCNPX Monte Carlo simulation software. In general, there are two methods: shift and moderator methods. The shift method aims to reduce the flux of neutrons made by moderator materials only, while the moderator method performs neutron flux reduction using moderator component in the collimator. In this simulation, we use both methods for such a simulation.

As we have mentioned, the SAMOP collimator design uses NG as its neutron source, in which the D-T reaction is used, for an output energy of 14.1 MeV. The design requires particular techniques in the process to conform to the requirements of the SAMOP system. Material for such a collimator is varied, so that a large initial energy can be reduced. Figure 2 shows the material variation of the SAMOP collimator design. The design of the collimator is given a variety of materials in such a way on each part of the collimator it is done in order to achieve the purpose of SAMOP system requirement.

![Figure 2. Collimator design geometry on MCNPX software.](image)

4. Results and discussion
The moderator material structure is used to slow down the fast neutron from the DT reactor as the neutron source for the SAMOP. This is actually a neutron generator having a tritium target bombarded by deuterium gas. The process is very important for filtering the resulting gamma rays and slowing down it fast neutrons output. The fast and epithermal neutrons must be filtered, due to the thermal neutron
energy being the only one designed to be used in the SAMOP system. Therefore, the first process of moderation is to moderate neutrons to rapidly reduce neutron energy into thermal neutrons, and then to filter the thermal neutrons in order to support the SAMOP work system and to ensure that its intensity does not pose risk for the human body. In Table 1, the variation of materials used at SAMOP collimator system are compared based on the results of the thermal neutron flux and energy output of each material combination.

Table 1. The variation of material using DT source.

| Material in cell | The average of neutron flux (neutron/cm².s) at cell 20 | The average of photon flux (particle/cm²) at surface 47 | The average of deposition energy at cell 20 (MeV/g) |
|------------------|--------------------------------------------------------|------------------------------------------------------|--------------------------------------------------|
| 1 Al AlF₃ Fe Pb Pb | (3.1E+04)±0.0399 | (6.4E+08)±0.1144 | (7.3E+07)±0.0230 |
| 2 Al₂O₃ AlF₃ Fe PbF₂ PbF₂ | (4.1E+06)±0.0072 | (1.0E+07)±0.0884 | (3.8E+07)±0.0301 |
| 3 Al LiF Pb PbF₂ PbF₂ | (2.3E+05)±0.0090 | (2.2E+07)±0.0173 | (7.3E+08)±0.3569 | (4.2E+07)±0.0263 |
| 4 C₂₃H₅₂ LiF Pb PbF₂ PbF₂ | (1.5E+07)±0.0038 | (9.7E+06)±0.0148 | (2.1E+08)±0.0747 | (3.8E+07)±0.0301 |
| 5 Al₂O₃ AlF₃ Fe PbF₂ PbF₂ | (8.5E+06)±0.0038 | (3.0E+05)±0.0341 | (8.8E+09)±0.7014 | (4.1E+07)±0.0250 |
| 6 Al₂O₃ AlF₃ LiF PbF₂ PbF₂ | (1.8E+06)±0.0286 | (2.3E+05)±0.0341 | (1.3E+06)±0.0747 | (3.8E+07)±0.0301 |
| 7 Al C₂₃H₅₂ Pb PbF₂ PbF₂ | (2.0E+07)±0.0033 | (2.0E+05)±0.0201 | (1.0E+07)±0.0678 | (4.1E+07)±0.0250 |
| 8 Al C₂₃H₅₂ AlF₃ PbF₂ PbF₂ | (9.4E+06)±0.0201 | (9.2E+06)±0.0241 | (3.1E+08)±0.3818 | (7.3E+07)±0.0230 |
| 9 Al C₂₃H₅₂ LiF PbF₂ PbF₂ | (5.6E+06)±0.0065 | (5.2E+06)±0.0039 | (3.1E+07)±0.0241 | (1.8E+08)±0.0187 |

Preparation of the moderator materials is required to reduce the fast and epithermal neutrons fluxes, while constantly prevent the epithermal neutrons slightly absorbed. The characteristic of material section is having a high absorption cross section and a high scattering cross section for fast neutrons and epithermal neutrons, as presented in Table 2. In this research, several moderator materials, namely C₂₃H₅₂, Al, and Al₂O₃, have been simulated. The effective material value is of Al as it is adequate to produce high thermal neutron flux, as shown in Table 1. Aluminum has a great inelastic scattering cross section to the neutron energy of 1 MeV and fast neutrons, and it has a low absorption at cross section for thermal neutron in particular energies. The suitable material characteristic for moderator is a material with small atomic mass, which thus produces inelastic collisions. In the process, half of the neutron energy can be transferred into atomic nucleus and will be smaller at the collision.

The filter material was selected based several characteristics, namely material’s cross section using quantity of neutrons that interact with the atomic nucleus in the surface. The cross section is divided into two parts: microscopic and macroscopic. The microscopic cross-section is sum of two parts, the collision cross section and the absorption cross section. The collision cross section is the sum of of elastic collision and inelastic collision cross sections. The cross section is divided to fission and radiation
capture cross sections. Elastic collisions occur due to a collision between fast neutrons and low mass of atomic nucleus. Inelastic collisions occur between neutrons and high atomic number atoms.

Neutrons will provide most of the energy in the atomic nucleus material which the core material will be in the excited state and γ-ray radiation in the inelastic collision. Excessive γ radiation should be avoided as it has a significant effect on the production of Mo-99 on SAMOP systems.

The higher flux of thermal neutron is the combination of collimator materials number nine, it consists of Al, C_{25}H_{52}, LiF, and PbF_{2} of 1.24\times10^{7}\text{ n/cm}^{2}\text{.s}. The selected material on the filter is C_{25}H_{52} (paraffin) which produces thermal neutrons and neutrons faster than epithermal neutrons. Figure 3 shows that paraffin (filter in material number nine) can prevent the reduction of neutrons during the moderation process so that the expected number of neutrons remains stable [8]. In addition to paraffin, AlF_{3} and LiF (shown in Table 1) are also good candidates for filling material in the filter section, but the materials produce higher epithermal flux mean in material number three and six (as shown in Figure 3). This causes the precedence of paraffin in filling the filter parts. Paraffin (C_{25}H_{52}) also has a low atomic number because it contains more hydrogen atoms than carbon atoms, so this material is more effective for lowering thermal energy compared to AlF_{3} and LiF. In Figure 3, it shown that the combination of collimator materials number one, i.e., Al, AlF_{3}, Fe, and Pb, produces the lowest thermal neutron flux of 1.31\times10^{4}\text{ n/cm}^{2}\text{.s}.

Table 2. Material characteristic based on neutron variety [7].

| Neutron variety | Energy range | Material characteristics                        |
|-----------------|--------------|-----------------------------------------------|
| Thermal         | 0 – 5 eV     | High absorption cross section and high scatter cross section |
| Epithermal      | 5 eV – 10 KeV| Low absorption cross section and low scatter cross section |
| Fast            | 10 KeV – 20 MeV | Low absorption cross section and high scatter cross section |

Figure 3. Neutron flux at material sample.

The material characteristics of the reflector section should have a high neutron scattering cross-section. The materials used in this section are Pb and PbF_{2}. In this research, the DT neutron source is directed to the material so that it can filter fast neutrons and gamma rays (photons) in the composition of the reflector material with PbF_{2} material. The reflector part easily resonates where elastic scattering occurs because neutrons react with atomic nucleus of high atomic numbers. This resonance caused largely dissipated energy neutrons.
Figure 4. Average of deposition energy at the material sample.

Table 3. Energy range for neutrons and required conditions of neutron fluxes [9].

| No. | Neutron   | Energy      | Neutron flux (n/cm².s) |
|-----|-----------|-------------|------------------------|
| 1.  | Thermal   | ~0.5 eV     | < 5×10⁷                |
| 2.  | Epithermal| 0.5 eV-10 keV | >1.5×10⁹             |
| 3.  | Fast      | 10 keV      | < 10¹²                |

The best result of neutron thermal flux is 1.24×10⁷ n/cm².s (Figure 3) and average energy deposition of 1.88×10⁶ MeV/g (Figure 4). The thermal neutron flux results are still smaller than expected for SAMOP that of 10⁸ neutrons/cm², but the result is less than the value set by the IAEA of 5×10⁷ as shown in Table 3. In the other hand, the minimum acceptable value from others research measurements of thermal and epithermal neutron flux on the neutron extractor is 10⁶ to 10⁷ (n/cm².s) [10, 11].

5. Conclusion
The best result of neutron flux is produced by the material combination of Al, C₂₅H₅₂, LiF, dan PbF₂, which resulted in a thermal neutron flux of 1.24×10⁷ neutron/cm².s and an average energy deposition of 1.88×10⁶ MeV/g with D-T neutron source. Thus this research needs to be developed for subsequent experiments to optimize the combination of materials and geometry systems to be used.

Acknowledgment
The authors of the paper would like to thank the Head of PSTA-BATAN and Professor Syarip who have allocated research fund from INSINAS 2016 so that this research can be implemented. They are also grateful to Alzero, Made, and Airlangga University students who have helped the authors in this research. Hopefully they are all given reward by Allah SWT, Aamiin.

References
[1] Setiadipura T and Saragi E 2007 Int. Conf. on Adv. in Nucl. Sci. and Engin. 2007 23–6
[2] Sakai M Fujimoto N Ishii K Murata I and Awazu K 2011 Nucl. Sci. and Tech. 1 513–6
[3] Sinha A Roy T Kashyap Y Ray N Shukla M Patel T Bajpai S Sarkar P S Bishnoi S and Adhikari P S 2015 Annals of Nucl. Energy 75 590–4
[4] Fauziah N 2013 J. Tech. Reactor Nucl. 15 112–9
[5] Bavarnegin E Kasesaz Y and Wagner F M 2017 J. Instrum. 12(5) P05005
[6] Laara S H 2014 Islam State Univ. Sunan Kalijaga (in Indonesia)
[7] de Boer J J 2008 New Filter Design with Monte Carlo Calculation TU Delft
[8] Gulo D P Suryasatriya T Santosa S and Sardjono Y 2015 *Indonesian J. of Appl. Phys.* 5(2) 25–33
[9] Sato A Takizawa Y Hiraga F and Kiyanagi Y 2014 *Phys. Proc.* 60(C) 15–22
[10] Leal A Campolina D Horizonte B and Revay Z 2011 *Proc. IAEA-CN-188-A19* 1–8
[11] Sutondo T 2015 *Ganendra J. Nucl. Sci. Technol.* 18 107–13