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Compact Neutron Generator as External Neutron Source of Subcritical Assembly for Mo-99 Production (SAMOP)

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Abstract. Subcritical assembly for Mo-99 production (SAMOP) has been developed at The Center for Accelerator Science and Technology (CAST), National Nuclear Energy Agency (BATAN). The objective of the SAMOP development is to provide a prototype of a subcritical assembly which produces Mo-99 isotope. In the nuclear medical application, Mo-99 plays an important role as a generator of isotope Tc-99m which is the most widely used radioisotope for diagnostic purposes. SAMOP use uranyl nitrate solution $UO_2(NO_3)_2$ as fuel as well as target material for the process of Mo-99 production. This process is based on the fission reaction of uranium U-235 contained in the uranyl nitrate solution. The U-235 fission reactions occur as long as there are still neutrons from external neutron source and it will stop whenever the external neutron source is removed. Presently SAMOP uses external neutron source from the radial beam port of the Kartini research reactor which emerges thermal neutrons in the order of 10^8 n/cm^{2}s. For further development, SAMOP will be equipped with a compact neutron generator as an external neutron source. Compact neutron generator is a neutron source device that contains a compact ion accelerator and that produces neutrons by fusing isotopes of hydrogen. The fusion reactions take place in the device by accelerating either deuteron, triton, or a mixture of these two isotopes into a metal hydride target which also contains deuterium, tritium or a mixture of these isotopes. The fusion of deuterium atoms (D + D) results in the formation of a He-3 ion and a neutron with a kinetic energy of approximately 2.5 MeV. The fusion of a deuterium and a tritium atom (D + T) results in the formation of a He-4 ion and a neutron with a kinetic energy of approximately 14.1 MeV. SAMOP needs an external neutron source which can provide neutrons with an average neutron flux of 10^8 n/cm^{2}s for maintaining the sustainability of U-235 fission reactions. The results of the study on compact neutron generator as an external neutron source of SAMOP show that a compact neutron generator has a chance to substitute a nuclear reactor as external neutron source of SAMOP. A compact neutron generator can produce fast neutrons which neutron yield is sufficient to meet the need of neutron for SAMOP. It depends on the type of the compact neutron generator, one of which is the compact neutron generator produced by LBNL i.e. the axial compact neutron generator which can produce neutron yield up to 10^9 n/s. The benefit of using a compact neutron generator as an external neutron source of SAMOP is that the operational cost will reduce since the neutron production cost using a compact neutron generator is cheaper than a nuclear reactor. In addition, the operational procedure of a compact neutron generator is simpler than a nuclear reactor.

INTRODUCTION

It has been proven that nuclear technology is useful for human welfare. Currently, the application of nuclear technology covers various fields such as agriculture, environment, industry, health, energy, etc. In health, nuclear technology is applied for diagnosing and therapy of diseases as well as for nuclear medicine research. Nuclear medicine is a branch of medicine that uses radioisotope to study the change of physiology, anatomy and biochemical. The rapid progress of nuclear medicine has been supported by the production of various radioisotopes and radiopharmaceutical as well as by the development of medical nuclear instrumentation[1].

Among the various radioisotopes used in nuclear medicine, metastable technetium-99 (Tc-99m) is the most widely used radioisotope for medical applications. Around 80% of the whole diagnoses in the world are conducted by using Tc-99m. This is due to the ideal characteristic of Tc-99m as a diagnostic radioisotope. Tc-99m is a pure γ

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emitting radioisotope that has a half-life of 6 hours and γ energy of 140.5 keV. Therefore it does not give a negative impact on the patient body, but it capable to penetrate the body tissue and easily detected from outside of the body using a radiation detector to measure the Tc-99m distribution inside the patient body[2].

Tc-99m can be obtained from the decay product of the radioisotope molybdenum-99 (Mo-99) with a half-life of 66 hours. In general, Mo-99 is produced by a fission reaction of enriched uranium U-235 in a nuclear reactor. The global need of Mo-99 is supplied mostly by the six nuclear reactors in the world, i.e. the High Flux Reactor (HFR) in Netherland, the National Research Universal (NRU) in Canada, the Belgium Reactor 2 (BR2) in Belgium, the Reactor Safari 1 in South Africa Selatan, the Reactor Osiris in French, and the Reactor Maria in Poland. The average age of these reactors is 47 years so that the possibility of operation shutting down is great. It can affect the supply of the Mo-99 global need[3]. The crisis supply of Mo-99 has occurred from 2008 through 2009 and lead to a shortage of Mo-99. Its impact was many postponements of the diagnostic procedure that should be done to the patients throughout the world[4].

The shutdown of the Mo-99 producing nuclear reactors has led to an alternative method of producing Mo-99 without using a nuclear reactor. A subcritical assembly for Mo-99 production (SAMOP) experimental facility is being developed at the Center for Accelerator Science & Technology (CAST), National Nuclear Energy Agency (BATAN)[5]. The SAMOP experimental facility for Mo-99 production is a subcritical reactor system with a neutron multiplication factor (sub-criticality level) of 0.98 to 0.99 which needs an external neutron source for its operation[6][7]. In this developing phase, the SAMOP experimental facility will use an external neutron source from the radial beam-port of the Kartini TRIGA reactor[8][9]. It has been identified that the external neutron source from the radial beam-port of Kartini TRIGA reactor is suitable for this purpose[10][11].

In the future, the SAMOP production process will use an external neutron source from a compact neutron generator or a particle accelerator that generates neutrons in the core of subcritical assembly[10]. As predicted, that the best known uses of particle accelerators, including the compact neutron generator, probably are for the cancer therapy, medical isotope production, and food irradiation[12]. This paper presents the result of the study associated with the plan of using a compact neutron generator as an external neutron source of SAMOP.

METHODOLOGY

This study was carried out using the library research method. Data sources were obtained from journals, articles, and relevant information. The literature was accessed using Elsevier, Science Direct, and Google Scholar. The keywords included Mo-99, SAMOP, neutron sources, and compact neutron generator. This study clearly describes an overview of using a compact neutron generator as an external neutron source of SAMOP. The objective of this study is to explore the chance as well as the benefit of a compact neutron generator used as an external neutron source for SAMOP.

RESULT AND DISCUSSION

Description of SAMOP Experimental Facility

The SAMOP experimental facility uses the external neutron source from the radial beam-port of Kartini TRIGA reactor which has been identified as a thermal neutron in order of 10^8 n/cm^{2}s[13]. The SAMOP reactor core consists of an annular cylindrical tube containing uranyl nitrate [UO_2 $(NO_3)_2$] or UN as fuel and target, surrounded by a ring of $UO_2(NO_3)_2$ tubes. The TRIGA fuel elements are loaded in the ring together with $UO_2(NO_3)_2$ tubes to increase the neutron multiplication factor. The enrichment of all UN used in SAMOP is 19.75% U-235[14].

The SAMOP reactor core is a cylinder made of stainless steel with a diameter of 40.4 cm and a high of 43 cm. The reactor core is surrounded by a neutron reflector made of graphite with a thick of 20 cm. The neutron reflector has a hole for an external neutron source. The core and neutron reflector are located in the cooling tank with diameter and height of 120 cm and 400 cm respectively. The SAMOP core, neutron reflector, boral rod neutron absorber, and neutron collimator are described in Figure 1[5]. The SAMOP experimental facility is provided by an instrumentation and control system in such that if there is a criticality indication, the boral control rod neutron absorber will dropped automatically inserted to the SAMOP reactor core. The technical design specification of SAMOP experimental facility is shown in Table 1[14].

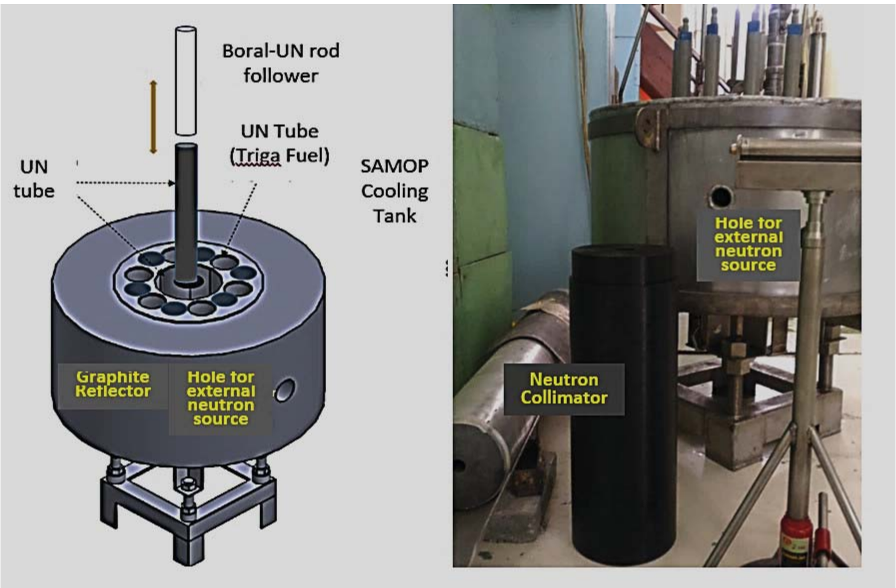


FIGURE 1. The core and neutron reflector configuration of SAMOP experimental facility[5].

TABLE 1. Design specification of SAMOP experimental facility[14]

Parameter	Material/Value/Dimension
Fuel solution	U-nitrate $UO_2(NO_3)_2$
Uranium enrichment	9.75 w/o U-235 enrichment
Uranium concentration	300 g U/L
Core volume	23 L
U-235 total	3.8 kg
Core height	35 cm
Core diameter	30.7 cm
Reflector thickness	20 cm, graphite
Radiation shielding	60 cm, heavy concrete
Neutron multiplication factor	0.98 – 0.99
Average neutron flux	10^{10} n cm^{-2} s^{-1}
Max. fuel temperature	55 °C
Thermal power	600 W
Mo-99 production	111 GBq/batch per week (3000 mCi/batch)

SAMOP requires a cooling system to remove heat in the core generated by the U-235 fission reaction. The cooling system of SAMOP is a cylindrical tank with a diameter of 120 cm and a high of 190 cm. The tank is filled with water and it serves as a primary cooling system. The heat removal takes place in the manner of natural convection by the water. Then by using a pump the water is circulated to the secondary cooling system that uses the water from bulk shielding of Kartini TRIGA reactor in a box of 112 cm x 230 cm x 374 cm dimensions. From the secondary cooling system, the water is returned to the primary cooling system, and part of the water is passed through a filter and a demineralizer (see Figure 2)[13].

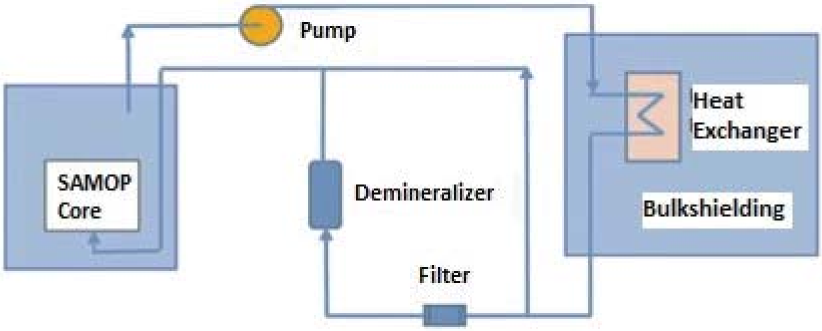


FIGURE 2. The schematic cooling system of SAMOP[13].

SAMOP uses an aqueous fuel i.e. uranyl nitrate solution with the U enrichment of 19.75% and concentration of 300 g U/L[14]. The uranyl nitrate is also used as a target material for Mo-99 production. The use of aqueous target material is very beneficial because it does not need a dissolution process so that the cycle of extraction of Mo-99 is being shorter[15]. The uranyl nitrate $UO_2(NO_3)_2$ is a yellow uranium salt and it is soluble in water. Therefore, the uranyl nitrate solution can be prepared by dissolving uranyl nitrate $UO_2(NO_3)_2$ into water solvent. The physical properties and compositions of uranyl nitrate are presented in Table 2[16].

TABLE 2. Physical properties and compositions of uranyl nitrate $UO_2(NO_3)_2$[16]

Physical Form	**Density**	**Melting point**	**Boiling point**	**Molecular weight**
Padat	2.087 g/cc	60 °C	118 °C	394.04
		Compositions		
Nuclide	Mass number	Number of atom	Molar mass (g/mol)	
U - total	238.028	1	238.028	
Nitrogen	14.007	2	28.014	
Oxigen	16	8	128	
Total molecular weight of uranyl nitrate			394.040	

SAMOP is developed based on the concept of a subcritical reactor system. So that the neutron multiplication factor (the ratio of the neutron production rate with the neutron absorption and neutron leakage rates) k_{eff} should be less than one (usually 0.98 to 0.99). To achieve the value of $k_{eff} \sim 0.99$), the content composition of $UO_2(NO_3)_2$ solution should be analyzed carefully. The analysis has been carried out using the WIMS-D/4 computer code. After the analysis result, it is shown that for achieving the k_{eff} value of ~ 0.99, the content composition of $UO_2(NO_3)_2$ solution are as follows: ^{235}U: 4.9715%, ^{238}U: 20.2009%, N: 5.3448%, O: 37.4138%, and H: 32.069%[17].

The formation of Mo-99 in SAMOP is based on the fission reactions of U-235 contained in uranyl nitrate fuel. The isotope Mo-99 produced by the fission reaction is about 6.1% of the total fission products, and it can be directly extracted from the uranyl nitrate solution. The extraction process is not expensive, and it has an embedded passive safety. Besides that, the treatment and purification of the fuel are simple.

Description of Compact Neutron Generator

Neutron generator is a charged particle accelerator used for producing neutrons. The charged particle that is accelerated in a neutron generator is deuteron ion. Neutron generator produces fast neutrons through D-D or D-T fusion reactions as expressed bellow[18].

$$D + D \rightarrow n + {}^{3}_{2}He \; (E_n = 2.45 \; MeV) \tag{1}$$

$$D + T \rightarrow n + {}^{4}_{2}He \; (E_n = 14.1 \; MeV) \tag{2}$$

The fusion reactions occur between deuteron ions and the target contains deuterium, tritium or mixed of them. The neutron energy produced from each fusion reaction is 2.45 MeV and 14.1 MeV respectively. The voltage used for accelerating deuteron ions is usually from 80 kV through 200 kV. The D-T reaction is used more often than the D-D reaction because the neutron yield of D-T reaction is higher than that of the D-D reaction (around 50 - 100 times). The neutrons produced from the D-T reaction are emitted nearly isotropic, while the neutrons from the D-D reaction are peaked in the forward direction[18].

The CAST BATAN has utilized neutron generator since the eighties for various research using neutrons, particularly for elemental analysis using fast neutron activation analysis method. For example, this method has been used for analyzing nitrogen, phosphor, and potassium in chemical and natural fertilizers, analyzing protein in rice, soybean, and corn as well as analyzing pollution level in rivers. The neutron generator has been also utilized for the study of neutron attenuation on various neutron shielding materials and for nuclear data measurement related to the reevaluation of scattered neutron and the cross section of nuclear activation particularly the (n,p), (n,α) and (n,2n) reactions[19].

There are many kinds of field applications that need a small size neutron source such as the detection of explosive materials and drugs at airport and harbors, the detection of corrosion, crack and impact damage using neutron radiography method, the analysis and mapping of mining minerals, the exploration of oil and uranium, the quality control of coal using PGNAA technique, the cancer therapy using BNCT method, etc.[20].

Components of Compact Neutron Generator

The requirements of a neutron generator used for field applications are that it should be easily transportable so that the dimension should be small; it has a high neutron yield and a long lifetime. The small dimension of a compact neutron generator can be realized by constructing the components of compact neutron generator in a compact tube so that it is frequently named as neutron tube or sealed tube. Neutron tubes have been developed and produced commercially by several companies such as Adelphi Technology (USA), EADS Sodern (France), Hotwell GmbH (Austria), Thermo Fischer Scientific (USA), VNIIA All Rusia Research Institut of Automatics (Rusian)[21].

The main components of a compact neutron generator consist of ion source as a generator of positive deuteron ions, accelerator system for accelerating deuteron ions, and target system made of metal hybrid material implanted with deuterium, tritium, or mixed deuterium and tritium. In addition, there are vacuum system, power supply, and gas replenisher that serve as supporting components. The schematic diagram of a compact neutron generator is as shown in Figure 3[22].

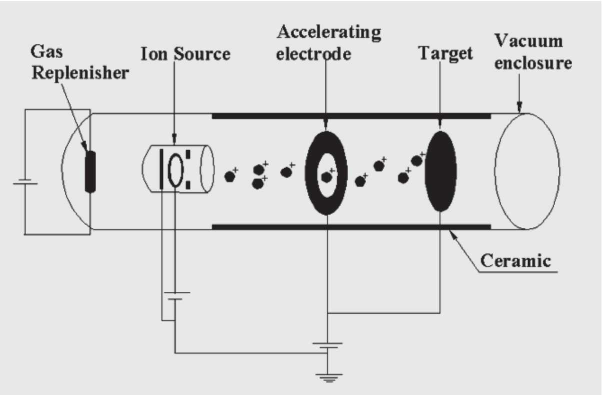


FIGURE 3. Schematic diagram of compact neutron generator[22].

Ion Source

An ion source in compact neutron generator is required to form and extract deuteron ions for bombarding the target. The type of ion sources usually used in compact neutron generators among others are the Penning ion source and the RF ion source[20]. They have each advantages and disadvantages. The Penning ion source has a simple structure, easy instrumentation, filament less operation, and long lifetime[22]. But its efficiency is low because the ions produced are mostly molecular ions. The efficiency can be enhanced by increasing the ions beam current and energy. By this technique, the power on the target surface rises significantly, but it can induce the sputtering of implanted deuterium/tritium ions on the target and leads to a decreasing of neutron yield. While the RF ion source has high power consumption, but its efficiency is high because it produces high-density plasma and the ions produced are mostly atomic ions. This leads to higher neutron yield with small beam spot size and without overheating on the target[23].

Accelerator System

To produce neutrons from D-D or D-T reactions, the deuteron ions should have a kinetic energy of around 100 to 200 keV. Therefore an accelerator system is required to accelerate deuteron ions so that its kinetic energy increases. To accelerate deuteron ion, an accelerating electrode is mounted in front of the extraction aperture of the ion source. The kinetic energy of accelerated deuteron ion is proportional to the voltage applied on the accelerating electrode. Beside to accelerate deuteron ions, the purpose of accelerator is also to shape a deuteron beam in such way that all deuteron ions in the beam are impinging on the target. In addition, the accelerating electrode should prevent the secondary electrons emitted from the target[24].

Target System

The target system plays an important role in the production of neutrons. The material commonly used as a target material in a compact neutron generator is copper (as backing material) coated with a thin film of titanium. Film thickness can range from 10 to 50 μm. Then, the titanium film is implanted with deuterium or tritium ions to form titanium hydrides. It is effectively trapping the ions into the titanium matrix. When the compact neutron generator is operated, the temperature of the target material will increase, and causing the diffusion of deuterium or tritium ions toward the surface of the titanium. The collision between the incoming deuteron ions and deuterium or tritium ions leads to the reaction that produces neutrons[25].

Neutron Yield of Compact Neutron Generator

The performance of a compact neutron generator can be seen from its neutron yield (neutron produced per second). The neutron production efficiency of target materials mainly depends on their capacity to retain deuterium and tritium, and on their stopping power. If the target materials can retain more deuterium and tritium, then there is more fusion reaction can occur between incoming deuteron ions and trapped deuterium or tritium. The lower the stopping power of the material, the less energy the ions will lose interacting with it. Titanium is a metal that has a low atomic number (Z = 22) so that its stopping power is relatively low compared to other higher-Z metals[24].

The calculation of neutron yield for Ti target has to consider the complexity of deuterium and tritium concentration buildup inside the Ti target. It is believed that in the steady-state condition, their concentrations reach a certain saturation value which is depend on the beam current density, target material, and the target temperature. It is assumed that the saturated concentration is constant over the particle range and that the particle range is shorter than the target thickness. The total number of neutron yield from a thick Ti target loaded with deuterium and bombarded with deuteron beam of current I and energy E composed of monoatomic species, can be computed using the equation[26]:

$$Y = \frac{I}{e} . \frac{ar.N_A}{A} \rho . \int_0^R \sigma[E(x)] dx \tag{3}$$

I is the deuteron beam current, *e* is the electronic charge, *ar* is the ratio of hydrogen atoms to metal atoms of the target (for titanium target, AR can be as high as 2.0), N_A is the Avogadro's number, *A* is the Ti mass number (47.9 g/mol), *ρ* is Ti density (4.51 g/cm^3), *σ[E(x)]* is the neutron production cross section for D-D reaction, and *R* is the range ion ions in the target. Since the cross section *σ* is the function of the energy range, so the equation (3) can be written in the form[26]:

$$Y = \frac{I}{e} . \frac{arN_A}{A} \rho . \int_0^{E_B} \frac{\sigma(E)}{dE/dx} dE \tag{4}$$

dE/dx is the stopping power of the target and E_B is the beam energy. After the Bragg's law of additivity, the stopping power of deuteron ions in the Ti target loaded with deuterium is the sum of the stopping power of Ti and D, can be written in the form[26]:

$$\frac{dE}{dx} = \frac{A}{A + 2\,ar} \left(\frac{dE}{dx} \right)_{Ti} + \frac{2\,ar}{A + 2\,ar} \left(\frac{dE}{dx} \right)_D \tag{5}$$

$(dE/dx)_{Ti}$ *and* $(dE/dx)_D$ are the stopping power of Ti and D respectively which can be calculated using SRIM (**S**topping and **R**ange of **I**ons in **M**atter) computer code. Then by using the stopper power of Ti and D, the neutron yield can be calculated using the equation (4). The results are shown in Figure 4 and Figure (5) respectively[26].

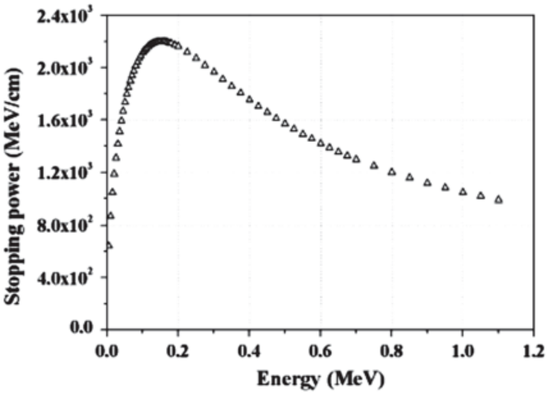


FIGURE 4. Stopping power of deuteron ions in Ti target loaded with deuterium[26].

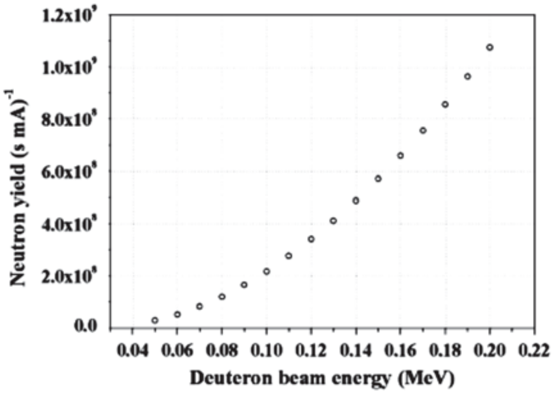


FIGURE 5. Neutron yield per mA vs. beam energy from deuteron beam impinging on Ti target[26].

The neutron yield per mA of deuteron ions beam is proportional to the energy of deuteron ions beam. It means that the neutron yield per mA is proportional to the accelerating voltage. The higher the accelerating voltage, the higher the neutron yield will be produced by accelerated deuteron ions. However, if the beam energy is higher than around 200 keV, the compact neutron generator has some problems related to high voltage isolation due to its compact size. Hence, for further increasing of neutron yield, it is more convenient to increase the beam current instead of the accelerating voltage. Even so, it is important to consider the power density deposited on the target caused by the high beam current density. The high power density deposited on the target will cause a cooling problem of the target.

The Chance of Compact Neutron Generator as Neutron Source for SAMOP

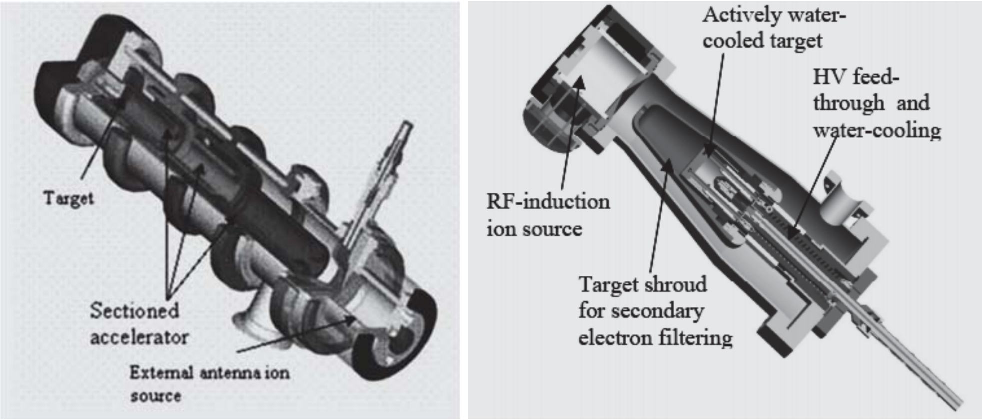
SAMOP needs an external neutron source to maintain the sustainability of the U-235 fission reactions in the uranyl nitrate solution for Mo-99 production. After the technical specification, SAMOP needs the neutron flux of 10^6 - 10^7 n/cm^2.s in 100 hours. The neutron need for SAMOP experimental facility is provided by the neutrons from the radial beam port of the Kartini TRIGA reactor[13]. However, for the future development SAMOP will use a compact neutron generator to meet the need of neutrons.

The compact neutron generator has a chance to substitute the external neutron source from the Kartini research reactor. Compact neutron generator produces fast neutrons with the energy of 2.45 MeV from the D-D reaction and 14.1 MeV from the D-T reaction. While the neutron required for SAMOP is thermal neutrons with the energy of around 0.5 eV. Therefore the neutrons from compact neutron generator should be slowed down using a moderator to match the neutrons requirement for SAMOP.

The compact neutron generator has several advantages over the neutron source from a nuclear reactor. Compact neutron generator does not have a safety criticality, it can be operated in steady and pulse mode, it does not have a radiation hazard when switched off, and it is easily transportable[22]. Therefore, from the safety point of view, compact neutron generator is safer than a nuclear reactor. Also, the operation cost of a compact neutron generator is cheaper than a nuclear reactor.

Now compact neutron generators are available in the global market. Compact neutron generators are produced by several companies (fabricants) and national laboratories. One of them is the Plasma and Ion Source Technology Group at the Lawrence Berkeley National Laboratory (LBNL) that has been developing compact neutron generators for over ten years. The compact neutron generators produced by LBNL use RF-induced discharge ion sources. This type of ion source can produce a high current density with atomic deuterium or tritium ion percentage of more than 90%. Hence, the compact neutron generator produced by LBNL can be operated at lower beam energy (~ 100 keV) and produce higher neutron yield than other commercial neutron generators. For the beam power of 100 kV, 10 mA the compact neutron generator can produce a neutron yield of ~ 2 x 10^9 n/s (from D-D reaction) and of ~ 3 x 10^{11} n/s (from D-T reaction). Also, the compact neutron generator can be operated either in CW or pulse mode by setting the RF power supply of the ion source[27].

The type of compact neutron generator that will be used for SAMOP has to adjust to the SAMOP core and reflector configuration. As shown in Figure 1, there is a hole across the core and reflector of SAMOP for external neutron source. Hence, the compact neutron generator that will be used as external neutron source should be installed in front of that hole. The LBNL developed axial compact neutron generators as shown in Figure 6[25].



(a) (b)

FIGURE 6. The axial compact neutron generator produced by Lawrence Berkeley National Laboratory (LBNL): (a) Sectioned accelerator axial compact neutron generator, (b) Fully high voltage shielded compact neutron generator[25].

There are two types of axial compact neutron generator. The first type is the sectioned accelerator axial compact neutron generator as shown in Figure 6a. In this type of compact neutron generator, the accelerator voltage is split into three segments evenly. The purposes of the segmented voltage are to shield the high voltage insulator from charging due to the travelling ion beam, and to minimize the electrostatic field stress. The second type is the fully high voltage shielded compact neutron generator as shown in Figure 6b. In this type of compact neutron generator, the high voltage element is shielded by a grounded vacuum/target chamber. The accelerator structure is a single gap with biased target shroud to filter the secondary electrons. The high voltage feed through integrates the HV cable and the water feeds for the target. The fully high voltage shielded compact neutron generator is designed for up to 10^9 n/s neutron yield using D-D reaction[25].

CONCLUSION

Based on the results of the study it is concluded that a compact neutron generator has a chance to substitute a nuclear reactor as external neutron source of SAMOP. A compact neutron generator can produce fast neutrons which neutron yield is sufficient to meet the need of neutron for SAMOP. It depends on the type of the compact neutron generator, one of which is the compact neutron generator produced by LBNL i.e. the axial compact neutron generator which can produce neutron yield up to 10^9 n/s. The benefit of using a compact neutron generator as an external neutron source of SAMOP is that the operational cost will reduced since the neutron production cost using a compact neutron generator is cheaper than a nuclear reactor. In addition, the operational procedure of a compact neutron generator is simpler than a nuclear reactor.

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