A study on radiation dose estimation of argon-41 and nitrogen-16 airborne released from the Kartini research reactor

M Salam and Elisabeth S
Center for Accelerator Science and Technology - National Nuclear Energy Agency (BATAN), Jl. Babarsari, Caturtunggal, Sleman, Yogyakarta, 55281, Indonesia
Email : mahrus.salam@batan.go.id

Abstract. A study on radiation doses estimation of Argon-41 and Nitrogen-16 airborne released from the Kartini research reactor is already carried out. Kartini reactor is a research reactor with the operating license of 100 KW. Estimation of radiation doses due to the radionuclide released in the airborne from the operating reactor is absolutely needed. This study is performed in order to estimate the internal dose contributions of the Argon-41 and N-16 received by radiation workers as well as the society in the reactor vicinity and to ensure that their dose contributions to the environment are still below the allowable limit value. Argon-41 and Nitrogen-16 are the activation products of gases containing Argon and Oxygen dissolved into primary coolant and released through the ventilation system. The estimated doses were calculated based on the fraction of Ar-41 and N-16 released to the environment through a reactor stack. The discharge rate was calculated by the normal condition for 400 hours of reactor operating in one year. The results showed that the estimated dose for the worker in the reactor building was 0.403 µSv/year, while the society in the reactor building vicinity was 4.45 x 10^-3 µSv/year. This value is very small compared to the dose constraint that appointed at Center for Accelerator Science and Technology which is 15 mSv/year for the radiation worker and 0.3 mSv/year for the society surrounding Yogyakarta Nuclear Facility.

1. Introduction
Nuclear Energy Acts. No.10, 1997, article 15 states that every nuclear utilization should not neglect the safety aspects in order to protect workers, society and the environment [1]. During the operation of nuclear reactors, in order to ensure that the value of received doses by radiation workers and the community is still below the Dose Limit Value (NBD) set by the Nuclear Energy Regulatory Agency (BAPETEN), an estimation of received dose have to be performed [2]. The increasing of received doses during the operation of the reactor could be contributed by external and internal radiation exposures. There would be a possible released of radioactive substances in the form of activation and fission products under normal and abnormal conditions when there is a leakage or damage in the fuel element cladding happen, while the radionuclide products from material contained in or dissolved in the reactor coolant can be released into the air in the reactor building, and then spread into the atmosphere through the reactor ventilation system.

According to the Government Regulation No. 02 of 2014 concerning on the Licensing for Installation and Utilization of Nuclear Materials [3], Kartini Reactor should conduct a study of received doses for both workers and the community on the reactor vicinity due to operating reactor. The received doses could be through direct radiation exposure by gamma transmitter radionuclides and through inhalation...
of released radionuclides air when the reactor operates. The Ar-41 and N-16 are the results of activated argon and oxygen that dissolved into primary coolant and radionuclides source terms that released to the atmosphere during the normal operating condition of the reactor.

The internal dose assessment of the Kartini reactor has been performed by using radioactivity measurement of air sampled in the reactor hall, and there was natural radionuclide only that has been detected [4]. This paper contains an evaluation method for estimating internal radiation doses contributed by inhalation of Ar-41 and N-16 as an activation product of the Argon and Oxygen gases on the Kartini reactor during normal operation. Under normal conditions Ar-41 and N-16 are formed and released continuously to the reactor building, mixed with air in the building and will be released into the environment through the ventilation system and reactor stack. When the radionuclide is released through the reactor stack, it will be dispersed into the environment as the local air dispersion and will be distributed and deposited so that potentially increasing the received dose to the community on the vicinity of the reactor. The potential increase of the received dose is caused by both beta and gamma radiation exposure when Ar-41 and N-16 inhaled by workers and people around the Center for Accelerator Science and Technology. The half-life of Ar-41 and N-16 are quite short, i.e. 109.43 minutes and 7.1 seconds, respectively [5], when compared to the average characteristics of atmospheric conditions as a function of time, it will decrease rapidly as a function of the reactor distance. Inside the reactor building Ar-41 is the result of activation of argon gas in channels or activation space around the reactor core and argon gas dissolved in reactor coolant, so that the Ar-41 gas before being released into the environment will contribute to increasing the worker radiation doses. While N-16 is an activation product of O-16 that composed of reactor coolant water.

On the calculation of Ar-41 and N-16 for internal dose contributed by the radioactive effluent that released through the stack taking into consideration wind speed, wind directions and other atmosphere parameters, such as temperature, humidity and solar radiation intensity. These parameters affect radionuclides dispersion and their pathway from the reactor stack. For gas fission products, likes Xenon, Krypton and Iodine, even though they were significantly contributing to the dose during abnormal conditions but during normal condition the dose is lower than the internal dose by Ar-41. In this study, radiation dose estimation from external and internal exposures of Ar-41 and N-16 as a result of operating the Kartini reactor would be performed.

2. Methodology

The flowchart for Ar-41 and N-16 dose calculation is shown in figure 1, Ar-41 dose was determined by multiplying radionuclide concentration of Ar-41 in the air and the effective dose rate per unit integrated air concentration of Ar-41 based on GSR part 3 [8], Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards. Ar-41 radioactivity concentration in the air was calculated based on the activation product of argon gas at the Kartini reactor. In this source term calculation, several parameters inputs are needed such as thermal neutron flux [6], argon flowrate and Kartini reactor operating time. N-16 radiation dose was calculated by multiplying concentration of N-16 in the air and the effective dose rate per unit integrated air concentration of N-16, as has been done on Ar-41 dose calculation.

2.1. The Ar-41 dose estimation for radiation worker and the public on the Kartini reactor vicinity

During the normal conditions, the Ar-41 is formed as the result of argon (Ar-40) activation that dissolved on the primary coolant in the reactor, as shown in Equation (1).

\[ \text{Ar}^{40} + n = \text{Ar}^{41} + \gamma \] (1)
Argon in the primary coolant of reactor vessel will be released to the air as a function of the changing of Ar-41 concentrations on the 3 critical points which are reactor core, primary coolant and air on the reactor building as given in the equations as follows [6]:

\[
V_1 \frac{dN_{41}^{41}}{dt} = V_1 \phi N_1^{40} \sigma^{40} - N_4^{41} (v_1 + V_1 \phi \sigma^{41} + \lambda^{41} V_1) + N_2^{41} V_1
\]  \hspace{1cm} (2)

\[
V_2 \frac{dN_{41}^{41}}{dt} = v_1 (N_1^{41} - N_2^{41}) - N_2^{41} \lambda^{41} V_2 - (f_{2-3} N_2^{41} V_2 - f_{3-2} N_3^{41} V_3)
\]  \hspace{1cm} (3)

\[
V_3 \frac{dN_{41}^{41}}{dt} = (f_{2-3} N_2^{41} V_2 - f_{3-2} N_3^{41} V_3) - N_3^{41} (\lambda^{41} V_3 + q)
\]  \hspace{1cm} (4)

where:

- index 1: reactor region (water/primary coolant inside the reactor core)
- index 2: primary coolant outside the reactor core
- index 3: reactor building region (reactor hall)
- Index 40: Argon-40
- Index 41: Argon-41
- \( V \): volume area, cm\(^3\)
- \( N \): atom density, atom/cm\(^3\)
- \( \lambda \): the decay constant, sec\(^{-1}\)
- \( \sigma \): neutron absorption cross section
- \( q \): volume flow rate from reactor building ventilation system, cm\(^3\)/sec.
$\nu_1$: volume flow rate of the coolant through the core, cm$^3$/sec.

$\varphi$: average thermal neutron flux in region no.1, n/cm$^2$.sec.

$\lambda_{i\rightarrow j}$: the fraction of Ar-41 atoms in region $i$ that escape to region $j$ per unit time, sec.

While the average volume flow rate of primary coolant through the reactor core was calculated using the equation follows:

$$\nu_1 = \frac{\Omega}{C_p \delta T \rho}$$

where:

$\Omega$: reactor power, $1.10^5$ W ($100$ kW)

$C_p$: specific heat of water, $4.19$ W.sec./g.°C

$\delta T$: Temperature rise across the core, $75$ °C (conservatively the highest value)

$\rho$: $0.958$ g/cm$^3$ exit water density

The calculated volume flow rate of coolant ($\nu_1$) is $3.32 \times 10^2$ cm$^3$/sec. The solution of Equation (2) to (4) is obtained by using several parameters as follows [7]:

$\nu_1$: $3.32 \times 10^2$ cm$^3$/sec.

$V_1$: $1.45 \times 10^9$ cm$^3$

$\varphi$: $1.2 \times 10^{12}$ n/cm$^2$.sec. (average of neutron flux)

$\sigma^{40}$: $0.047 \times 10^{-24}$ cm$^2$

$\lambda^{41}$: $1.06 \times 10^{-4}$ sec$^{-1}$

Therefore, $\nu_1 + V_1 \varphi \sigma^{41} + \lambda^{41} V_1 \approx \nu_1$ and Equation (2) could be simplified as follows:

$$V_1 \frac{dN_1^{41}}{dt} = V_1 \varphi N_1^{40} \sigma^{40} - (N_1^{41} - N_2^{41}) \nu_1$$

On equilibrium condition, Equation (2), (3) and (4) could be stated as follows:

$$V_1 \varphi N_1^{40} \sigma^{40} = (N_1^{41} - N_2^{41}) \nu_1$$

$$N_2^{41} (\lambda^{41} V_2 + f_{2\rightarrow 3} V_2) = (N_1^{41} - N_2^{41}) \nu_1 + f_{3\rightarrow 2} N_3^{41} V_3$$

$$N_3^{41} (\lambda^{41} V_3 + q + f_{3\rightarrow 2} V_3) = f_{2\rightarrow 3} N_2^{41} V_2$$

By combined of Equations (7) and (8) then obtained:

$$N_2^{41} = \frac{V_1 \varphi N_1^{40} \sigma^{40}}{\lambda^{41} V_2 + f_{2\rightarrow 3} V_2} + \frac{f_{3\rightarrow 2} N_3^{41} V_3}{\lambda^{41} V_2 + f_{2\rightarrow 3} V_2}$$

By substituting the value of $N_2^{41}$ from the Equation (10) to the Equation (9), it will be obtained equation as follows:

$$N_3^{41} \frac{\lambda^{41} V_3 + q + f_{3\rightarrow 2} V_3}{f_{2\rightarrow 3} V_2} - \frac{f_{3\rightarrow 2} V_3}{\lambda^{41} V_2 + f_{2\rightarrow 3} V_2} = \frac{V_1 \varphi N_1^{40} \sigma^{40}}{\lambda^{41} V_2 + f_{2\rightarrow 3} V_2}$$

Some of the constant values used in Equation (11) is as the following:

$V_2$: $1.8 \times 10^7$ cm$^3$ (volume of primary coolant region/H$_2$O)

$V_3$: $4.45 \times 10^9$ cm$^3$ (air volume in reactor building)

$q$: $6.6 \times 10^6$ cm$^3$/sec. (volume flow rate on the ventilation system)

$\sigma^{40}$: $0.047 \times 10^{-24}$ cm$^2$
By entering the several constants above, only the magnitude \( N_1^{40}, f_{2\rightarrow 3}, f_{3\rightarrow 2} \) and \( N_3^{41} \) will be reviewed. The concentration of saturated Argon gas in water, according to Henry's law can be expressed by the equation as follows:

\[
X = \frac{P}{k}
\]  

where :

\( X \) : Argon molecule fraction in the water
\( P \) : Argon partial pressure above water, mmHg
\( k \) : Argon constant (2.9 x 10^7 mmHg per molecule fraction in solution at 30 °C temperature)

Argon content in the air is 0.94 % of total volume, therefore partial pressure of Ar above coolant water is 6.9 mm Hg, by this value, Ar molecule fraction in the water is 2.4 x 10^-7, while the value of \( N_1^{40} \) is 8.06 x 10^{15} Ar-atomic/cm³ of water. Parameter of \( f_{2\rightarrow 3} \) (as part of Ar atomic in the coolant that released to the air per second) could be estimated by testing the mobility of ions in aqueous solution. Those ions mostly have velocity in the order of \( 3 - 8 \times 10^{-4} \) cm/s below potential gradient which is 1 volt/cm. Therefore, Argon atoms that have velocity smaller than \( 3 \times 10^{-4} \) cm/s that were not affected by the potential gradient. Argon atoms in the area of \( 3 \times 10^{-4} \) cm from the surface of primary coolant only that will be able to escape from the water every second even though the volume of this source is very large. Thus the highest limit for the total fraction of Argon atoms that would be released from water is 4.9 x 10^-7/s.

In equilibrium conditions and by assuming there are no significant differences between the velocity of Argon-40 and Ar-41 atoms, the number of Argon atoms released into the air is equal to the number of Argon atoms that enter from air to water. So that it can be stated:

\[
f_{2\rightarrow 3}N_2^{A}V_2 = f_{3\rightarrow 2}N_3^{A}V_3
\]  

By using \( N_2^{A} \) is 2.1 x 10^{17} Ar atom/cm³ of air, while \( N_3^{A} \) is 8.06 x 10^{15} Ar atom/cm³ of air, therefore the value of \( f_{3\rightarrow 2} \) could be calculated as follows:

\[
f_{3\rightarrow 2} = \frac{f_{2\rightarrow 3}N_2^{A}V_2}{N_3^{A}V_3}
\]  

So \( f_{3\rightarrow 2} \) is \( 7.61 \times 10^{11} \) per second. It is shown that \( \lambda^{41} \gg f_{2\rightarrow 3} \gg f_{3\rightarrow 2} \), therefore the equation (11) could be stated as follows:

\[
N_3^{41} = \frac{V_3f_{2\rightarrow 3}\varphi N_1^{40}a^{40}}{\lambda^{41}(A^{41}V_3 + q)}
\]  

\( N_3^{41} \) value could be found is 4.31 x 10^2 atom/cm³, while the radioactivity concentration of Ar-41 in the air of reactor building could be stated as follow:

\[
A^{41} = \frac{\lambda^{41}N_3^{41}}{\lambda^{41}N_3^{41}}
\]  

Ar-41 concentration is 4.57 x 10^6 Bq/cm³. The release rate of Ar-41 from the reactor building was obtained by multiplying the radioactivity concentration Ar-41 with \( q \) (volume flowrate), where the Ar-41 release rate is 30.2 Bq/s.

When it is known that the effective dose rate per integrated air concentration unit for Ar-41 is 5.3.10^-9 Sv.day^-1/Bq.m³, based on GSR part 3 [8], Radiation Protection And Safety Of Radiation Sources: International Basic Safety Standards, by assuming that the Kartini reactor operates for 400 hours in 1 year, so the amount of internal radiation dose received by radiation workers from Argon gas inhalation
is 0.403 µSv/year. The Ar-41 radiation dose received by the people in the Kartini reactor vicinity was calculated based on the concentration of Argon released through the reactor stack. So that the concentration of argon and nitrogen radionuclides deposited on the ground must be lower than the effluent concentration. To determine radionuclide concentration that deposited on the ground could be used the following equation[9]:

\[ X(0,0,0) = \frac{1}{0.5x4x\bar{u}} Q \]  (17)

where:

\[ X(0,0,0) \]: concentration at location x,y,z (Bq/cm³)
\[ A \]: area of the reactor building, when the ventilating system is not operating (cm²)
\[ \bar{u} \]: wind speed diluting the plume (cm/s)
\[ Q \]: Ar-41 released rate (Bq/s)

By assuming that the Kartini reactor operates for 400 hours in 1 year, Ar-41 released rate is 30.2 Bq/s. If the average wind speed around the Kartini reactor \( \bar{u} \) is 3 x 10^2 cm/s and reactor building area is 4 x 10^6 cm², the amount of radioactivity that reaches the ground is 5,033 x 10^-2 Bq/m³. The effective dose rate per unit of integrated air concentration for Ar-41 is 5.3 x 10^-9 Sv.day^-1/Bq.m^-3 based on GSR part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards [8], the amount of internal radiation dose received by the society in the reactor building vicinity from Argon gas inhalation is 4.45 x 10^-3 µSv/year.

This calculation uses several data inputs such as neutron flux[12,13], fuel material composition and also operation time of the Kartini reactor. The output from this calculation is the radioactivity of several fission and activation products. The flowchart of radiation source-term analysis is described in Figure 1.

2.2. Determination of N-16 Dose for Radiation Worker and Public in Kartini Reactor Vicinity

Oxygen naturally consists of stable isotopes of O-16, O-17 and O-18, with O-16 is the most commonly found in nature (99.72% natural abundance). N-16 is formed when the primary coolant passes through the reactor core so it is activated by neutrons. N-16 is the result of an activation reaction of oxygen (O-16) which dissolves into primary coolant as shown in Equation (18).

\[ O^{16} + n = N^{16} + p + \gamma \]  (18)

The cross-section threshold for the Oxygen-16 reaction is at the energy of 9.4 MeV. However, the minimum neutron energy is 10 MeV. It is only about 0.1% of all fission neutrons have energy exceeding 10 MeV, so this threshold value causes N-16 production is limited. The effective cross-sectional measurement for O-16 (n, p) N-16 reactions is 2.13 x 10^-29 cm^2 or 0.213.10^-6 barn. The concentration of N-16 atoms per volume of water released from the reactor core can be determined by the following equation [10]:

\[ N^N = \frac{\varphi N^o \sigma^o (1 - e^{-N^\lambda t})}{A^N} \]  (19)

where:

\[ N^N \]: N-16 atoms per cm³ of water
\[ N^o \]: Oxygen atoms per cm³ of water (3.34 x 10^22 atom/cm³)
\[ \varphi \]: 1.2 x 10^{12} n/cm².second(average of neutron flux)
\[ \sigma^o \]: Oxygen cross absorption(2.13 x 10^{-29} cm²)
\( \lambda \): N-16 decay constant (9.7 \times 10^{-2} \text{ second}^{-1})

\( t \): Average exposure time in the reactor

Average exposure time in Kartini reactor could be calculated as follows:

\[
t = \frac{V_1}{v_1}
\]

(20)

The average exposure time was obtained at 43.6 seconds, and the number of N-16 atoms per cm of water was 7.01 \times 10^{10} \text{ atoms/cm}. With a flowrate of 3.32.10^{-2} \text{ cm/second}, the N-16 velocity that escapes from the reactor core is 2.33 \times 10^{15} \text{ atoms/second}. Only a small portion of N-16 atoms near the coolant water surface that transferred into the air of reactor building. N-16 atoms are formed as a recoil with various degrees of ionization. For high purity water (2 Molar), all N-16 practically combines with Oxygen and Hydrogen atoms of water. Most of them are in the form of anions which tend to remain in the water. In this calculation it is assumed that at least half of all the ions formed are anions.

N-16 atoms will not stay long enough because of their short half-life (only 7.4 seconds) to reach a homogeneous concentration in the coolant. It can be assumed that N-16 atoms will be spread in 30 cm of water in the top surface of the reactor coolant by operating a diffuser. Although the highest contribution of the dose value is radiation exposure directly from the core, the most concern review in this case is the number of N-16 atoms that released into the air per second. The maximum part of the N-16 atom that released to air per second \( f_{2 \rightarrow 3} \) can be estimated in the same way as the estimation for an Argon atom of \( 5 \times 10^{-6} \text{ per second.} \) When the speed of N-16 accumulation in the reactor building (hall) is given by the equation as follows:

\[
\frac{d(V_3 N^N)}{dt} = S - (\lambda^N + \frac{q}{V_3}) N^N V_3
\]

(21)

where:

\( S \): N-16 atoms that released from primary coolant to the air in reactor building per second

\( V_3 \): Reactor building volume (4.45 \times 10^9 \text{ m}^3)

\( q \): the volume flow rate of the ventilating system of reactor building (6.6 \times 10^6 \text{ cm}^3/\text{second})

For saturated conditions, equation (21) can be stated as follows:

\[
N^N V_3 = \frac{S}{(\lambda^N + \frac{q}{V_3})}
\]

(22)

\[
A^N = \lambda^N N^N
\]

(23)

Therefore, N-16 radioactivity concentration was obtained at \( 5.57 \times 10^{-4} \text{ Bq/m}^3 \). The release rate of N-16 from the reactor building was obtained by multiplying the radioactivity concentration N-16 with \( q \) (volume flowrate), where the N-16 release rate is \( 3.7 \times 10^{-3} \text{ Bq/sec.} \)

3. Result and Discussion

In this paper the Ar-41 and N-16 dose calculations in the Kartini reactor were carried out. By assuming that the Kartini reactor operates for 400 hours in 1 year, so the amount of internal radiation dose received by radiation workers from Argon gas inhalation is 0.403 \( \mu \text{Sv/year.} \) When the reactor building is ventilated, the argon and nitrogen concentrations in the air would be considered to be released through the ventilation system and reactor stack with a fixed concentration. From the calculation the amount of internal radiation dose received by the society in the reactor building vicinity contributed by Argon gas inhalation is 4.45 \times 10^{-3} \text{ \( \mu \text{Sv/year.} \)
By using the same method, the N-16 radioactivity was calculated. The result shows that the N-16 radioactivity is very small compared to the Ar-41 radioactivity, so that N-16 radioactivity would not be contributed significantly to the increasing dose, therefore it can be ignored. The flowrate of N-16 radioactivity released from the reactor building is obtained by multiplying the concentration of N-16 radioactivity with the debit of the ventilation system, which is equal to 3.7 x 10^3 Bq/sec. By assuming that the Kartini reactor operates for 400 hours in 1 year, the average released rate of N-16 is 8.39 x 10^3 Bq/s. If the average wind speed around the Kartini reactor (\(u\)) is 3 x 10^2 cm/s and the reactor building area is 4 x 10^6 cm^2, the amount of radioactivity that reaches the ground is 1.4 x 10^7 Bq/m^3. These results indicate that the dose value of the N-16 radiation received by workers and also the community in the Kartini reactor vicinity are very small.

The result of calculation shows that the radiation dose received by radiation workers and the public from N-16 and Ar-41 contribution it is still far below the dose constraint value that appointed in Center of Accelerator Science and Technology (CAST), where the dose constraint that appointed in the CAST is 15 mSv/year [11] for radiation workers and 0.3 mSv/year for the general public respectively [2]. The internal radiation dose from Ar-41 contributions received by radiation workers by assuming the reactor operates for 400 hours/year is 0.403 μSv/year, while for the community only 4.45 x 10^3 μSv/year.

From these results it can be concluded that the contribution of the internal dose of Ar-41 and N-16 from the operation of the Kartini reactor received by both radiation workers and the general public is very small. However, to validate the results of this calculation, in the next study it is necessary to estimate the internal radiation doses from Ar-41 and N-16 by using direct measurement.

4. Conclusions
An evaluation of the Ar-41 and N-16 internal dose calculations received by the radiation workers and the public contributed by the operation of the Kartini reactor has been carried out. It was assumed that the Kartini reactor operates for 400 hours a year, the results show that the received dose for the radiation workers is 0.403 μSv/year and for the public in the reactor vicinity is 4.45 x 10^3 μSv/year. The internal dose calculation of N-16 obtained based on the N-16 radioactivity rate in the reactor hall is very small, which is 3.7 x 10^3 Bq/sec, while the amount of radioactivity that reaches the ground is 1.4 x 10^7 Bq/m^3. Both Ar-41 and N-16 radiation dose contributions received by radiation workers and the public are very small compared to the dose constraint that appointed in CAST which is 15 mSv/year for radiation workers and 0.3 mSv/year for the public in the Kartini reactor vicinity.

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