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Neutron Dose Assessment Using ²⁴Na in Blood for Korean Radiation Workers

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Rapid and easy dose assessment must be provided following criticality accident to permit medical staff to determine the appropriate initial treatment for each victim. The KBADA was developed for more rapid and easy neutron dose assessment using ²⁴Na activity in blood following well-established methods at ORNL. In this study, the results of neutron dose evaluations by the KBADA are shown to be capable of applying to neutron accident dose. Experiments were conducted in the neutron fields of Yonggwang unit 1, 2 and 4, Wolsung unit 3 and the MC-50 proton accelerator. The PMMA slab phantom was used for a human phantom and neuron spectra were also measured. The KBADA program was used to evaluate the neutron dose of experiments using the activity of ²⁴Na and the neutron spectra. The results showed that the neutron dose assessment using ²⁴Na in blood could apply to an over-exposure person at a criticality accident.

KEYWORDS: neutron dose assessment program, KBADA, ²⁴Na, absorbed dose

I. Introduction¹

Korea Hydro & Nuclear Power Co., Ltd. (KHNP) is the only company generates electricity by nuclear power and operates 20 nuclear power plants in Korea. The internal and external doses of all radiation workers in KHNP have been systematically managed by Radiation Safety Management System (RSMS) of KHNP with elaborate safety procedures and guidelines. Generally, during the normal operation, the doses of radiation workers are evaluated by thermo luminescent dosimeters (TLD) for external irradiation and whole body counter (WBC) for the intake of radionuclides. In accidental case including TLD missing, other dose assessment methods like biological dosimetry, electron spin resonance (ESR) and radiation transport codes are added for the dose reconstruction. Whatever accidents may happen, rapid and accurate dose assessment is also very important to give appropriate medical treatments for the victims in early stage.

In case of neutron over-exposure such as criticality accident, measurement of the activated sodium in the blood is widely used with many other methods for the neutron dosimetry of the victims without personal dosimeters. Stable element, ²³Na distributed in a whole human body is activated to ²⁴Na through ²³Na(n,γ)²⁴Na reaction by neutron irradiation. So, if the neutron spectrum is known or assumed, it is possible to assess the accident neutron dose by the analysis of ²⁴Na in the blood. This method was first implemented by Hempelmann in 1952¹⁾ and developed through many experiments conducted on phantoms using different critical assemblies. Although the experiments for accident neutron dosimetry were limited to a certain degree by a lack of information on accident neutron spectra, the principles and procedures of bioassay for accident neutron dosimetry were established at Oak Ridge National Laboratory (ORNL) of the United States²⁾ and applied to the victims of JCO accident by National Institute of Radiological Sciences (NIRS) of the Japan³⁾.

The KBADA (KHNP Body Activation Dose Assessment) program developed by Radiation Health Research Institute (RHRI) of KHNP adopted this activated sodium analysis method and improved the dose calculation process by computerization for the convenience of users. In this program, the accident neutron dose is calculated theoretically using the selected neutron spectrum of neutron spectrum database on the program with the neutron body capture probability or estimated by the measured bioassay data from the neutron irradiated person. However, in most cases, neither the measured data nor the theoretical values can give the reliable results alone. So, the comparison of evaluated doses and the analysis of the factors result in the differences should be conducted for the verification.

In this study, for the application of KBADA to neutron over-exposure accident, measurements of neutron spectra and neutron activation experiments at the same places in practical neutron fields were carried out. Then, the results were compared with those of theoretically calculated with the measured neutron spectrum only. In case of experiments, it is almost impossible to make similar accidental conditions like criticality accidents due to the high neutron intensity. So, extended irradiation times for nuclear power plants in normal conditions were adopted instead. High neutron intensity but different from general critical neutron spectra were also considered by using a proton accelerator.

II. Experiments

1. Neutron dose assessment program

The KBADA was coded by Visual Basic[®] program to assess the neutron dose using ²⁴Na in blood through the rapid selection of the spectrum harmonized with the exposure situation based on the neutron spectrum database. There are three logics in the KBADA program, but this study needs only two logics (Logic 1 and 2) shown in **Fig 1**. The Logic 3 is used for the neutron dose assessment using ³²P of hair.

Logic 1 is a method used for general neutron dose assessment with the neutron spectrum ($\Phi_E(E)$), measured with a Bonner Multi-sphere Spectrometer(BMS) at a nuclear

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Fig. 1 Flowchart of the neutron dose assessment program by Logic 1 and Logic 2 in the KBADA.

power plant or a neutron-generating facility by using equation (1). In this paper, Logic 1 provides the reference dose to be compared against the neutron dose assessment outcome (Logic 2) by activation of 24 Na in blood.

The normalized spectrum, b(E) and the total fluence, Φ are essential information to assess a neutron dose, and every Logic is programmed to calculate or input them. The Logic 1 is used in case there are two factors $(b(E), \Phi)$. Also, it is used for the demonstration of neutron dose assessment using ²⁴Na in blood with the following equations:

$$D = \sum R(E)\Phi_E(E) \tag{1}$$

where D = neutron dose (mSv or mGy); R(E) = dose conversion coefficients (mSv cm² or mGy cm²); $\Phi_E(E)$ = fluence per unit energy (# cm⁻²),

$$\Phi_F(E) = \Phi \times b(E) \tag{2}$$

where, Φ = total fluence (# cm⁻²); b(E) = normalized spectrum.

In the Logic 2, it is assumed that the b(E) is known but Φ is unknown. The Logic 2 assesses the neutron dose using radioactivity of ²⁴Na in blood and calculates the neutron dose with following equation:

$$D = \frac{23F_a \sum_t V}{N_{avo} R_t \lambda \sigma_{Na} S} \frac{A_{HPGe}}{m_{Na} A_A} \frac{\sum R(E)b(E)}{\sum \xi(E)b(E)}$$
(3)

Experiments were performed at the Weapons Neutron Research (WNR) facility⁴⁾ in Los Alamos Neutron Science where 23 = atomic weight of ²³Na; F_a = Decay correction factor during the irradiation period; Σ_t = total absorption cross section for thermal neutron (cm⁻¹); V = total volume of the human body (cm³); N_{avo} = Avogadro's number (# mole⁻¹); R_t = metabolic correction factor; $\lambda = {}^{24}$ Na decay constant (s⁻¹); $\sigma_{Na} = {}^{23}$ Na(n, γ)²⁴Na cross section (cm²); S = the projected area in human body (cm²); A_{HPGe} = radioactive concentration of 24 Na measured by HPGe (Bq ml⁻¹); $m_{Na,AA} =$ 23 Na concentration in a blood sample (g ml⁻¹); R(E) = dose conversion coefficients (mGy cm² or mSv cm²); $\zeta(E)$ = neutron capture probability.

Using the shape and the size of a BOMAB⁴ for neutron irradiation in AP (anterior to posterior) direction, the projected area (S) in the body to the incident neutron beam is $5.69 \times 10^3 \text{ cm}^2$ and the total volume (V) of the body is $6.828 \times 10^4 \text{ cm}^3$. If the blood sample is collected several days after

the accident, it is necessary to correct for the excretion of ²⁴Na from the body. The biological decay of ²⁴Na activity in the body can be represented by the sum of three exponential terms. The metabolic correction factor (R_i) of ²⁴Na retained in the body is given by following equation²)

 $R_t = 0.487e^{-0.0815t} + 0.510e^{-0.0513t} + 0.0027e^{-0.0015t}$ (4)

Where, t = the elapsed time from incident to collection of blood sample (days). Of the total number of neutrons striking the phantom, a fraction $\xi(E)$ (capture probability) will be captured in the phantom. The moderation of the body to the incident neutrons is so strong that the interaction of ²³Na(n, γ)²⁴Na is mainly determined by the body capture probability. In this study, the neutron capture probabilities in a BOMAB were used.

There are five dose conversion coefficients R(E) in the KBADA; fluence to absorbed dose of tissues or organs and fluence to effective dose from ICRP 74⁵), ambient dose equivalent H^{*}(10) and personal dose equivalent H_P(10, α) of IAEA TRS 403⁶), maximum absorbed dose of IAEA TRS 211⁷). Mean value of tissues or organs absorbed dose of AP direction of ICRP 74 is used in this study. The incident direction of radiation to the body also affects the dose to the whole body⁸). However, only the AP direction is being considered by KBADA at the moment, and all directions being provided by ICRP 74 shall be considered in the future.

Uncertainty factors which affect the neutron dose assessment using the neutron activation of blood are A_{HPGe} , $m_{Na,AA}$ and assume to comply with the normal distribution. After producing each random number in the " $N(\mu, \sigma^2)$ " normal distribution of A_{HPGe} and $m_{Na,AA}$ using Monte Carlo

simulation, neutron dose is calculated by Logic 2. *N* means normal distribution, μ and σ^2 are mean value and variance of A_{HPGe} and $m_{Na,AA}$. It is repeatedly performed 10,000 times and calculated neutron doses are arranged in ascending order. Neutron dose is described by representative dose and dose range with 95% confidence level. Neutron dose range is distributed from the 250th (minimum) to the 9750th (maximum) values and representative dose is the 5000th (median) value.



Fig. 2 A schematic showing irradiation position of blood and NaCl solution samples.

It is difficult to estimate the spectrum at exposure circumstances because neutron spectra in nuclear power plants are differently distributed according to exposure locations and fuel cycles. Collecting of many neutron spectra into database in many locations of nuclear power plant is good way to improve the dose assessment accuracy but it takes a long time. Therefore the method for the rapid estimation of neutron spectra is needed to establish based on the former measured neutron spectrum.

Around a hundred measured neutron spectra in Korean nuclear power plants are collected into the database of KBADA. In consideration of additional convenience for continuously measuring neutron spectra in Korean nuclear power plants at present, the database was developed by $Excel^{\textcircled{B}}$. The data types of collected neutron spectra were standardized to normalized spectrum (*b*(*E*)/lethargy) of IAEA TRS 403. Also, the energy bins of capture probability, cross section and dose conversion coefficients were adjusted to those of IAEA TRS 40

2. Phantom and measurement system

Because the radioactivity of neutron induced ²⁴Na in blood is proportional to total number of absorbed neutrons and the concentration of ²³Na, it is important to measure the accurate concentration. In this study, the ²³Na concentrations of blood samples were measured by Atomic absorption spectrometer (Thermo Co.).

The water slab phantom was fabricated by Korea Research Institute of Standards and Science (KRISS). Its material is polymethyl methacrylate (PMMA) and the size is $30 \text{cm} \times 30 \text{cm} \times 15 \text{cm}$. The thickness of front side is 3 mm and other sides are 10mm and the phantom was filled with pure water. The mass of this PMMA phantom is close to that of the ICRU sphere, and its backscatter characteristics are acceptably close to those of the human trunk for both photon and neutron irradiations⁹.

A 20ml vial of blood sample and NaCl solution samples with various concentrations were placed on the front surface of the phantom for each neutron irradiation (Fig 2). These irradiation methods were performed in accordance with the ORNL procedure²⁾. The activation experiment method in Fig 2 cannot describe the actual level of activation for ²⁴Na within the human body compared to Bottle Manikin absorption (BOMAB). A simulation of the activation level of ²³Na for the BOMAB phantom and the method described in Fig 2 by using MCNP revealed that the activation by the method in Fig 2 was higher than that by BOMAB at low energy. Also, it can be confirmed from other studies that the dose varies depending on the samples' depth in the trunk¹⁰. However, in this study, certain limitations exist when performing an experiment using a BOMAB phantom due to the neutron activation experiment at high-gamma radiation fields of the nuclear power plant and to poor experimental conditions such as narrow workspace, low neutron activation rates, etc., and thus the activation experiment was performed in accordance with the ORNL method as an alternative.

Neutron activated ²⁴Na decays by emitting 1.369 MeV gamma rays with 14.8 h radioactive half life. So, the measurement of ²⁴Na activities from neutron irradiated samples were carried out by high purity Germanium detectors (HPGe, Canberra Co.).

3. Experiments

Irradiation experiments were performed with the samples attached PMMA water slab phantom in the CV(Containment

Vessel) of Wolsung nuclear power plant (NPP) unit 3, Yonggwang NPP unit 1, 2, 4 and a treatment room of MC-50 proton accelerator in Korea institute of radiological & medical science (KIRAMS). These experiments were TLD performed for the purpose of preparing against any abnormal dosimetry such as losing dosimeter or TLD reading failure in case of accident or normal operation of nuclear facilities. NPPs are different types; Wolsung unit 3 is CANDU (Canada Deuterium Uranium) type, Yonggwang unit 1 & 2 are Westinghouse type and Yonggwang unit 3 is CE (Combustion Engineering) type. The experiment places are selected in the high possibility areas of radiation work and relatively high neutron flux areas in the CV during normal operation. Every neutron activation experiment except MC-50 is performed after measuring the neutron spectrum using a Bonner Multi-sphere Spectrometer (BMS) with 6 polyethylene spheres and a ⁶Li(Eu) scintillation counter by Nuclear engineering & technology institute (NETEC) of KHNP.

The neutron spectrum from thick target gantry of MC-50 was simulated by MCNP-X 2.5¹¹). In the thick target gantry, accelerated protons bombard 10.5 mm Be target and yield neutrons. To obtain the information of neutron spectrum produced by 35 MeV incident protons on the Be target from the gantry, the number of protons correspond to 35 MeV are transported through the Be target. Then, the neutron beams produced by (p, n) reactions were adjusted by additional modeling of some filters and collimators inside the gantry to make same irradiation geometry of the experiment. In the experiment, the blood sample (²³Na 2.17 mg ml⁻¹) and the NaCl solution (²³Na 1.91 mg ml⁻¹) were irradiated by the neutron beam produced in same condition modeled by simulation for 1 hour at 100 cm from the gantry

III. Result and discussion

Table 1 shows the ²³Na concentrations of the blood and NaCl solution samples, measured activities and the results of the neutron dose assessment. The neutron dose ranges resulted from the uncertainty factors of A_{HPGe} and $m_{Na,AA}$. The absorbed doses are mean value of tissue or organs absorbed doses calculated with dose conversion coefficients from ICRP 74.

Table 2 shows the results of dose calculation using the neutron spectrum alone. The absorbed doses, the mean value of tissue or organs absorbed doses, could be directly calculated since the flux and total fluence were known. From the **Fig 3** and **4**, these directly calculated absorbed doses are well agreement with those from the measured data. In a critical accident, the neutron spectrum shape may be similar to those in NPPs but has different intensity.

In a critical accident, the neutron spectrum shape may be similar to those in NPPs but has different intensity. So, it can be considered that the extended irradiation time and the correction of production and decay rate of ²⁴Na can simulate critical accident cases as the experiments in this study. Also, other neutron over-exposure cases from the different neutron source can be applied from the result of MC-50 proton accelerator.

NPP	Sample ID	²³ Na concentration (mg ml ⁻¹)	²⁴ Na specific Activity (Bq ml ⁻¹)	Time ¹⁾ (min)	Dose range (mGy)	Absorbed Dose (mGy)
Wolsung unit 3	Blood	2.13 ± 0.19	0.03 ± 0.01	193	$0.22 \sim 0.90$	0.52
	NaCl	7.02 ± 0.61	0.04 ± 0.01	398	$0.11 \sim 0.41$	0.25
Yonggwang unit 4	Blood	2.24 ± 0.01	< MDA	147	-	-
	NaCl	5.78 ± 0.01	0.11 ± 0.05	340	0.19 ~ 1.71	0.89
Yonggwang unit 1	Blood	1.91 ± 0.10	3.89 ± 0.02	30	45.5 ~ 65.6	54.8
	NaCl	1.91 ± 0.10	3.85 ± 0.02	197	45.5 ~ 65.5	54.7
Yonggwang unit 2	Blood	1.91 ± 0.10	2.51 ± 0.13	354	$28.0 \sim 43.5$	35.1
	NaCl	1.91 ± 0.10	2.69 ± 0.13	379	$30.1 \sim 46.5$	37.6
MC 50	Blood	2.17 ± 0.11	0.29 ± 0.03	1462	$54.8 \sim 98.1$	74.7
	NaCl	1.91 ± 0.10	0.22 ± 0.01	273	$48.8\sim74.9$	60.8

Table 1. The results of neutron dose assessment using blood activation

¹⁾ The time from the end of exposure to the beginning of detecting with HPGe

Table. 2 The results of total fluence

NPP	Flux (#cm ⁻² s ⁻¹)	Total fluence (#cm ⁻²)	Absorbed Dose (mGy)
Wolsung unit 3	$3.40 \times 10^{2} \pm 1.2 \times 10^{1}$	$5.75 \times 10^{7} \pm 2.0 \times 10^{6}$	0.20 ± 0.007
Yonggwang unit 4	$1.48 \times 10^{3} \pm 6.2 \times 10^{1}$	$\begin{array}{c} 1.28 \text{ x } 10^8 \\ \pm 5.4 \text{ x } 10^6 \end{array}$	0.65 ± 0.027
Yonggwang unit 1	$\begin{array}{c} 1.38 \ x \ 10^5 \\ \pm \ 1.0 \ x \ 10^4 \end{array}$	$\begin{array}{c} 2.24 \text{ x } 10^{10} \\ \pm 1.6 \text{ x } 10^9 \end{array}$	52.2 ± 3.8
Yonggwang unit 2	$\begin{array}{c} 8.28 \text{ x } 10^4 \\ \pm 4.0 \text{ x } 10^3 \end{array}$	$\begin{array}{c} 1.24 \ x \ 10^{10} \\ \pm \ 6.1 \ x \ 10^{8} \end{array}$	29.4 ± 1.5
MC-50	3.98 x 10 ⁵	1.43 x 10 ⁹	76.3



Fig. 3 Neutron absorbed dose of Wolsung unit 3 & Yonggwang unit 4



Fig. 4 Neutron absorbed dose of Yonggwang units 1, 2 & MC-50

IV. Conclusion

Rapid and easy dose assessment must be provided following criticality accident to permit medical staff to determine the necessary initial treatment for each victim. The KBADA was developed for the more rapid and easy neutron dose assessment using ²⁴Na activity in blood following well-established methods at ORNL. From this study, the results of neutron dose evaluations by the KBADA are shown to be capable of applying to neutron accident dose assessment.

Neutron spectra in Korea NPPs are still measured by research institutions such as NETEC, KRISS. Now RHRI is continuously collecting those neutron spectra into database in the KBADA. It will be useful for the neutron dosimetry research using TLD as well as for the neutron dose assessment using ²⁴Na in blood.

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