Development of Rapid Dose Assessment Program from Activated Sodium in Human Body for Criticality Accident

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A dose assessment program, called RADAPAS (Rapid Dose Assessment Program from Activated Sodium in Criticality Accidents), was developed to evaluate absorbed dose to an exposed person from induced activity of sodium-24, $^{24}\text{Na}$, in human body. A generally used personnel computer is available to run the developed program. RADAPAS prepares databases of energy spectra and dose conversion coefficients, which had been derived beforehand from Monte Carlo calculations with the MCNP-code. Information necessary for dose calculation is given by a dialogue method with interface displays. The dose to human body can be estimated from measured specific activity of $^{24}\text{Na}$ using the incorporated data and the interactively specified condition in RADAPAS. In order to validate its availability, RADAPAS was implemented to assess radiation dose in an experiment carried out at the Transient Experiment Critical Facility (TRACY) and for heavily exposed persons in the first criticality accident in Japan. The results show that RADAPAS is applicable to estimation of the magnitude of absorbed dose to a victim to determine initial medical treatment soon after a criticality accident.

KEY WORDS: dose assessment program, RADAPAS, activated sodium (sodium-24), criticality accident, MCNP-code, energy spectrum, exposed dose, specific activity of $^{24}\text{Na}$, Transient Experiment Critical Facility (TRACY).

I INTRODUCTION

A criticality accident brings about a severe exposure by large amount of neutrons and gamma-rays due to uncontrolled nuclear excursion in fissile material, even though sophisticated technology and appropriate procedures can restrain the possibility of its occurrence to almost zero. Decisions for medical treatment to heavily exposed persons require information for magnitude of absorbed doses to their bodies.\(^1\)\(^-\)\(^3\) Especially, early dose assessment and medical treatment significantly contribute to improvement of health for a victim in a criticality accident.\(^1\)\(^,\)\(^5\) The International Atomic Energy Agency (IAEA) recommends a dosimetry system using detectors with several kinds of activated foil.\(^1\) A system with combining dosimeters is being developed to estimate high radiation doses from neutron and gamma-rays separately.\(^4\)

In addition to these techniques, methods should be established for dosimetry in a serious situation, where no reliable information is available from any detectors.\(^2\) In such an unexpected case, absorbed dose to a person can be estimated from activity of sodium-24 ($^{24}\text{Na}$) induced in human body through neutron capture reaction of sodium-23, $^{23}\text{Na}(n, \gamma)\, ^{24}\text{Na}$.\(^1\)\(^,\)\(^5\)\(^-\)\(^7\) Some past accident events have proved effectiveness of this technique.\(^2\)\(^,\)\(^5\)\(^-\)\(^7\) However, dose assessment by this method alone is regarded as useful only for screening heavily exposed persons,\(^1\) because no systematic data had been prepared for conversion from specific activity of induced $^{24}\text{Na}$ to absorbed dose in human body.\(^2\) Thus, a previous study\(^5\) had clarified essential data for this dose assessment technique for some hypothesized accident situations by Monte Carlo calculations using the MCNP-code.\(^9\)

The dose assessment should be performed within certain accuracy even for initial stage soon after an outbreak of accident. The dosimetric quantity and its unit may be also argued for an acute exposure. From the view points of these discussions, a program, Rapid Dose Assessment Program from Activated Sodium (RADAPAS), was developed to meet the requirement in a rapid dose assessment from activity of induced $^{24}\text{Na}$ in human body. In RADAPAS, the absorbed dose to body of an exposed person can be derived with the incorporated data, which had been obtained in the previous
analyses. The dose conversion can be executed with information of source configuration or characteristics of radiations in an accident field. The availability was examined by an application of RADAPAS to neutron dose evaluations in an experiment at the Transient Experiment Critical Facility (TRACY) in the Japan Atomic Energy Agency (JAEA). RADAPAS was implemented to derive absorbed doses to heavily exposed persons in the first criticality accident at the nuclear fuel processing facility in Japan from the $^{24}$Na specific activity.

II ISSUES FOR RAPID DOSE ASSESSMENT WITH INDUCED $^{24}$Na

Two issues were taken into account to develop RADAPAS, which carries out a rapid dose assessment based upon quantity of induced $^{24}$Na in human body.

1. Data for dose conversion from induced $^{24}$Na activity

The previous study had derived the coefficients for dose conversions from quantity of induced $^{24}$Na in human body with Monte Carlo calculations using the MCNP-code. It had been clarified that the relation between quantity of induced $^{24}$Na and radiation dose to human body significantly depends on energy of incident neutrons to human body. Energy distributions and a mixture condition of neutrons and gammarays incident to a human body are influenced by compositions and thickness of materials between fissile material (radiation source) and the exposed person in a criticality accident. Then, calculations had been performed to obtain energy distributions of neutrons and gamma-rays escaping from the spherical configurations, which had consisted of fissile fuel, a container and surrounding shielding material.

In the following, a mathematical human model had been assumed to receive uniform exposure from neutrons or gamma-rays with each calculated energy distribution or mono-energetic neutrons. Some of the derived data are depicted in Figs. 1 and 2. The ‘Neutron dose’ in both figures does not include the dose component due to energy deposition by gamma-rays emitted through capture reaction of thermalized neutrons by some elements within human body. The dose component from gamma-rays emitted within human body is given with the term of ‘2nd gamma-ray dose’ in Fig. 2. It had been stated that the contribution of external gamma-rays could not directly relate to induction of $^{24}$Na from neutron exposure. Thus, conversion coefficients had been prepared for dose from external gamma-rays (hereinafter, ‘Gamma-ray dose’) for each supposed configuration.

Specific activity of $^{24}$Na induced within human body is a ‘reference value’ in this dose assessment technique. Activity of $^{24}$Na induced in body may be measured by a Ge semi-conductor detector for blood sample or a whole body counter. Thus, previous calculations had derived data of absorbed dose to a person per specific activity of induced $^{24}$Na averaged over soft tissue or a whole body. It was also clarified that the radiation dose per specific activity of $^{24}$Na induced in human body was larger in lateral irradiation geometry than in frontal irradiation geometry, as summarized in Table 1. The data derived by the previous analysis were incorporated as a database in RADAPAS.

2. Dosimetric quantity used in a criticality accident

Figure 2 shows that a person mainly receives dose from gamma-rays emitted within human body for an external exposure of low energy neutrons. The value of radiation weighting factor, $w_R$ for incident neutron is applied for 2nd gamma-ray dose to estimate effective dose for low dose exposure. Difference can be observed in early radiation health effect between neutrons and gamma-rays as well as stochastic effect, even if absorbed doses are same for the two types of radiation. Thus, RADAPAS was set up to provide separately the ‘2nd gamma-ray dose’ and the ‘neutron dose’ in a neutron exposure for a judgment of medical treatment.

Fig. 1 Conversion coefficients from $^{24}$Na specific activity induced in body soft tissue to absorbed dose in whole body for different configurations with fissile uranium in the frontal irradiation geometry. $^{1}$: Contained in Stainless container ($r=3 \text{ mm}$).

Fig. 2 Dependence of induction of $^{24}$Na in human body soft tissue and absorbed dose in whole body on neutron energy incident to human body in the frontal irradiation geometry. $^{8}$ $^{4}$: Specific activity induced in soft tissue over a whole body.
In addition, there is a room for discussions about the dosimetric quantity and its unit in the dose assessment for a criticality accident. The quantities in the unit of Sv, such as dose equivalent and effective dose, are not suitable for an acute exposure, because these quantities include implicitly quality factor, Q or wr. These factors are defined with consideration of appearance of stochastic effect. The values used in weighting neutron dose can be different between radiation effects of concern. From these considerations, the absorbed dose in the unit of Gy is adopted as the dosimetric quantity in RADAPAS.

### III DOSE ASSESSMENT METHOD IN RADAPAS

RADAPAS is an interactive dose assessment program, which is available for a generally used personnel computer running the Windows-XP operation system. Information to be required is set by a dialogue method with an interface display at each step. Dose calculation in RADAPAS is based upon the given information and incorporated data, which are described in Section 2.1. Two methods can be selected according to resources informed soon after an outbreak of a criticality accident.

The source configuration is one of the most influential factors in dose conversion from specific activity of $^{24}$Na in human body tissue. For example, neutron dose ranged more than 5 times between the configurations in Fig. 1 even for same $^{24}$Na specific activity. Gamma-ray dose depends more significantly on the condition around the source because of transmission of neutrons and gamma-rays in shielding. Thus, RADAPAS was set up to provide rapidly dose within certain accuracy based upon the source configuration. In the method of Fig. 3, the criticality system closest to the accident condition should be firstly selected from the fissile fuel and surrounding shielding in Table 2. Seven values were prepared

![Flowchart of dose assessment with incorporated data for the previously hypothesized criticality accident configurations in Table 2.](image)
for the radius of solution fuel. Among them, the solution fuel with radius of 20.2 cm was based upon the condition in the past criticality accident. The values of 17.4 cm, 18.6 cm, 23.8 cm and 42.4 cm were determined by the ratio of hydrogen to fissile material ($\text{H}/^{235}\text{U}=100$, 200, 500 and 1,000). The interval of thickness is small for all shielding materials below 20 cm, since conversion coefficients for neutron dose significantly decreases for the region, as depicted in Fig. 1. The ratios of gamma-ray doses to neutron dose were also taken into account to determine the thicknesses of concrete and heavy-type concrete.

RADAPAS can show energy spectra of neutrons and photons corresponding to the defined system in a graphic style. In the following, the conversion coefficients for each dose component are automatically selected from the database. Since the initially induced activity of $^{24}\text{Na}$ in human body, $A_{\text{Na}-24}$, plays a role as a reference value in dose calculation with RADAPAS, a correction should be made for decay of $^{24}\text{Na}$ ($T_{1/2}=14.96$ hours). The measuring time is also to be taken into account for the correction, because about 5% of $^{24}\text{Na}$ disintegrates, for example, during one hour measurement. The value of $A_{\text{Na}-24}$, however, can be obtained by correcting with the time interval between the outbreak of accident and the middle time of measurement, if a measurement is carried out within 2 hours. In addition, correction should be made for biokinetics of the isotope with biological half-life of $^{24}\text{Na}$, 10 days. A certain dose conversion from quantity of induced $^{24}\text{Na}$ also requires the method of gamma-ray spectrometry. The differences between the two geometries are about 40% and 90% for neutron dose and gamma-ray dose, respectively. Then, radiation doses can be appropriately converted from quantity of induced $^{24}\text{Na}$ in RADAPAS, if irradiation geometry is assumed from certain information, such as activation of elements in clothes. Absorbed doses averaged over a whole body and red bone marrow are calculated with the given information as Eqs. (1) to (3). The results of dose calculation are saved as a form of table.

$$D_n = A_{\text{Na}-24} \times C_{F_n}$$  \hspace{1cm} (1)
$$D_{2\gamma} = A_{\text{Na}-24} \times C_{F_{2\gamma}}$$  \hspace{1cm} (2)
$$D_{\gamma} = A_{\text{Na}-24} \times C_{F_{\gamma}}$$  \hspace{1cm} (3)

$D_n$, $D_{2\gamma}$, and $D_{\gamma}$: Neutron, 2nd gamma-ray and gamma-ray doses (Gy)

$A_{\text{Na}-24}$: Initially induced specific activity of $^{24}\text{Na}$ in human body ($^{24}\text{Na}\text{-Bq}/^{23}\text{Na}\text{-g}$)

$C_{F_n}$, $C_{F_{2\gamma}}$ and $C_{F_{\gamma}}$: Conversion coefficients for neutron, 2nd gamma-ray and gamma-ray doses ($\text{Gy}/^{24}\text{Na}\text{-Bq}^{23}\text{Na}\text{-g}$)

The energy spectra of neutrons and photons in a field can be obtained by other radiation transport calculations or measurements with dosimeter systems. It is preferable to apply the energy spectrum clarified by a certain method to dose assessment. Thus, RADAPAS can utilize such energy spectra with the flowchart in Fig. 4. A neutron energy spectrum should be
Fumiaki TAKAHASHI, Akira ENDO, Yasuhiro YAMAGUCHI and Keiji ODA

set with arbitrary energy boundaries and differential fluence in each bin for this method. A user can confirm the defined energy spectra with a graphic indication. The specific activities of $^{24}$Na is then converted two components of doses for neutron exposure based upon the given energy spectrum with the Eqs. (4) and (5). The conversion coefficient for each energy bin is calculated by interpolation of data plotted in Fig. 2. The interpolation is carried out using a 4-point Lagrangian interpolation formula on a log-log scale.

\[ D_n = A_{Na^{24}} \times \left( \sum_{i=1}^{N} f(i) \times CF_n(i) \right) \]  

\[ D_{2nd-y} = A_{Na^{24}} \times \left( \sum_{i=1}^{N} f(i) \times CF_{2nd-y}(i) \right) \]  

$D_n$, $D_{2nd-y}$ and $D_I$: Neutron, 2nd gamma-ray and gamma-ray doses (Gy)

$A_{Na^{24}}$: Initially induced specific activity of $^{24}$Na in human body ($^{24}$Na-Bq/$^{23}$Na-g)

$f(i)$: Fraction of differential neutron fluence in the i-th bin for neutron spectrum

$CF_n(i)$ and $CF_{2nd-y}(i)$: Interpolated conversion coefficients in the i-th energy bin for neutron dose and 2nd gamma-ray dose (Gy/$^{24}$Na-Bq/$^{23}$Na-g))

$N$: The number of energy bins for neutron energy spectrum

IV DOSE ASSESSMENT WITH RADAPAS

The data for dose conversions in RADAPAS had been obtained by the calculations with ideal situations, as described in Section 2.1. Then, the availability of RADAPAS was verified for the dose assessment in a realistic field and a heterogeneous exposure situation.

4.1 Dose assessment in a realistic field (Experiment in TRACY)

a) Examination for activation of sodium in human body

An experiment had been carried out at TRACY in JAEA to check applicability of the conversion coefficient derived by the previous analysis to neutron dose assessment from induced $^{24}$Na. TRACY can simulate a transient criticality accident condition and the reactor room is shielded with thick concrete walls. After sodium chloride solution contained in a slab phantom with polymethyl-methacrylate (PMMA) walls had been exposed to neutrons from the core tank in the reactor room, specific activity of $^{24}$Na in the solution had been measured with gamma-ray spectrometry using a Ge semi-conductor detector.

Following, calculations using the MCNP-code had quantitatively clarified the influences of differences in material and geometry between the mathematical human model and the irradiated slab phantom on activation of sodium. Some phantom models had been located at the position of irradiated slab phantom in the room model for the simulations. The existence of the 1cm thickness PMMA walls of the slab phantom affected the induction of $^{24}$Na, as summarized in Table 3. The measured activity of $1.58 \times 10^5$ ($^{24}$Na-Bq/$^{23}$Na-g) had been corrected to $1.33 \times 10^4$ ($^{24}$Na-Bq/$^{23}$Na-g) with the factor of 0.84 in the bottom column.

b) Neutron dose assessment based upon specific activity of the induced $^{24}$Na

Neutron dose had been assessed from the corrected $^{24}$Na specific activity with a conversion coefficient, which had been selected based upon the structure of the TRACY core tank among the data derived in the previous study, as listed in Table 4. This procedure was same as the dose calculation of Fig.3 in RADAPAS. Dose conversion coefficient had been derived with a mathematical human model in the above calculation to correct the measured $^{24}$Na specific activity. Measurements using two types of tissue-equivalent dosimeters had clarified distribution of neutron dose in the TRACY reactor room. Table 4 also presents the neutron doses assessed with these different approaches.

The flowchart in Fig. 4 was newly applied to the neutron dose assessment for the experiment. Neutron energy spectra were calculated at the slab phantom position with an elliptic cylindrical volume tally with the size of $20.2 \times 10.2 \times 175$ (h) cm by the MCNP-code. The tally volume was near to the external form size of the mathematical human model. Two models were defined in the calculation. One model (here-

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| Table 3 Effect of phantom material and geometry on induction of $^{24}$Na. |
|------------------------------|-----------------|-----------------|
| Material | Phantom geometry | The ratio of $^{24}$Na specific activity*1 |
| NaCl solution | Contained in a slab phantom with walls | 1.00 |
| Soft tissue | Contained in a slab phantom with walls | 0.96 |
| Soft tissue | Averaged over a slab phantom | 0.84 |
| Soft tissue | Mathematical human model | 0.84 |

*1: The value of experiment condition is set to 1.00.
*2: Size, 30 cm x 30 cm x 15 cm
*3: 1 cm thickness PMMA walls

| Table 4 Assessed neutron dose in the experiment at TRACY. |
|----------------|----------------|----------------|
| Method | Conversion coefficient ($^{24}$Na-Bq/$^{23}$Na-g) | Neutron dose (Gy) |
| RADAPAS | | |
| a) in Fig.3 method | $6.37 \times 10^{-5}$ | 8.5 |
| b) in Fig.4 method | | |
| Spectrum (A) in Fig.5 | $6.82 \times 10^{-5}$ | 9.1 |
| Spectral (B) in Fig.5 | $4.96 \times 10^{-5}$ | 6.6 |
| MCNP calculation | $4.69 \times 10^{-5}$ | 6.2 |
| Dosimeter system | | 7.2 |

*1: Calculated with a mathematical human model
Development of Rapid Dose Assessment Program from Activated Sodium in Human Body for Criticality Accident

inafter, (model A') took into account only the reactor tank. The model A is supposed for a dose assessment in a situation, where scattered radiations do not significantly contribute to an external exposure, such as a person near the tank. The room walls were added to this model in the other model (hereinafter, 'model B'). The model B can be defined after a criticality accident, if resources are informed to determine the position of exposed person and an accident field. Fig. 5 depicts the calculated neutron spectra with the two models. These spectra were applied to neutron dose assessment in the method of Fig. 4.

Table 4 presents neutron dose converted from the corrected $^{24}$Na specific activity in RADAPAS. The selected neutron spectrum was shifted to higher energy region than that in the calculated energy spectrum in the model A. The neutron dose, however, was larger in the method of Fig. 4 using the spectrum calculated with the model A than the dose given by the selected coefficient in the Fig. 3 method, which is given by calculation using the continuous function with neutron energy. This discrepancy is due to interpolation of coefficients at energy regions for thermal energy (<0.4 eV) and higher energy region more than 1 MeV. The difference of neutron doses between the two methods, however, is less than 10%. The neutron dose is 6.6 Gy, if the neutron spectrum calculated by using the model B was applied to the dose assessment in RADAPAS. This value is close to the doses obtained by other calculations and measurements, which are 6.2 Gy and 7.2 Gy, respectively. This result clarifies that precise consideration for energy distribution of incident neutrons improves the precision in assessment.

Some notices, however, should be mentioned here to perform dose assessment with the method of Fig. 4 in RADAPAS. The computation time of the Monte Carlo calculation using the model B was about 6 hours to obtain the energy spectrum in Fig. 5. It is not always assured that sufficient resources can be immediately informed to estimate energy characteristic after an accident. On the contrary, the method of flowchart in Fig. 3 requires only the information for a source configuration. The absorbed dose to an exposed person can be rapidly provided in this method soon after quantification of $^{24}$Na specific activity. Table 4 shows that the dose derived by the flowchart in Fig. 3 agrees with the doses by other estimation methods within about 30%. The difference is smaller than 50%, which is the value indicated as an acceptable uncertainty published by IAEA in dosimetry within 48 hours after an outbreak of accident.

4.2 Dose assessment for heterogeneous exposure

In a criticality accident, a victim can be exposed heterogeneously to large amount of radiations near fissile material. Two workers were heavily exposed to neutrons and gamma-rays near the precipitation tank, into which fissile uranium in unapproved quantity was poured, in the first criticality accident in Japan. Absorbed doses and specific activity of $^{24}$Na averaged over whole bodies had been analyzed for the heavily exposed workers by Monte Carlo calculations, in addition to precise dose distribution within their bodies. The heterogeneous exposure had been taken into account to establish geometry condition in the analysis and the posture of each worker is described in References 2) and 11).

The doses to the heavily exposed persons were assessed with RADAPAS based upon the value of $^{24}$Na specific activity given by the precise analysis. As the two persons worked beside the fissile uranium at the time of accident, the contribution of scattered radiations was small. Then, the method in Fig. 3 was carried out in this study. The criticality configuration

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**Fig. 5** Calculated neutron energy spectra at the irradiated slab phantom position in TRACY reactor room ((A): using model A and (B): using model B) and (C): the spectrum selected from database in RADAPAS.

**Fig. 6** Interface display for selecting the criticality configuration.
tion in the accident was closest to the assembly with only solution fuel in a 23.8 cm radius and stainless container among the systems, which can be selected with the display of Fig. 6 in RADAPAS. The exposure condition was set to the frontal irradiation geometry, as assumed in the initial dose assessment after the Tokai-mura criticality accident. The interface display to give $^{24}\text{Na}$ specific activity and irradiation geometry for the worker A. The calculated dose was given with the format of Fig. 8, which was indicated within a few minutes after defining the recourses in Figs. 6 and 7.

Table 5 compares the doses given by RADAPAS with the doses previously obtained with the analysis. The absorbed doses averaged over whole body agree well with each other except the gamma-ray dose of the worker A. Difference can be seen only for the gamma-ray dose of the worker A between the two methods. The posture of the worker A was slightly different from the frontal irradiation condition, assuming from the skin dose distribution derived in the previous analysis. Incident direction of radiations to body affects more significantly on the conversion coefficient for the gamma-ray dose than that for the neutron dose, as presented in Table 1. Then, the ratio of the gamma-ray dose to neutron dose is different between the results by RADAPAS and the previous analysis.

Table 5 Dose assessment with RADAPAS for the heavily exposed persons in the first criticality accident in Japan.

<table>
<thead>
<tr>
<th>Dose component</th>
<th>Assessed dose with RADAPAS (Gy)</th>
<th>Dose by the precise analysis (Gy) $^{11}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Conversion coefficient (Gy/$^{24}\text{Na-Bq}^{23}\text{Na-g}$))</td>
<td>Worker A $^{*1}$</td>
</tr>
<tr>
<td>Neutron</td>
<td>$6.23 \times 10^{-5}$</td>
<td>5.55</td>
</tr>
<tr>
<td>2nd gamma-ray</td>
<td>$1.26 \times 10^{-5}$</td>
<td>1.12</td>
</tr>
<tr>
<td>Gamma-ray</td>
<td>$1.04 \times 10^{-4}$</td>
<td>9.27</td>
</tr>
</tbody>
</table>

$^{*1}$: The worker supporting the tank; Specific activity of $^{24}\text{Na}$ : $8.91 \times 10^4$ (24Na-Bq/23Na-g)$^{11}$

$^{*2}$: The worker pouring solution fuel; Specific activity of $^{24}\text{Na}$ : $5.23 \times 10^4$ (24Na-Bq/23Na-g)$^{11}$
Table 3 indicates, however, that RADAPAS can be applied to judge the magnitude of exposure for medical decision-making soon after an accident regardless of homogeneity in exposure.

V CONCLUSIONS

A dose assessment program, RADAPAS, was developed to give medical staff information for degree of radiation dose to an exposed person in a criticality accident even in a serious situation, where no certain information is available from any dosimeters. RADAPAS can assess both neutron and gamma-ray absorbed doses to a heavily exposed person from the specific activity of induced $^{24}\text{Na}$ within body. The dose conversion can be performed soon after an occurrence of accident, if source configuration is informed and $^{24}\text{Na}$ specific activity is quantified. In addition, any other reliable radiation spectrum can be also utilized in dose estimation with RADAPAS. The necessary information can be interactively set by an interface display without any complicated procedures. The validity of RADAPAS was examined with dose assessments in the experiment at TRACY and the heavily exposed persons in the first criticality accident in Japan. The results show that RADAPAS meets the requirement for early dose assessment following a criticality accident. RADAPAS can be executed in a generally used personnel computer and will be provided by requesting the use of the program to the author (F. T.).

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