An Evaluation Method for Activation Analysis using Pre-evaluated Contribution of Nuclides with Impurity

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1. Introduction

Recently, one of the significant issues in nuclear industry is a residual radiation, which is emitted a long period. Nuclides in radiation facilities become unstable from nuclear reaction. It emits residual radiation to be stable. Some unstable nuclides remain after operation in the material. It continuously emits the radiation, which has a harmful effect to worker when they try maintenance and plant decommissioning.

It is known that residual radiation from impurity occupies a large portion of the radiation dose [1]. If impurity concentration is higher than expectation, the effects of residual radiation could be underestimated. Therefore, estimation of residual radiation is repeatedly calculated according to impurity concentration. In this study, an approach estimating the activation was proposed using pre-evaluated nuclide's contribution to reduce the calculation time and effort of worker.

2. Methods and Results

2.1 Overview of Activation Calculation

Abundances of radioactive nuclide with consideration of decay chain can be estimated by Bateman equation as given in Eq. (1)

$$\frac{dN_m(t)}{dt} = -\beta_m N_m(t) + \sum_{k\neq m} N_k(t)\gamma_{k\to m} \quad (1)$$

where $N_m(t)$ is atom density nuclide of '*m'* nuclide, $-\beta_m N_m(t)$ represents disappearance term for *m* nuclide, and $\sum_{k\neq m} N_k(t)\gamma_{k\to m}$ represents production term for *m* nuclides from other nuclides. Each terms of the Eq. (1) is calculated in consideration of spontaneous decay and reactions with neutron, as follows:

$$\beta_m = \lambda_m + < \phi(E)\sigma_a^m > \quad (2)$$
$$\gamma_{k\to m} = \lambda_{k\to m} + < \phi(E)\sigma_a^m > \quad (3)$$

where

λ_m = decay constant of *m* nuclide,

$\gamma_{k\to m}$ = decay constant of *k* nuclide into *m* nuclide,

$\phi(E)$ = neutron flux.

The material compositions after neutron irradiation can be estimated by solving a matrix, which elements are consists of Bateman equation with a given initial conditions. The matrix form of eq. (1) can be expressed as:

$$\overrightarrow{N'} = [A]\vec{N} \quad (4)$$

The solution of Eq. (4) can be express as follows [2]:

$$\vec{N} = [V][\Lambda][V]^{-1}\vec{N}_0 \quad (5)$$

where

$\vec{N}$ = atom density at given time t,

$\vec{N}_0$ = initial nuclide distribution in the material

$[V] = (\boldsymbol{v}_1|\boldsymbol{v}_2|\ldots|\boldsymbol{v}_N)$

$[\Lambda] = \mathrm{Diag}[e^{\Lambda_1 t}, e^{\Lambda_2 t}, \ldots, e^{\Lambda_n t}]$

Λ_n = eigenvalues of matrix [*A*]

If the RHS of Eq. (5) is set to certain nuclide '*m*' and $\vec{N}_0$ is set to initial nuclide '*k*', $[V][\Lambda][V]^{-1}$ means contribution to '*m*' nuclide from '*k*'. Therefore, the number of nuclide '*m*' can be obtained by following equation.

$$\mathrm{N}_m(t) = \sum_k^{all} C_{k\to m}(t) N_k(t=0) \quad (6)$$

where, $C_{k\to m}(t)$ is contribution to *m* nuclide number of *k* nuclide at given time t.

2.2 Proposed method

Intensity of gamma produced by unstable nuclide can be described by following expression:

$$N_m(t)Q_m(E_\gamma) = I_m(E_\gamma, t) \quad (7)$$

where $Q_m(E_\gamma)$ is the gamma production rate, and $I(t,E_\gamma)$ is gamma intensity come from '*m'* nuclide at given time *t*.

By substituting Eq. (7) into Eq. (1), variation of gamma with time can be obtained as follows:

$$\frac{dI_m(E_\gamma,t)}{dt} = -\beta_m(t)N_m(t)Q_m(E_\gamma) + \sum_{k\neq m} N_k(t)\gamma_{k\to m}(t)Q_m(E_\gamma) \quad (8)$$

In the same manner of Eq. (6), the gamma intensity, which is solution of Eq. (8), can be written as follows:

$$\mathrm{I}(E_\gamma, t) = \sum_{k}^{all} N_k(t=0) f_k^{+}(E_\gamma, t) \qquad (9)$$

where, $f_k^{+}(E_\gamma, t)$ is residual gamma contribution come from *k* nuclide.

It was assumed that residual gamma intensity follow exponentially decrease because radioactive nuclides are decay with exponential form. Thus, the gamma intensity in certain time can be calculated based on pre-evaluated contribution:

$$\mathrm{I}(E_\gamma, t^*) = \sum_{k}^{all} N_k(t=0)\{y_i \exp(-\ln(\frac{y_i}{y_{i+1}})(\frac{t^* - t_i}{t_{i+1} - t_i})\} \qquad (10)$$

To solve the Eq. (10), An evaluation procedure was proposed as follows: (1) calculate incident neutron spectrum to target material; (2) by using the Bateman equation, evaluate nuclide's contribution for specific response such as residual gamma spectrum; (3) Using Eq. (10), estimate the response at time *t*. Fig. 1 shows procedure of estimation approach.

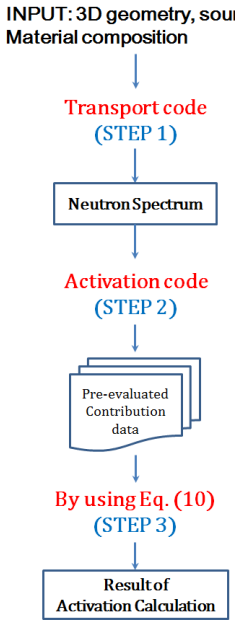


Fig. 1 Procedure of proposed method

2.3 Validation of Proposed Method

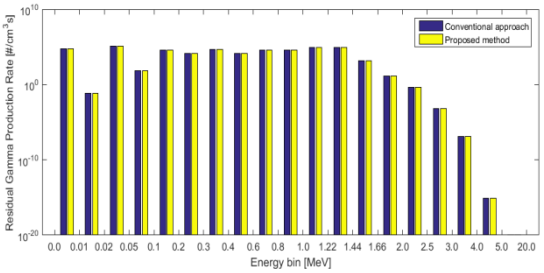
Residual gamma calculation in a part of nuclear power plant was performed to validate the proposed approach. Transport calculation for getting neutron spectra was simulated by using MCNPX 2.7 [3] code and activation calculation was performed with FISPACT 2010 [4] with EAF-2010 activation library [5]. The calculation conditions are as follows:

I. The contribution was stored (evaluated) at time of 1d, 7d, 1m, 6m, 1y, 10y and 30y after shutdown.
II. The nuclear reactor is operates during 40 years with 18 months operating and 60 days cooling per period.
III. The contribution $f_k^{+}(E_\gamma, t)$ was calculated at 1 sec, 1 day, 30 day, 180 day, 1 year, 10 year, and 30 year after shutdown
IV. Concreate density is 2.30 g/cm^3
V. The concreate composition is given in Table I

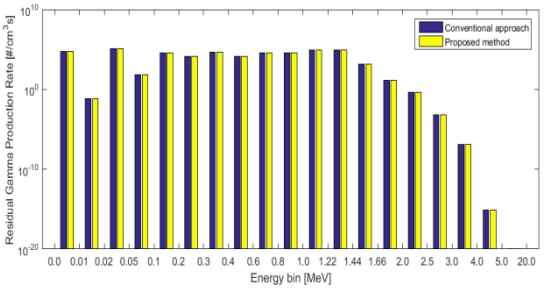
Table I: Concreate composition used in the calculation

Nuclide	Density (g/ cm^3)	Nuclide	Density (g/ cm^3)
H-1	1.30000E-02	Ca-43	2.79363E-04
Li-6	6.52508E-06	Ca-44	4.42518E-03
O-16	1.17100E+00	Ca-46	9.70010E-06
Na-23	4.00000E-02	Ca-48	4.32624E-04
Mg-24	4.68588E-03	Fe-54	1.65271E-03
Mg-25	6.12660E-04	Fe-56	2.66423E-02
Mg-26	7.01460E-04	Fe-57	6.20890E-04
Al-27	1.07000E-01	Fe-58	8.41000E-05
Si-28	6.81704E-01	Co-59	3.20816E-05
Si-29	3.57499E-02	Ni-58	9.46136E-05
Si-30	2.45456E-02	Ni-62	7.41684E-06
K-39	4.18217E-02	Cs-133	8.67835E-06
K-41	3.17295E-03	Eu-151	3.03797E-06
Ca-40	1.87544E-01	Eu-153	3.32779E-06
Ca-42	1.30951E-03		

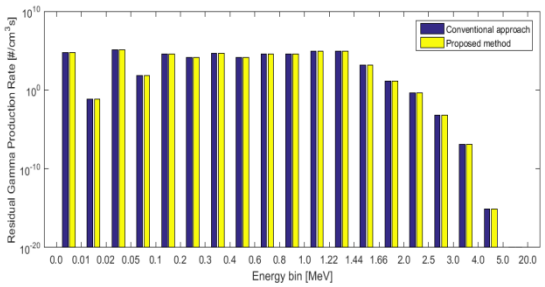
Fig. 2 shows gamma spectrum emitted from concreate at 1 year after shutdown. This result shows that there is good agreement within 7.3 % between the conventional approach and proposed method. Therefore, it was expected that proposed method can accurately estimate the gamma intensity without additional activation calculation.



(a) 1 year after shutdown (pre-evaluated point)



(b) 11 months after shutdown (near the pre-evaluated points)



(c) 9 months after shutdown (middle of pre-evaluated points)

Fig. 2. Gamma spectrums obtained by proposed method and the conventional approach

3. Conclusions

In this study, in order to reduce the calculation time and effort of worker, activation analysis method based on pre-evaluated nuclide contribution was proposed. This method was verified using concreate activation problem, which is located in nuclear power plant. The results show that our proposed method has good agreement with Bateman equation. Hence, it is expected that proposed method can efficiently evaluate an activation problem according to concentration of impurity.

Acknowledgement

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