Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 19-20, 2022

Analyses on the Important Fission Products for Source Term Estimation during a Severe Accident

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1. Introduction

The importance of source term evaluation under severe accidents was more widely recognized shortly after the Three-Mile Island accident as it was observed that only relatively small amounts of iodine were released to the environment compared to the amount from TID-14844 [1]. Therefore, an attempt was made to revise the source term by comparing the major relevant insights available from recent severe accident research on the fission product behavior. The resulting source term is provided in NUREG-1465 [2]. In NUREG-1465, the source term is categorized into several groups based on the similarity of chemical behaviors. However, it requires additional information such as the amount of the radioactive materials in the reactor, in order to evaluate the radiation dose due to a severe accident, which is essential for consequence analysis.

In order to perform such an analysis, the amount of radioactive materials produced in the reactor and released into the containment/environment. In this study, we select important fission products considering the amount of the radioactive materials in the reactor obtained via DEGETION (Depletion Generation and Transmutation of Isotopes on Nuclear application) [3], release fractions derived from NUREG-1465, and effective dose conversion coefficients taken from ICRP 119 [4].

For the important fission products, adjoint solutions of Bateman equations [5], are calculated in order to analyze the importance of the precursors producing the aforementioned fission products, which will provide the insights into the assumption used in the decay chain of MACCS2 for level 3 PSA analysis [6].

2. Calculational Framework on Important Fission Products during a Severe Accident

2.1 Selection of Important Fission Products

From the depletion calculation via DEGETION, we can obtain the change of concentrations of fission products in the reactor during normal operation. Then the release fraction of the fission products should be considered in the source term analysis. The fraction can be obtained from via severe accident analysis code and it can vary depending on the accident scenario. In this paper, as discussed in the previous section, we use the values listed in NUREG-1465 [2], which are regarded as standard values derived from the analyses of various accident scenarios. The values that we consider are the release fractions of the PWR and they are listed in the following table.

Table 1. Release fraction into containment for PWR

Fission product group	Fraction
Noble gas	1.0
Iodine	0.75
Cesium	0.75
Tellurium	0.305
Strontium	0.12
Barium	0.12
Ruthenium	0.0055
Lanthanum	0.0055
Cerium	0.0052

Then, effective dose conversion coefficients for each individual fission products are considered to select important fission products since the fission products show different radiological effects on the human body depending on their physical and chemical properties and biological effects from their affinity to human organs. The normalized effective dose for fission products generated for reactor operation time of Δt_b and time of Δt_c passed after release from the reactor, in vector form as follows:

$$\vec{E_d}\left(\Delta t_b, \Delta t_c\right) = E_{dose} \cdot \vec{R}\left(\vec{X}\left(\Delta t_b\right), \Delta t_c\right), \quad (1)$$

where

E_{dose} : matrix for the effective dose conversion coefficient for fission products,

$\vec{R}\left(\vec{X}\left(\Delta t_b\right), \Delta t_c\right)$: normalized concentration vector for fission products that are generated for reactor operation time of Δt_b and time of Δt_c passed after release from the reactor, calculated by the following equation:

$$\vec{R}\left(\vec{X}\left(\Delta t_b\right), \Delta t_c\right) = \exp\left(A_{dec} \cdot \Delta t_c\right) \cdot R_f \cdot \vec{X}\left(\Delta t_b\right), \quad (2)$$

A_{dec} : matrix on the radioactive decay (cooling) of isotopes,

$\vec{X}\left(\Delta t_b\right)$: normalized concentration of fission products generated for reactor operation time of Δt_b,

R_f : matrix form of release fractions of the isotopes during a severe accident.

Note that we use the effective dose conversion coefficients when the size of aerosol particle consisting of each fission product is 5 μm, since it is regarded as the typical size of aerosol particles for dose conversion

calculation [4]. In the case of iodine, we use the dose conversion coefficients for the elemental gaseous form, since it is known to be the most conservative in terms of dose conversion, as discussed in TID-14844 [1]. From the numerical results from the above calculation, we select important fission products showing higher contributions than 10% to the total effective dose in the source term during a severe accident.

2.2 Backward calculations for importance of the precursors for the fission products

The important fission products selected from the aforementioned procedures can be continuously produced by radioactive decay of precursors after release from the reactor. Therefore, we should also consider the amount of the released precursors. In order to consider the importance of such precursors with regard to the radioactive decay, we perform backward calculations to obtain the adjoint solutions of Bateman equation.

For the arbitrary parameter α, which may cause change of the concentration vector on the fission products $\vec{X}$, and/or matrix A_{dec}, we have the following :

$$\frac{d}{d\alpha}\left(A_{dec}\cdot\vec{X}-\frac{d\vec{X}}{dt}\right)=\frac{\partial A_{dec}}{\partial\alpha}\cdot\vec{X}+A_{dec}\cdot\frac{d\vec{X}}{d\alpha}-\frac{d}{dt}\cdot\frac{d\vec{X}}{d\alpha}. \quad (3)$$

If we define an auxiliary equations as follows :

$$A_{dec}^{*}\cdot\vec{X}^{*}=-\frac{d\vec{X}^{*}}{dt}-\frac{\partial P}{\partial\vec{X}}, \quad (4)$$

where

A_{dec}^{*} *:* transpose matrix on A_{dec},

P : response function in the auxiliary equation.

With some algebra, we can obtain the following :

$$\left\langle\frac{\partial}{\partial t}\left(\frac{d\vec{X}}{d\alpha}\cdot\vec{X}^{*}\right)\right\rangle+\left\langle\frac{dP}{d\vec{X}}\cdot\frac{d\vec{X}}{d\alpha}-\vec{X}^{*T}\cdot\frac{\partial A_{dec}}{\partial\alpha}\cdot\vec{X}\right\rangle=0, \quad (5)$$

where

< > : integration over time interval of interest.

Solving for the second term of Eq. (5), we have

$$\left\langle\frac{dP}{d\vec{X}}\cdot\frac{d\vec{X}}{d\alpha}\right\rangle=\left\langle\vec{X}^{*T}\cdot\frac{\partial A_{dec}}{\partial\alpha}\cdot\vec{X}\right\rangle. \quad (6)$$

In order to determine the meaning of Eq. (6), we make an additional equation. Let K be a response function and for the arbitrary parameter α, which may cause change of the concentration vector on the fission product, we obtain the sensitivity of *K* to α as follows :

$$\frac{\delta K}{K}=\left[S_k\cdot\frac{\delta\alpha}{\alpha}\right]+\text{higher order terms}, \quad (7)$$

where Sk is sensitivity coefficient

Using the Taylor series expansion on the function K and with some algebra, we have the following relationship :

$$S_k=\frac{\alpha}{K}\cdot\frac{\partial K}{\partial\alpha}+\vec{X}^{*T}\cdot\frac{\partial A_{dec}}{\partial\alpha}\cdot\vec{X}, \quad (8)$$

If there is no change in the initial condition, we have the following :

$$\delta K=\left\langle\vec{X}^{*T}\cdot\frac{\partial A_{dec}}{\partial\alpha}\cdot\vec{X}\right\rangle. \quad (9)$$

Since Eq. (4) is equivalent to the adjoint time-dependent Boltzmann transport equation, the solution $\vec{X}^{*}$ can be interpreted as the importance vector of the isotope concentrations at time t to the response at time t_f.

For the best-estimation of the importance, a normalized amount of the fission product release should be considered due to various volatilities of the nuclides depending on their chemical characteristics as discussed in previous section. Then the importance is expressed as

$$W\left(\Delta t_b,\Delta t_c\right)=\vec{R}^{T}\left(\vec{X}\left(\Delta t_b\right),\Delta t_c\right)\cdot\vec{X}^{*}\left(\Delta t_c\right)\cdot \quad (10)$$

For efficient calculation, the initial condition perturbation can be excluded [5]. Then Eq. (3) becomes the following final value problem which is equivalent to the initial value problem of forward calculation :

$$A_{dec}^{*}\cdot\vec{X}^{*}=-\frac{d\vec{X}^{*}}{dt},\quad \vec{X}^{*}\left(t_f\right)=\vec{X}_f^{*}. \quad (11)$$

Note that Eq. (11) is compatible with a system of Bateman equations in a matrix form. Therefore, it can be solved by the conventional matrix exponential method. In this work, we use the CRAM method.

3. Numerical Results

In order to identify the important fission products in source term estimation during a severe accident of PWR, we apply the aforementioned scheme to the analyses of the PWR fuel. In the analyses, we consider 3.17w/o UO_2 fuel which is typical enrichment in PWR. In the calculation, CRAM implemented in DEGETION [3] is used with a time step of 1.0 days. 32 GWD/MTU is considered as the burnup of fuel when the release of fission products starts, which corresponds to 180 days of depletion, respectively.

With the results of the depletion calculation, we also calculate the normalized effective dose from the fission products released from the reactor core. In this calculation, we assume that the released fission products are completely inhaled by human in the off-site for the conservative estimation. We also assume that the fission products released from the reactor undergo radioactive decay for three days since the containment integrity is assumed to be maintained for three days in the severe accident analyses. The following Fig. 1 shows contribution of the fission products to the source term.

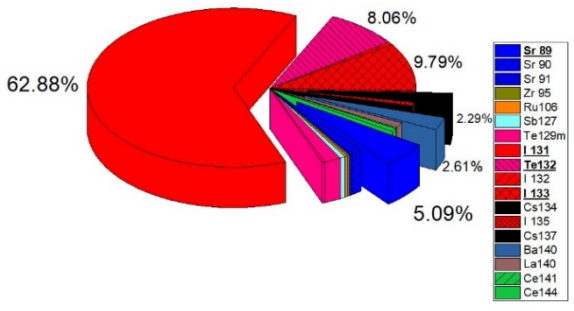

Fig. 1. Contribution of the fission products to total effective dose during a severe accident

As shown in Fig. 1, I-131 shows largest contribution to the total effective dose, i.e., it is larger than 40% of the total effective dose, which corresponds to the body of knowledge on the radioactive fission products in the source term evaluation. I-133 shows the second largest contribution to the total effective dose, i.e., it is 10~12 % of the total dose. Considering that the iodine undergoes various chemical reactions in the containment and the effective dose of iodine can vary depending on the chemical forms, the results stress the importance of the analyses of the chemical behaviors of iodine during a severe accident for the best-estimation of the source term.

Meanwhile fission products showing top 20 contributions but not discussed in Fig. 1 are listed in Table 2. Note that most of the fission products listed in the table have longer half-life than those of the aforementioned fission products. The list includes some of well-known fission products such as Ru-106, Cs-137, and etc.

Table 2. List of top 20 contributors to the total effective dose

Fission product	Contribution [%]	Half-lfie [sec]
Sr-88	5.09	4.366E+06
Sr-90	1.24	9.079E+08
Sr-91	0.35	3.467E+04
Zr-95	0.28	5.532E+06
Ru-106	0.31	3.210E+07
Sb-127	0.33	3.326E+05
Te-129m	1.82	1.062E+03
Te-131m	0.31	1.197E+05
I-131	62.89	6.934E+05
Te-132	8.06	2.768E+05
I-132	0.60	8.262E+03
I-133	9.79	7.488E+03
Cs-134	2.29	6.512E+07
I-135	0.72	2.365E+04
Cs-136	0.29	1.137E+06
Cs-137	0.99	9.486E+08
Ba-140	2.61	4.022E+08
La-140	1.14	5.293E+07
Ce-141	0.61	2.815E+06
Ce-144	0.22	2.462E+07

For I-131 and I-133, we perform adjoint calculations in order to determine the effect of precursors on the productions of the aforementioned important fission products. The calculations are also done by DEGETION with the same computation conditions as considered in the depletion calculation except that the burnup matrix in the adjoint calculation is constructed considering only radioactive decay. In addition, the final concentrations of the important fission products are set to be zero since in this calculation, we focus on the production of the three aforementioned isotopes. The results of the adjoint calculations are shown in the following figures. Note that from Figs. 2~3, the importance of precursors of the two aforementioned important fission products are very low compared to those of the fission products themselves. Therefore, we can conclude that the contribution of the precursors of the aforementioned fission products to the effective dose from the fission products are low in the source term during a severe accident.

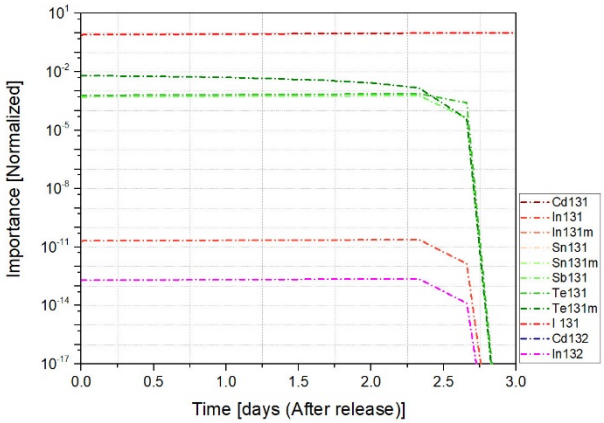

Fig. 2. Importance of precursors producing I-131

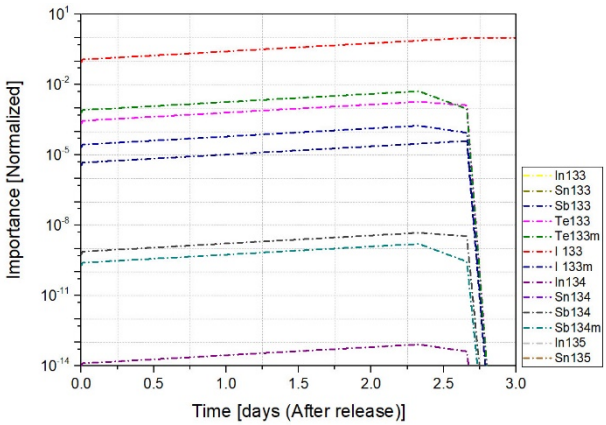

Fig. 3. Importance of precursors producing I-133

We also calculate the normalized amount of I-131 after release of the reactor assuming that there are no precursors of the aforementioned fission products if their adjoint are less than 10^{-2} and the results are compared with those with consideration of all precursors of the isotopes as shown in Figs. 4~5. Note that there are no significant differences between the results of calculations. Therefore, we can conclude that the assumption on the decay chain used in MACCS2 [6] code for the level 3 PSA analysis is reasonable with reduced computing time for effective calculations.

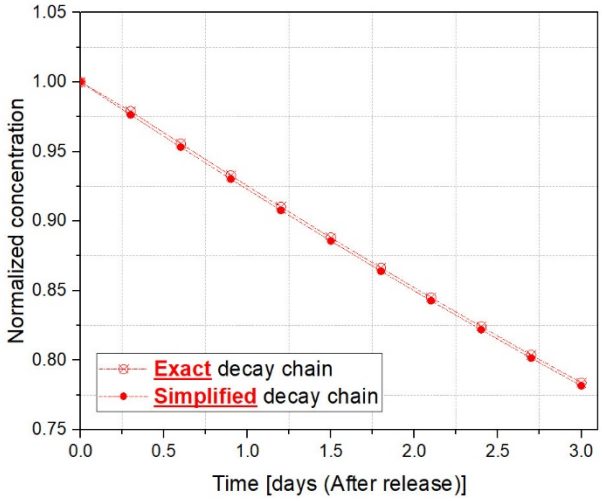

Fig. 4. Comparison of normalized concentration of I-131 for the calculations with exact decay chain and those with simplified decay chain

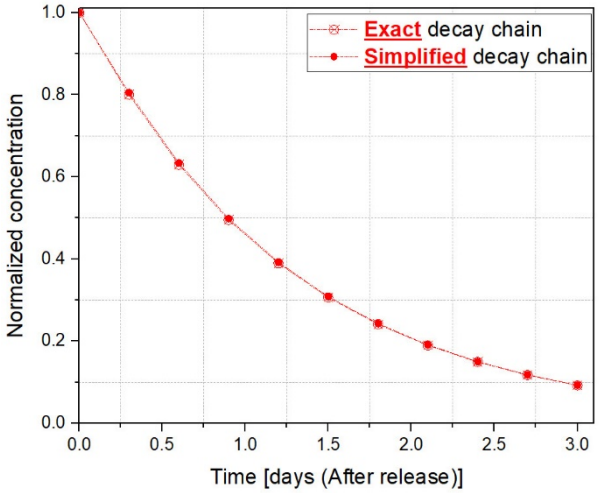

Fig. 5. Comparison of normalized concentration of I-133 for the calculations with exact decay chain and those with simplified decay chain

4. Conclusions

In this paper, first, we selected the important fission products for the estimation of source term during a severe accident with depletion calculations on the typical PWR fuel via DEGETION, the release fraction obtained from NUREG-1465, and the effective dose conversion coefficients from ICRP 119. We also performed adjoint calculations on the Bateman equation in order to determine the importance of precursors on the productions of the important fission products.

We found that I-131 shows the highest contribution to the total effective dose, i.e., more than 60 % contribution. We also found that I-133 shows the second highest contributions i.e., the contribution is 10~12% of the total effective dose. Te-132 and Sr-89 also showed considerable contributions, i.e., 5~9% contributions. Therefore, the dose contribution from the iodine species is larger than 70 %.

From the backward analysis, we found that the contributions to the total effective dose from the aforementioned fission products are mainly from the products directly. Therefore, we concluded that the assumption on the decay chain used in MACCS2 code for the level 3 PSA analysis is reasonable with reduced computing time for effective calculations.

As perspective on the source term studies during a severe accident, it is important to perform analyses of the iodine behaviors in the containment since we found that the iodine isotopes are dominant contributors to the total effective dose. In addition, the iodine isotopes are found to undergo various physical and chemical reactions in the containment, which leads to various effective dose.

ACKNOWLEDGEMENTS

This work was supported by the Nuclear Safety Research Program through the Korea Foundation Of Nuclear Safety (KoFONS) using financial resources granted by Nuclear Safety and Security Commission (NSSC), Republic of Korea (No. 2106007).

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