A Prediction Method for the Dose Rate of Fuel Debris Depending on the Constituent Elements

Kenichi Terashima1,2*, Keisuke Okumura1,2

1Collaborative Laboratories for Advanced Decommissioning Science, Japan Atomic Energy Agency (JAEA)
2International Research Institute for Nuclear Decommissioning (IRID)

* terashima.kenichi@jaea.go.jp

Abstract. In 2021, fuel debris samplings are planned to start as part of a step-by-step process at the Fukushima Daiichi nuclear power station. The dose rate of the fuel debris for safety treatments of the fuel debris should be predicted. However, various elements are mixed in the fuel debris, and thus predicting the dose rate will be challenging. Therefore, we conducted a large number of Monte Carlo radiation transport simulations for cases where parameters such as fuel debris size, composition, and density were significantly changed. Consequently, we obtained a simple and analytical formula that can predict the dose rate using a minimum number of parameters.

Keywords: Fuel debris, Dose rate, Fukushima Daiichi, PHITS, Computer simulation

1. Introduction

The 2011 Great East Japan Earthquake and the resulting tsunami caused a fuel-melting accident at the Fukushima Daiichi nuclear power station (hereafter “1F”). Consequently, nuclear fuel (UO2), fuel cladding material (zirconium alloy), structural material (stainless steel), neutron absorbers (B4C, Gd2O3), etc. were melt-mixed, and a large amount of fuel debris was generated in units 1 to 3. It is assumed that some of the melted mixtures were dropped through the reactor vessel and subsequently mixed with concrete at the bottom of the primary containment vessel.

At 1F, several internal investigations have been conducted by robots, whereas fuel debris retrieval is planned to start in 2021 [1]. The retrieval will begin on a small scale before being gradually expanded. In the early stage of the retrieval up to about 2030, it is predicted that the retrieved fuel debris will be limited to a size of a few centimeters. Since the retrieved fuel de-
bris will be analyzed and stored, the range of the attendant dose rate should be predicted.

In this study, we proposed a simple method to predict the surface dose rate of small-sized fuel debris (<10 cm in diameter) depending on numerous constituent elements. The method was obtained by combining theoretical formulas and photon transport simulations with the Particle and Heavy Ion Transport code System PHITS [2].

2. Evaluation method

A dose rate produced by a photon source is proportional to the photon intensity of the source. Therefore, the surface dose rate can be expressed as follows:

\[ D = P \cdot R \]  

(1)

where \( D \) denotes the surface dose rate [mSv/h], \( P \) denotes the function expressing the photon intensity [photons/s], and \( R \) denotes the function expressing the surface dose rate caused by the unit photon source [(mSv/h)/(photons/s)]. In this equation, the effects of nuclide weight and decay time are addressed via the \( P \) function. Since both \( P \) and \( R \) functions depend on nuclides \((i)\) in fuel debris, \( D \) can be expressed according to a summation of all nuclides as follows:

\[ D = \sum_i P_i \cdot R_i \]  

(2)

2.1. Photon intensity function \( P_i \)

In this study, we assumed that the fuel debris is a homogeneous sphere with radius \( r \) and considered the function for expressing \( P_i \) (\( P_i \) function). Assuming that the source nuclides are accompanied by uranium (U) in the fuel debris, the photon intensity can be expressed as follows:

\[ P_i (r, \rho, u, B, t) = \left( \frac{4}{3} \pi r^3 \right) \cdot \rho \cdot \left( \frac{u}{100} \right) \cdot p_i (B, t) \]  

(3)

where \( r \) denotes the radius of the fuel debris [cm], \( \rho \) denotes the bulk density of the fuel debris [g/cm\(^3\)], \( u \) denotes the U concentration of the fuel debris [wt\%], and \( p_i \) denotes the function expressing the photon intensity per U mass in the fuel debris from a nuclide \( i \) [(photons/s)/(g-U)]. This depends on the fuel burnup degree \( B \) [GWd/t] and the elapsed time \( t \) [years] from the 1F accident. The bulk density \( \rho \) [g/cm\(^3\)] was set as follows according to the true density \( \rho_0 \) [g/cm\(^3\)] and porosity ratio \( \alpha \) [%]:

74
However, in terms of the actual situation, noble gas and volatile fission products (FPs), such as krypton (Kr) and cesium (Cs), were released to the outside of the fuel debris at the time of the 1F accident, and this should be addressed. Since the actual release ratio of each nuclide in the 1F debris is unclear, it was set based on the results of the FP release test, Phebus-FPT4 [3], as shown in Table 1. Following the FP release, the residue ratio was set to $f_i$ for each nuclide $i$ and the $P_i$ function was corrected as follows:

$$P_i(r, \rho, u, B, t, f_i) = f_i \cdot \left(\frac{4}{3} \pi r^3\right) \cdot \rho \cdot \left(\frac{u}{100}\right) \cdot p_i(B, t)$$  \hspace{1cm} (5)

| Type           | Element | Release ratio | Residue ratio ($f_i$) |
|----------------|---------|---------------|-----------------------|
| Noble gas      | He      | 0.99          | 0.01                  |
|                | Ne      | 0.99          | 0.01                  |
|                | Ar      | 0.99          | 0.01                  |
|                | Kr      | 0.99          | 0.01                  |
|                | Xe      | 0.99          | 0.01                  |
|                | Rn      | 0.99          | 0.01                  |
| Volatility FPs | I       | 0.97          | 0.03                  |
|                | Cs      | 0.84          | 0.16                  |
|                | Te      | 0.80          | 0.20                  |
|                | Mo      | 0.77          | 0.23                  |
|                | Rb      | 0.53          | 0.47                  |
|                | Cd      | 0.44          | 0.56                  |
|                | Ba      | 0.35          | 0.65                  |
|                | Sb      | 0.30          | 0.70                  |
|                | Pd      | 0.27          | 0.73                  |
|                | Ag      | 0.092         | 0.908                 |
|                | Tc      | 0.071         | 0.929                 |
|                | Ru      | 0.018         | 0.982                 |
|                | Sr      | 0.014         | 0.986                 |
2.2. Radioactive nuclides contributing to dose rate

Specifically, in Eq. (2), we should consider all the radioactive nuclides produced during the 1F reactor operation. However, the number of nuclides currently contributing to the surface dose rate is expected to be limited. To elucidate the contributing nuclides, we compared the dose rate $D_{i}^{H_{i}10}$ at a 1.0 m distance from a point source of each nuclide $(i)$ in the air using the following equation:

$$D_{i}^{H_{i}10} = H_{i}^{*}(10) \cdot f_{i} \cdot p_{i}(B, t)$$  \hspace{1cm} (6)

where $H_{i}^{*}(10)$ is the 1 cm dose equivalent rate constant of nuclide $i$, $f_{i}$ is given in Table 1 (otherwise = 1.0), and $p_{i}(B, t)$ was estimated according to the fuel burnup calculation for unit 2 [4] until the accident and the radioactive decay calculation following the accident up to March 2021 (10 years after the 1F accident). Table 2 shows the results.

It was found that $^{137m}\text{Ba}$, $^{154}\text{Eu}$, and $^{134}\text{Cs}$ are major contributing nuclides, with the sum of these nuclides accounting for 96% of the total dose rate. Note that $^{137m}\text{Ba}$ is the radiative equilibrium nuclide of $^{137}\text{Cs}$.

Table 2: Major nuclides contributing to the dose rate in the case of a point source in air.

| Nuclide | Dose rate $[\mu\text{Sv/h}]$ | Ratio [%] |
|---------|-----------------------------|-----------|
| $^{137m}\text{Ba}$ | $3.52E+07$ | 67 |
| $^{154}\text{Eu}$ | $1.07E+07$ | 20 |
| $^{134}\text{Cs}$ | $4.47E+06$ | 9 |
| $^{125}\text{Sb}$ | $8.34E+05$ | 2 |
| $^{106}\text{Rh}$ | $4.15E+05$ | 1 |

2.3. Parameters of dose rate response function $R_{i}$

The function $R_{i}$ is defined as the surface dose rate per unit photon source of the nuclide $i$ in the fuel debris. The fuel debris can contain numerous elements from melted materials such as fuel (U, Pu, and O), zirconium alloy (Zr and Sn), stainless steel (Fe, Cr, Ni, and Mn), neutron absorbers (B, C, and Gd), and concrete (Si, Al, Fe, Ca, Mg, O, and H). Since a large amount of seawater was poured into the reactor vessel to eliminate the decay heat at the time of the accident, a certain amount of salt (NaCl) can also be contained in the fuel debris. If we con-
consider the concentrations of these elements as parameters for the $R_i$ function, the expression of the $R_i$ function would be proven difficult to obtain. As such, we aimed to reduce the number of parameters as far as possible.

Figure 1 shows the photon energy dependences of the mass attenuation coefficients for the constituent elements of the fuel debris, except for tungsten (W) and lead (Pb), which are shown as typical heavy elements for comparative purposes. As shown in Fig. 1, the values of the mass attenuation coefficients are approximately the same in the energy range from several hundred kiloelectronvolts to several megaelectronvolts, except for the heavy elements such as U, W, and Pb. In this energy region, the Compton scattering effect is dominant. Meanwhile, in the low-energy region, where the photoelectric effect dominates, the difference in the mass attenuation coefficients is large. However, as shown in Fig. 2, the dose rate conversion coefficient becomes smaller in the low-energy region. Moreover, a cross section of the Compton scattering is proportional to an atomic weight $Z$ with the same number of extra-nuclear electrons. Thus, it was considered that the $R_i$ function could be expressed as a function of the electron number density, except in the case of heavy elements such as U or plutonium (Pu). In other words, the $R_i$ function can be expressed by the U concentration ($u$) and the electron number density ($N_e$), without any consideration of the individual concentration of other elements. Although the 1F fuel debris may contain a little Pu, its effect is expected to be approximately equal from the perspective of photon transport. As such, the $R_i$ function is assumed to be approximated according to radius $r$, U concentration $u$, and electron number density $N_e$.

![Figure 1: Mass attenuation coefficients dependent on photon energy. Since hydrogen (H) has no neutrons in an atomic nucleus, the value of the mass attenuation coefficient was divided by 2.0.](image)
2.4. Photon transport simulation

Assuming that the unit photon source of the \(i\)-th nuclide distributed in the fuel debris is homogeneous, the \(R_i\) value can be directly obtained using the photon transport calculation as a dose rate near the surface of a sphere of fuel debris in the air. The photon transport calculations were conducted for many cases with different radii and elemental compositions to identify the concrete expression of the \(R_i\) function. For these calculations, we used the PHITS [2] based on the Monte Carlo continuous energy method. Following this, the results were arranged for each parameter, and the \(R_i\) function was set using the curve fitting to the quadratic polynomial on \(u\) and \(N_z\) as in Eq. (7).

\[
R_i(r, u, N_z) = \left\{ b_0^i(r) + b_1^i(r)u + b_2^i(r)u^2 \right\}
+ \left\{ c_0^i(r) + c_1^i(r)u + c_2^i(r)u^2 \right\} N_z
+ \left\{ d_0^i(r) + d_1^i(r)u + d_2^i(r)u^2 \right\} N_z^2
\]  

(7)

where \(N_z\) denotes the electron number density [electrons/cm\(^3\)] in the fuel debris, and \(b_0^i, c_0^i, d_0^i\) are fitting coefficients based on the least squares method. The electron number density \(N_z\) can be defined as follows:
where \( j \) denotes the index of the elements in the fuel debris, \( L \) denotes the total number of elements in the fuel debris, \( Z_j \) denotes the atomic number of the \( j \)-th element, \( M_j \) denotes the atomic weight of the \( j \)-th element, \( w_j \) denotes the concentration [wt\%] of the \( j \)-th element in fuel debris, and \( N_A \) denotes the Avogadro number. As a sample of the results, the \( R_i \) values obtained via PHITS for the cases where \( i = ^{137}\text{Ba} \) and \( r = 5.0 \text{ cm} \) are shown in Fig.3a–d, where the fitting functions obtained via Eq. (7) are also shown by the smooth lines. The U concentration \( u \) was (a) 0 wt\%, (b) 35 wt\%, (c) 58 wt\%, and (d) 88 wt\%. Note that \( u = 88 \text{ wt\%} \) corresponds to 100 wt\% of \( \text{UO}_2 \) and that \( u = 100 \text{ wt\%} \) equates to pure U metal. Since the \( (\text{U}, \text{Zr})\text{O}_2 \) is considered to be one of the typical chemical forms of the fuel debris, we believe that 88 wt\% can be taken as a maximum \( u \) value.

From Fig.3a–d, the PHITS results agree well with the fitted \( R_i \) function given by Eq. (7) in the case of \( u = 0 \text{ wt\%} \). However, in the case of higher concentrations of U, there was a tendency to deviate from Eq. (7). A similar tendency was observed in the results for the cases with different radii and photon sources \((i)\).

![Figure 3a: Results of \( R_{^{137}\text{Ba}} \) via PHITS (markers) and fitting function (solid line) for the case where \( u = 0 \text{ wt\%}, r = 5 \text{ cm} \)](image_url)
Figure 3b: Results of $R_{\text{Ba137m}}$ via PHITS (markers) and fitting function (solid line) for the case where $\nu = 35$ wt\%, $r = 5$ cm

Figure 3c: Results of $R_{\text{Ba137m}}$ via PHITS (markers) and fitting function (solid line) for the case where $\nu = 58$ wt\%, $r = 5$ cm
2.5. Error estimation for dose rate due to the $R_i$ function fitting

We had to consider the $P_i$ function as a weighting to estimate the error of the dose rate due to the $R_i$ function fitting, given that the final dose rate $D$ was calculated via Eqs. (2), (5), and (7) and that the values are listed in Table 1. Thus, the dose rate error in this method was estimated by comparing the dose rates via direct PHITS calculations for 138 cases with different conditions within the probable ranges of fuel debris: $r \leq 5$, $u \leq 88$, $\rho \leq 11$, and $N_z \leq 2.64 \times 10^{24}$.

Figure 4a–b shows the results, where the relative standard deviations are plotted depending on the radius and U concentration, respectively. Here, the standard deviations increased as the fuel debris radius and U concentration increased. This is because the self-shielding effect of the photons in fuel debris became larger, whereas the quadratic fitting accuracy of the $R_i$ function became poor. Although improvements can be expected by introducing more complex functional forms or table interpolations, given that the fuel debris composition is currently unclear, a maximum error of approximately 8% ($1\sigma$) is considered acceptable.

In the range of $0 \leq r < 1$ in Fig.4a, the relative standard deviation was not evaluated because of no PHITS calculations in this range. However, it is expected that the error due to the $R_i$ function becomes smaller when the radius is close to 0 cm, because the radiation source of the fuel debris becomes like a point source and the self-shielding effect disappears.
Figure 4a: Debris radius dependence of relative standard deviation (1σ)
for the dose rate prediction accuracy by this method

Figure 4b: U concentration dependence of relative standard deviation (1σ)
for the dose rate prediction accuracy by this method

3. Prediction of dose rates for two types of fuel debris

Although the actual characteristics of the 1F fuel debris remained unclear at this point, as a
demonstration, we applied this method to each of the two types of fuel debris shown in Table
3. One is molten debris in the form of \((\text{U}_{0.5}\text{Zr}_{0.5})\text{O}_2\), and the other is a form of molten core–
concrete interaction (MCCI) product. Features such as the bulk densities shown in Table 3
were assumed on the basis of the fuel debris observed in the TMI-2 reactor [5] and the MCCI
The dose rate of the molten debris (Fig.5a) had higher values than those of the MCCI product (Fig.5b) across all radii. This can be attributed to the assumption that dominant nuclides accompany U in fuel debris and that the U concentration is higher in molten debris. Moreover, as a larger radius, the dose rate of the molten debris tended to saturate more than that of the MCCI product. This is because the higher bulk density and electron number density provide a greater self-shielding effect. Note that the value of the dose rate is highly sensitive to not only the parameters of the \( R_i \) function but also to other parameters related to the \( P_i \) function, such as the burnup degree and the Cs residue ratio.

Table 3: Assumed characteristics of two types of fuel debris

| Fuel debris type | Molten debris | MCCI products |
|------------------|---------------|---------------|
| Burnup degree [Gwd/t] in Unit 2 | 23.1 | 23.1 |
| Elapsed time from 1F accident [years] | 10 | 10 |
| Radius \( r \) [cm] | 1~5 | 1~5 |
| Bulk density \( \rho \) [g/cm\(^3\)] | 6.28 | 2.69 |
| Uranium concentration \( u \) [wt\%] | 57.92 | 7.88 |
| Electron number density \( N_z \) [electrons/cm\(^3\)] | 1.58E+24 | 7.82E+23 |

Elemental composition

| Element | Concentration [wt\%] |
|---------|---------------------|
| B       | 0.00                |
| O       | 16.69               |
| Na      | 0.00                |
| Mg      | 0.00                |
| Al      | 0.00                |
| Si      | 0.00                |
| K       | 0.00                |
| Ca      | 0.00                |
| Cr      | 0.00                |
| Fe      | 0.00                |
| Zr      | 25.39               |
| La      | 0.00                |
| Ce      | 0.00                |
| U       | 57.92               |

88
Figure 5a: Predicted surface dose rate and nuclide-wise contribution via this method for (U, Zr)O₂ debris ($u = 58$ wt%, $\rho = 6.3$ g/cm³, $N_z = 1.58 \times 10^{24}$ electrons/cm³)

Figure 5b: Predicted surface dose rate and nuclide-wise contribution via this method for an MCCI product ($u = 7.9$ wt%, $\rho = 2.7$ g/cm³, $N_z = 7.82 \times 10^{23}$ electrons/cm³)
4. Conclusions

From this study we can conclude the following:

1) The major FPs that will contribute to the dose rate around the fuel debris in 2021 are considered to be $^{137}$mBa (radiation equilibrium daughter of $^{137}$Cs), $^{154}$Eu, $^{134}$Cs, $^{125}$Sb, and $^{106}$Rh.

2) From the various gamma-ray transport simulations, we found that the self-shielding effect of finite-sized fuel debris that comprised various elements can be expressed according to the U concentration and the electron number density of other elements.

3) We elucidated that the following are the dominant parameters for the dose rate of sampled fuel debris: fuel debris radius ($r$), bulk density ($\rho$), U concentration ($u$), burnup degree ($B$), elapsed time ($t$) from the 1F accident, $^{137}$Cs residue ratio in fuel debris ($f_{cs137}$), and electron number density ($N_e$).

4) Finally, we obtained a simple analytical formula for predicting the dose rate near the surface of the 1F fuel debris. This will help enhance our understanding of the characteristics of various fuel debris and estimate the sensitivity of the parameters to the dose rate.

We believe that the above knowledge will be elucidated when applied to the chemical analyses of retrieved fuel debris conducted in the near future.

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