Development of Evaluation Method of Uncertainty of Radioactivity by Propagating Nuclear Data Covariance for Clearance Verification in Decommissioning of Nuclear Power Plants

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To optimize the disposal of low-level radioactive wastes originating from the decommissioning of nuclear facilities, required are 1) a reliable assessment of the radioactivity level by calculations and measurements and 2) a rational estimate of uncertainties of those results for the classification of the radioactive waste. We established a procedure to estimate uncertainties of a radioactivity concentration required in the clearance verification by evaluating and propagating covariance of nuclear data properly. For this aim, we estimated covariance of neutron cross sections of nuclides that account for over 90 % in the sum of D/C of concrete materials and carbon steels. Here, D stands for the radioactivity concentration, and C stands for the clearance level. The covariance of nuclear data was estimated by employing a propagation of uncertainties of nuclear model parameters in the resonance and smooth regions by a combination of nuclear model codes implemented in T6. Then, we developed a new method to calculate uncertainty of the radioactivity by "Total Monte Carlo" method by connecting randomly perturbed ENDF-6 format files generated by T6 to NJOY2016, and random 1-group activation cross sections were inserted to ORIGEN2 in ORLIBJ40. It was concluded that the uncertainty of the radioactivity due to that of nuclear data for nuclides which dominate the ΣD/C is of the order of 0.12 to 1.2%, which is sufficiently smaller than the uncertainty of the radioactivity originating from that of the neutron flux.

1. Background and Purpose

At present, more than 20 nuclear power plants are either in the process of decommission or planned to be decommissioned in near future in Japan. Therefore, decommissioning will become an important activity in nuclear industry, so the procedure must be standardized. Decommissioning of nuclear power plants produces huge amount of low-level radioactive waste. Radioactive wastes with extremely low radioactivity concentrations and therefore having negligible impact to general public are referred to as clearance wastes. They can be reused and disposed of in the same way as general industrial waste with the confirmation and permission of regulation authority. Promotion of clearance reduces the environmental burdens and hence accomplishes streamlining and facilitating decommissioning in a proper way.

To be classified as clearance wastes, ΣD/C, sum of D/C by each specified nucleus, must be lower than 1. Here, D stands for radioactivity concentration, and C stands for clearance level. In near future, Nuclear Regulation Authority will stipulate that the uncertainty of radioactivity concentration must be considered as a part of clearance procedure [1]. Therefore, it is required to establish a method to evaluate the radioactivity concentration having rational uncertainty.

Uncertainty of radioactivity concentration depends on uncertainty of three factors, (1) nuclide composition of material, (2) neutron irradiation condition and (3) nuclear data. Uncertainty of nuclear data, namely, covariance data, was not considered in previous clearance studies. On the other hand, while nuclear data library JENDL-4.0 that is frequently used for radioactivation calculation in Japan has nuclear data for 406 nuclides, only 95 of them have covariance data. Evaluation of uncertainty of radioactivity cannot be performed correctly without covariance data. For example, it is known that simple square sum propagation of variance data gives underestimation of the uncertainty when the off-diagonal elements of the covariance matrix are positive, while it gives overestimation when the off-diagonal elements are negative. Accurate estimation of the uncertainty of the radioactivity is possible only when the full covariance matrix is taken into consideration. For this reason, considering covariance data in uncertainty evaluation is essentially important.

The purpose of this study is to establish a method to estimate the covariance data for nuclides without covariance data in JENDL-4.0, and also to construct a method to quantitatively evaluate the effect of uncertainty of nuclear data in the calculation of radioactivity by connecting to radioactivation calculation.

2. Developed evaluation method

As a method to quantitatively evaluate the influence of uncertainty in nuclear data, we developed an evaluation method using the TMC (Total Monte Carlo) method, which incorporates calculations by the nuclear data evaluation code group T6 [2]. Figure 1 shows our evaluation method.

T6 consists of 6 codes (TASMAN, TALYS, TARES, TEFAL, TANES, TAFIS) and produces nuclear data library. In this study, we used 4 codes in T6, TALYS, TARES, TASMAN and TEFAL (Table 1). T6 calculation generates perturbed cross sections in ENDF-6 format file. These files are denoted as random files. TMC calculation is the process of performing radioactivation calculation using ORLIBJ40 with these random files as input, and then statistically processing the result.

As preparation of T6 calculation, (1) reproduction of cross section in JENDL-4.0 and (2) adjustment of distributions of model parameters for random calculation are needed. For (1), resonance parameters in JENDL-4.0 were used to calculate cross sections in the resonance region with TARES and parameters including optical model parameters are adjusted manually to calculate cross sections in the continuous region with TALYS. For (2), uncertainties of resonance parameters evaluated by Mughabghab [3, 4] were employed in the resonance region. Uncertainties of negative resonances were adjusted to reproduce uncertainty of the thermal cross section. On the other hand, TALYS improves parameter distributions for each random calculation based on Bayesian Monte Carlo (BMC) method. The posterior distribution obtained by the BMC method starting from a uniform distribution given as the prior distribution was used as the parameter distribution in the main calculation. After configurating the above setting, 1,000 random files were produced by 1,000 random calculations as the main calculation.

In the TMC calculation, the random files were processed by NJOY2016 and then effective one-group cross section was generated for each random file using neutron flux data. Then, it was incorporated into the one-group cross section library set ORLIBJ40, and ORIGEN2 was used to calculate the radioactivity. This procedure was repeated for the number of random files, and the results were statistically processed to obtain the mean value and standard deviation.

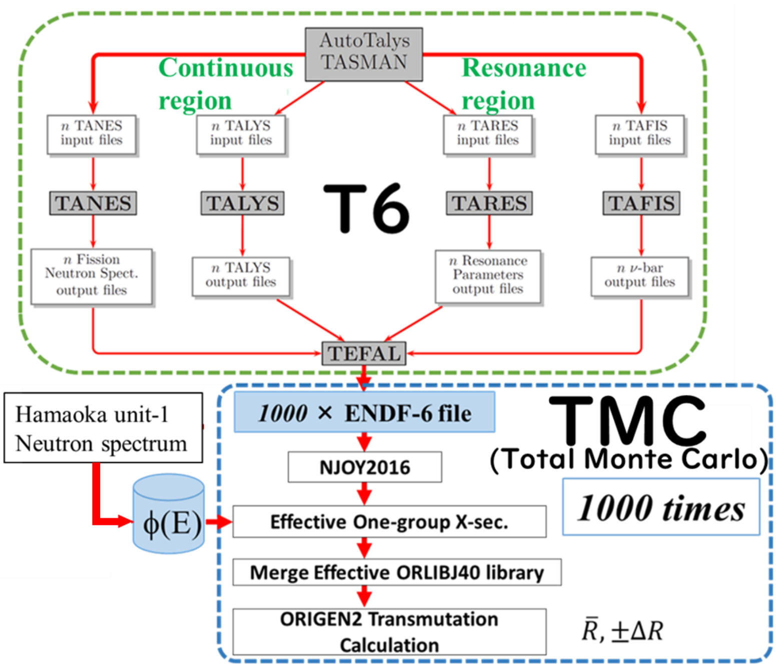


Figure 1 Overview of developed evaluation method

Table 1 Codes in T6 used in this study

TALYS	a nuclear reaction code to calculate cross sections in the continuous region.
TARES	a code to generate resonance cross sections.
TASMAN	a code for production of covariance data using result of TALYS and TARES.
TEFAL	a code for the translation of the nuclear reaction results from TALYS, TARES and TASMAN into ENDF-6 formatted nuclear data libraries.

3. Calculation Result of Covariance with T6

The nuclides to be evaluated were selected by performing calculation of radioactivity by ORLIBJ40. The assumed plant was Hamaoka unit-1. Evaluation position was located at 30cm depth from the surface of the concrete of the RSW (Reactor Shielding Wall) at the center height of the reactor core. Materials were general concrete material (density = 2.3 g/cm^3) and carbon steel of primary containment vessel (PCV) (density = 7.86 g/cm^3). Material composition data and neutron flux data ware provided from Chubu Electric Power Co., Inc. Figure 2 shows neutron flux at the evaluation point. Effective full power year (EFPY) is 16.2 years. As a result of activation calculation, ^{60}Co and ^{152}Eu were found to be the dominant radionuclides. Therefore, their parent nuclides, ^{59}Co and ^{151}Eu, were selected as the nuclides to be evaluated.

Random calculations were carried out by reproducing the cross sections of JENDL-4.0 for the evaluated nuclides and adjusting the parameter distribution according to the procedure shown in Section

2. The top graph in figure 3 shows perturbed ^{151}Eu(n, γ)^{152}Eu cross sections generated T6. The bottom graph in Figure 3 shows the relative standard deviation when the generated cross sections were grouped by the VITAMIN-B6 structure and statistically processed.

Figure 4 shows correlation matrix of (n, γ) cross section of ^{151}Eu. In 1/*v* region, correlation is strongly positive since the energy-dependence of the cross section is fixed. On the other hand, correlation is weak and complex in resonance region. Correlation between continuous and resonance regions is zero because TALYS and TARES are independent calculations of each other. Similarly, we also calculated correlations between different reactions for calculations, including neutron transport calculation, other than this study.

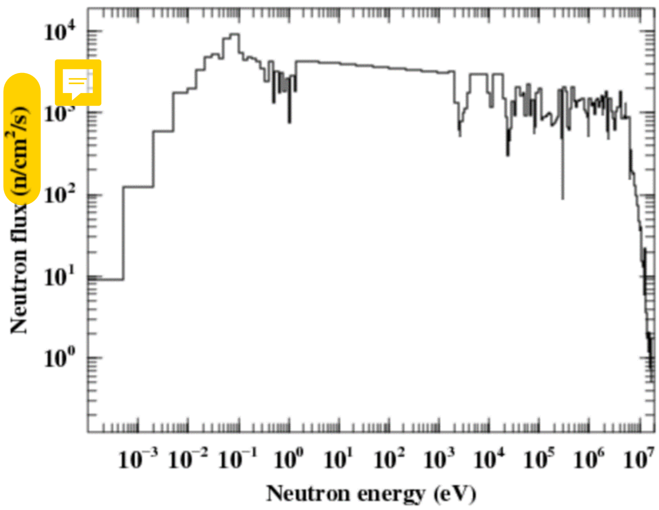


Figure 2 Neutron flux in the concrete 30 cm deep from the surface of the RSW at the center height of the reactor core (VITAMIN-B6 structure)

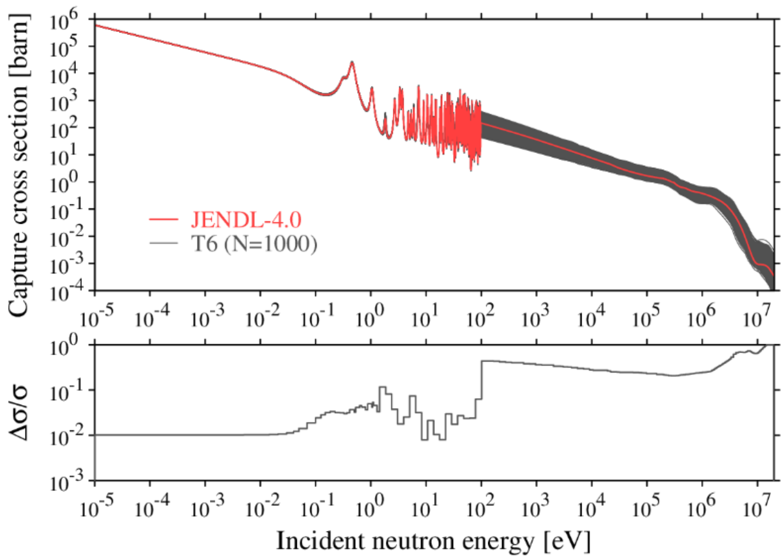


Figure 3 Perturbed ^{151}Eu(n, γ)^{152}Eu cross sections generated by 1000 times T6 calculation and relative standard deviation

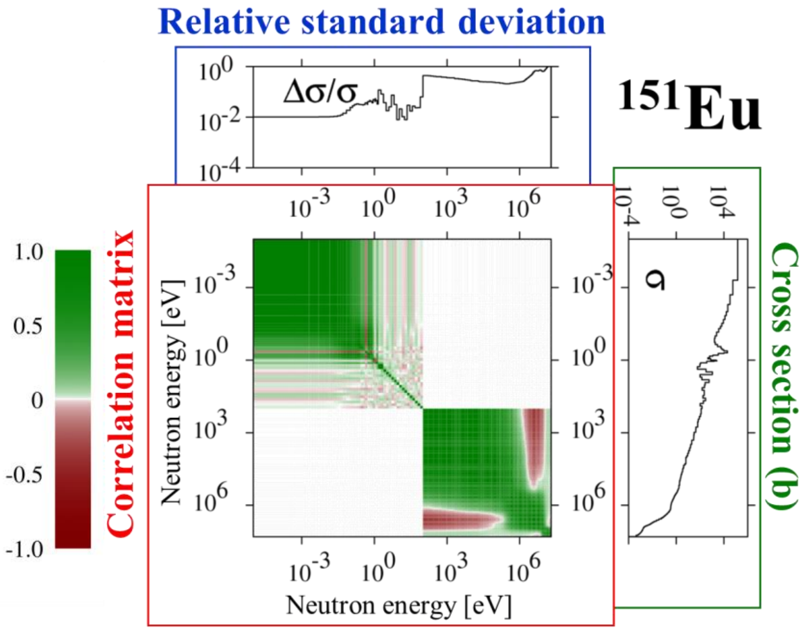
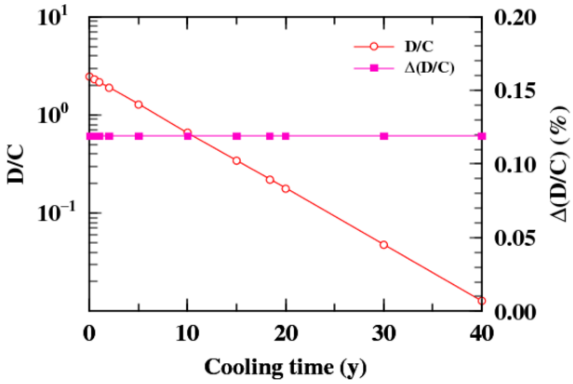


Figure 4 Correlation matrix of (n, γ) cross section of ^{151}Eu calculated with T6

4. Evaluation of Uncertainty of Radioactivity

We calculated D/C uncertainty due to nuclear data by TMC calculation. Figure 5 shows calculated D/C and relative standard deviation of that of dominant nuclides in concrete. From the result, Δ(D/C) of ^{60}Co is 0.12 % and that of ^{152}Eu is 1.2 %. Figure 6 shows similar calculation result in carbon steel (PCV). From the result, Δ(D/C) of ^{60}Co is 0.12 %. These results are smaller than uncertainty due to neutron flux data, which is dozens % [5]. Therefore, it can be concluded that the uncertainty due to nuclear data is not major factor in the uncertainty of the radioactivity.



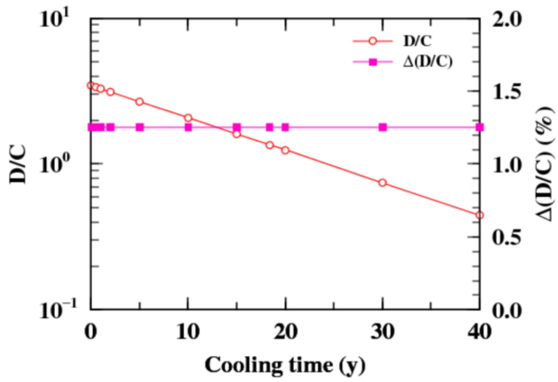


Figure 5 D/C and relative standard deviation of D/C (Δ(D/C)) of dominant nuclides in concrete (left: ^{60}Co, right: ^{152}Eu)

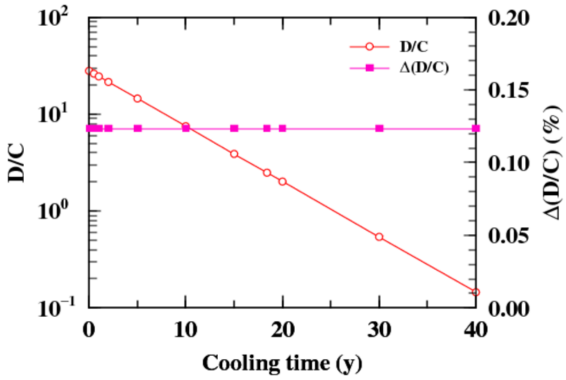


Figure 6 D/C and Δ(D/C) of dominant nuclide, ^{60}Co, in carbon steel

5. Conclusion

In order to contribute to the rationalization of the disposal of dismantled waste generated in the decommissioning of nuclear power plants, we developed quantitative evaluation method of the uncertainty in radioactivity due to uncertainty of nuclear data. Then, we applied our evaluation method to Hamaoka unit-1 and evaluated the uncertainty of D/C of important nuclides in clearance verification, ^{60}Co and ^{152}Eu.

Our evaluation method consists of T6 calculation and TMC calculation. T6 calculation generates perturbed cross sections in ENDF-6 format file for data that reproduces JENDL-4.0. TMC calculation is the process of performing activation calculation using ORLIBJ40 with generated cross sections as input, and then statistically processing the result.

We applied developed method to Hamaoka unit-1 and calculated uncertainty of D/C of dominant nuclides in concrete material and carbon steel. From the result, uncertainty of ^{60}Co due to nuclear data is 0.12 % and that of ^{152}Eu is 1.2 %. They are smaller than uncertainty due to neutron flux data, which is dozens %. Therefore, it can be concluded that the uncertainty due to nuclear data is not major factor in the uncertainty of the radioactivity.

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