Radioisotope Assessment of CANDU Spent Fuel Packages through Least Square Fitted Scaling Factors

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1. Introduction

The CANDU reactor emits lots of spent nuclear fuels via online operation with an average burnup of 7,000 MWD/MTU. Usually, radioactive waste drums store active materials including fission product of spent fuel or activation products. In the previous study, the least square fitted scaling factor method was proposed in order to quantify difficultly measuring (DTM) radioactive isotopes by using the key (KEY) isotopes in the PWR waste drum.[1] The scaling factor is defined as the relative inventory ratio based on the key isotopes such as Co-60 Cs-137, and Ce-144.[2][3] In CANDU reactor, the situation is similar to that of PWR and activation products(Co-58, Co-60, Fe-55, Nb-94, Ni-59, Ni-63), fission products(H-3, C-14, I-129, Ce-144, Sr-90, Tc-99, Cs-137), and actinides must be clearly quantified before when a radioactive waste drum is stored in the storage site.

In this paper, a simplified simulation test for scaling factors is carried out by using the ORIGEN-S code[3] for CANDU reactor. Fuel burnup is a main variable and explicit formula for scaling factors as a function of burnup is obtained by the generalized least square fitting (LSF) method.[5] After obtaining scaling factors from the LSF method, the decay effects are also implemented by multiplying exponential decay term including decay constant of each isotope. The CANDU results of scaling factors are compared with those of PWR simulation results, too.

2. Simulation Conditions for Scaling Factor for CANDU Reactor

For the application of the general scaling factor, a typical CANDU fuel assembly library in the ORIGEN-S code is used such as 'candu37'. The fuel burnp varies from 7,400 MWD/MTU to 7,800 MWD/MTU with 100 MWD/MTU step. The cooling time is given up to 10 years. One metric ton uranium is loaded and 30 kg stainless steel is added for considering the activation of structural materials. The element composition in ORIGEN-S calculation is given in Table I.

Table I: Element Composition for Simulation of Scaling Factor

Element	U	Fe	Cr	Ni
Weight (kg)	1,000	20.64	5.7	2.67

The total number of target isotopes is 12 and a group of alpha emission isotopes including about 35 actinide isotopes described in the IAEA guide.[6] From the results of ORIGEN-S, the inventories of 47 isotopes are evaluated as a function of decay time. Figs. 1 and 2 show scaling factors of various isotopes based on the Cs-137 and Co-60, respectively, when the cooling time is 10 years. After 7,600 MWD/MTU burnup, the activities of Cs-137 and Co-60 are estimated with 10 years cooling as 7.46E+14 Bq, 1.23E+11 Bq, respectively. Due to low range of burnup, the scaling factors exhibit almost constant. Depending on the characteristics of isotopes, the trends of some isotopes are quite different as a function of decay time as shown in Fig.3. In Fig.3, the scaling factor Sr-90 (half-life=28.8 years) of Co-60 decreases as burnup increases. But due to the relatively short half-life of Co-60 (half-life= 5.27years), Sr-90 decays slow into Y-90 and Zr-90. Thus the scaling factor of Sr-90 increases as a function of cooling time.

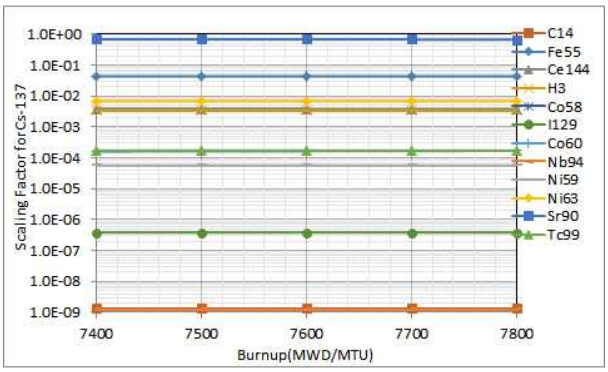

Fig. 1. Scaling factor change based on the Cs-137

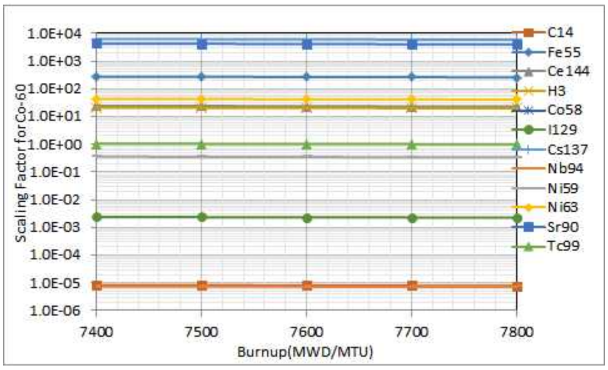

Fig. 2. Scaling factor change based on the Co-60

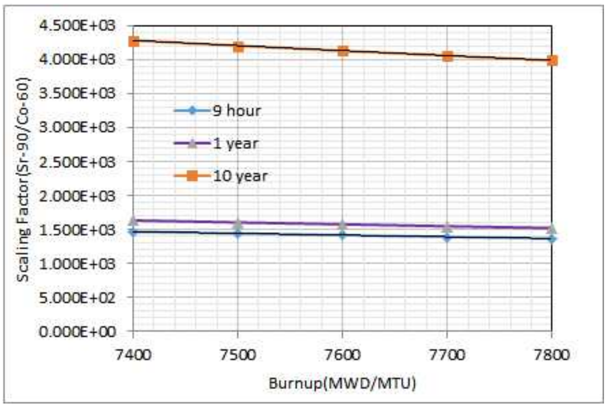

Fig. 3. Scaling factor of Sr-90 based on the Co-60

3. Least Square Fitted Scaling Factors

The scaling factors of specified isotopes are obtained as a function of burnup for CANDU spent fuel. Three decay times of 9hr, 1 year and 10 years are chosen and the behavior of scaling factor is quite different as shown in Fig. 3.

Then by using of a generalized least square fitting method, the scaling factor can be expressed in the linear form as follows.

$$\begin{aligned} y &= (A_1 \pm \sigma_{A_1})x + (A_0 \pm \sigma_{A_0}) \\ &= A_1 x + A_0 \pm (\sigma_{A_1} x + \sigma_{A_0}) \end{aligned} \quad (1)$$

where x is burnup and y is scaling factor. When compared the previous study in PWR case, the linear form is more adequate due to small variations of scaling factors.

From the data, a least square fitting in the first order polynomial can be applied such as

$$\begin{pmatrix} 1 & x_0 \\ 1 & x_1 \\ \vdots & \vdots \\ 1 & x_n \end{pmatrix} \begin{pmatrix} A_0 \\ A_1 \end{pmatrix} = \begin{pmatrix} y_0 \\ y_1 \\ \vdots \\ y_n \end{pmatrix} \quad (2)$$

where x_i is a i-th burnup, A_j is a coefficient of x^j polynomial to be determined, and y_i is a scaling factor. The equation is written as in the matrix form as follows

$$XA = Y \quad (3)$$

Equation (3) is solved easily with the standard least square fitting by multiplying the transpose matrix of X on both terms.

$$X^T XA = X^T Y \quad (4)$$

Then the coefficients are obtained as

$$A = \left(X^T X\right)^{-1} X^T Y \quad (5)$$

and their variances are also obtained as

$$Var(A_j) \approx \frac{R}{n-m}\left(X^T X\right)^{-1}_{jj} \quad (6)$$

In this case, the coefficients and their errors are easily obtained and are given in Table II.

Table II: Coefficients of Least Square Fitted Scaling Factor for Sr-90 based on Co-60 after 10 years cooling

A1	-9.50E-01	σ(A1)	1.27E-01
A0	1.14E+04	σ(A0)	9.69E+02

Additionally, a decay time is corrected simply by multiplying exponential decay term as follows

$$f(B,t) = \frac{A_i e^{-\lambda_i t}}{A_k e^{-\lambda_k t}} = (A_i / A_k) e^{-(\lambda_i - \lambda_k)t} \approx f(B) e^{-(\lambda_i - \lambda_k)t} \quad (7)$$

where t is decay time and λ_i and λ_k are decay constants for DTM and KEY istotopes, respectively.

Fig. 4 depicts the least square fitted scaling factor of Sr-90 as a function of burnup by comparing scaling factors evaluated directly by the ORIGEN-S. The difference of two scaling factors is not significant and the fitted scaling factors predicts well enough. Table III shows fitted scaling factors of various isotopes based on the Co-60 with the burnup of 7,550 MWD/MTU, and 10 year decay time is given.

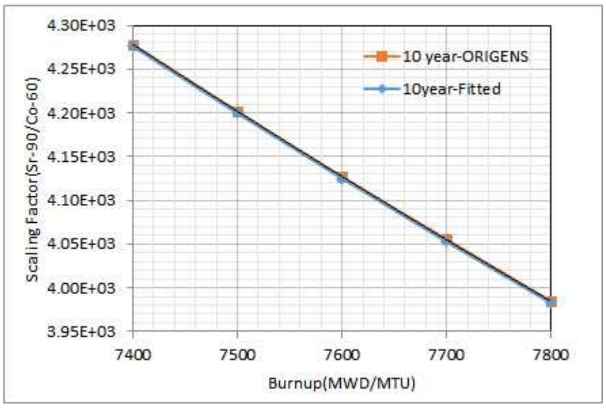

Fig. 4. Comparison of least square fitted scaling factors of Sr-90 as a function of burnup.

Table III: Least Square Fitted Scaling Factor based on Co-60 of 7,550 MWD/MTU and 10 year cooling time.

Isotope	Scaling Factor	Error
C14	8.16E-06	1.55E-07
Ce144	2.40E+01	6.06E-01
Co58	1.34E-13	8.92E-15

Fe55	2.69E+02	6.06E+00
H3	2.05E+01	5.18E-01
I129	2.36E-03	4.63E-05
Nb94	7.43E-06	7.97E-08
Ni59	3.48E-01	8.02E-03
Ni63	4.26E+01	9.22E-01
Sr90	4.20E+03	1.93E+03
Tc99	1.04E+00	1.52E-02

4. Conclusions

By using the ORIGEN-S code, scaling factors of CANDU spent fuels and their contaminated wastes are evaluated with a function of burnup through the first order least square fitting method. The fitted results are confirmed by comparing with the direct results of the ORIGEN-S and their errors are provided by utilizing the least square fitted results. By considering the previous approaches for PWR spent fuel case, it requires various different initial conditions such as different fuel type, enrichment, fuel burnup, and cooling time in order to apply accurately scaling factors of various isotopes. In addition, the waste treatment procedure should be also taken into consideration to explain isotopic different behaviors. In order to validate the measured data for the radioactive waste drum, the least square fitting approach is expected to provide fruitful and valuable contributions.

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