# Development of Inverse Estimation Program of Burnup Histories for Nuclear Spent Fuel Based on ORIGEN-S

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## 1. Introduction

There have been increasing possibilities that international terrorist groups could use nuclear materials for their purposes rather than nuclear bomb because the nuclear are difficult to be manufactured by small group of terrorists. So, the ability to identify the perpetrators of some attack by nuclear materials like spent nuclear fuels and the origin of the nuclear materials is critical and the methodology which can find the burnup history and characteristics of the original nuclear materials based on the analysis of the post-event materials has been considered as a necessary and effective tool for international nuclear safeguards.

The purpose of this work is to develop a computer program which can accurately estimate burnup histories of spent fuels based on the environmental sample measurements. The burnup histories of spent fuels include initial uranium enrichment, discharge burnup, cooling time after discharge, and nuclear reactor type in which the spent fuel was burnt. The methodologies employed in our program are based on the formulations developed by M. R. Scott[1] but we developed a stable bi-section method to correct initial uranium enrichment and used a simplified algorithm without burnup correction. Also, ORIGEN-S[2] rather than ORIGEN-2[3] was used in our program to improve the accuracies by using the new capabilities of burnup dependent cross section libraries of ORIGEN-S. Our program is applied to several benchmark problems including realistic Mihama-3 problems to test the accuracies.

## 2. Methods and Results

### *2.1 Theory and Methodologies*

The equation which can give the estimation of burnup of spent fuel is derived by considering balance on the number of atoms and the amount of the generated energy. The following equation gives the balance relation[1] :

$$N_0^U = F + N^{U235}(T) + N^{U236}(T) + N^{U238}(T) + N^{PU238}(T) + N^{PU239}(T) + ....., \quad (1)$$

where $T$ is the measurement time of the spent fuel sample, $F$ is the number density of accumulated fissions, $N_0^U$ is the number density of the initial uranium atoms, and $N^X(T)$ the number density of atoms of the nuclide $X$ at the measurement time $T$. The definition of the burnup gives the following simple relation between burnup and the fission number density :

$$F = \frac{\rho_o^U}{E_R} BU(T), \quad (2)$$

where $\rho_0^U$ is the initial heavy metal density and $E_R$ is the average released energy per fission. The substitution of Eq.(2) into Eq.(1) gives

$$N_0^U = N^U(T) + N^{PU238}(T) + N^{PU239}(T) + N^{PU240}(T) + ..... + \frac{N_0^U M_U}{N_a E_R} BU(T), \quad (3)$$

where $N_a$ and $M_U$ are the Avogadro's number and atomic weight of uranium, respectively. The division of Eq.(3) by $N^{U238}(T)$ gives the equation relating the measured quantities with burnup as follows :

$$\frac{N_0^U}{N^{U238}(T)} = \frac{\frac{N^U(T)}{N^{U238}(T)} + \frac{N^{PU238}(T)}{N^{U238}(T)} + \frac{N^{PU298}(T)}{N^{U238}(T)} + ...}{1 - \frac{M_U}{N_a E_R} BU(T)}. \quad (4)$$

In this equation, it is noted that all the quantities in the numerator are known from the measurements but the quantity in the left hand side is not measurable quantity. So, we need one additional equation to determine the burnup. Before deriving the additional equation, we have to consider the monitor nuclides for burnup estimation. Actually, any fission product produced directly proportional to the burnup can be used as a burnup monitor and it has been known that burnup can be measured within a one percent accuracy coupled with mass spectrometry. But for our problems, the reactor type is not given before the problem is solved and so the burnup monitors which produced at the same rate regardless of reactor type should be chosen. Also, a constant fission yield across reactor types and

long half-life are desirable for burnup monitor, which simplifies the formulation. The additional equation for the estimation of the burnup is derived by considering the following simple relation for a burnup monitor fission product $N^B$:

$$\frac{dN^B}{dt} = Y_B[N^{U235}(t)\bar{\sigma}_f^{U235}\phi(t) + N^{PU239}(t)\bar{\sigma}_f^{PU239}\phi(t) + ...] \quad (5)$$

where $Y_B$ means the fission product yield of the burnup monitor $N^B$. In Eq.(5), the radioactive decay of the burnup monitor is neglected by considering its long half-live and also the spectrum change of the core over time is neglected for simplicity. The time integration of Eq.(5) from 0 to irradiation time gives

$$\begin{aligned} N^B(T) &= Y_B\int_0^T dt[N^{U235}(t)\bar{\sigma}_f^{U235}\phi(t) + N^{PU239}(t)\bar{\sigma}_f^{PU239}\phi(t) + ...] \\ &= Y_B F = \frac{\rho_0^U}{E_R} Y_B BU(T) \end{aligned} \quad (6)$$

Division of Eq.(6) by the initial uranium atom density gives the additional equation for the burnup estimation given by

$$.BU(T) = \frac{N^B(T)}{N^{U238}(T)} \frac{N^{U238}(T)}{N_0^U} \frac{E_R N_a}{Y_B M_U}. \quad (7)$$

In Eq.(7), the first term in the right hand side is a measured quantity and so, Eq.(4) and Eq.(7) can be used to estimate burnup.

The equation for the estimation of initial uranium enrichment is derived by considering the balance equation on the atom density of initial $^{235}$U. The balance equation is given by

$$\begin{aligned} N_0^{U235} = N^{U235}(T) &+ \bar{\sigma}_f^{U235}\int_0^T N^{U235}(t)\phi(t)dt \\ &+ \bar{\sigma}_\gamma^{U235}\int_0^T N^{U235}(t)\phi(t)dt. \end{aligned} \quad (8)$$

For simplicity, we omitted the detailed procedure of the equation for the initial uranium enrichment estimation which is given by

$$\begin{aligned} e_0 \equiv \frac{N_0^{U235}}{N_0^U} &= \frac{N^{U235}(T)}{N_0^U} + \frac{N^{U236}(T)}{N_0^U} + \frac{\rho_0^U}{E_R N_0^U} BU(T) \\ &- \frac{1}{N_0^U}[\frac{\bar{\sigma}_f^{U238}}{\bar{\sigma}_a^{U238}}[N_0^{U238} - N^{U236}(T)] \\ &+ \frac{\bar{\sigma}_f^{PU239}}{\bar{\sigma}_a^{PU239}}[N_0^{PU239} - N^{PU239}(T) + F^{U238}] \\ &+ \frac{\bar{\sigma}_f^{PU240}}{\bar{\sigma}_a^{PU240}}[N_0^{PU240} - N^{PU240}(T) + F^{PU239}] \\ &+ \frac{\bar{\sigma}_f^{PU241}}{\bar{\sigma}_a^{PU241}}[N_0^{PU241} - N^{PU241}(T) + F^{PU240}]]. \end{aligned} \quad (9)$$

In Eq.(9), the following definitions for $F$ are used :

$$\begin{aligned} F^{U238} &= \frac{\bar{\sigma}_\gamma^{U238}}{\bar{\sigma}_a^{U238}}[N_0^{U238} - N^{U238}(T)], \\ F^{PU239} &= \frac{\bar{\sigma}_\gamma^{PU239}}{\bar{\sigma}_a^{PU239}}[N_0^{PU239} - N^{PU239}(T) + F^{U238}], \\ F^{PU240} &= \frac{\bar{\sigma}_\gamma^{PU240}}{\bar{\sigma}_a^{PU240}}[N_0^{PU240} - N^{PU240}(T) + F^{PU239}], \end{aligned} \quad (10)$$

If we assume that initial plutonium isotope masses are zero, Eq.(9) can be simplified to

$$\begin{aligned} e_0 = \frac{N^{U238}(T)}{N_0^U}[&\frac{N^{U235}(T)}{N^{U238}(T)} + \frac{N^{U236}(T)}{N^{U238}(T)}] \\ &+ \frac{M_0^U}{N_a E_R} BU(T) - G^{U238} - G^{PU239} \\ &- G^{PU240} - G^{PU241}, \end{aligned} \quad (11)$$

where

$$\begin{aligned} G^{U238} &= \frac{\bar{\sigma}_f^{U238}}{\bar{\sigma}_a^{U238}}[1 - e_0 - \frac{N^{U238}(T)}{N_0^U}], \\ G^{PU239} &= \frac{\bar{\sigma}_f^{PU239}}{\bar{\sigma}_a^{PU239}}[-\frac{N^{PU239}(T)}{N^{U238}(T)} \cdot \frac{N^{U238}(T)}{N_0^U} \\ &\quad + G^{U238}\frac{\bar{\sigma}_\gamma^{U238}}{\bar{\sigma}_a^{U238}}\frac{\bar{\sigma}_a^{U238}}{\bar{\sigma}_f^{U238}}], \\ G^{PU240} &= \frac{\bar{\sigma}_f^{PU240}}{\bar{\sigma}_a^{PU240}}[-\frac{N^{PU240}(T)}{N^{U238}(T)} \cdot \frac{N^{U238}(T)}{N_0^U} \\ &\quad + G^{PU239}\frac{\bar{\sigma}_\gamma^{PU239}}{\bar{\sigma}_a^{PU239}}\frac{\bar{\sigma}_a^{PU239}}{\bar{\sigma}_f^{PU239}}], \\ G^{PU241} &= \frac{\bar{\sigma}_f^{PU241}}{\bar{\sigma}_a^{PU241}}[-\frac{N^{PU241}(T)}{N^{U238}(T)} \cdot \frac{N^{U238}(T)}{N_0^U} \\ &\quad + G^{PU240}\frac{\bar{\sigma}_\gamma^{PU240}}{\bar{\sigma}_a^{PU240}}\frac{\bar{\sigma}_a^{PU240}}{\bar{\sigma}_f^{PU240}}] \end{aligned} \quad (12)$$

It is noted that Eq.(12) contains the ratio $N^{U238}(T)/N_0^U$ and this quantity is already determined during the estimation of burnup. Our program uses an iterative algorithm to solve Eq.(11) and Eq.(12), and the algorithm is always rapidly convergent. However, the estimated values of burnup and initial enrichment using Eq.(4), Eq.(7), Eq.(11), and Eq.(12) are

generally not accurate. In particular, the estimated value of initial uranium enrichment is much less accurate than the one of burnup and so the correction of initial uranium enrichment is required to improve the accuracies. For this purpose, in Ref. 2, a correction method coupled with ORIGEN-2 forward calculation was suggested both for burnup and initial uranium enrichment. But ORIGEN-2 is a very old code but the accuracies of burnup calculation by ORIGEN-2 are limited due to use of burnup-independent cross section libraries. In particular, we have experienced that the algorithm given in Ref. 2 to correct burnup can be unstable. In our program, we developed a stable bi-section algorithm to correct the initial uranium enrichment and we employed ORIGEN-S rather than ORIGEN-2 to improve the accuracies because ORIGEN-S provides a variety of new capabilities including the burnup dependent cross section library.

Next, we describe the enrichment correction method by using the bi-section method. The bi-section method starts with the initial estimation of uranium enrichment by setting

$$X = XL = XR = e_0. \tag{13}$$

Then, our program automatically prepares an ORIGEN-S input file by using the initial uranium enrichment and burnup estimated previously. At present, the ORIGEN-S input assumes an initial uranium mass of 1000kg and a specific power of 37.4W/g. Then, our program executes ORIGEN-S to perform depletion calculation and then calculates the following function value by using the results of ORIGEN-S output :

$$f(X) = R_m A_{U238}^{ORIGEN} - A_{U235}^{ORIGEN}. \tag{14}$$

In Eq.(14), $R_m$ is the ratio of the number of $^{235}$U atoms to the number of $^{238}$U atoms obtained from sample measurement. We hope that the function given by Eq.(14) is nearly zero when the enrichment correction is completed because the function $f(X)$ means the difference between the numbers of $^{235}$U atoms obtained from ORIGEN-S calculation and obtained from the measurement ratio $R_m$ multiplied by the ORIGEN-S estimation of the number of $^{238}$U atoms. Then, if the function $f(X)$ is positive, then $XL$ is increased by a specified value $\Delta$ until $f(XL)$ becomes negative. Then, the last $XL$ at which $f$ is negative is set to $XR$. Otherwise, if the function $f(X)$ was negative at first, then $XR$ is decreased by a specified value $\Delta$ until $f(XR)$ becomes positive. Then, the last $XR$ at which $f$ is positive is set to $XL$. Once the initial values of $XL$ and $XR$ at which $f$ s have different signs are determined, the conventional bi-section method is used as follows :

$$\begin{aligned}
&1)\ X = (XL + XR)/2.0,\\
&2)\ if\ \ f(X) < 0.001 \Rightarrow STOP\\
&\quad \text{else}\\
&\qquad if\ \ f(X) * f(XR) \Rightarrow XR = X\\
&\qquad else\ \ XL = X,\\
&\qquad \text{Go To 1)}
\end{aligned} \tag{15}$$

Actually, the above enrichment corrections are done for all of the candidate reactor types.

After the enrichment corrections are completed, the cooling time after discharge is estimated. For this, the users can select the age monitors for age (or cooling time) determination. To get good accuracy of cooling time, the age monitors having similar half-life to cooling time is desirable but the cooling time is unknown. Generally, the age monitors having half-lives of 1year ~ 30years are recommended to be chosen. ORIGEN-S calculations are performed again for each type of reactors and the number densities of the age monitor nuclides at the time of 30 days after discharge are extracted from the ORIGEN-S outputs. And these values are set to $N_{0,i}^C$. If the age monitor nuclides are not produced from decay of the other nuclides, the number densities of the age monitor nuclides at cooling time $T_C$ after discharge is given by

$$\begin{aligned}
&N_i^C(T_C) = N_{0,i}^C \exp(-\lambda_i T_C)\\
&\Rightarrow T_C = -\frac{T_{1/2,i}}{\ln(2)} \ln\left(\frac{N_i^C(T_C)}{N_{0,i}^C}\right).
\end{aligned} \tag{16}$$

where $\lambda_i$ and $T_{1/2,i,}$ are the decay constant and half-life of the i'th age monitor, respectively. To use the measurable quantities, Eq.(16) is changed into

$$T_C = -\frac{T_{1/2,i}}{\ln(2)} \ln\left(\frac{N_i^C(T_C)/N^{U238}(T)}{N_{0,i}^C/N^{U238}(T)}\right). \tag{17}$$

In Eq.(17), it is noted that the denominator is the calculated one from the ORIGEN-S calculations while the numerator is from the measurement. When several numbers of age monitors are used, some monitors can give relatively larger errors in cooling time than the others and so the final cooling time is carefully determined. We first calculate the average of the cooling times for all the age monitors and select two values that are closest to the average value. The average values of these two values of cooling time are finally selected as the cooling times for each of the reactor types.

The estimation of reactor type where the spent fuel was burnt is also done with ORIGEN-S calculations. The monitor nuclides for estimation of reactor type should be chosen such that their depletion

characteristics are distinctly different for different types of reactors. To differentiate the reactor types from each other, isotopes with cross sections and yields their change significantly from reactor type to reactor type are needed. To avoid the complication of decay, stable or long-lived isotopes are needed. The method for determining reactor type depends upon the accuracy of ORIGEN-S, and as the decay chain becomes more complicated the accuracy is diminished. In particular, it is difficult to accurately determine reactor type for the spent fuels having low burnup cases. Specially, the discrimination between PWR and BWR is difficult for low burnup cases. To determine reactor type, ORIGEN-S inputs are prepared for all the reactor type candidates using their previously estimated initial uranium enrichment, burnup, and cooling time. Then, the ORIGEN-S depletion calculations are done for all the inputs mentioned above. After that, the ratios of the atomic densities of the reactor type monitors to that of $^{238}U$ at the estimated measurement times and the differences between these ratios and their measured values are calculated and the reactor type having a minimum difference is selected as the final reactor type.

### *2.2 Applications to Test Problems and Results*

Test and verification of our program consist of two steps. The first step is to test if our program gives the correct estimation of the initial uranium enrichment, burnup, cooling time, and reactor type for the test problems that are made by using ORIGEN-S forward depletion calculations. We set up six test problems for ORIGEN-S forward depletion. Table I summarizes the monitor nuclides used in our tests and verification. We were selected monitor nuclides based on the ones given in Ref. 1. Table II specifies the six test problems and summarizes the test results obtained with our program. As shown in Table II, we considered two PWR test problems (PWR-1, and -2) that have 18MWD/kg and 30MWD/kg burnups, respectively. Their cooling times are 5.0 and 10.0 years while the initial uranium enrichments are 3.0wt% and 4.0wt%, respectively. Two BWR problems[4] of which their enrichments are 2.93wt% have nearly the same burnups of 33.9MWD/kg and 31.04MWD/kg, respectively while they have the same cooling time of 5.35 years. The last two CANDU problems use natural uranium of 0.711wt% $^{235}U$ and they have the same cooling time of 5.0 years but they have different burnups of 5.0MWD/kg and 3.0MWD/kg, respectively. For these test problems, the input data for our program are prepared by extracting atomic number densities of the monitor nuclides from the ORIGEN-S outputs and the goal of the tests is to certify if our program gives the specified values of the uranium enrichment, burnup, cooling time, and reactor type. As shown in Table II, it is noted that our program correctly identified the reactor types for all the test problems. For the PWR and BWR test problems given in Table II, our program gives very accurate estimations of all parameters for burnup history while our program gives larger errors for the CANDU test problems but these levels of errors are low to be acceptable. These large errors for CANDU test problem are due to the small burnup, and short cooling time. The maximum error for CANDU test problems is ~5.7% for enrichment.

For verification, we applied to the known series of the Mihama-3 problem. For the Mihama-3 problems[5], uranium enrichment is ~3.25wt% for all the cases and their cooling time is 5.0 years while there are large number of different burnups in Mihamam-3. For these verification problems, our program all correct estimation of reactor type. Table III shows that our program gives very accurate estimation of burnup of which the maximum error is 2.54% and accurate estimation of enrichment less than 4.2%. On the other hand, the errors for cooling time are relatively larger than errors in the burnup and enrichment. The maximum error for the cooling time is 10.3% for the case of Mihama3-5.

Table I : Summary of Monitor Nuclides

| *Monitors* | *Specification* |
|---|---|
| Burnup Estimators | U-235, U-236, U-238, Np-237, Pu-239, Pu-240, Pu-241, Pu-242 |
| Burnup Monitors | Nd-148 |
| Enrichment Monitors | U-234, U-235, U-236, U-238, Pu-239, Pu-240, Pu-241 |
| Reactor Type Monitors | Nd-148, Pu-240, Nd-143 |
| Cooling Time Monitors | Ru-106, Sb-125, Cs-134, Cs-137 |

## 3. Summary and Conclusion

We developed a computer program to determine the burnup history such as initial uranium enrichment, burnup, cooling time, and reactor type by using the results of sample measurements as input. Our methodologies are based on the methodologies given in Ref. 1 but we devised a new stable bisection method for the correction of initial uranium enrichment and we used ORIGEN-S rather than ORIGEN-2 to utilize the new capabilities of ORIGEN-S such as burnup dependent cross sections which can be prepared by using SCALE6. For test and verification, our program was applied to the basic test problems prepared using ORIGEN-S and to the well-known Mihama-3 problems which have realistic measurement data. The results of test show that our program gives accurate estimation of the burnup history parameters and currently we are developing and implement a burnup correction method for further improving accuracies.

## REFERENCES

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[4] Post Irradiation Examination for Spent Fuel Sample (Cooper), "http://www.oecd-nea.org/sfcompo/Ver.2/Eng/Cooper/index.html".
[5] Post Irradiation Examination for Spent Fuel Sample (Cooper), "http://www.oecd-nea.org/sfcompo/Ver.2/Eng/Mihama-3/index.html".

Table II: Test problem from the ORIGEN-S

| Promblem | Is reactor type predicted correctely ? | Burnup (MWD/kg) | | Enrichment (MWD/kg) | | Cooling Time (Yr) | |
|---|---|---|---|---|---|---|---|
| | | Reported value | Predicted value (error) | Reported value | Predicted value (error) | Reported value | Predicted value (error) |
| PWR-1 | Yes | 18 | 18.3 (1.64%) | 3.0 | 3.03 (0.99%) | 5.0 | 4.96 (0.81%) |
| PWR-2 | Yes | 30 | 30.4 (1.32%) | 4.0 | 4.06 (1.48%) | 10.0 | 9.94 (0.60%) |
| BWR-1 | Yes | 33.94 | 34.3 (1.05%) | 2.93 | 2.95 (0.68%) | 5.35 | 5.31 (0.75%) |
| BWR-2 | Yes | 31.04 | 31.4 (1.15%) | 2.93 | 2.96 (1.01%) | 5.35 | 5.32 (0.56%) |
| CANDU-1 | Yes | 5.0 | 5.3 (5.66%) | 0.711 | 0.736 (3.40%) | 5.0 | 5.19 (3.66%) |
| CANDU-2 | Yes | 3.0 | 3.2 (6.25%) | 0.711 | 0.728 (2.34%) | 5.0 | 5.22 (4.21%) |

Table III: Mihama problem from the OECD/NEA

| Promblem | Is reactor type predicted correctely ? | Burnup (MWD/kg) | | Enrichment (MWD/kg) | | Cooling Time (Yr) | |
|---|---|---|---|---|---|---|---|
| | | Reported value | Predicted value (error) | Reported value | Predicted value (error) | Reported value | Predicted value (error) |
| Mihama3-1 | Yes | 8.3 | 8.11 (2.34%) | 3.25 | 3.30 (1.52%) | 5.0 | 5.09 (1.77%) |
| Mihama3-2 | Yes | 6.9 | 6.8 (1.47%) | 3.25 | 3.31 (1.81%) | 5.0 | 4.81 (3.95%) |
| Mihama3-3 | Yes | 15.3 | 15.03 (1.80%) | 3.24 | 3.32 (2.41%) | 5.0 | 5.16 (3.10%) |
| Mihama3-4 | Yes | 21.2 | 20.8 (1.92%) | 3.24 | 3.29 (1.52%) | 5.0 | 4.78 (4.60%) |
| Mihama3-5 | Yes | 14.6 | 14.3 (2.10%) | 3.24 | 3.25 (0.31%) | 5.0 | 4.53 (10.3%) |
| Mihama3-6 | Yes | 29.44 | 28.9 (1.87%) | 3.25 | 3.20 (1.56%) | 5.0 | 5.39 (7.24%) |
| Mihama3-7 | Yes | 32.3 | 31.5 (2.54%) | 3.25 | 3.24 (0.31%) | 5.0 | 4.82 (3.73%) |
| Mihama3-8 | Yes | 33.7 | 32.99 (2.15%) | 3.25 | 3.12 (4.17%) | 5.0 | 5.03 (0.60%) |
| Mihama3-9 | Yes | 34.1 | 33.5 (1.79%) | 3.25 | 3.22 (0.93%) | 5.0 | 5.16 (3.10%) |