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Original Article

iBEST: A PROGRAM FOR BURNUP HISTORY ESTIMATION OF SPENT FUELS BASED ON ORIGEN-S

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ABSTRACT

In this paper, we describe a computer program, iBEST (inverse Burnup ESTimator), that we developed to accurately estimate the burnup histories of spent nuclear fuels based on sample measurement data. The burnup history parameters include initial uranium enrichment, burnup, cooling time after discharge from reactor, and reactor type. The program uses algebraic equations derived using the simplified burnup chains of major actinides for initial estimations of burnup and uranium enrichment, and it uses the ORIGEN-S code to correct its initial estimations for improved accuracy. In addition, we newly developed a stable bisection method coupled with ORIGEN-S to correct burnup and enrichment values and implemented it in iBEST in order to fully take advantage of the new capabilities of ORIGEN-S for improving accuracy. The iBEST program was tested using several problems for verification and well-known realistic problems with measurement data from spent fuel samples from the Mihama-3 reactor for validation. The test results show that iBEST accurately estimates the burnup history parameters for the test problems and gives an acceptable level of accuracy for the realistic Mihama-3 problems.

1. Introduction

The danger is growing that international terrorist groups could use nuclear materials from spent nuclear fuels rather than a nuclear bomb because it would be difficult for a small group of terrorists to manufacture a nuclear bomb [1–3]. Therefore, the ability to identify the perpetrators of such attacks and the origin of any such nuclear material is critical. The methodology, finding the burnup history and characteristics of the original nuclear materials based on an analysis of the postevent materials, is considered a necessary and effective tool for international nuclear safeguards. A similar methodology can also identify the initial uranium enrichment and burnup of spent nuclear fuels.

The purpose of this work is to develop a computer program that can accurately estimate the burnup histories of spent nuclear fuels based on sample measurement data within a reasonably short time. Gamma-ray isotopic analysis gives relative isotopic ratios in spent fuel samples, and thus the estimation of burnup history depends on the relative isotopic ratios in the samples rather than on the absolute isotopic masses. The burnup history parameters of spent

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fuels include initial uranium enrichment, discharge burnup, cooling time after discharge from a nuclear reactor, and the type of nuclear reactor in which the spent fuel was burnt. Actually, the isotopic compositions of spent nuclear fuel in nuclear fuel assemblies can be accurately calculated using numerous lattice calculation codes, such as HELIOS [4], CASMO [5], KARMA [6], and DeCART [7]. In those lattice codes, the isotopic compositions of fuel regions after burnup are determined by alternatively solving the neutron transport equation and the Bateman equation describing the change in isotopic composition over a specified number of time steps. On modern computers, those codes typically take several tens of minutes to complete the isotopic composition determination for a single-fuel assembly model. But the inverse problem, to determine the burnup history parameters such as initial uranium enrichment and burnup, can require many iterations. The use of the lattice codes for this purpose can take a considerable amount of computing time. Also, the lattice codes are typically applicable only for Pressurized Water Reactor (PWR) because their cross-section libraries were produced using the PWR spectrum. In 2005, M.R. Scott [1] proposed a method that used simplified depletion chains to derive simple algebraic equations to estimate the initial uranium enrichment and burnup of spent fuel from its isotopic ratios. He also devised a simple iterative algorithm coupled with ORIGEN-2 [8] to correct the initial estimation of the burnup history parameters. With his methods, burnup could be found within 5% accuracy, enrichment within 2.5% accuracy, and age within 10% accuracy for the nine samples taken from the Mihama-3 reactor. However, he reported that the reactor type (i.e., PWR) could not be correctly predicted, and his application was confirmed only by the Mihama-3 spent fuel problems.

We developed a program called iBEST [9,10] for the initial estimation of burnup and uranium enrichment based on the simple algebraic equations developed by M.R. Scott, but we newly developed a stable bisection method to correct the initial uranium enrichment and burnup because we found that the simple correction method used by Scott [1] can be unstable. Moreover, we used ORIGEN-S [11] rather than ORIGEN-2 in our program to fully take advantage of the burnup-dependent cross-section libraries of ORIGEN-S for improving accuracy and developed a graphic user interface (GUI) for input and output visualizations, whereas the previous work presented by Scott [1] used the old ORIGEN2 code combined with a simple correction method. For verification of iBEST, we devised benchmark problems using ORIGEN-S for several different types of reactor, and the results of iBEST that were obtained with the input parameters extracted from ORIGEN-S outputs were compared with the initial conditions of ORIGEN-S. Then, we tested the iBEST program using the well-known realistic Mihama-3 problems [12], which have measurement data from spent fuel assays, for validation. In Section 2, we review the methodologies for the initial estimation of burnup and enrichment and describe the stable bisection method developed in this work. Section 2 also gives the correction methods for uranium and burnup, as well as the computational procedure used in iBEST. The verification and validation of iBEST are given in Section 3. Section 4 gives the summary and conclusions.

2. Theory and computational method

2.1. Initial estimations of uranium enrichment and burnup

In this section, we first review the method and formulations for the initial estimation of burnup and enrichment. The equation that can estimate the burnup of spent fuel is derived by considering that the initial atom number density of uranium is equal to the sum of the remaining atom number densities of uranium isotopes and all of the uranium reactions undergone during irradiation at the measurement time. The following equation gives the balance relation [1]:

$$\begin{aligned} N_0^U = {} & N^{U235}(T) + N^{U238}(T) + \sigma_f^{U235}\int_0^T N^{U235}(t)\varphi(t)dt \\ & + \sigma_\gamma^{U235}\int_0^T N^{U235}(t)\varphi(t)dt + \sigma_f^{U238}\int_0^T N^{U238}(t)\varphi(t)dt \\ & + \sigma_\gamma^{U238}\int_0^T N^{U238}(t)\varphi(t)dt, \end{aligned} \tag{1}$$

where T is the measurement time of the spent fuel sample, σ_f^{U235} is the microscopic one-group effective fission cross section, σ_γ^{U235} is the microscopic one-group effective capture cross section, N_0^U is the number density of the initial uranium atoms, and $N^X(T)$ is the number density of atoms of nuclide X at measurement time T. Eq. (1) also assumes that the neutron spectrum does not change over time. The capture terms of Eq. (1) can be decomposed into the capture and fission rates of the corresponding capture products. With those decompositions, Eq. (1) can be rewritten as:

$$\begin{aligned} N_0^U = {} & N^{U235}(T) + N^{U238}(T) + N^{U236}(T) + N^{Pu239}(T) + N^{Pu240}(T) + \cdots \\ & + \sigma_f^{U235}\int_0^T N^{U235}(t)\varphi(t)dt + \sigma_f^{U238}\int_0^T N^{U238}(t)\varphi(t)dt \\ & + \sigma_f^{U236}\int_0^T N^{U236}(t)\varphi(t)dt + \sigma_f^{Pu239}\int_0^T N^{Pu239}(t)\varphi(t)dt + \cdots, \end{aligned} \tag{2}$$

where the delayed production of ^{239}Pu from the decay of ^{239}U and ^{239}Np is ignored. At this point, the burnup monitor nuclides are considered to estimate the burnup. Actually, any fission product produced directly proportional to the burnup can be used as a burnup monitor, and it is known that burnup can be measured within a one percent accuracy coupled with mass spectrometry. For our problems, however, the reactor type is not given before the problem is solved, and so burnup monitors that are produced at the same rate regardless of reactor type should be chosen. Also, fission products that have a constant fission yield across reactor types and a long half-life are desirable for burnup monitoring, to simplify the formulation. For a good burnup monitor, the following equation is satisfied:

$$\frac{dN^B}{dt} = Y_B\left[N^{U235}(t)\bar{\sigma}_f^{U235}\varphi(t) + N^{Pu239}(t)\bar{\sigma}_f^{Pu239}\varphi(t) + \cdots\right], \tag{3}$$

where the cumulative yield (Y_B) for the burnup monitor is assumed to be the same for all fissionable nuclides and to be

independent of the incident neutron energy. In Eq. (3), the right-hand side should include all of the fissions contributed from all the fissionable nuclides, and the radioactive decay of the burnup monitor is neglected by considering its long half-life. Also, in Eq. (3), production from radioactive decay and the radiative capture of the preceding nuclides are not considered. The integration of Eq. (3) over time gives:

$$N^{B}(T) = Y_B \int_0^T dt\left[N^{U235}(t)\bar{\sigma}_f^{U235}\varphi(t) + N^{Pu239}(t)\bar{\sigma}_f^{Pu239}\varphi(t) + \cdots\right] = \frac{\rho_0^U}{E_R} Y_B BU(T), \tag{4}$$

where ρ_0^U is the initial density of uranium in the fuel, BU(T) is the burnup of the fuel, and E_R is the average recoverable energy per fission. The substitution of Eq. (4) into Eq. (1) gives:

$$N_0^U = N^U(T) + N^{Pu238}(T) + N^{Pu239}(T) + N^{Pu240}(T) + \cdots + \frac{N_0^U M_U}{N_a E_R} BU(T), \tag{5}$$

where N_a and M_U are Avogadro's number and the atomic weight of uranium, respectively. The division of Eq. (5) by $N^{U238}(T)$ after solving for N_0^U gives the equation relating the measured quantities with burnup as follows:

$$\frac{N_0^U}{N^{U238}(T)} = \frac{\frac{N^U(T)}{N^{U238}(T)} + \frac{N^{Pu238}(T)}{N^{U238}(T)} + \frac{N^{Pu298}(T)}{N^{U238}(T)} + \cdots}{1 - \frac{M_U}{N_a E_R} BU(T)}. \tag{6}$$

In this equation, all the quantities in the numerator can be known from the measurements of a spent fuel sample, but the quantity on the left-hand side is not measurable. Therefore, we need one additional equation to determine the burnup. Dividing Eq. (4) by the initial uranium atomic number density gives the additional equation needed for burnup estimation:

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

In Eq. (7), the first term on the right-hand side is a measured quantity and so, Eqs. (6) and (7) can be used to estimate the burnup.

Next, the equation for the estimation of initial uranium enrichment is derived by considering the balance of the initial atomic density of ^{235}U. The balance equation is given by:

$$N_0^{U235} = N^{U235}(T) + \bar{\sigma}_f^{U235}\int_0^T N^{U235}(t)\varphi(t)dt + \bar{\sigma}_\gamma^{U235}\int_0^T N^{U235}(t)\varphi(t)dt. \tag{8}$$

In deriving the equation for initial uranium enrichment estimation, it was assumed that ^{239}Np and ^{239}U decay instantaneously to ^{239}Pu. The decay of all fissionable nuclides was neglected for simplicity because the initial uranium enrichment will be improved by using the correction step. The capture term of Eq. (8) is replaced with the atomic number density of ^{236}U if the fission of ^{236}U is neglected, which gives:

$$N_0^{U235} = N^{U235}(T) + N^{U236}(T) + \bar{\sigma}_f^{U235}\int_0^T N^{U235}(t)\varphi(t)dt. \tag{9}$$

Then, Eq. (4) is used to eliminate the fission term from Eq. (9), which gives:

$$N_0^{U235} = N^{U235}(T) + N^{U236}(T) + \frac{\rho_0^U}{E_R} BU(T) - \left[\bar{\sigma}_f^{U238}\int_0^T N^{U238}(t)\varphi(t)dt + \bar{\sigma}_f^{Pu239}\int_0^T N^{Pu239}(t)\varphi(t)dt + \cdots\right]. \tag{10}$$

Dividing Eq. (10) by the initial uranium atomic number density gives the following equation:

$$e_0 = \frac{N_0^{U235}}{N_0^U} = \frac{N^{U235}(T)}{N_0^U} + \frac{N^{U236}(T)}{N_0^U} + \frac{\rho_0^U}{N_0^U E_R} BU(T) - \frac{1}{N_0^U}\left[\bar{\sigma}_f^{U238}\int_0^T \varphi(t)N^{U238}(t)dt + \bar{\sigma}_f^{Pu239}\int_0^T \varphi(t)N^{Pu239}(t)dt + \cdots\right]. \tag{11}$$

The next step is to use the depletion equation without consideration of radioactive decay for the actinides that appear in the last term of Eq. (11). For example, the depletion equation for ^{238}U is given by:

$$\frac{dN^{U238}}{dt} = -\sigma_a^{U238}\varphi(t)N^{U238}(t) \Rightarrow \int_0^T dt\varphi(t)N^{U238}(t) = \frac{-N^{U238}(T) + N_o^{U238}}{\sigma_a^{U238}}. \tag{12}$$

This equation can then be substituted into Eq. (11), and a similar procedure can be successively done for the other actinides that appear in the last term of Eq. (11). This procedure gives the following equation for the determination of the initial uranium enrichment of the spent fuel:

$$\begin{aligned} e_0 \equiv \frac{N_0^{U235}}{N_0^U} = {} & \frac{N^{U235}(T)}{N_0^U} + \frac{N^{U236}(T)}{N_0^U} + \frac{\rho_0^U}{E_R N_0^U} BU(T) \\ & - \frac{1}{N_0^U}\left[\frac{\bar{\sigma}_f^{U238}}{\bar{\sigma}_a^{U238}}\left[N_0^{U238} - N^{U238}(T)\right]\right] \\ & + \frac{\bar{\sigma}_f^{PU239}}{\bar{\sigma}_a^{PU239}}\left[N_0^{PU239} - N^{Pu239}(T) + F^{U238}\right] \\ & + \frac{\bar{\sigma}_f^{Pu240}}{\bar{\sigma}_a^{PU240}}\left[N_0^{PU240} - N^{Pu240}(T) + F^{Pu239}\right] \\ & + \frac{\bar{\sigma}_f^{Pu241}}{\bar{\sigma}_a^{Pu241}}\left[N_0^{Pu241} - N^{Pu241}(T) + F^{Pu240}\right]. \end{aligned} \tag{13}$$

In Eq. (13), the following definitions for F were used:

$$\begin{aligned} F^{U238} &= \frac{\bar{\sigma}_\gamma^{U238}}{\bar{\sigma}_a^{U238}}\left[N_0^{U238} - N^{U238}(T)\right], \\ F^{Pu239} &= \frac{\bar{\sigma}_\gamma^{PU239}}{\bar{\sigma}_a^{Pu239}}\left[N_0^{Pu239} - N^{Pu239}(T) + F^{U238}\right], \\ F^{Pu240} &= \frac{\bar{\sigma}_\gamma^{Pu240}}{\bar{\sigma}_a^{Pu240}}\left[N_0^{Pu240} - N^{Pu240}(T) + F^{Pu239}\right]. \end{aligned} \tag{14}$$

If we assume that initial plutonium isotope masses are zero, Eq. (14) can be simplified to:

$$e_0 = \frac{N^{U238}(T)}{N_0^U}\left[\frac{N^{U235}(T)}{N^{U238}(T)} + \frac{N^{U236}(T)}{N^{U238}(T)}\right] + \frac{M_0^U}{N_a E_R} BU(T) - G^{U238} - G^{Pu239} - G^{Pu240} - G^{Pu241}, \tag{15}$$

where

$$
\begin{aligned}
G^{U238} &= \frac{\overline{\sigma}_f^{U238}}{\overline{\sigma}_a^{U238}}\left[1 - e_0 - \frac{N^{U238}(T)}{N_0^U}\right],\\
G^{Pu239} &= \frac{\overline{\sigma}_f^{Pu239}}{\overline{\sigma}_a^{Pu239}}\left[-\frac{N^{Pu239}(T)}{N^{U238}(T)}\cdot\frac{N^{U238}(T)}{N_0^U} + G^{U238}\frac{\overline{\sigma}_\gamma^{U238}}{\overline{\sigma}_a^{U238}}\frac{\overline{\sigma}_a^{U238}}{\overline{\sigma}_f^{U238}}\right],\\
G^{Pu240} &= \frac{\overline{\sigma}_f^{Pu240}}{\overline{\sigma}_a^{Pu240}}\left[-\frac{N^{Pu240}(T)}{N^{U238}(T)}\cdot\frac{N^{U238}(T)}{N_0^U} + G^{Pu239}\frac{\overline{\sigma}_\gamma^{Pu239}}{\overline{\sigma}_a^{Pu239}}\frac{\overline{\sigma}_a^{Pu239}}{\overline{\sigma}_f^{Pu239}}\right],\\
G^{Pu241} &= \frac{\overline{\sigma}_f^{Pu241}}{\overline{\sigma}_a^{Pu241}}\left[-\frac{N^{Pu241}(T)}{N^{U238}(T)}\cdot\frac{N^{U238}(T)}{N_0^U} + G^{Pu240}\frac{\overline{\sigma}_\gamma^{Pu240}}{\overline{\sigma}_a^{Pu240}}\frac{\overline{\sigma}_a^{Pu240}}{\overline{\sigma}_f^{Pu240}}\right].
\end{aligned}
\tag{16}
$$

Eq. (16) contains the ratio $N^{U238}(T)/N_0^U$, and this quantity was already determined during the estimation of burnup. Our program uses an iterative algorithm to solve Eqs. (15) and (16), and we found that the algorithm is always rapidly convergent.

2.2. Correction of uranium enrichment and burnup

The methods for the initial estimation of uranium enrichment and burnup described in Section 2.1 are relatively simple and computationally efficient. However, the estimated values found using Eqs. (6), (7), (15), and (16) are generally inaccurate. In particular, the estimated value of initial uranium enrichment is much less accurate than the estimated burnup, and so correction of those initial estimations is required to improve the accuracy. For this purpose, a correction method coupled with the ORIGEN-2 forward depletion calculation was suggested by Scott [1] for both burnup and initial uranium enrichment. However, ORIGEN-2 is a very old code, and the accuracy of its depletion calculations is limited by its burnup-independent cross-section libraries. Furthermore, we have found that the algorithm given by Scott [1] to correct the uranium enrichment can be unstable. In this work, we newly developed a stable bisection algorithm to correct the initial uranium enrichment and burnup. In addition, we used ORIGEN-S rather than ORIGEN-2 to improve the accuracy because ORIGEN-S provides several new capabilities, including a burnup-dependent cross-section library.

Next, we describe enrichment correction using the bisection method. The bisection method starts with the initial estimation of uranium enrichment by setting:

$$X = XL = XR = e_0. \tag{17}$$

Then, our program automatically prepares an ORIGEN-S input file using the initial uranium enrichment and burnup estimations. At present, the ORIGEN-S input assumes an initial uranium mass of 1,000 kg and a specific power of 40 W/g. Next, our program executes ORIGEN-S to perform depletion calculation up to the initially estimated burnup and then calculates the following function value using the results of the ORIGEN-S output:

$$f(X) = R_m A_{U238}^{ORIGEN}(X) - A_{U235}^{ORIGEN}(X), \tag{18}$$

where X represents uranium enrichment. In Eq. (18), R_m is the ratio of the number of ^{235}U atoms to the number of ^{238}U atoms obtained from sample measurement, and A_{U238}^{ORIGEN} is the number of ^{238}U atoms from the ORIGEN-S output at the burnup. We hope that the function given by Eq. (18) is nearly zero when the enrichment correction is completed because the function $f(X)$ indicates the difference between the numbers of ^{235}U atoms obtained from the ORIGEN-S calculation and from the measurement ratio R_m multiplied by the number of ^{238}U atoms estimated by ORIGEN-S. Thus, if the function $f(X)$ is positive, then XL is increased by a specified value Δ until $f(XL)$ becomes negative. The last XL value at which the function is negative is then set to XR. By contrast, if the function $f(X)$ is negative, then XR is decreased by a specified value Δ until $f(XR)$ becomes positive. The last XR value at which the function is positive is then set to XL. Once the initial values of XL and XR at which the function has different signs are determined, the conventional bisection method is used as follows:

$$
\begin{aligned}
&(1)\ X_{old} = XL \text{ or } XR \text{ depending on the initial sign of } f(e_0)\\
&(2)\ X = (XL + XR)/2.0,\\
&(3)\ \text{if} \left|\frac{X - X_{old}}{X_{old}}\right| < \varepsilon \Rightarrow \text{STOP}\\
&\text{else}\\
&\quad X_{old} = X\\
&\quad \text{if } f(X)*f(XR) < 0 \Rightarrow XR = X\\
&\quad \text{else } XL = X,\\
&\quad \text{Go To } (2)
\end{aligned}
\tag{19}
$$

In the first step of the above algorithm, if the initial sign of $f(e_0)$ is positive, XR is set to X_{old} and otherwise, XL is set to X_{old}. In Eq. (19), ε is a convergence criterion. The above enrichment corrections are done for all of the candidate reactor types that have a corresponding one-group cross-section library. After the correction of uranium enrichment, a similar procedure using the bisection method is applied to correct the burnup. In the correction of burnup, the function given in Eq. (18) is replaced with:

$$f(X) = R_{m,BM} A_{U238}^{ORIGEN}(X) - A_{BM}^{ORIGEN}(X), \tag{20}$$

where $R_{m,BM}$ is the ratio of the number of burnup monitor atoms to the number of ^{238}U atoms, A_{BM}^{ORIGEN} is the number of burnup monitor atoms calculated with ORIGEN-S at the measurement time, and X is the burnup. These sequential corrections of enrichment and burnup are actually performed two or three times in our program.

2.3. Determination of cooling time and reactor type

After the enrichment corrections are completed, the cooling time after discharge from the reactor is estimated. In general, fission products with a half-life value similar to the cooling time are desirable as the age monitors, but the cooling time is unknown before the problem is solved. Generally, fission products with half-lives of 1–30 years are recommended as age monitors. ORIGEN-S calculations are performed again for each type of reactor, and then the atomic number densities of the age monitor nuclides at 30 days after discharge are extracted from the ORIGEN-S outputs to wait the decay out of the short-lived fission products that lead to the age monitor. Those values are set to $N_{0,i}^C$. If the age monitor nuclides are not produced from the decay of other nuclides, their atomic number densities at cooling time T_C after discharge are given by:

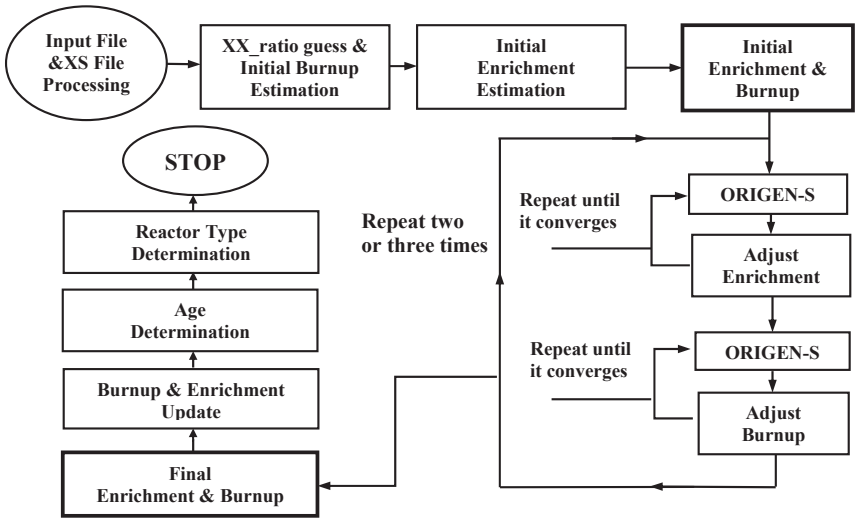

Fig. 1 – Calculation procedure in iBEST (invest Burnup Estimator).

$$T_C = -\frac{T_{1/2,i}}{\ln(2)} \ln\left(\frac{N_i^C(T_C)}{N_{0,i}^C}\right), \tag{21}$$

where λ_i and $T_{1/2,i}$ are the decay constant and half-life of the ith age monitor, respectively. To use measurable quantities, Eq. (21) is changed into:

$$T_C = -\frac{T_{1/2,i}}{\ln(2)} \ln\left[\frac{N_i^C(T_C)/N^{U238}(T)}{N_{0,i}^C/N^{U238}(T)}\right]. \tag{22}$$

In Eq. (22), the denominator is calculated from the ORIGEN-S calculations, and the numerator is from the measurements. When several age monitors are used, some monitors can give larger errors in cooling time than others; therefore, the final cooling time should be carefully determined. We first calculate the average of the cooling times for all the age monitors and select the two values closest to the average value. Then, we select the average value of those two values as the cooling time for each reactor type. Finally, 30 days should be added to the cooling time determined by Eq. (22) to give the final cooling time because the atomic number densities at 30 days after discharge were used in Eq. (22).

The estimation of the reactor type in which the spent fuel was burnt is also done using ORIGEN-S calculations. The reactor type monitor nuclides should be chosen such that their depletion characteristics are distinctly different for different types of reactors. Also, to avoid the complication of decay, stable or long-lived isotopes are desirable as reactor type monitors. The method for determining reactor type also depends on the accuracy of the depletion calculations, which diminishes as the decay chain becomes more complicated. In particular, it is difficult to accurately determine the reactor type for spent fuels with low burnup. The discrimination between PWR and Boiling Water Reactor (BWR) at low burnup is much more difficult than in other cases. To determine the reactor type, ORIGEN-S inputs are automatically prepared for all reactor type candidates using the previously estimated uranium enrichments, burnups, and cooling times. Then, the ORIGEN-S depletion calculations are done with all the inputs for all the candidate reactor types. After the depletion calculations, the atomic number density ratios of the reactor type monitors to ^{238}U are calculated using the results of the depletion calculations at the measurement time. Then, the differences between those ratios and their measured values are calculated, and the reactor type with the minimum difference is selected.

2.4. *Computational procedure in iBEST*

We developed a computer program called iBEST using the methodologies described in the previous sections to estimate the burnup history of spent fuel. We wrote this program using C++ and developed a GUI program for user convenience. The computational procedure using the methodologies given in Sections 2.1, 2.2, and 2.3 is shown in Fig. 1. First, the program estimates the burnup and xx_ratio($=N_0^U/N^{U238}(T)$)by iteratively solving Eqs. (6) and (7). Note that only the fission yield values for the burnup monitors and their isotopic ratios to ^{238}U are required to solve those equations. Then, uranium enrichment is estimated by iteratively solving Eqs. (15) and (16). Solving these two equations requires the effective one-group fission, absorption, and capture cross sections for the actinides. Therefore, iBEST requires an "xs.file" that contains the effective one-group cross sections of the actinides for all of the candidate reactor types. Uranium enrichment estimation is done for each of the candidate reactor types.

Uranium enrichment and burnup correction are followed by the bisection method described in Section 2.2. The uranium enrichment and burnup corrections are repeated two or three times in iBEST for all candidate reactor types. The next step is to estimate the cooling times for all the candidate reactor types using Eq. (22), and 30 days are added to the cooling times, as explained in Section 2.3. Finally, the reactor type is determined after the ORIGEN-S depletion calculations for all the candidate reactor types using their burnup, uranium enrichment, and cooling time estimates. Users can prepare the input file for iBEST using the GUI, which also provides

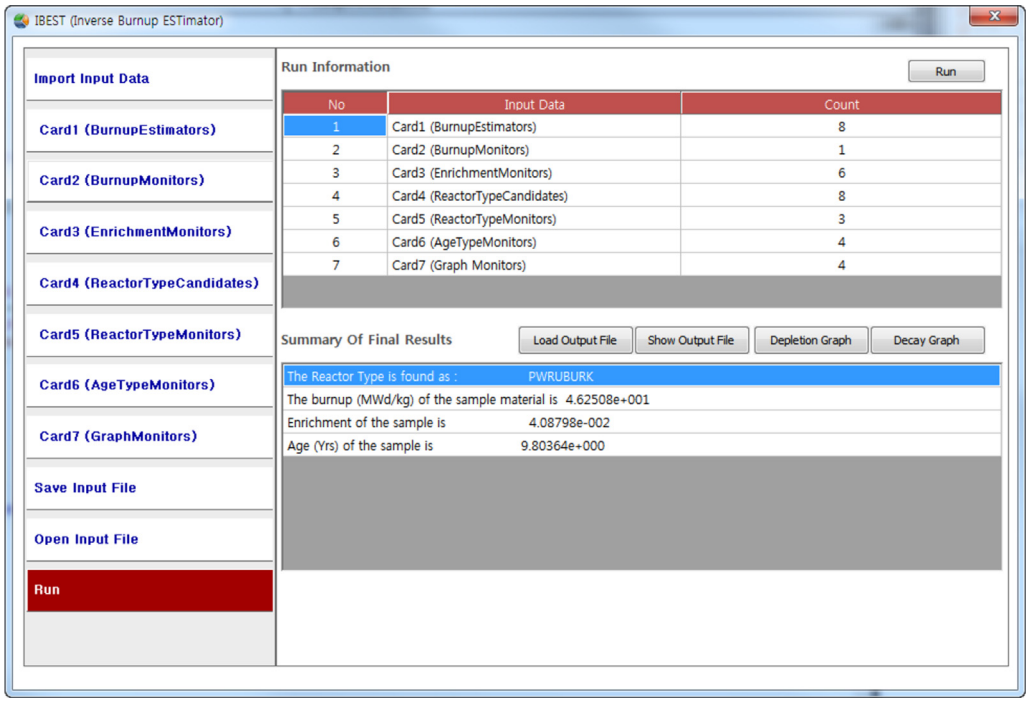

Fig. 2 – Main window of iBEST (invest Burnup Estimator) graphic user interface.

output visualization. Fig. 2 shows the main window of the iBEST GUI.

3. Numerical verification

3.1. *Numerical tests with ORIGEN-S*

For verification, we first applied iBEST to several test problems prepared using ORIGEN-S. The purpose of these test problems was to show whether iBEST gave the correct estimations of burnup history parameters. In these test problems, the burnup history parameters are initially specified, and the input parameters for iBEST were prepared by extracting the atomic number densities of the monitor nuclides from the ORIGEN-S output files. We considered four different types of test problems: (1) PWR test problems, (2) BWR test problems, (3) CANDU (Canada Deuterium Uranium) test problems, and (4) MAGNOX test problems. The PWR tests were based on the Mihama-3 problems [12], which model nine spent fuel samples from the Mihama-3 reactor. Table 1 specifies the atomic

Table 1 – Atomic number density ratios of the monitor nuclides for the Mihama-3 PWR test problems.

Monitor type	Nuclides	Problem no.								
		1	2	3	4	5	6	7	8	9
Burnup	^{148}Nd	[a]1.57E−04	1.31E−04	4.01E−04	2.89E−04	2.76E−04	5.59E−04	6.14E−04	6.41E−04	6.49E−04
Enrichment	^{235}U	[a]2.44E−02	2.57E−02	1.48E−02	1.87E−02	1.92E−02	1.04E−02	9.15E−03	8.57E−03	8.42E−03
	^{236}U	1.62E−03	1.53E−03	3.45E−03	2.78E−03	2.69E−03	4.14E−03	4.32E−03	4.40E−03	4.42E−03
	^{238}U	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{237}Np	6.23E−05	5.29E−05	2.59E−04	1.64E−04	1.54E−04	4.02E−04	4.51E−04	4.75E−04	4.81E−04
	^{239}Pu	3.30E−03	2.92E−03	5.27E−03	4.64E−03	4.54E−03	5.70E−03	5.79E−03	5.83E−03	5.84E−03
	^{240}Pu	4.62E−04	3.50E−04	1.56E−03	1.06E−03	1.00E−03	2.20E−03	2.39E−03	2.48E−03	2.51E−03
	^{241}Pu	1.27E−04	8.44E−05	6.59E−04	4.06E−04	3.75E−04	9.73E−04	1.07E−03	1.11E−03	1.12E−03
	^{242}Pu	1.17E−05	6.33E−06	1.84E−04	7.62E−05	6.67E−05	4.11E−04	5.06E−04	5.56E−04	5.70E−04
Reactor type	^{132}Ba	[b]1.24E−07	1.00E−07	3.51E−07	2.45E−07	2.33E−07	5.03E−07	5.55E−07	5.81E−07	5.88E−07
	^{148}Nd	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
Age	^{106}Ru	[a]2.73E−06	2.16E−06	8.73E−06	5.89E−06	5.56E−06	1.28E−05	1.41E−05	1.48E−05	1.50E−05
	^{125}Sb	1.08E−06	8.83E−07	3.02E−06	2.12E−06	2.02E−06	4.26E−06	4.68E−06	4.88E−06	4.94E−06
	^{134}Cs	2.94E−06	2.02E−06	1.81E−05	9.79E−06	8.95E−06	3.30E−05	3.88E−05	4.18E−05	4.27E−05
	^{137}Cs	5.07E−04	4.22E−04	1.29E−03	9.34E−04	8.92E−04	1.80E−03	1.98E−03	2.06E−03	2.09E−03

PWR, Pressurized Water Reactor.
[a] Atomic density ratio to ^{238}U.
[b] Atomic density ratio to ^{148}Nd.

Table 2 – Test results for the Mihama-3 PWR test problems.

Problem No.	RT[a]	Burnup (MWD/kg)		Enrichment (%)		Cooling time (y)	
		Reference	Predicted	Reference	Predicted	Reference	Predicted
1	Yes	8.3	8.31[b]/0.11[c]	3.208	[a]3.21/[b]0.19	5.0	[a]5.06/[b]1.17
2	Yes	6.9	6.91/0.17	3.208	3.22/0.26	5.0	5.07/1.39
3	Yes	21.2	21.15/0.22	3.203	3.20/0.05	5.0	5.05/0.94
4	Yes	15.3	15.28/0.11	3.203	3.20/0.01	5.0	5.04/0.84
5	Yes	14.6	14.58/0.16	3.203	3.21/0.08	5.0	5.04/0.81
6	Yes	29.44	29.47/0.11	3.210	3.21/0.14	5.0	5.03/0.59
7	Yes	32.3	32.32/0.06	3.210	3.20/0.24	5.0	5.02/0.44
8	Yes	33.7	33.71/0.02	3.210	3.20/0.17	5.0	5.02/0.34
9	Yes	34.1	34.11/0.02	3.210	3.21/0.14	5.0	5.02/0.31

PWR, Pressurized Water Reactor; RT: Reactor Type.
[a] Is reactor type predicted correctly?
[b] Predicted value.
[c] Discrepancy between the predicted and reference values (%).

number density ratios of the monitor nuclides calculated using the atomic number densities from the ORIGEN-S outputs for these test problems. Table 2 summarizes the test results and the specifications of burnups, uranium enrichments, and cooling times denoted as the reference values for the nine test problems. As shown in Table 2, these nine test problems cover a burnup range from 8.3 MWD/kg to 34.1 MWD/kg. The considered cooling time is 5.0 years. Three enrichments of 3.208%, 3.203%, and 3.210% are considered for these problems.

The reactor type monitors used in the PWR tests were ^{132}Ba and ^{148}Nd, and PWR and BWR were the two candidate reactor types considered. The cross-section libraries for PWR and BWR provided by SCALE6.1 [13] for ORIGEN-S are "w15x15" and "ge7x7-0," respectively. Our test problems were prepared using ORIGEN-S calculations with the "w15x15" library. As shown in Table 2, iBEST correctly identified the reactor type as PWR in all cases. In general, discrimination between PWR and BWR is quite difficult for small burnup values. The maximum errors in uranium enrichment, burnup, and cooling time were 0.26%, 0.22%, and 1.39%, respectively. The results of these tests show that iBEST quite accurately predicted the specified burnups, uranium enrichments, and cooling times.

The test problems for BWR were prepared based on the Cooper reactor, a BWR in the United States [14]. This reactor uses a 7 × 7 fuel assembly. The spent fuel isotopic data obtained through postirradiation examination (PIE) of this reactor are given by the Organisation for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA) [14]. For this reactor, six sample datasets are available. Unfortunately, the isotopic data of the spent fuel for this reactor are insufficient to be used in the validation of iBEST; however, we prepared the six test problems corresponding to the six samples using ORIGEN-S depletion calculations with the "ge7x7-0" library. Table 3 specifies the atomic number density ratios for the monitor nuclides. Table 4 shows the test results and the specifications of the burnups, uranium enrichments, and cooling times denoted as reference values.

Table 3 – Atomic density ratios of the monitor nuclides for the Cooper BWR test problems.

Monitor type	Nuclides	Problem no.					
		1	2	3	4	5	6
Burnup	^{148}Nd	6.41E−04[a]	6.26E−04	3.58E−04	5.86E−04	5.51E−04	3.35E−04
Enrichment	^{235}U	5.03E−03[a]	5.34E−03	1.30E−02	6.21E−03	7.02E−03	1.38E−02
	^{236}U	4.14E−03	4.10E−03	3.00E−03	3.99E−03	3.88E−03	2.86E−03
	^{238}U	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{237}Np	3.55E−04	3.45E−04	1.66E−04	3.18E−04	2.95E−04	1.50E−04
	^{239}Pu	3.91E−03	3.91E−03	3.81E−03	3.92E−03	3.92E−03	3.72E−03
	^{240}Pu	2.41E−03	2.37E−03	1.35E−03	2.24E−03	2.12E−03	1.24E−03
	^{241}Pu	7.53E−04	7.40E−04	4.15E−04	7.06E−04	6.71E−04	3.76E−04
	^{242}Pu	5.94E−04	5.62E−04	1.34E−04	4.80E−04	4.14E−04	1.12E−04
Reactor type	^{132}Ba	4.67E−07[b]	4.56E−07	2.54E−07	4.25E−07	3.99E−07	2.35E−07
	^{148}Nd	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
Age	^{106}Ru	1.15E−05[a]	1.12E−05	5.75E−06	1.09E−05	1.01E−05	5.55E−06
	^{125}Sb	4.43E−06	4.32E−06	2.40E−06	4.11E−06	3.86E−06	2.28E−06
	^{134}Cs	3.45E−05	3.30E−05	1.16E−05	2.99E−05	2.67E−05	1.04E−05
	^{137}Cs	2.05E−03	2.00E−03	1.15E−03	1.88E−03	1.77E−03	1.08E−03

BWR, Boiling Water Reactor.
[a] Atomic density ratio to ^{238}U.
[b] Atomic density ratio to ^{148}Nd.

Table 4 – Test results for the Cooper BWR test problems.

Problem No.	RT[a]	Burnup (MWD/kg)		Enrichment (%)		Cooling time (y)	
		Reference	Predicted	Reference	Predicted	Reference	Predicted
1	Yes	33.94	33.9[b]/0.12[c]	2.93	[a]2.91/[b]0.69	5.35	[a]5.38/[b]0.56
2	Yes	33.07	33.1/0.09	2.93	2.92/0.34	5.35	5.38/0.56
3	Yes	18.96	18.98/0.11	2.93	2.95/0.68	5.35	5.40/0.93
4	Yes	31.04	30.97/0.23	2.93	2.92/0.34	5.28	5.31/0.56
5	Yes	29.23	29.22/0.03	2.93	2.94/0.34	5.28	5.31/0.56
6	Yes	17.84	17.78/0.34	2.93	2.95/0.68	5.28	5.34/1.12

BWR, Boiling Water Reactor.
[a] Is reactor type predicted correctly?
[b] Predicted value.
[c] Discrepancy between the predicted and reference values (%).

The enrichment for these six cases is 2.93%, and the considered cooling times are 5.35 years and 5.28 years. The burnups range from 17.8 MWD/kg to 33.9 MWD/kg.

For these test problems, PWR and BWR were also used as the reactor type candidates to see whether iBEST can correctly discriminate between PWR and BWR. Table 4 shows that iBEST accurately estimated the burnups and enrichments, with maximum errors of 0.34% and 0.69%, respectively, for all the cases. The maximum error for the cooling time was 1.12%, which was larger than those of the burnup and enrichment. However, this level of maximum error is still small and acceptable. Also, using ^{132}Ba and ^{148}Nd as reactor type monitors, iBEST correctly identified the reactor type as BWR in all cases.

The next test problems modeled CANDU reactors. We prepared six test problems using depletion calculations with the "candu37" library provided by SCALE6.1 for ORIGEN-S. Table 5 describes the atomic number density ratios for the monitor nuclides prepared using the ORIGEN-S outputs with the "candu37" library after the depletion calculations. Table 6 summarizes the test results and the specifications of burnups, uranium enrichments, and cooling times for these CANDU test problems. In general, the discharge burnup of the CANDU fuels is much lower than that of PWR fuels, about 6.5–7.5 MWD/kg. Also, CANDU fuel uses natural uranium, in which only 0.711% of the total uranium is ^{235}U. As shown in Table 5, the burnups range from 1.5MWD/kg to 10.0MWD/kg. The cooling times include 5 years and two extremely short cases of 0.1 and 0.3 years.

In these test problems, we considered a larger number of candidate reactor types than in the PWR and BWR test cases: PWR, BWR, CANDU, and MAGNOX, whose libraries (provided by SCALE6.1 for ORIGEN-S) are "w15x15," "ge7x7-0," "candu37," and "magnox," respectively. The monitors for the reactor type are ^{240}Pu, ^{148}Nd, and ^{143}Nd for these test problems. Table 6 shows that iBEST correctly identified the reactor type as CANDU in all cases, even the very low burnup cases. The burnups were quite accurately predicted, within 0.7%, and the uranium enrichment error was within 1.4%. The uranium enrichment errors are larger than those in the BWR and PWR cases because of the much lower ^{235}U content than in the previous cases. The cooling times were also accurately predicted, within 1.8%, but the errors for the two extreme cases with very short cooling times were large, up to 16.7%. The difficulty in predicting the short cooling time is because

Table 5 – Atomic density ratios of the monitor nuclides for the CANDU test problems.

Monitor type	Nuclides	Problem no.					
		1	2	3	4	5	6
Burnup	^{148}Nd	2.85E−05[a]	9.58E−05	1.53E−04	1.92E−04	1.92E−04	1.92E−04
Enrichment	^{235}U	5.70E−03[a]	3.34E−03	2.08E−03	1.50E−03	1.50E−03	1.50E−03
	^{236}U	2.73E−04	6.36E−04	8.25E−04	9.10E−04	9.09E−04	9.09E−04
	^{238}U	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{239}Pu	1.16E−03	2.35E−03	2.71E−03	2.82E−03	2.82E−03	2.82E−03
	^{240}Pu	8.95E−05	6.08E−04	1.11E−03	1.41E−03	1.41E−03	1.41E−03
	^{241}Pu	5.98E−06	8.80E−05	1.87E−04	2.51E−04	3.18E−04	3.14E−04
	^{242}Pu	3.20E−07	1.85E−05	7.04E−05	1.26E−04	1.26E−04	1.26E−04
Reactor type	^{143}Nd	3.09E+00[b]	2.57E+00	2.23E+00	2.02E+00	1.99E+00	2.02E+00
	^{148}Nd	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{240}Pu	3.14E+00	6.35E+00	7.21E+00	7.37E+00	7.37E+00	7.37E+00
Age	^{106}Ru	5.06E−07[a]	2.82E−06	5.33E−06	7.11E−06	2.00E−04	1.75E−04
	^{125}Sb	1.80E−07	7.99E−07	1.42E−06	1.84E−06	6.39E−06	6.08E−06
	^{134}Cs	1.02E−07	1.44E−06	3.68E−06	5.67E−06	2.94E−05	2.75E−05
	^{137}Cs	9.01E−05	3.02E−04	4.85E−04	6.07E−04	6.79E−04	6.76E−04

CANDU, Canada Deuterium Uranium.
[a] Atomic density ratio to ^{238}U.
[b] Atomic density ratio to ^{148}Nd.

Table 6 – Test results for the CANDU test problems.

Problem no.	RT[a]	Burnup (MWD/kg)		Enrichment (%)		Cooling time (y)	
		Reference	Predicted	Reference	Predicted	Reference	Predicted
1	Yes	1.5	1.502[b]/0.13[c]	0.711	[a]0.719/[b]1.11	5	[a]5.09/[b]1.77
2	Yes	5.0	5.03/0.60	0.711	0.719/1.11	5	5.09/1.77
3	Yes	8.0	8.01/0.12	0.711	0.720/1.25	5	5.05/0.99
4	Yes	10.0	9.93/0.70	0.711	0.720/1.25	5	5.05/0.99
5	Yes	10.0	9.93/0.70	0.711	0.721/1.39	0.1	0.12/16.67
6	Yes	10.0	9.93/0.70	0.711	0.720/1.25	0.3	0.32/6.25

CANDU, Canada Deuterium Uranium; RT, Reactor Type.
[a] Is reactor type predicted correctly?
[b] Predicted value.
[c] Discrepancy between the predicted and reference values (%).

the predicted cooling time can be significantly influenced by the contribution of the preceding fission products to the formation of age monitors.

The last test problems modeled MAGNOX type reactors [15], which are gas (CO_2)-cooled reactors with graphite moderators. The fuel is natural uranium in metallic form, canned with a magnesium alloy called Magnox. In MAGNOX reactors, the fuel burnup is typically low because they use natural uranium, as in CANDU. So, we considered four different burnup cases of 2 MWD/kg, 3 MWD/kg, 4 MWD/kg, and 8 MWD/kg. The considered cooling time was 5 years, and a very short cooling time of 0.1 years was also considered. Table 7 describes the atomic number density ratios for the monitor nuclides prepared using ORIGEN-S depletion calculations from the "magnox" library. The test results and specifications of the burnups, uranium enrichments, and cooling times for these problems are summarized in Table 8. We used the same reactor type candidates and their corresponding cross section libraries for ORIGEN-S as in the CANDU test problems. As shown in Table 8, iBEST correctly predicted the reactor type as MAGNOX in all cases except for the one with 0.1 years of cooling time. Also, the program accurately estimated the burnups and uranium enrichments within 0.4% and 1.1%, respectively. The cooling time had slightly larger errors than the burnup and uranium enrichment, and the short cooling time (0.1 year) could not be accurately predicted, as in the CANDU cases.

3.2. *Mihama-3 test problems with PIE data*

For this section, we applied iBEST to the nine samples from the Mihama-3 reactor. To our knowledge, these nine cases are the only available problems with sufficient PIE measurement data for the validation of iBEST. The Post Irradiation Examination (PIE) were conducted in JAERI. Nine samples were taken from three fuel assemblies irradiated in the Mihama Unit 3 PWR reactors. Table 9 describes the atomic number density ratios for the monitor nuclides calculated using the data given by the OECD/NEA [12]. Table 10 summarizes the test results and the specifications, such as the burnups, uranium enrichments, and cooling times, which are exactly the same as those in Table 1. For these nine samples, we used ^{148}Nd as the burnup monitor and considered ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, and ^{241}Pu as the enrichment monitors. The reactor type

Table 7 – Atomic ratios of the monitor nuclides for the MAGNOX test problems.

Monitor type	Nuclides	Problem no.				
		1	2	3	4	5
Burnup	^{148}Nd	1.45E−04[a]	7.26E−05	5.46E−05	3.64E−05	7.26E−05
Enrichment	^{235}U	2.43E−03[a]	4.13E−03	4.71E−03	5.39E−03	4.13E−03
	^{236}U	8.01E−04	5.38E−04	4.45E−04	3.35E−04	5.38E−04
	^{238}U	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{239}Pu	2.45E−03	1.98E−03	1.74E−03	1.39E−03	1.98E−03
	^{240}Pu	1.25E−03	5.44E−04	3.65E−04	1.98E−04	5.44E−04
	^{241}Pu	2.27E−04	8.07E−05	4.81E−05	2.12E−05	1.02E−04
	^{242}Pu	9.15E−05	1.50E−05	6.52E−06	1.86E−06	1.50E−05
Reactor type	^{143}Nd	2.41E+00[b]	2.83E+00	2.96E+00	3.10E+00	2.73E+00
	^{148}Nd	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{240}Pu	8.64E+00	7.49E+00	6.70E+00	5.44E+00	7.49E+00
Age	^{106}Ru	[a]5.62E−06	2.29E−06	1.55E−06	8.82E−07	6.45E−05
	^{125}Sb	1.45E−06	6.40E−07	4.51E−07	2.74E−07	2.21E−06
	^{134}Cs	4.42E−06	1.11E−06	6.10E−07	2.51E−07	5.78E−06
	^{137}Cs	4.84E−04	2.41E−04	1.81E−04	1.20E−04	2.70E−04

[a] Atomic density ratio to ^{238}U.
[b] Atomic density ratio to ^{148}Nd.

Table 8 – Test results for the MAGNOX test problems.

Problem no.	RT[a]	Burnup (MWD/kg)		Enrichment (%)		Cooling time (y)	
		Reference	Predicted	Reference	Predicted	Reference	Predicted
1	Yes	8	8.020[b]/0.25[c]	0.711	[a]0.715/[b]0.56	5	[a]5.06/[b]1.19
2	Yes	4	4.016/0.40	0.711	0.719/1.11	5	5.09/1.77
3	Yes	3	3.004/0.13	0.711	0.717/0.84	5	5.05/0.99
4	Yes	2	2.002/0.10	0.711	0.718/0.97	5	5.04/0.79
5	No	4	4.016/0.25	0.711	0.719/1.11	0.1	0.17/41.18

RT, Reactor Type.
[a] Is reactor type predicted correctly?
[b] Predicted value.
[c] Discrepancy between the predicted and reference values (%).

Table 9 – Atomic number density ratios of the monitor nuclides for the Mihama-3 problems with PIE sample measurement data.

Monitor type	Nuclides	Problem no.								
		1	2	3	4	5	6	7	8	9
Burnup	^{148}Nd	1.50E−04[a]	1.25E−04	3.88E−04	2.79E−04	2.65E−04	5.42E−04	5.92E−04	6.20E−04	6.31E−04
Enrichment	^{235}U	2.55E−02[a]	2.68E−02	1.58E−02	2.00E−02	1.99E−02	1.08E−02	9.92E−03	8.67E−03	8.96E−03
	^{236}U	1.51E−03	1.31E−03	3.34E−03	2.68E−03	2.69E−03	3.73E−03	3.88E−03	4.09E−03	4.14E−03
	^{238}U	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{237}Np	7.01E−05	5.78E−05	—	1.65E−04	1.65E−04	—	—	—	4.98E−04
	^{239}Pu	3.13E−03	2.93E−03	5.31E−03	4.84E−03	4.93E−03	5.57E−03	5.79E−03	5.26E−03	5.62E−03
	^{240}Pu	4.35E−04	3.54E−04	1.55E−03	1.08E−03	1.10E−03	2.20E−03	2.40E−03	2.46E−03	2.56E−03
	^{241}Pu	1.12E−04	8.50E−05	6.75E−04	4.21E−04	4.39E−04	9.96E−04	1.12E−03	1.04E−03	1.13E−03
	^{242}Pu	9.71E−06	6.17E−06	1.82E−04	7.60E−05	7.72E−05	4.24E−04	5.19E−04	5.67E−04	5.94E−04
Reactor type	^{143}Nd	3.10E+00[b]	3.14E+00	2.60E+00	2.82E+00	2.83E+00	2.37E+00	2.30E+00	2.21E+00	2.20E+00
	^{148}Nd	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
	^{240}Pu	2.91E+00	2.82E+00	3.99E+00	3.86E+00	4.14E+00	4.06E+00	4.02E+00	3.93E+00	4.05E+00
Age	^{106}Ru	2.61E−06[a]	2.07E−06	7.85E−06	5.80E−06	5.77E−06	1.09E−05	1.27E−05	1.23E−05	1.27E−05
	^{125}Sb	9.48E−07	9.02E−07	2.54E−06	1.79E−06	1.77E−06	3.06E−06	3.59E−06	3.59E−06	3.59E−06
	^{134}Cs	3.08E−06	2.21E−06	1.98E−05	1.10E−05	1.11E−05	3.34E−05	4.02E−05	4.05E−05	4.28E−05
	^{137}Cs	4.86E−04	3.91E−04	1.28E−03	8.93E−04	8.88E−04	1.73E−03	1.93E−03	2.01E−03	2.03E−03

PIE, Post Irradiation Examination; RT, Reactor Type.
[a] Atomic density ratio to ^{238}U.
[b] Atomic density ratio to ^{148}Nd.

Table 10 – Test results for the Mihama-3 problems with PIE sample measurement data (without burnup correction).

Problem No.	RT[a]	Burnup (MWD/kg)		Enrichment (%)		Cooling time (y)	
		Reference	Predicted	Reference	Predicted	Reference	Predicted
1	Yes	8.3	8.11[b]/2.36[c]	3.208	3.30[b]/2.90[c]	5.0	5.25[b]/4.69[c]
2	Yes	6.9	6.80/1.46	3.208	3.32/3.28	5.0	4.96/0.71
3	Yes	21.2	20.82/1.80	3.203	3.31/3.34	5.0	4.97/0.52
4	Yes	15.3	15.03/1.80	3.203	3.34/4.01	5.0	5.31/5.85
5	Yes	14.6	14.32/1.99	3.203	3.26/1.88	5.0	4.69/6.67
6	Yes	29.44	28.90/1.87	3.210	3.22/0.46	5.0	5.58/10.42
7	Yes	32.3	31.51/2.52	3.210	3.29/2.31	5.0	5.00/0.01
8	Yes	33.7	32.99/2.16	3.210	3.16/1.74	5.0	5.22/4.13
9	Yes	34.1	33.52/1.74	3.210	3.25/1.34	5.0	5.32/6.03

PIE, Post Irradiation Examination; RT, Reactor Type.
[a] Is reactor type predicted correctly?
[b] Predicted value.
[c] Discrepancy between predicted and reference values (%).

monitors were ^{143}Nd, ^{148}Nd, and ^{240}Pu, exactly the same as those given by Scott [1], and we used ^{106}Ru, ^{125}Sb, ^{134}Cs, and ^{137}Cs as age monitors. The results given in Table 10 were obtained without burnup correction (only enrichment correction) to see the effects of burnup correction coupled with enrichment correction.

In this test, we first considered only two reactor type candidates, PWR and BWR. Their corresponding ORIGEN-S libraries are "PWR_W15x15" and "BWR_GE7x7-0," respectively. For all the cases, iBEST correctly identified the reactor type as PWR, whereas M.R. Scott reported that his program, NEMASYS, predicted PWR correctly only twice out of the nine samples. We attribute our better performance in predicting the reactor type to our use of ORIGEN-S and its burnup-dependent libraries rather than ORIGEN-2. Table 10 shows that iBEST accurately predicted uranium enrichment within 4.01% and burnup within 2.5%. However, the cooling time was predicted with larger errors (maximum error of 10.42% for the sixth sample). This larger error does not reflect the inability of iBEST, but it might stem from measurement uncertainties in the monitors because it gave quite accurate predictions for all the test problems in Section 3.1. For the first and second samples of the nine, we tested again with four reactor candidates (PWR, BWR, CANDU, MAGNOX) to show iBEST's ability to correctly predict the reactor type. iBEST again correctly identified the reactor type as PWR. We considered only the first two samples, which have low burnups, because the CANDU and MAGNOX libraries do not contain cross section data for higher burnups.

Next, we tested the samples with corrections for both uranium enrichment and burnup, with results given in Table 11. A comparison of the results in Tables 10 and 11 shows that with the corrections, iBEST returned larger discrepancies between the predicted and reported values of burnups than without corrections. The burnups predicted with the corrections are within 5.0%, and that error is considered acceptable for forensic applications. By contrast, as shown in Table 11, the discrepancies of uranium enrichment and cooling time predicted with the corrections are reduced compared with the no correction cases. Table 11 also reports the predicted values from Scott [1] for comparison with our results. This comparison shows that iBEST predicted the burnups and cooling times with similar discrepancies to those reported by Scott [1], whereas the discrepancies for uranium enrichments predicted by iBEST are slightly larger than those given by Scott [1], but they remain within 3.4%.

4. Conclusions

We successfully developed a program called iBEST to predict burnup history parameters, such as uranium enrichment, burnup, and cooling time, using isotopic measurement data from spent nuclear fuel. This program uses simple algebraic equations for the initial estimation and burnup to reduce computing time. Then, it corrects those initial estimations using a newly developed stable bisection method coupled with ORIGEN-S depletion calculations to improve the accuracy. The use of ORIGEN-S, with its burnup-dependent libraries, rather than ORIGEN-2, provides flexibility in the applicable reactor types and improves the accuracy. The validation of iBEST was done using a two-step procedure. In the first step, we tested iBEST with various test problems for different reactor types: PWR, BWR, CANDU, and MAGNOX. The test problems were prepared by extracting the atomic number densities of the monitor nuclides from ORIGEN-S output files following depletion calculations with the given burnups, uranium enrichments, and cooling times. The extracted atomic number densities were then used as input files for iBEST. The results show that iBEST correctly predicted the reactor types in all cases except for one MAGNOX case with an extremely short cooling time. iBEST also estimated the given burnups, uranium enrichments, and cooling times quite accurately for all test cases. In the second step, iBEST was applied to the nine samples of spent fuel from the Mihama-3 reactor with PIE isotopic measurement data. With those realistic problems, iBEST estimated the burnups, uranium enrichments, and cooling times within 5.1%, 3.4%, and 10.3%, respectively. In the results of test problems with known solutions, discrepancies between the values iBEST estimated and the measured values are partially caused by uncertainties

Table 11 – Test results for the Mihama-3 problems with PIE sample measurement data (with the burnup correction).

Problem no.	RT[a]	Burnup (MWD/kg)		Enrichment (%)		Cooling time (y)	
		Reference	Discrepancy[b]	Reference	Discrepancy[b]	Reference	Discrepancy[b]
1	Yes	8.3	4.38[c]/5.03[d]	3.208	0.37[c]/2.28[d]	5.0	3.09[c]/2.37[d]
2	Yes	6.9	3.32/4.19	3.208	1.90/2.66	5.0	3.95/2.27
3	Yes	21.2	3.93/3.48	3.203	0.09/2.44	5.0	6.38/3.14
4	Yes	15.3	4.37/4.01	3.203	2.05/3.40	5.0	4.60/3.37
5	Yes	14.6	3.97/4.03	3.203	0.09/0.96	5.0	10.62/10.21
6	Yes	29.44	3.68/3.21	3.210	0.00/0.49	5.0	0.60/5.55
7	Yes	32.3	4.43/3.63	3.210	2.43/1.40	5.0	3.09/1.53
8	Yes	33.7	4.11/3.22	3.210	0.31/2.42	5.0	1.38/2.69
9	Yes	34.1	3.58/2.78	3.210	0.00/0.70	5.0	0.20/2.20

iBEST, inverse Burnup ESTimator; PIE, Post Irradiation Examination; RT, Reactor Type.
[a] Is reactor type predicted correctly?
[b] Discrepancy between the predicted and reference values (%).
[c] Texas A&M University.
[d] iBEST.

in the measurements of the spent fuel samples. The results of the test problems and realistic Mihama-3 samples show that iBEST can successfully estimate burnup history parameters and be applied to spent nuclear fuels from various nuclear reactor types.

Conflicts of interest

The authors declare no conflict of interest.

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