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Original Article

Integral nuclear data validation using experimental spent nuclear fuel compositions

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ABSTRACT

Measurements of the isotopic contents of spent nuclear fuel provide experimental data that are a prerequisite for validating computer codes and nuclear data for many spent fuel applications. Under the auspices of the Organisation for Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA) and guidance of the Expert Group on Assay Data of Spent Nuclear Fuel of the NEA Working Party on Nuclear Criticality Safety, a new database of expanded spent fuel isotopic compositions has been compiled. The database, Spent Fuel Compositions (SFCOMPO) 2.0, includes measured data for more than 750 fuel samples acquired from 44 different reactors and representing eight different reactor technologies. Measurements for more than 90 isotopes are included. This new database provides data essential for establishing the reliability of code systems for inventory predictions, but it also has broader potential application to nuclear data evaluation. The database, together with adjoint based sensitivity and uncertainty tools for transmutation systems developed to quantify the importance of nuclear data on nuclide concentrations, are described.

1. Introduction

Integral benchmark experiments provide global measures of data performance for applications and are a valuable resource for nuclear data testing and evaluation efforts. Integral testing of the US ENDF/B-VII.1 nuclear data library [1] and several international nuclear data libraries rely heavily on testing using critical benchmarks as compiled in the International Criticality Safety Benchmark Experiment Project (ICSBEP) Handbook [2]. Integral testing of ENDF/B-VII.1 included more than 2,000 critical benchmarks for compounds, metals, solutions, and other mixed systems in thermal, intermediate, and fast neutron energy spectra. The use of criticality experiments as a centerpiece of data-testing efforts in the United States reflects both the investment in nuclear data development by the criticality community and the availability of high-quality and well-documented ICSBEP benchmarks.

Under the auspices of the Organisation for Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA), physics benchmarks have been compiled and documented in the International Reactor Physics Benchmark Experiment (IRPhEP) Handbook [3] that exercise nuclear data in broader range of applications. As of 2014, the IRPhEP Handbook contained 136 evaluations and measurements of critical buckling, spectral characteristics, reactivity coefficients, kinetics, reaction rates, and power distribution measurements. Both the ICSBEP and IRPhEP benchmarks are applied to realistic applications, including uncertainty analysis and cross section adjustment, uncertainty evaluation for reactor core design methods, criticality safety problem validation, and cross section data testing [4]. However, an intrinsic limitation of both the ICSBEP and the IRPhEP benchmarks is that their focus, for the most part, is on static systems of unirradiated nuclear materials. Thus, many of these benchmarks exercise data for predominantly uranium and plutonium metals and compounds, moderators/reflectors materials, and absorber (criticality control) materials. There are relatively few benchmarks for irradiated nuclear materials.

Interim dry fuel storage research and licensing activities are increasing as spent fuel pools at nuclear plants in many countries

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are reaching capacity. In addition, geological repositories in Finland and Sweden are expected to be operational in the next decade. Recognizing the need for improved experimental benchmarks to support international activities in spent fuel management, the Expert Group on Assay Data for Spent Nuclear Fuel (EGADSNF), under the guidance of the Working Party on Nuclear Criticality Safety of the OECD/NEA, has developed and expanded a database of spent fuel isotope compositions obtained by destructive radiochemical analysis of irradiated fuel samples from commercial nuclear reactors. Under this initiative, the Spent Fuel Compositions database, SFCOMPO 2.0, originally developed by the Japan Atomic Energy Research Institution [5], has been expanded from 246 samples to more than 750 samples from 44 reactors representing eight different international reactor technologies.

Because of the design complexities and uncertainties in the actual conditions of an operating reactor, applying these data to obtain realistic estimates of the accuracy of calculated results can be extremely difficult. In addition, there can be large uncertainties in the measurement data attributed to the complex radiochemical measurement processes not present in criticality and reactor physics experiments. Under the OECD/NEA, these data have been more completely documented and peer reviewed for accuracy by international experts. Although measurement accuracies and the quality and extent of documentation often fall short of the high standards set by the ICSBEP and IRPhEP benchmarks, these measurements nevertheless represent valuable validation tests and have been widely used to evaluate the performance characteristics of computer codes, models, and nuclear data used for calculating spent fuel compositions [6,7].

Because of the significant complexity of spent fuel measurements and past inconsistent documentation of reactor design, operating data, and the design and measurement uncertainties, these measurements have not been widely adopted for integral testing of nuclear data. France is an exception, where measurements of irradiated fuel compositions from the French Gravelines and Bugey reactors, and the Swiss Beznau reactor, were used in the development and validation of the European JEF-2.2 libraries [8]. More recently, additional assay measurements, including data made available in SFCOMPO, have been used in the validation of JEFF-3.1 [9,10].

Unlike the ICSBEP and IRPhEP benchmarks involving primarily steady-state systems, spent fuel experiments involve time-dependent isotope depletion and decay as defined by the transmutation equations. Therefore, identifying the relationships between computational bias and nuclear data is particularly challenging. Tools to help understand and quantify the complex relationships between nuclear data and measured compositions are therefore needed in order to better utilize databases such as SFCOMPO 2.0 for nuclear data testing and evaluation.

This paper describes the current status of the new SFCOMPO 2.0 database and benchmark development efforts, and it summarizes and demonstrates new nuclear data sensitivity and uncertainty analysis tools for depletion problems being developed and targeted for future release in the SCALE nuclear systems analysis code system [11].

2. SFCOMPO database

2.1. Database content

SFCOMPO 2.0 is a database of well-documented, peer-reviewed measurements of spent fuel assay data [12] that can be used by the nuclear community for validating computational systems and nuclear data in applications of reactor physics, nuclear fuel cycle, used fuel storage and disposal, as well as advanced fuels and reactor design concept evaluations. The measured assay data have been compiled and reviewed by international radiochemistry experts and modeling practitioners as coordinated through the activities of the OECD/NEA EGADSNF [13].

Since 2007, the database has been expanded beyond pressurized water reactor (PWR) and boiling water reactor (BWR) fuel types that were included in earlier versions. The database now includes eight different reactor designs: light water PWR, BWR, water–water energetic reactor (VVER)-440, and VVER-1000; heavy water CANDU (CANada Deuterium Uranium), graphite-moderated Magnox, AGR (advanced gas-cooled reactor), and RBMK (Reaktor Bolshoy Moshchnosti Kanalnyy) reactor designs. Measurements have been acquired from 67 different fuel assemblies and 291 fuel rods. The updated database, SFCOMPO 2.0, contains more than 20,000 measurement entries, with associated uncertainties as reported by the measurement laboratories.

Each reactor design class includes a range of fuel assembly designs. The diversity of designs, coolants, moderators, enrichments, and fuel burnup values provides a wide range of neutron spectral conditions during irradiation that can be used to test the performance of nuclear data over different neutron energy regions. For example, the database includes spent fuel measurements for samples with enrichments from 0.71 wt.% (natural U) to 4.9 wt.%, and burnup values from 0.85 GWd/tU to more than 70 GWd/tU. Coolant and moderator materials include light water, heavy water, gas, and graphite. The reactors, country of origin, and the number of measured fuel samples available for each reactor are summarized in Table 1.

The database is designed with a top-to-bottom hierarchical structure organized by reactors, fuel assemblies, fuel rods, and samples, with the measurement data usually being associated at the sample level. However, some experiments involving dissolution of entire assemblies from a reprocessing plant are included and are therefore associated with the assembly level.

The database includes measured nuclide concentrations, activities, isotopic atom and mass ratios, measurement method, uncertainty, laboratory, and measurement date. Measurements for 91 isotopes are included (Fig. 1). Measurement data are always included as the values and units reported by the laboratory or experimental program. Where possible, the measurement data are automatically converted by the program to a common mass basis of mg/g fuel or mg/g initial U.

With the addition of newer experimental program data that include high-precision measurements and improved design and operating data documentation, the quality of the database has improved significantly compared to earlier versions. Moreover, as part of the data review process, multiple independent cross checks of the information have been performed. Also, preliminary benchmark models have been developed for many of the experiments.

An important caveat on the database is that these reviews have been performed to ensure that the database accurately reflects the experimental data *as reported* and are traceable to primary reference reports. The reviews do not constitute a formal evaluation of the data to assess quality and accuracy of the measurements, consistency of the data, completeness of the documentation, development of benchmarks, or an analysis of uncertainties. However, procedures for such evaluations have been developed [14], and preliminary detailed evaluation reports have been initiated for several datasets.

2.2. Database application interface

SFCOMPO 2.0 is a structured query language Java H2 relational database accessible through a graphical application interface developed by the NEA. The database has a similar interface and functionality as the IRPhEP Database and Analysis Tool [15] and

Table 1
Summary of SFCOMPO 2.0 database content.

Design	Reactor	Country	Samples
AGR	Hinkley—3	United Kingdom	21
	Hinkley—4	United Kingdom	36
	Hunterston B—1	United Kingdom	6
BWR	Cooper	United States	17
	Dodewaard	The Netherlands	5
	Forsmark—3	Sweden	2
	Fukushima Daiichi—3	Japan	36
	Fukushima Daini—1	Japan	13
	Fukushima Daini—2	Japan	44
	Garigliano—1	Italy	26
	Gundremmingen—1	Germany	18
	JPDR	Japan	30
	Monticello	Italy	30
	Quad Cities—1	United States	18
	Tsuruga—1	Japan	10
CANDU	Bruce—1	Canada	3
	NPD	Canada	27
	Pickering A—1	Canada	1
Magnox	Bradwell—1	United Kingdom	1
	Hunterston A—1	United Kingdom	3
PWR	Beznau—1	Switzerland	6
	Calvert Cliffs—1	United States	33
	Genkai—1	Japan	2
	Gösgen—1	Switzerland	4
	H. R. Robinson—2	United States	7
	Mihama—3	Japan	9
	Neckarwestheim—2	Germany	1
	Obrigheim—1	Germany	33
	Ohi—1	Japan	1
	Ohi—2	Japan	5
	Takahama—3	Japan	16
	Three Mile Island—1	United States	24
	Trino Vercellese—1	Italy	49
	Turkey Point—3	United States	18
	Vandellos—2	Spain	9
	Yankee	United States	78
RBMK	Leningrad—1	Russia	41
VVER-1000	Balakovo—2	Russia	3
	Balakovo—3	Russia	2
	Kalinin—1	Russia	5
	Novovoronezh—5	Russia	10
VVER-440	Kola—3	Russia	12
	Novovoronezh—3	Russia	7
	Novovoronezh—4	Russia	28

AGR, advanced gas-cooled reactor; BWR, boiling water reactor; CANDU, CANada Deuterium Uranium; PWR, pressurized water reactor; RBMK, Reaktor Bolshoy Moshchnosti Kanalnyy; SFCOMPO, Spent Fuel Compositions database; VVER, water–water energetic reactor.

Database for the ICSBEP [16] programs, also developed by the NEA. Information in the database can be accessed by browsing the data in a hierarchical organization structure by reactor technology type (e.g., BWR, Magnox), reactor name, assembly ID, fuel rod, and measured fuel sample level. Fig. 2 illustrates the database structural organization as viewed in the application interface browser, expanded to show the data for one of the Fukushima-Daini-1 reactor data sets. Fig. 3 provides a screen shot of the database browse panel showing the organizational structure (left side) and the display of design and operating information at the assembly level.

A major advancement is the ability to store and display more complete and detailed fuel and assembly design information and time-dependent operating history data for the reactor, assembly, fuel rod, and measured sample. SFCOMPO 2.0 also provides references for all the data it displays. These references are linked to primary reports such as radiochemistry laboratory reports, open journal papers, and assay data evaluation reports, providing full traceability of the data. Primary experimental and reference reports are accessible as electronic portable document format (PDF) versions of the reports that can be accessed through a built-in viewer.

2.3. Search capabilities

Many database fields can be queried using the *Basic* and *Advanced* search panels. The *Basic* search query includes reactor type, measured fuel type (i.e., U-metal, UO_2 or MOX, UO_2-Gd_2O_3), sample burnup range, and measured nuclide (see the nuclide search panel in Fig. 1). *Advanced* query parameters include the following:

- Reactor name
- Coolant type
- Moderator type
- Assembly identifiers
- Lattice type
- Lattice dimension
- Fuel rod pitch (range)
- Fuel rod diameter (range)
- Enrichment (range)
- Axial position or height of the sample (range)
- Measurement type
- Measurement uncertainty (range)

These search features allow the user to rapidly identify measurement data for isotopes, the reactor, and fuel design parameter characteristics of interest. The search panel returns the nuclide concentration values and uncertainties of experimental measurements as queried by the user for inspection and comparison purposes. SFCOMPO 2.0 allows the user to visualize measurement data of selected samples and provides the numeric measurements data, along with other information queried. The search results list the sample level information for all samples matching the search criteria and the measurement data in separate panels. A graphical plotter displays nuclide concentrations provided in normalized units of initial (unirradiated) fuel mass as a function of sample burnup. A more general plotting capability can be used to display most of the measurement data against other fields used as the *x* axis. Fig. 4 shows the results of a database query for ^{239}Pu measurements for all uranium-based fuels and a plot of the results and uncertainties as a function of sample burnup.

3. Applications of the database

Destructive radiochemical assay data are one of the primary means used to validate depletion codes and nuclear data libraries for spent fuel applications. Measured isotopic data in SFCOMPO 2.0 include many nuclides that dominate applications in nuclear criticality safety (burnup credit), gamma radiation sources, passive neutron sources, decay heat, and long-lived radiological isotopes important to geological repository disposal. A widely used approach to burnup code and data validation is to apply the code to calculate the same nuclide inventories for which measurements are available. This technique has underpinned uncertainty analysis for many spent fuel applications, including burnup credit, in nuclear criticality safety. In addition to measured nuclide concentration data, these applications also require the detailed design and reactor operating conditions needed to model the fuel assembly irradiation and predict the nuclide concentrations in the fuel.

Another application of the data includes the use of measured data alone without the need to model individual experiments. SFCOMPO 2.0 has been used recently to develop uncertainty bands for waste categorization in instances where detailed information on the waste is not available [17]. For this application, there is no requirement for detailed operating history information.

Comparing calculated-to-measured nuclide concentrations provides information necessary to validate codes and estimate the

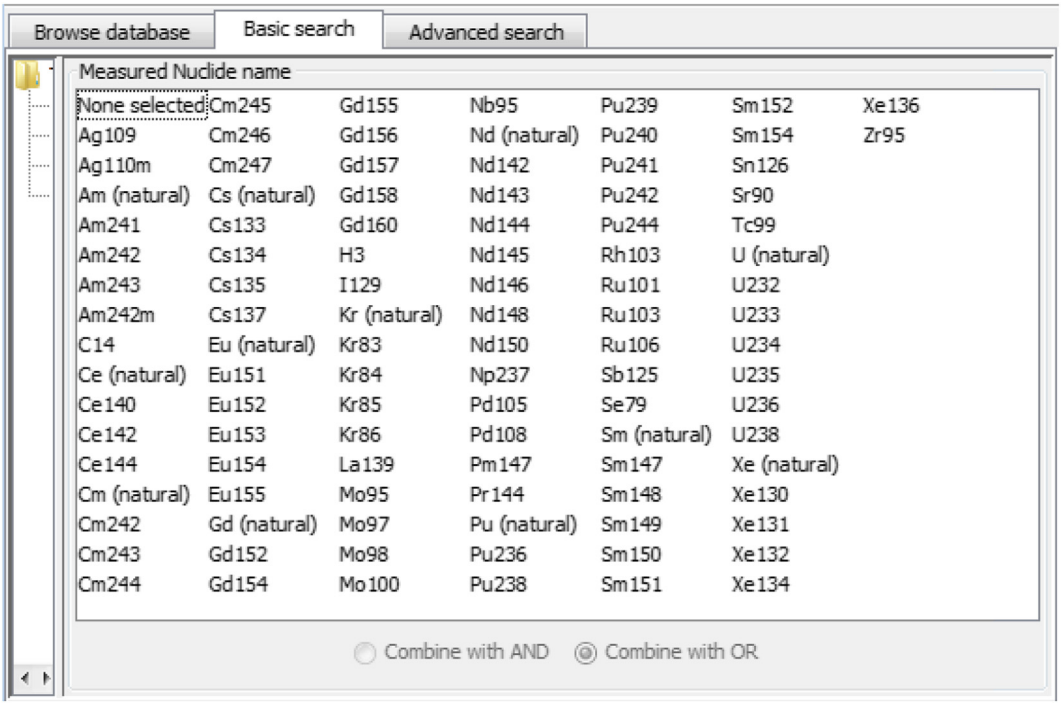

Fig. 1. Measured isotopes available in SFCOMPO 2.0. SFCOMPO, Spent Fuel Compositions database.

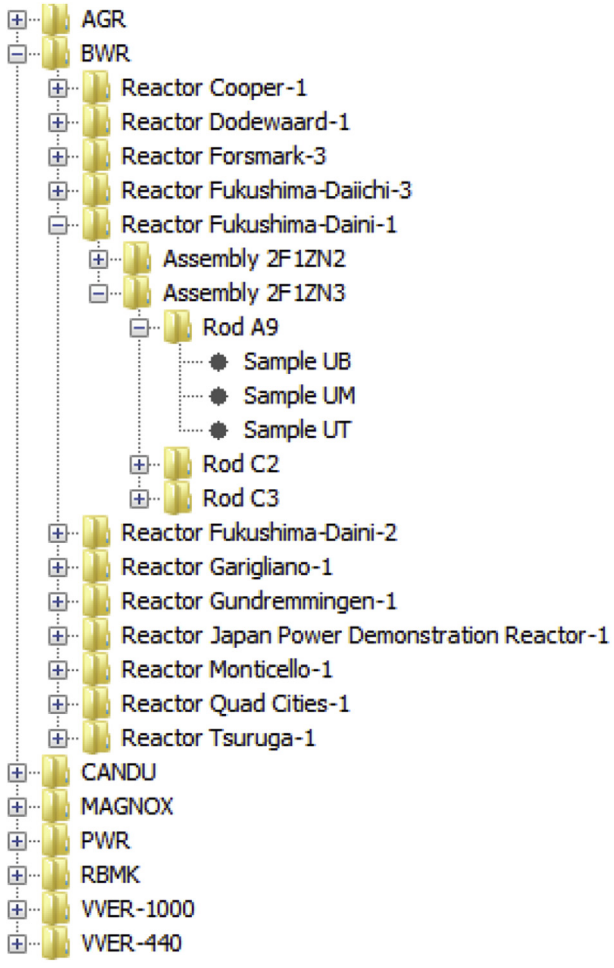

Fig. 2. Screen capture of the SFCOMPO 2.0 organizational structure showing information for the Fukushima-Daini-1 samples of assembly 2F1ZN3 rod A9. SFCOMPO, Spent Fuel Compositions database.

bias and uncertainty in isotope predictions and other integral quantities. However, these comparisons do not provide direct information on the accuracy of the specific nuclear data used by the codes. The time-dependent relationship between nuclide concentrations, N_i, and nuclear data is given by the transmutation equation

$$\frac{\partial N_i}{\partial t} = \underbrace{-\lambda_i N_i + \sum_j \lambda_j^{j\to i} N_j}_{\text{Decay}} + \underbrace{\sum_{j'} N_{j'} \langle \sigma_{j'} \rangle^{j\to i} \langle \phi \rangle - N_i \sum_{\forall r} \langle \sigma_i \rangle^{(r)} \langle \phi \rangle}_{\text{Reactions}} \quad (1)$$

where decay constants λ, energy-averaged neutron cross sections σ and energy-integrated neutron flux ϕ define the transition rates. Provided that the system description is given such that accurate estimates of $\phi(E)$ can be obtained by transport calculations of the fuel assembly, and the nuclide content is measured with low uncertainty, spent fuel measurements can be used for nuclear data testing because the results of computation simulations of these experiments rely directly on the accuracy of the data used by the codes.

Application to nuclear data evaluation is complicated by the large amount of data used by the codes for burnup simulations and time dependence of data importance in transmutation analysis. The production of any nuclides usually depends on cross sections and half-lives for many different nuclides in the production chain, and fission products also depend on their fission yields. Software tools to aid in the evaluation of nuclear data uncertainty in transmutation systems are being developed at Oak Ridge National Laboratory (ORNL). The application of these tools to nuclear data evaluation and testing using spent fuel databases such as SFCOMPO is briefly summarized below.

4. Tools for nuclear data assessment

Computational analysis of irradiated fuel compositions can provide a wealth of information concerning bias and uncertainty in the nuclear data used in modeling and simulation codes, including

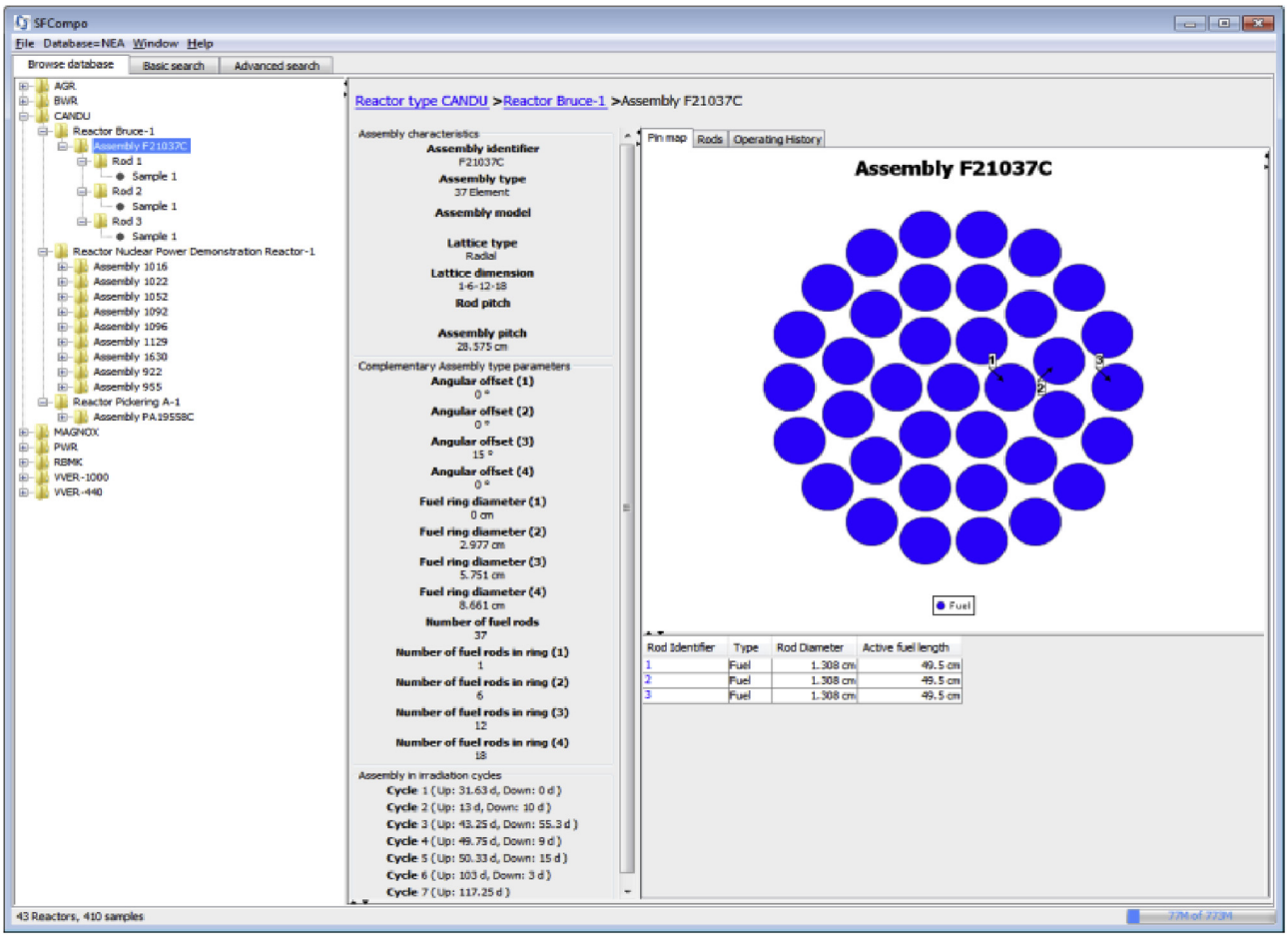

Fig. 3. Browser panel showing CANDU reactor design data (37-element design) and fuel bundle map of measured fuel rod locations. CANDU, CANada Deuterium Uranium.

neutron cross section data, fission product yields, and nuclear decay data. Transmutation calculations use a large amount of nuclear data, and it is essential in nuclear data testing to identify which data have the largest sensitivity on the responses and which data are therefore most responsible for biases in the calculated results. The transmutation matrix used by ORIGEN contains more than 50,000 transition element coefficients that represent energy-averaged neutron reactions, decay branch transitions, and fission product yields. The cross sections alone include data for 774 nuclides and 14,000 neutron reactions. Therefore, direct perturbation of nuclear data for these complex systems to determine the importance of nuclear data on different observables is often not efficient or practical.

An efficient approach to calculating nuclear data sensitivities and uncertainties from nuclear data is to use adjoint-based perturbation theory. Adjoint techniques for criticality safety analyses have been developed and are now widely adopted for nuclear data sensitivity analysis, which is required for numerous applications including experiment similarity analysis [18] and nuclear data adjustment for eigenvalue problems [19]. However, similar techniques are not widely available to analyze time-dependent transmutation or depletion systems. The development of the adjoint equations for depletion was first published by Gandini [20] and implemented in the ORIGEN code by Williams and Weisbin [21]. This implementation of perturbation theory assumes that the nuclide field is uncoupled from the neutron flux field, i.e., a perturbation of the nuclide field does not significantly affect the flux field, and each can be varied independently. A more general perturbation formulation for the space–energy-dependent burnup equations in a coupled neutron–nuclide field, such as in a reactor core, has also been developed [22].

In the uncoupled formalism, the absolute sensitivity of a calculated response R to nuclear data element α can be shown to be,

$$S_\alpha^R = \left\langle N_R^*(t)\frac{\partial A(\alpha)}{\partial \alpha}N^T(t)\right\rangle, \tag{2}$$

where N^{T} is the transpose of the forward solution (column) of the nuclide composition vector and N_R^* is the adjoint solution of the nuclide vector solved for a user defined response R, and $A(\alpha)$ is the transmutation matrix containing transition elements α that define the energy-average neutron cross sections, the fission product yields, and decay transition data [18]. This expression is integrated from $t = 0$ to $t = T_f$. Note that the partial derivative of $A(\alpha)$ with respect to α results in zeros in the matrix locations where the data element α does not contribute. The sensitivity coefficients in Eq. (2) give the absolute change in a defined response R to a change in data parameter α. The response can be defined for an individual isotope or aggregate isotopes that contribute to an integral response such as decay heat, gamma emission, or other spent fuel properties.

The forward transmutation equation is expressed in matrix notation as

$$\frac{dN(t)}{dt} = A\,N(t), \quad t\in\left(0, T_f\right), \tag{3}$$

with the solution

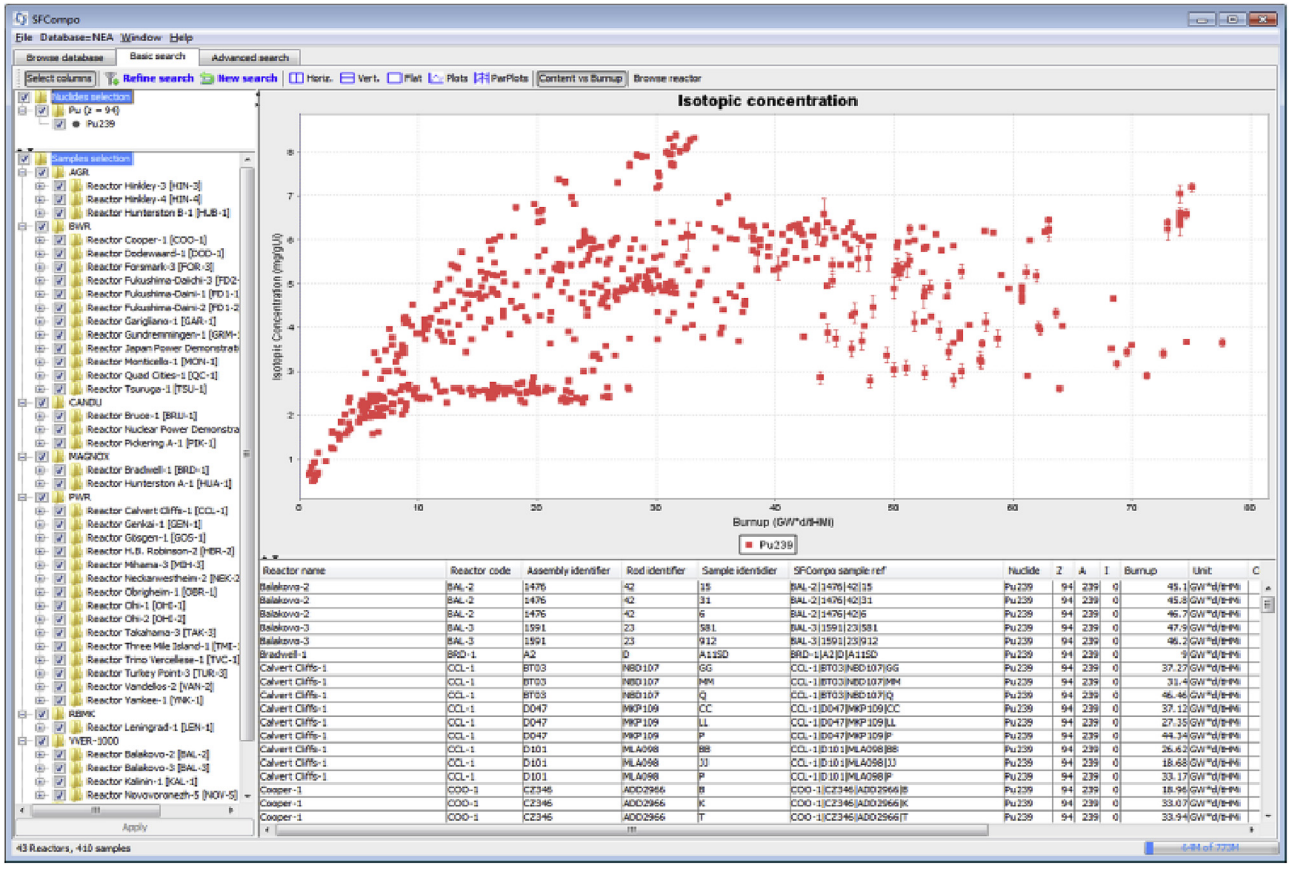

Fig. 4. Query results and plot of measured ^{239}Pu concentration data for all uranium-type fuels.

$$N(t) = N_0 e^{At}, \tag{4}$$

where N_0 is the known initial value of the nuclide concentration vector at $t = 0$. The adjoint transmutation equation, in the same notation, is

$$-\frac{dN(t)^*}{dt} = A^T N(t)^* \quad t \in \left(T_f, 0\right) \tag{5}$$

and $N(t)^*$ is defined at the end of irradiation, $t = T_f$ for the nuclide response of interest. The adjoint equation is very similar to the forward transmutation equation, except the time derivative is negative, and the transmutation matrix A is transposed. As indicated by the negative derivative in Eq. (5), the adjoint essentially solves the transmutation equation backward in time as a final-valued problem, where the final condition is equal to the response-function nuclide vector. In addition to using the adjoint solution to determine nuclear data sensitivities as defined in Eq. (2), the adjoint function at t=0 is the absolute sensitivity coefficient for the initial condition.

$N(t)^*$ is a nuclide importance vector that carries the nuclide contribution to the final response at time t.

ORNL recently developed and implemented an adjoint depletion solution in the ORIGEN code [23], which was released in SCALE 6.2. A new solver applies the Chebyshev Rational Approximation Method (CRAM) to approximate the exponential matrix function,

$$e^{At} \approx a_0 + 2Re\left[\sum_{i=1}^{k/2} a_i (At + \theta_i I)^{-1}\right] \tag{6}$$

where a_0 is the limiting value of the approximation at infinity, a_i and θ_i are the residues and the poles of the rational function, and k is the order of the polynomials used in the CRAM method. This method allows the transmutation equation to be solved using a single numerical method, without the need, as in previous implementations in the ORIGEN code, to remove short-lived isotope chains from the matrix that are then solved separately using Bateman solutions. The single CRAM solution has substantially facilitated the implementation of the adjoint solution.

The forward solution N and adjoint solution N^* can be used in perturbation theory-based sensitivity and depletion uncertainty analysis using Eq. (2) to calculate sensitivity coefficients that relate nuclear data to uncertainty in nuclide concentrations after a specified irradiation and decay period. Using this approach, the importance of all 50,000 nuclear data elements in transmutation matrix on the production of any nuclide, group of nuclides, or integral response of interest can be quantified using a single forward and adjoint ORIGEN calculation. This capability provides an important tool for evaluation of spent fuel isotopic composition experiments, enabling the identification of important nuclear data responsible for formation of measured isotopic concentrations.

Although sensitivities provide the importance of nuclear data, they do not reflect the uncertainties in the nuclear data. Uncertainties in the nuclear data propagate through the transmutation calculations and produce uncertainties in the calculated

nuclide concentrations and other responses of interest. Sensitivities can be used to obtain the response uncertainty because

$$\Delta R = S_\alpha^R \Delta\alpha \tag{7}$$

Uncertainty contributions to response R from all nuclear data elements can be determined if data uncertainties or covariances, $C_{\alpha\alpha}$, are available as

$$Var(R) = \left\langle S_\alpha^T C_{\alpha\alpha} S_\alpha \right\rangle = \tag{8}$$

$$\underbrace{[S_{\alpha1} S_{\alpha2} S_{\alpha3}]}_{S_\alpha^T} \underbrace{\begin{bmatrix} \alpha_{\alpha1}^2 & \sigma_{\alpha1}c_{1,2}\sigma_{\alpha3} & \sigma_{\alpha1}c_{1,3}\sigma_{\alpha3} \\ \sigma_{\alpha2}c_{2,1}\sigma_{\alpha1} & \alpha_{\alpha2}^2 & \sigma_{\alpha2}c_{2,3}\sigma_{\alpha3} \\ \sigma_{\alpha3}c_{3,1}\sigma_{\alpha1} & \sigma_{\alpha3}c_{3,2}\sigma_{\alpha2} & \alpha_{\alpha3}^3 \end{bmatrix}}_{\text{Reactions}} \underbrace{\begin{bmatrix} S_{\alpha1} \\ S_{\alpha2} \\ S_{\alpha3} \end{bmatrix}}_{S_\alpha} \tag{9}$$

A prototype code for nuclear data sensitivity analysis using ORIGEN is being developed and tested. This code, Orsen (ORIGEN sensitivity analysis), automates the process of performing the forward and adjoint depletion calculations and processing the output to provide nuclear data sensitivities for all data used in the calculation. This capability will be extended to include data uncertainty analysis using nuclear data covariance data. The basic steps in the procedure are shown in Fig. 5.

As a demonstration case, the sensitivity coefficients that relate data perturbations to perturbations in nuclide concentrations after some specified burnup and decay period are calculated using Orsen for the production of ^{238}Pu from low-enriched uranium fuel with a burnup 30 GWd/t. The relative sensitivity coefficients (S) for all nuclear data are obtained and ranked. The data with the largest sensitivities are listed in Table 2. These coefficients represent the magnitude of changes in the nuclear data on the concentration of ^{238}Pu. A coefficient of unity means that a 1% change in the nuclear data will result in a 1% change in the ^{238}Pu concentration. A negative coefficient means that an increase in the nuclear data quantity will decrease the predicted ^{238}Pu concentration.

In this example, the production of ^{238}Pu is most sensitive to the neutron capture cross sections of ^{237}Np, ^{235}U, and ^{236}U. This indicates that the major production path starts with ^{235}U through the chain:

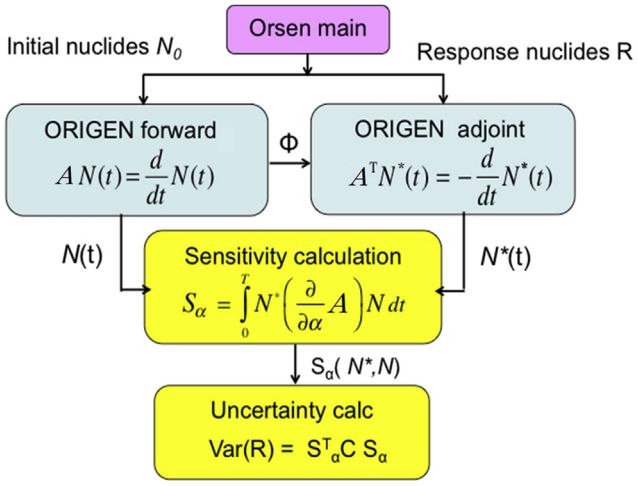

Fig. 5. Orsen code for sensitivity and uncertainty analysis. Orsen, ORIGEN sensitivity analysis. ORIGEN, Oak Ridge Isotope Generation code; A, matrix of nuclear data transition elements α; N, nuclide concentration vector; S, sensitivity coefficients; C, covariance matrix; Var(R), variance of nuclide response R.

Table 2
Relative sensitivity coefficients for ^{238}Pu production data.

Parent nuclide	Product nuclide	Data type	S
^{237}Np	^{238}Np	(n,γ)	0.6136
^{235}U	^{236}U	(n,γ)	0.5928
^{236}U	^{237}U	(n,γ)	0.5877
^{241}Pu	^{241}Am	Half-life	0.2132
^{238}U	^{239}U	(n,γ)	0.2155
^{242}Cm	^{238}Pu	Decay branch	0.2182
^{241}Pu	^{241}Am	Decay branch	0.2176
^{242}Am	^{242}Cm	Decay branch	0.2175
^{239}Pu	^{240}Pu	(n,γ)	0.1740
^{238}U	^{237}U	(n,2n)	0.1576
^{238}Pu	^{239}Pu	(n,γ)	−0.1475

S, Relative sensitivity coefficients of the nuclide concentration to the nuclear data.

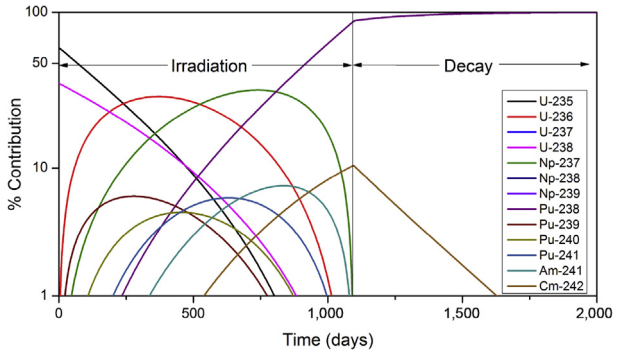

Fig. 6. Application of the adjoint solution to identify the contribution pathways for the example of ^{238}Pu production. The calculation assumes a 3-year irradiation time of PWR fuel (33 GWd/tU) and 3-year decay time. PWR, pressurized water reactor.

$$^{235}\mathrm{U}(\mathrm{n},\gamma) \rightarrow {}^{236}\mathrm{U}(\mathrm{n},\gamma) \rightarrow {}^{237}\mathrm{U}\left(\beta^-\right) \rightarrow {}^{237}\mathrm{Np}(\mathrm{n},\gamma) \rightarrow {}^{238}\mathrm{Np}\left(\beta^-\right) \rightarrow {}^{238}\mathrm{Pu}. \tag{10}$$

High sensitivities for decay branching fractions (fractions for each mode of decay) of ^{242}Cm, ^{241}Pu, and ^{242}Am indicate that the ^{238}Pu buildup from the chain beginning with ^{238}U is also important. The relative importance of all nuclear data to any measured nuclide concentration can similarly be quantified.

Another application of the sensitivity analysis tools is pathway analysis, shown in Fig. 6, using the function $N_R^*(t)N^T(t)$. This plot, for the production of ^{238}Pu, shows the contribution of each precursor nuclide to the final response as a function of time. For this example of a final response of ^{238}Pu atoms produced, this figure shows where the atoms that contribute to final ^{238}Pu reside in the transmutation chain for times less than T_f. The figure, at $t = 0$, also shows that about 62% of the ^{238}Pu concentration comes from ^{235}U and 38% comes from ^{238}U.

5. Conclusions

Increasing international interest in spent fuel isotopic data to validate computation systems extends to many scientific applications involving spent fuel, including nuclear criticality safety, fuel cycles, spent fuel storage, transportation, geological disposal, and advanced reactor and fuel design.

The OECD/NEA, with support from many international organizations and experts, has led efforts to compile and expand experimental spent fuel measurement data representative of the world's current and past fleet of commercial reactors. This database, SFCOMPO 2.0, was developed to serve as a central resource of reliable, well-documented experimental assay data, with design

specifications and operating information for use by the international scientific community, industry, and regulatory authorities. The efforts of the EGADSNF have resulted in a 3-fold increase in the number of measurements as well as improved quality and documentation of the data through the contributions of experts in NEA member countries engaged in compiling and reviewing data. The EGADNSF plans to progressively expand the database and initiate development of benchmark models for each dataset with recommended model inputs and calculated results. Through the contributions and effort of NEA member countries, SFCOMPO 2.0 is today's largest, freely accessible open source of experimental isotopic assay data for international reactor designs.

Application of spent fuel measurements has not been universally adopted for integral nuclear data testing, and isotopic assay data are a largely underutilized resource in many data evaluation activities, including ENDF/B. Because nuclear data largely define the accuracy of modeling and simulation codes used for fuel composition calculations, results from depletion simulations can provide important integral information on data performance. Recent and ongoing developments of adjoint-based depletion sensitivity and uncertainty methods can be used to provide additional insight into the complex relationships between nuclear data and nuclide inventories and can help define a quantitative approach to data evaluation.

With the expansion of data representing a wide range of enrichments, burnups, fuel types, moderator and coolant types, along with tools to help the evaluator identify nuclear data importance, SFCOMPO 2.0 can provide valuable information for nuclear data testing for materials, nuclear data and reaction processes that cannot be accomplished using traditional steady-state benchmarks.

Conflicts of interest

All authors have no conflicts of interest.

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