ADVANCES IN STUDSVIK'S SYSTEM FOR SPENT FUEL ANALYSES

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ABSTRACT

Studsvik's approach to spent nuclear fuel analyses combines isotopic concentrations, fluxes, and cross-sections, calculated by the CASMO5 neutron transport and depletion code, with irradiation history data from the SIMULATE5 reactor core simulator and tabulated isotopic decay data. These data sources are used and processed by the SNF code to compute the spent nuclear fuel characteristics. Recent advances in the system, including cross-sections and decay data from ENDF/B-VIII.R0, are presented in this paper, together with validation results against decay heat power and isotopic compositions measurements. Measurements conducted at the Swedish interim storage facility, CLAB, are used for validation of the decay heat power, while comparisons to the results of the international program ARIANE are used to demonstrate the capability of CMS5/SNF to accurately predict isotopic compositions. The paper shows the results calculated with ENDF/B-VIII.R0, and the effect on the spent fuel characteristics is evaluated by comparisons to the earlier ENDF/B-VII.R1 results.

KEYWORDS: SNF, ENDF, CLAB, ARIANE

1. INTRODUCTION

Studsvik's approach to spent nuclear fuel analyses combines isotopic concentrations, fluxes, and cross-sections, calculated by the neutron transport and depletion code CASMO5 [1], with irradiation history data from the reactor core simulator SIMULATE5 [2] and tabulated isotopic decay data. These data sources are used and processed by the SNF code [3,4] to compute the basic spent nuclear fuel characteristics: isotopic concentrations; radioactivity; decay heat power; photon sources and spectra; neutron source and spectra from spontaneous fission; neutron source and spectra from (α ,n) reactions.

This code system is fully integrated with Studsvik's Core Monitoring System (CMS5) and implicitly employs the three-dimensional (3D) discretization of the assemblies in the reactor core. Designed and implemented as a production tool, it provides comprehensive results per axial node of a fuel assembly or a fuel pin. In addition to the basic spent fuel characteristics, the SNF code is supplied with various optional modules intended to support and facilitate data preparation for analyses related to handling, transportation and storage of spent fuel, including accident analyses, burnup credit in criticality safety evaluations, and dry cask loading scenarios. The decay heat power, computed by the summation method, is complemented

by two standards for decay heat power in light water reactors: ANSI/ANS-5.1-2014 [5] and DIN-25463-1/2-2014 [6], implemented as integrated methodology or as stand-alone applications.

The latest set of decay data implemented in the SNF code is based on ENDF/B-VII.R1 [7]. With the release of ENDF/B-VIII.R0 [8] and following its implementation [9] in CASMO5, a new decay data set based on ENDF/B-VIII.R0 was generated. Its application to two validation benchmarks is presented in this work, along with a brief description of the methodology.

2. METHODOLOGY

The cross-section library, generated by the lattice physics code CASMO5, provides isotopic concentrations tabulated by exposure, moderator density history, control rod history, fuel temperature, and boron history. The nodewise exposure and accumulated history parameters, obtained from qualified operational reactor data or code simulation (SIMULATE5), are used as entry points in the interpolation routines and, with the power history model, are used to compute the end-of-life (EOL) isotopic concentration. The fuel assembly is divided into axial nodes (index: K) that may be further divided into sub-nodes (N). A sub-node is characterized by its associated lattice type (L). The concentration C_{base} of a given isotope (J) in sub-node (N) of node (K) is calculated as:

$$C_{base}^{J,K,N} = C_{tab}^{J,L}(E) + \sum_{h \in H} \Delta C_{tab}^{J,L,h}(E) * \Delta H_h, \quad (1)$$

where:

- $C_{tab}^{J,L}(E)$ is the concentration (g/t) of isotope J in lattice type L at exposure E;
- $\Delta C_{tab}^{J,L,h}(E)$ is the history-differential (difference between base and alternate depletion histories) concentration of isotope J in lattice type L at exposure E;
- ΔH_h is a nodal history effect (moderator density, control, boron, void, fuel temperature, etc.).

In this equation, all concentrations are obtained by interpolation in two-dimensional tables for lattice type L to the nodal exposure E. The nodal concentrations, $C_{base}^{J,K}$, are obtained by a weighted average over the sub-nodes in each node. Further, the individual isotope concentrations are affected by the nodal power variations, the effect of which can be significant for many isotopes. SNF makes use of the nodal power histories (from the 3D core simulator) to apply "power history correction factors", $C_{PHIST}^{J,K}$:

$$C_{EOL}^{J,K} = C_{PHIST}^{J,K} * C_{base}^{J,K} \quad (2)$$

to the nodal concentrations. The power history correction factor for isotope J in node K with burnup E is defined as the ratio of the final concentration, C_J , computed with the actual power history, to the reference concentration, C_J^{ref} , computed with the reference power level at the same burnup.

The EOL isotopic concentrations are then applied to the decay chains resolved by Bateman system. The Bateman system is solved analytically by a method from [10] to obtain the final concentration, $C_t^{J,K}$, at decay time t. The decay chains are built exactly as defined by the parent-daughter information from ENDF, which allows all other parameters (such as recoverable energies, decay constants, and emission spectra) to be applied implicitly. With the final isotopic concentrations, it is straightforward to compute the disintegration rates at cooling time t, and the recoverable energy is computed by:

$$Q^{J,K}(t) = \sum_{m=\alpha,\beta,\gamma} Q_m^{J,K} * R^{J,K}(t) \quad (3)$$

where:

$R^{J,K}(t)$ is the disintegration rate of isotope J in node K, after time t;

$Q_m^{J,K}$ is the average recoverable energy per disintegration for particle type m.

3. DECAY DATA

The SNF library includes basic data, such as decay constants, atomic masses, and nuclide transmutation chains; radiation emission spectra for photons from radioactive decay, (α ,n) reactions, bremsstrahlung, and spontaneous fission; electrons and alpha particles from radioactive decay; neutrons from radioactive decay, spontaneous fission, and (α ,n) reactions; decay heat production; and electro-atomic interaction data for bremsstrahlung production. These data are compiled from fundamental (ENDF, ENSDF [11], TENDL [12,13]) and processed (ESTAR and ASTAR [14]) sources. For consistency with CASMO5, ENDF/B-VII.R1 data are complemented with data from TENDL-2012 [12], and the ENDF/B-VIII.R0 data are complemented with data from TENDL-2017 [13]. The evaluation and validation procedure of the decay data implemented in the SNF library is reported elsewhere [15].

The basic decay data set per isotope - half-life, atomic mass, decay modes and branching ratios, Q-values, and daughter isotopes - is entirely derived from ENDF, as are the discrete emission spectra for α , γ , and x-rays. The uncertainty of line-energies for the discrete spectra is provided by ENDF. When populating the photons in the user-defined energy groups, the intensity of a given line may be split equally between the adjacent groups if the group-boundary falls within the corresponding line-uncertainty. For the continuum spectra for prompt photons emitted during spontaneous fission and α -decay, empirical correlations from [16] are adopted.

The (α ,n) and (α , γ) reaction data are processed from TENDL by NJOY-2012 [17], with the alpha particle stopping powers for the elements, including compounds and mixtures, derived from ASTAR. The TENDL data for the oxygen isotopes ^{17}O and ^{18}O is complemented by (α ,n) cross-sections from Perry and Wilson [18]. The approach used for these isotopes was to rescale the neutron resonance data of Perry and Wilson to match the TENDL data at the upper energy of the resonance range (5.404 MeV for ^{17}O and 5.18 MeV for ^{18}O); the rescaling increased the cross-sections below these energies by $\sim 25\%$ and $\sim 4\%$, respectively. This allowed the cross-section representation to be extended to the upper limit of the SNF α -decay data (~ 12 MeV). The photon production data were not rescaled. Compared to TENDL-2012, the TENDL-2017 based (α ,n)-yields are in average about 2% smaller.

The continuum spectra for electrons and positrons were calculated by the method of Stamatelatos and England [19], coupled with the Fermi function approximation of Schenter and Vogel [20], with the spectrum endpoint energies provided in the ENDF decay sub-library. The external bremsstrahlung cross-sections and production kernels were processed from the ENDF electron sub-library, while the electron stopping power data were calculated with the ESTAR application.

4. VALIDATION RESULTS

4.1. CLAB decay heat power measurements

The measurements were performed during 2003-2004 by SKB in the interim storage facility CLAB in Sweden [21]. Assemblies of nine different designs were measured, with exposures ranging from 20 GWd/tHM to 51 GWd/tHM and cooling times from 10 years to 22 years. The decay heat measurements were performed by means of an electrically calibrated calorimeter submerged in water. The measured decay heat powers have been corrected for the fraction of the released gamma heat absorbed outside the

calorimeter tank. The design goal for the measurement uncertainty is established to be less than $\pm 2\%$. Table I and Figure 1 present C/E results calculated with the two sets of decay data - E7R1 and E8R0.

Table I. C/E comparison between CMS5/SNF and CLAB measurements.

Assem ID	Meas. W	E7R1 C/E	E8R0 C/E	Assem ID	Meas. W	E7R1 C/E	E8R0 C/E	Assem ID	Meas. W	E7R1 C/E	E8R0 C/E
C01	415.8	1.011	1.006	0C9	491.2	1.025	1.020	14076	240.3	0.983	0.981
C12	410.3	1.015	1.010	0E2	587.9	1.002	0.997	3838	126.4	1.016	1.013
C20	426.6	1.009	1.005	0E6	487.8	1.006	1.003	KU0100	185.3	0.971	0.969
C42	445.3	0.980	0.976	1C2	417.7	1.020	1.016	KU0269	192.7	0.973	0.971
D27	456.1	0.991	0.987	1C5	499.2	1.010	1.005	KU0278	195.4	0.968	0.966
D38	442.3	1.010	1.007	1E5	468.8	0.999	0.997	KU0282	218.5	0.975	0.972
E38	375.3	0.994	0.993	2A5	233.8	1.033	1.029	5535	84.6	1.052	1.050
E40	381.3	0.989	0.985	2C2	466.5	1.019	1.014	11494	166.0	0.991	0.989
F14	381.8	0.995	0.993	3C1	470.2	1.009	1.005	11495	167.6	0.983	0.981
F21	420.9	0.993	0.990	3C4	497.3	1.005	1.000	13775	178.4	0.996	0.994
F25	396.7	1.005	1.002	3C5	501.4	1.006	1.001	13847	169.9	1.009	1.007
F32	692.0	1.008	1.000	3C9	468.4	1.012	1.008	13848	170.7	1.004	1.002
G11	416.4	0.988	0.986	4C4	422.0	1.009	1.005	12684	282.7	0.987	0.985
G23	420.6	0.995	0.993	4C7	498.7	1.011	1.006	12078	120.2	1.020	1.018
I09	507.9	1.016	1.013	5A3	239.3	1.007	1.003	13628	194.0	1.014	1.012
I20	403.5	1.001	0.999	5A3	230.6	1.027	1.023	13630	235.7	1.000	0.998
I24	410.1	0.993	0.991	5F2	714.1	0.993	0.988				
I25	445.8	0.994	0.991

Figure 1. Comparison of C/E between CLAB measurements and CMS5/SNF (BWR assemblies – red squares; PWR assemblies – blue diamonds).

The overall effect of E8R0 on the decay heat powers, presented in Table II, is considered very small as compared to E7R1. The average deviation decreases by about 0.4% with effectively identical standard deviation. Overall, the calculated decay heat powers are in very good agreement with the experimental results. The observed average deviation of 0.1–0.3% along with the standard deviation of $\sim 1.7\%$ demonstrates the ability of Studsvik's SNF system to reliably predict the assembly decay heat power for

assemblies irradiated in BWR or PWR. A deviation below the measurement uncertainty goal of 2.0% is observed for 40 out of 51 measurements, and 49 assemblies are within 3%.

Table II. Comparison summary of C/E between CMS5/SNF and CLAB measurements.

Lattice Type	# of meas. assemblies	ENDF/B-VII.R1 Decay heat power C/E		ENDF/B-VIII.R0 Decay heat power C/E	
		Average	2 σ (%)	Average	2 σ (%)
pwr15x15	18	0.999	2.0	0.996	2.0
pwr17x17	17	1.014	2.2	1.007	2.0
bwr	16	0.997	4.4	0.994	4.4
All	51	1.003	3.4	0.999	3.2

The BWR results are slightly more dispersed, with standard deviation 2.2%, compared to ~1% standard deviation for the PWRs. It should be noted that the set of BWR assemblies consists of 6 different assembly designs irradiated in 6 different reactors. However, the number of assemblies per type is insufficient to make confident conclusions about dependencies on the assembly type or on reactor type.

4.2. ARIANE, PIE of MOX fuel irradiated in PWR

The ARIANE program [22] (Actinides Research In A Nuclear Element) is an experimental program designed to improve the evaluation and prediction of actinides and fission products in MOX and UOX fuel elements. The program was conducted between 1995 and 2001, following irradiation of 10 fuel rods (17 samples, MOX and UOX) in three reactors - 2 PWRs (Beznau and Gösgen) and one BWR (Dodewaard). The PIE was performed independently in three laboratories – Paul Scherrer Institute (PSI) (Switzerland), SCK.CEN (Belgium), and ITU (Germany) - and the measurements were cross-checked for verification.

Comparisons of calculated vs. experimental results (C/E) for samples BM1 and BM5 are presented in this paper. The two samples were irradiated to about 46.5 GWd/tHM and 56.5 GWd/tHM in two different assemblies at Beznau NPP during normal operation between cycles 18 and 22. The results presented in this work are obtained by applying the irradiation histories from SIMULATE5 [23], two sets of cross-section data from CASMO5, and two sets of decay data based on E7R1 and E8R0, respectively.

The evaluation of the fuel samples is based on data generated for the corresponding fuel rods. The nuclear data required for the analyses of the selected fuel rod are computed by CASMO5 along with the nodal data for SIMULATE5 and tabulated by CMSLINK5. The rod exposures are computed by the pin-power reconstruction model in SIMULATE5, following detailed simulation of the reactor operation history [23]. The isotopic concentrations are computed for the axial node corresponding to where the samples were cut.

The experimental values recommended for code validation were subjected to cross-check analyses and validated either by combining the two measurements Equation 4, or as a unique recommended value. The C/E comparison for samples BM1 and BM5 is shown in Table III.

$$x_r = \frac{\frac{x_1}{\sigma_{x_1}^2} + \frac{x_2}{\sigma_{x_2}^2}}{\frac{1}{\sigma_{x_1}^2} + \frac{1}{\sigma_{x_2}^2}} \quad \text{with} \quad \sigma_{x_r}^2 = \frac{1}{\frac{1}{\sigma_{x_1}^2} + \frac{1}{\sigma_{x_2}^2}} \quad (4)$$

Table III. C/E comparison between CMS5/SNF calculation and BM1 and BM5 samples.

Nuclide	BM1				Note	BM5				Note
	mg/g fuel	E7R1	E8R0	2 σ , %		mg/g fuel	E7R1	E8R0	2 σ , %	
U-234	7.81E-02	0.995	0.990	7.85		4.94E-02	0.982	0.975	9.19	
U-235	8.28E-01	1.002	0.983	1.99		5.86E-01	1.034	1.004	2.05	
U-236	2.56E-01	0.957	0.969	4.14		2.69E-01	0.964	0.975	4.54	
U-238	8.07E+02	0.993	0.993	0.45		7.98E+02	0.995	0.995	0.45	
Np-237	1.50E-01	1.093	1.077	12.07		1.75E-01	1.016	1.002	0.43	
Pu-238	1.11E+00	0.989	0.983	3.05		9.95E-01	1.011	1.002	3.05	
Pu-239	1.19E+01	1.055	1.040	0.56		9.80E+00	1.070	1.049	0.57	
Pu-240	1.18E+01	0.992	1.004	0.56		1.01E+01	0.983	0.996	0.57	
Pu-241	6.06E+00	1.020	1.005	0.56		5.39E+00	1.013	0.997	0.57	
Pu-242	3.88E+00	0.990	0.986	0.56		3.67E+00	0.967	0.962	0.57	
Pu-244	6.85E-04	0.539	0.554	49.93		1.00E-03	0.449	0.461	50.10	
Am-241	2.17E+00	1.043	1.027	2.54		1.12E+00	1.184	1.165	2.78	
Am-242m	1.48E-02	1.051	1.031	9.46		1.06E-02	1.228	1.198	4.78	
Am-243	9.87E-01	0.976	0.971	3.51		9.09E-01	1.045	1.038	0.28	
Cm-242	1.70E-01	0.878	0.868	3.49		1.56E-01	0.869	0.857	3.46	
Cm-243	8.74E-03	0.938	0.928	6.45		7.42E-03	1.104	1.090	10.59	
Cm-244	6.15E-01	1.009	1.007	1.48	EOL	8.13E-01	0.907	0.903	2.48	EOL
Cm-245	5.99E-02	1.094	1.086	2.57		8.96E-02	0.979	0.970	3.42	
Cm-246	6.52E-03	0.924	0.928	3.57		1.47E-02	0.774	0.780	4.68	
Ce-144	2.50E-01	1.033	1.027	6.88		2.38E-01	1.017	1.010	5.13	
Nd-142	1.72E-02	1.030	1.032	6.98		2.91E-02	0.992	0.995	7.01	
Nd-143	9.00E-01	1.008	1.004	0.56		1.04E+00	0.990	0.986	0.56	
Nd-144	1.19E+00	0.992	0.991	0.56		1.54E+00	0.959	0.959	0.56	
Nd-145	7.01E-01	1.007	1.005	0.56		8.40E-01	0.982	0.979	0.56	
Nd-146	7.40E-01	1.009	1.008	0.56		9.58E-01	0.976	0.974	0.56	
Nd-148	4.43E-01	1.012	1.011	0.56		5.49E-01	0.984	0.984	0.56	
Nd-150	2.65E-01	1.015	1.013	0.58		3.28E-01	0.988	0.987	0.58	
Pm-147	1.85E-01	0.975	0.970	2.91		1.77E-01	0.977	0.973	10.45	
Sm-147	2.65E-01	0.972	0.969	1.05		2.35E-01	1.003	1.000	0.61	last date
Sm-148	1.89E-01	0.970	0.977	0.63		2.74E-01	0.960	0.966	0.59	analyses
Sm-149	5.03E-03	1.084	1.062	2.05		4.85E-03	0.998	0.974	9.25	analyses
Sm-150	3.62E-01	0.996	0.993	0.64		4.37E-01	0.991	0.986	0.60	analyses
Sm-151	2.47E-02	1.001	0.981	8.26		2.27E-02	1.047	1.021	0.77	EOL
Sm-152	1.64E-01	0.928	0.925	0.63		1.60E-01	0.979	0.975	0.61	analyses
Sm-154	7.95E-02	0.966	0.962	0.65		8.97E-02	1.030	1.028	0.63	analyses
Eu-151	1.27E-03	0.985	0.965	3.31		7.94E-04	0.784	0.764	2.07	
Eu-153	1.96E-01	0.980	0.977	0.66		2.32E-01	0.961	0.959	0.63	
Eu-154	5.38E-02	1.050	1.011	2.47	EOL	6.65E-02	1.076	1.027	1.94	
Eu-155	1.77E-02	0.842	0.855	3.84	EOL	1.94E-02	0.976	0.987	4.33	
Gd-154	2.71E-02	1.078	1.041	2.97	PSI	2.76E-02	1.007	0.968	3.62	PSI
Gd-155	9.36E-03	0.988	1.004	2.67		7.44E-03	0.981	0.991	2.00	
Gd-156	1.53E-01	0.940	0.952	1.37	PSI	2.60E-01	0.860	0.874	3.10	PSI
Gd-158	4.15E-02	1.067	1.067	2.21	PSI	5.75E-02	1.016	1.018	3.58	PSI
Gd-160	3.36E-03	0.977	0.974	6.88	PSI	4.31E-03	0.921	0.917	4.97	PSI
Sr-90	3.43E-01	0.920	0.920	0.83		3.96E-01	0.948	0.949	9.17	
Mo-95	6.06E-01	1.207	1.204	3.75		9.31E-01	0.932	0.931	3.61	
Tc-99	5.74E-01	1.613	1.608	2.80		1.07E+00	1.005	1.004	4.40	
Ru-101	5.74E-01	1.748	1.743	2.80		1.23E+00	0.965	0.964	4.14	analyses
Ru-106	1.38E-01	2.011	1.996	3.33		2.29E-01	1.153	1.146	7.62	EOL
Rh-103	4.41E-01	2.032	2.024	7.78		9.91E-01	0.989	0.984	2.57	analyses
Ag-109	8.70E-02	1.207	1.203	1.92		1.49E-01	1.130	0.825	17.33	analyses
Sb-125	6.73E-03	1.712	1.705	5.99		8.21E-03	1.446	1.442	3.69	EOL
I-129						2.24E-01	1.371	1.368	11.25	

Cs-133	1.34E+00	1.021	1.020	1.79	analyses	1.55E+00	1.016	1.014	2.14
Cs-134	1.53E-01	0.945	0.943	1.86	EOL	2.06E-01	0.940	0.939	2.30
Cs-135	8.58E-01	1.029	1.014	1.80	analyses	1.06E+00	0.980	0.964	2.08
Cs-137	1.52E+00	1.005	1.003	1.77	EOL	1.82E+00	0.998	0.997	2.16

The cross-check analyses did not provide a recommended value for several isotopes. Instead, the value for the following isotopes was marked as “*maintained but no recommendation*”:

BM1: ^{244}Cm , ^{154}Eu , ^{155}Eu , ^{133}Cs , ^{134}Cs , ^{135}Cs and ^{137}Cs
 BM5: ^{244}Cm , ^{147}Sm , ^{148}Sm , ^{149}Sm , ^{150}Sm , ^{151}Sm , ^{152}Sm , ^{154}Sm , ^{101}Ru , ^{106}Ru , ^{103}Rh , ^{109}Ag , ^{125}Sb

The above isotopes were included in the analyses presented here with their experimental value and uncertainties defined by Equation 4. The state of comparisons of those isotopes is notified in column "Note" in Table III as provided in the benchmark specification: EOL – end of life; analyses – at the time of analyses; last date – at last date available. Four gadolinium isotopes - ^{154}Gd , ^{156}Gd , ^{158}Gd and ^{160}Gd - were measured only by PSI (hence with "no maintained value") were included and marked as PSI, which stands for “PSI measurement at the time of analyses”.

In general, the effect of ENDF/B-VIII.R0 on the prediction of the selected isotopes, although small in most cases, is in the direction of improving the agreement. The overall tendency of decreasing the calculated concentrations is attributed to small variations in the irradiation history conditions for the reactor cores simulated by cross-sections based on E7R1 and E8R0. Although most changes are within 1%, noticeable improvement can be observed in:

BM1 : ^{239}Pu (1.5%), ^{154}Eu (3.7%), $^{154-156}\text{Gd}$ (2-3.5%)
 BM5 : ^{235}U (3%), ^{239}Pu (2%), ^{151}Sm (2.4%), ^{154}Eu (4.5%)

The calculations and the measurements are generally in very good agreement with 49 out of 57 isotopes within 10%. Excellent agreement can be observed for the uranium and plutonium isotopes, except ^{244}Pu . An underestimation of ~50% and ~12% is observed in ^{244}Pu and ^{242}Cm , respectively, for both samples; ^{246}Cm is underpredicted by ~22% for BM5, but within 10% for BM1. A similar, but reverse, result is observed in ^{241}Am and $^{242\text{m}}\text{Am}$ - both are overpredicted for BM5 by ~18% and ~22%, respectively. The measurement uncertainty of ^{244}Pu is ~50% and quite comparable to the observed deviation. However, given the good agreement in ^{241}Pu (~1.2%) and the underprediction in ^{242}Cm and ^{246}Cm , the overprediction in the two americium isotopes for BM5 is inconsistent and difficult to explain.

The prediction of the fission products - isotopes of Ce, Nd, Pm, Eu (except ^{151}Eu for BM5), Gd, and Cs - are in excellent agreement. It is observed that for BM5, most of the traditionally difficult-to-measure metallic fission products are also in excellent agreement: ^{90}Sr , ^{95}Mo , ^{99}Tc , ^{101}Ru , and ^{103}Rh . The overpredictions observed in the metallic fission products for BM1 are generally explained by incomplete dissolution, resulting in significant remaining residue.

5. CONCLUSIONS

The application of Studsvik's system for spent nuclear fuel analyses together with nuclear data libraries based on the latest release of ENDF/B-VIII.R0 is presented in this work. A comparison against decay heat powers measurements, CLAB, and the PIE program ARIANE were presented to demonstrate the applicability of E8R0 for spent fuel calculation.

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