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Isotopic Validation for PWR Actinide-Only Burnup Credit Using Mihama-3 Data

Meraj Rahimi (Johnson Associates Inc.)
Dale Lancaster (TRW Environmental Safety Systems)

Introduction

In May 1995, the Department of Energy's (DOE's) Office of Civilian Radioactive Waste Management submitted a Topical Report on Actinide-Only Burnup Credit for PWR Spent Nuclear Fuel Packages[1] to the Nuclear Regulatory Commission (NRC) on DOE's behalf. The purpose of this topical report is to obtain the NRC's approval on a generic burnup credit methodology.

A major part of this methodology is the validation of the neutronics model which is used for designing the criticality control system. SCALE 4.2 [2] was chosen to demonstrate the proposed methodology in the burnup credit topical report. To perform burnup credit criticality analysis using SCALE 4.2, only the isotopic generation/depletion and criticality control modules are used. Therefore, the methodology includes validation of SAS2H which is used for isotopic generation/depletion and CSAS25 used for criticality analysis.

The validation methodology presented in the burnup credit topical report consisted of establishing a set of isotopic correction factors (i.e., a bias factor with 95 percent confidence level) by comparison between a set of isotopic measurements and the corresponding calculated values. The number of isotopic samples used for the measurement and presented in the Topical Report was 19. The range of burnup and enrichment for these samples were from 16.02 to 46.46 GWd/MTU and 2.45 to 3.13 wt-% U-235, respectively.[1]

The NRC has commented on the topical report by indicating that the amount of experimental data presented in the topical report is very limited and needs to be augmented. In an effort to increase the number of data points for isotopic validation, nine samples from three spent fuel assemblies discharged from Mihama-3 reactor were identified.[3] The initial enrichment for the spent fuel assemblies from which the samples were taken was 3.2 wt-% U-235 and they range in burnup from 6.9 to 34.2 GWd/MTU.

Development of Isotopic Generation/Depletion Model

SAS2H control module performs isotopic generation and depletion in 1-D radial dimension under a set of specified reactor core conditions. The parameters needed to construct SAS2H input decks for Mihama-3 samples are: fuel density, temperature, and composition; moderator temperature, density, and boron content; fuel pin model for cross section averaging, fuel assembly physical characteristics, power as a function of time, and the composition of non-fuel material in fuel assemblies.

Most of the data needed came from Reference 3 but assumptions were required for the moderator density and soluble boron concentration. Since the fuel assembly heights from which the samples

were taken was not available, an average core temperature using inlet and outlet temperatures was used to calculate the moderator density. The moderator boron concentration was assumed to be 450 parts per million (ppm). This is the average boron concentration for reactors with annual cycles which Mihama-3 operating history indicates.

Using the pellet and rod information provided in Reference 3, an infinite lattice of fuel pins model (called path A model) was constructed. This model is used by SAS2H to process the cross sections using a resonance self-shielding calculation followed by a discrete-ordinates 1-D transport computation of the neutron flux in a unit cell. The cell-weighted cross sections produced by this model are then applied to the fuel region of the assembly model (called path B model). Path B accounts for the intra-assembly gap, instrumentation tube and the guide thimbles with or without burnable absorbers. This Path B model is used to collapse the cross sections for use in the ORIGEN-S module of SCALE which performs the depletion.

Comparison of Calculated to Measured Values

Table 1[3] provides the measured values of only the 10 actinides used in the Topical Report. All values are corrected for decay to represent the content at 5 years after the end of irradiation. Burnup was determined experimentally by measurement of Nd-148 concentrations. The expected isotopic content for each of the ten isotopes in the nine samples were calculated. The ratio of the measured to calculated concentrations are provided in Table 2.

The agreement between measurement and calculation is in general within a few percent. The largest disagreement is for Pu-242 (13%) and Pu-240 (8.6%). The agreement in the highest worth isotopes U-235 (4%), U-238 (0%), and Pu-239 (0.3%) is much better. The calculations were performed with the 27BURNUPLIB which is based on ENDF/B-IV.

The ratios for sample 5 appear to be further from unity than for the other samples. Samples 3, 4, and 5 come from the same rod at differing heights. Using Table 1 it is difficult to reconcile the reported concentrations for samples 3 and 5 with the reported burnups. All the isotopes are consistent with the measured burnup being too low. Eliminating sample 5 will reduce the standard deviation in the samples but unfortunately is not significant in changing the mean values (maximum 2% effect on Pu-238, Pu-241, and Pu-242).

References

1. U.S. Department of Energy, May 1995. *Topical Report on Actinide-Only Burnup Credit for PWR Spent Nuclear Fuel Packages*, Rev. 0, Office of Civilian Radioactive Waste Management, Washington D.C.

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3. Japan Atomic Energy Research Institute, Y. Naito, M. Kurosawa, and T. Kaneko, March 1994. *Data Book of the Isotopic Composition of Spent Fuel in Light Water Reactors*, JAERI-M 94-034, 163-171, Department of Fuel Cycle Safety Research.

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5. Y. Nakahara, et al. Amounts of Nuclides Constituting PWR Spent Fuels, Comparison of Observed with Calculated Values. *Radichimica Acta, International Journal for Chemical Aspects of Nuclear Science and Technology* Volume 50 (½) (1990): 9.

Table 1. Measured Isotopic Quantities for Mihama-3 Samples [3]
(% atom of isotope per initial uranium)

	1	2	3	4	5	6	7	8	9
Burnup[a]	8.30	6.90	15.30	21.20	14.60	29.44	32.30	33.70	34.10
u-234	2.720E-02						1.771E-02	1.769E-02	
u-235	2.420E+00	2.553E+00	1.878E+00	1.473E+00	1.868E+00	9.912E-01	9.180E-01	7.984E-01	8.136E-01
u-236	1.660E-01	1.474E-01	2.670E-01	3.300E-01	2.681E-01	3.861E-01	4.030E-01	4.216E-01	4.230E-01
u-238	9.618E+01	9.623E+01	9.563E+01	9.523E+01	9.570E+01	9.466E+01	9.439E+01	9.438E+01	9.424E+01
pu-238	4.508E-04	3.408E-04	2.588E-03	5.720E-03	2.684E-03	1.309E-02	1.591E-02	1.676E-02	1.855E-02
pu-239	3.009E-01	2.814E-01	4.632E-01	5.059E-01	4.716E-01	5.275E-01	5.448E-01	4.948E-01	5.293E-01
pu-240	4.185E-02	3.411E-02	1.026E-01	1.477E-01	1.050E-01	2.080E-01	2.252E-01	2.299E-01	2.408E-01
pu-241	1.072E-02	8.179E-03	4.029E-02	6.431E-02	4.192E-02	9.430E-02	1.043E-01	9.703E-02	1.063E-01
pu-242	9.335E-04	5.940E-04	7.270E-03	1.726E-02	7.385E-03	4.011E-02	4.821E-02	5.256E-02	5.602E-02
am-241[b]	2.989E+01	2.263E+01	1.174E+02	1.907E+02	1.168E+02	3.004E+02	2.998E+02	3.205E+02	3.360E+02

[a] GWd/MTU
[b] gram/MTU

Table 2. Measured to Calculated Ratios for Mihama-3 Samples

	1	2	3	4	5	6	7	8	9	Ave.
U-234	1.056						0.938	0.958		0.984
U-235	1.025	1.022	1.051	1.059	1.016	1.027	1.084	1.016	1.057	1.040
U-236	0.937	0.953	1.001	1.000	1.040	0.939	0.943	0.971	0.970	0.973
U-238	1.000	1.001	0.999	1.000	1.000	1.001	1.000	1.003	1.001	1.000
Pu-238	0.878	1.007	1.079	1.086	1.253	1.019	1.024	0.979	1.053	1.042
Pu-239	0.949	1.009	1.059	1.021	1.100	0.989	1.009	0.912	0.976	1.003
Pu-240	1.007	1.073	1.073	1.065	1.166	1.091	1.097	1.080	1.119	1.086
Pu-241	0.895	1.024	1.023	0.988	1.155	0.988	1.003	0.895	0.970	0.993
Pu-242	0.987	1.152	1.129	1.088	1.314	1.144	1.122	1.104	1.140	1.131
Am-241	0.877	0.990	0.997	0.983	1.074	1.007	0.929	0.953	0.986	0.977