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PREDICTION OF SPENT MOX AND LEU FUEL COMPOSITION AND COMPARISON WITH MEASUREMENTS

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ABSTRACT

This work examines the capabilities of simulation codes to predict the concentration of nuclides in spent reactor fuel, in particular mixed-oxide (MOX) fuel, via comparisons with destructive radiochemical analyses performed on irradiated samples. Two MOX samples from a pressurized-water reactor (PWR) and a boiling-water reactor (BWR), and one UO_2 sample from a BWR, are discussed. Both actinide and fission-product concentrations were available for comparison purposes. The actinides include isotopes of uranium, plutonium, americium, and curium. The fission products include isotopes of cesium, neodymium, samarium, and europium. For many of the actinides, the predictions are quite good when compared with the measured values; but concentrations for fissile species tend to be overpredicted. The cesium and neodymium concentrations are well predicted, but the samarium and europium isotopes show variable results. It is pointed out that an important difficulty in this kind of work is the accurate estimation of the burnup experienced by exposed samples. The sensitivity of some of the results to sample-burnup estimates is discussed. Work on this project is ongoing, and more samples together with a number of other nuclides B both actinides and fission products B will be discussed in later reports.

1. INTRODUCTION

The disposal of weapons-usable plutonium by irradiation in commercial reactors is a current policy objective of the U.S. Department of Energy. To that end, the plutonium would be part of a MOX fuel containing oxides of plutonium and uranium, with weapon-grade plutonium forming the majority of the fissile material.

Compliance with ANSI/ANS-10.5-1987 (R1998), various Nuclear Regulatory Commission regulations, and good engineering judgement requires that computer programs for nuclear analyses be verified and validated. Data for validation, including actinide and fission product inventories for MOX and low-enriched uranium (LEU) fuels, have been measured recently in a multinational research program. The Actinide Research in a Nuclear Element (ARIANE)

program examined irradiated MOX fuel samples from PWR and BWR fuel assemblies. Irradiated BWR and PWR UO_2 samples were also investigated as part of the program. The plutonium in the MOX samples was reactor-grade, containing ^{239}Pu in excess of 60 wt %; the ^{235}U was approximately 0.2 wt %. Burnups for the MOX samples were in the range of 38 to 60 GWd/t. Reported in this article are calculated-to-experimental (C/E) comparisons of isotopic measurements and general descriptions as to the burnup conditions in the assemblies studied.

Radiochemical analysis of the spent fuel samples determined the concentrations for actinides from ^{234}U to ^{246}Cm. Measurement of plutonium and uranium isotopes was needed to assess the level of accuracy in calculated, burnup-dependent reactivity worth. Measurement of the Cm isotopes was needed as a previous study[1] had shown that ^{244}Cm is the major contributor to the neutron dose from spent MOX fuel. Concentration values for a number of important fission products have also been determined. These include cesium, neodymium, and samarium isotopes as well as ^{155}Gd. The cesium isotope, ^{137}Cs, is the principal gamma source for irradiated MOX fuels for up to 100 years after discharge from the reactor **B** an important consideration in establishing that irradiated weapons plutonium is difficult to divert or use. Another study[2] shows that the Cs, Nd, Sm, Eu, and Gd isotopes are all significant neutron poisons and are important to burnup-credit analyses for spent nuclear fuel.

The relatively large number of nuclides investigated (as compared, for instance, to the studies reported by Fisher and Difilippo[3]) allows for the validation of both reactor physics computer programs (here the assembly analysis program, HELIOS[4]) and radionuclide source estimation programs (here the production and decay program ORIGEN-S[5]). Simulations were carried out with both a one-dimensional, discrete-ordinates methodology (the SAS2H sequence from the Oak Ridge National Laboratory SCALE system[5]) and a two-dimensional, collision-probability technique (the HELIOS system[4]). In the SCALE approach, a unit cell is modeled with the remainder of the fuel, clad, and moderator in the assembly being smeared over a cylindrical region that is equivalent in volume to the assembly being modeled. The results of a SCALE simulation are in terms of concentrations that are average values for this assembly-equivalent volume. In modeling the burnup of a sample with HELIOS, a more detailed and sample-specific rendition of the assembly is employed. In the HELIOS model, each fuel pin is treated as a distinct region for which atom densities can be calculated separately. Fuel pins, fuel-pin clad, moderator, etc., can all be specified as separate entities, having their own compositions, densities and temperatures. In

2. THE PWR MOX SAMPLES

A number of MOX samples were exposed in a PWR (Beznau). Eventually, data for four of these MOX samples will be reported. Below, we report C/E ratios for two samples. These C/E ratios are reported for both actinides and fission products. In these PWR experiments, the assemblies containing the experimental MOX samples were composed entirely of MOX fuel. Therefore, the results presented in this section are probably representative of the overall assembly containing each sample under discussion.

It is important to understand the experimental accuracy that applies to the measurements being reported in this work. Because of the ongoing nature of the work it is likely that experimental uncertainty estimates will be updated. However, in regards to the results being reported here, the following will give a sense as to the measurement precision that is being reported to date (the numbers refer to the 95% confidence limit): Uranium isotopes are quoted with uncertainties around 1%, except for ^{234}U where the uncertainty is estimated to be in the 5% to 10% range. The uncertainty in the ^{237}Np measurements is around 4%. For the plutonium isotopes, uncertainties are quoted to be around 1%, but for ^{244}Pu (whose concentrations are exceedingly low) the uncertainty is about 50%. The americium isotopes have uncertainties of a couple of percent. For ^{243}Cm the uncertainty is about 33%, and for the other curium isotopes, it is about 6%. The neodymium isotopes have a quoted uncertainty of about 1%. The samarium isotopes have an uncertainty of about 1%, except for ^{149}Sm, which is around 2%; the europium isotopes have uncertainties around 2%, although for ^{155}Eu it is about 5%.

Both of these MOX samples were exposed for four cycles in the PWR (Beznau), and they experienced burnups of 46.9 and 45.2 GWd/t, respectively. Figure 1 shows C/E ratios for the actinides from the first sample labeled BM1 (Beznau, MOX Sample 1), and Fig. 2 shows C/E ratios for the second sample labeled BM3 (Beznau, MOX Sample 3).

Figures 1 and 2 show the results of both the SCALE and HELIOS comparisons. The comparisons, in this instance, are with measurements performed at the SCK/CEN radiochemical laboratory at Mol, Belgium. Other laboratories are involved in the analysis of the ARIANE spent-fuel samples; however, the SCK/CEN measurements are the most complete to date for these two samples. In general, it can be stated that the trends in the comparisons are similar for BM1 and BM3. The burnup in both cases was determined by matching the calculated and measured concentrations for ^{148}Nd. It is generally considered that ^{148}Nd is a reliable indicator of burnup.[6] Each of the one- and two-dimensional calculations was matched, independently, to the reported ^{148}Nd concentration. Since two different simulation models were used for each one of the samples, it is possible that the burnup estimates would be different for the two different simulation approaches. In fact, in the case of both samples, the SCALE and HELIOS-related burnup estimates are within one percent of each another. Estimates of sample burnup were also obtained from the reactor operators, and these were found to be within 3% and 7% of the ^{148}Nd estimates for BM1 and BM3, respectively (operator-supplied estimates were higher).

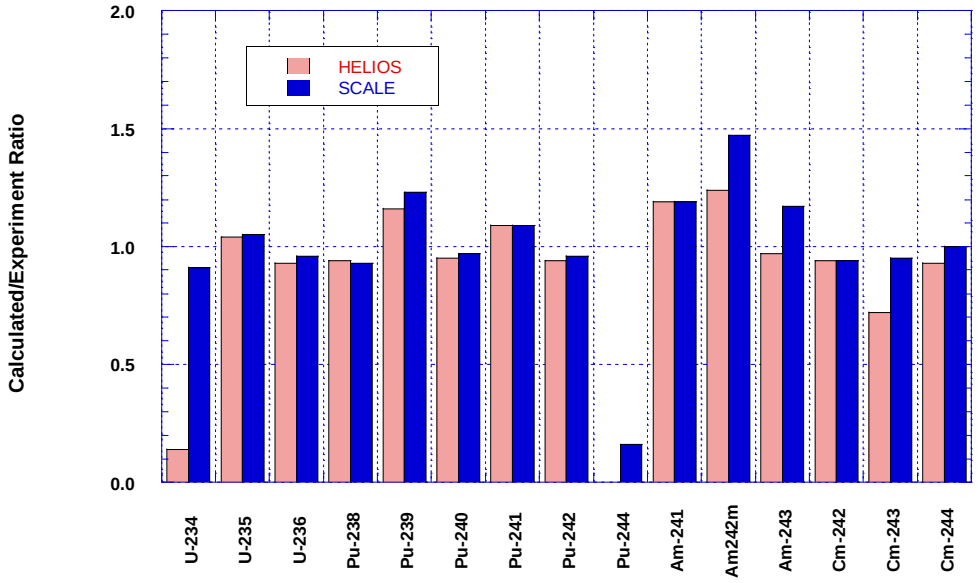


Fig. 1. Calculated to experimental (C/E) ratios for actinides from the Beznau PWR MOX sample BM1

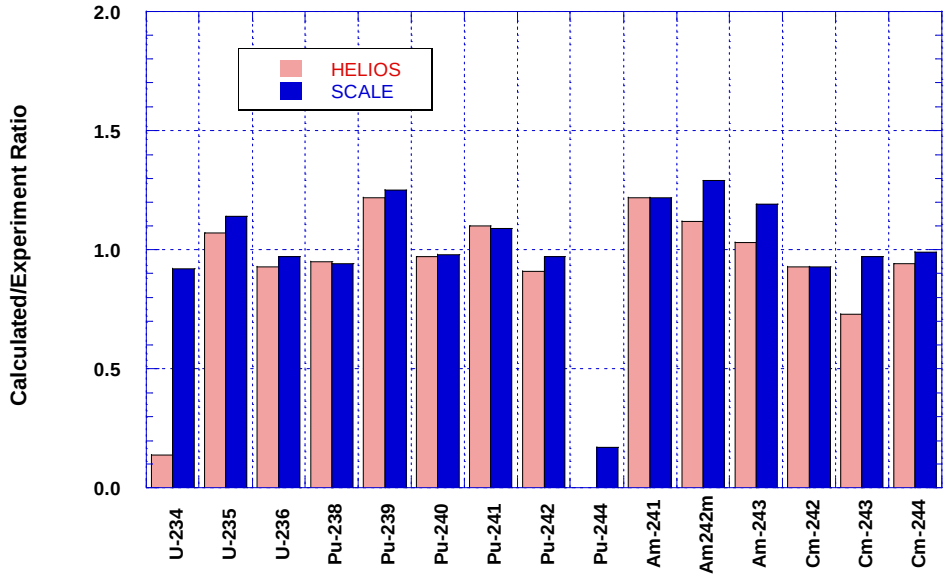


Fig. 2. C/E ratios for actinides from the Beznau PWR MOX sample BM3.

As noted above, the trends in Figs. 1 and 2 are similar. In regards to the low value for ^{234}U from HELIOS, this is due to the absence of the (n,2n) cross section for ^{235}U in the HELIOS library. The low ^{244}Pu value is possibly caused by measurement uncertainty because the concentration of ^{244}Pu was extremely low (a ^{244}Pu prediction was not obtained in the HELIOS case due to the absence of a capture cross section for ^{243}Pu). It is of particular interest that the predicted concentrations for the three fissile species, ^{235}U, ^{239}Pu, and ^{241}Pu are consistently high when compared with the measured values. Overprediction of fissile inventories by reactor-depletion programs could lead to significant overestimation of expected fuel cycle length with consequent economic penalties. The C/E ratios for these three nuclides are significantly different from values calculated with the same version of HELIOS and HELIOS data libraries for low-burnup MOX pins irradiated in the Quad Cities reactor.[3] Furthermore, the reported uncertainties in the analytical chemistry measurements of the three nuclides cannot account for such large C/E ratios. The locations of the pins in the fuel assembly were such that interface effects should not be significant.

Figures 3 and 4 show C/E ratios for the fission products from the BM1 and BM3 samples, respectively. To first approximation, the trends are similar for both of these samples. The cesium and neodymium isotopes are well predicted, but there is significant variability among the samarium and europium isotopes. There are HELIOS predictions for ^{155}Gd, and these are low. If verified by other analysts, the overprediction of the Sm and Eu nuclides would mean that uncompensated burnup credit analyses would be nonconservative. These overpredictions would also lead to underprediction of the multiplication factor for the reactor. The details on the extent of the agreement for individual nuclides are under active study and will be discussed in later reports.

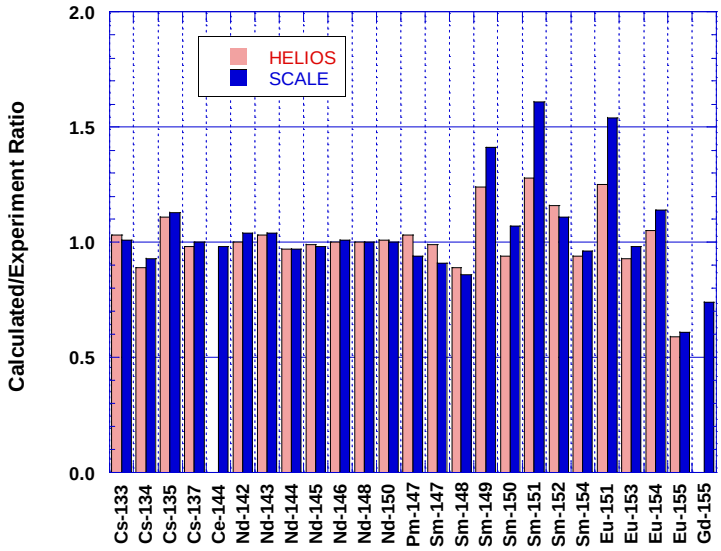


Fig. 3. C/E ratios for fission products from the Beznau PWR MOX sample BM1.

simulations are not expected to be optimal in the case of the BWR assembly. Furthermore, BWR simulations are more difficult than are PWR ones for two reasons: (1) BWR assemblies are less homogeneous than are PWR ones because the former have some pins that contain uranium/gadolinium, thus producing considerable spatial variability in the neutron spectrum; (2) For a BWR there is both an axial and a temporal variability in the moderator (water) density. (Note that the burnup simulations reported here refer to the particular axial height at which the experimental samples were located.)

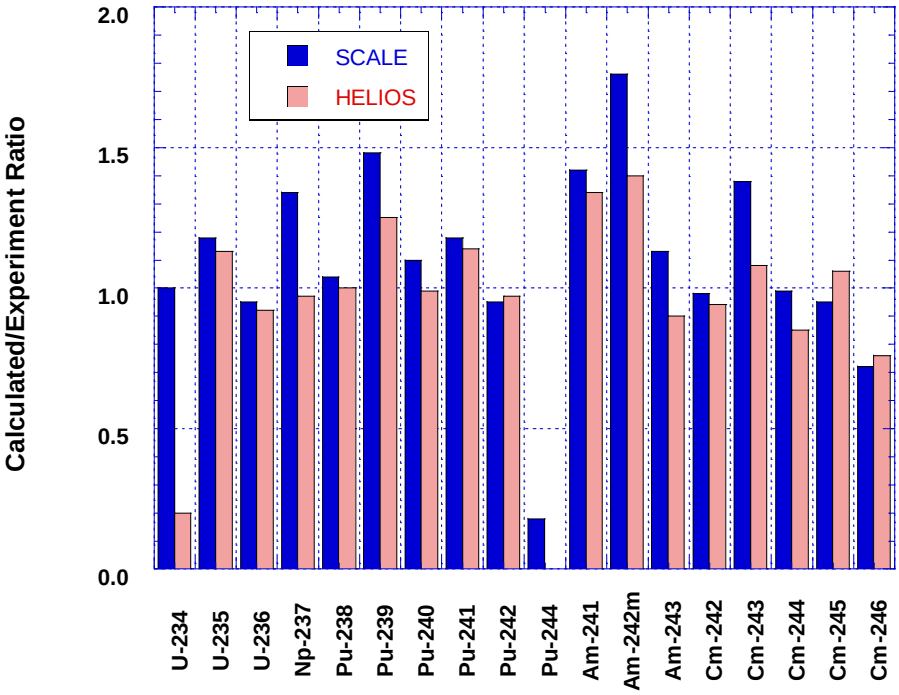


Fig. 5. C/E ratios for actinides from the Dodewaard BWR MOX sample DM1.

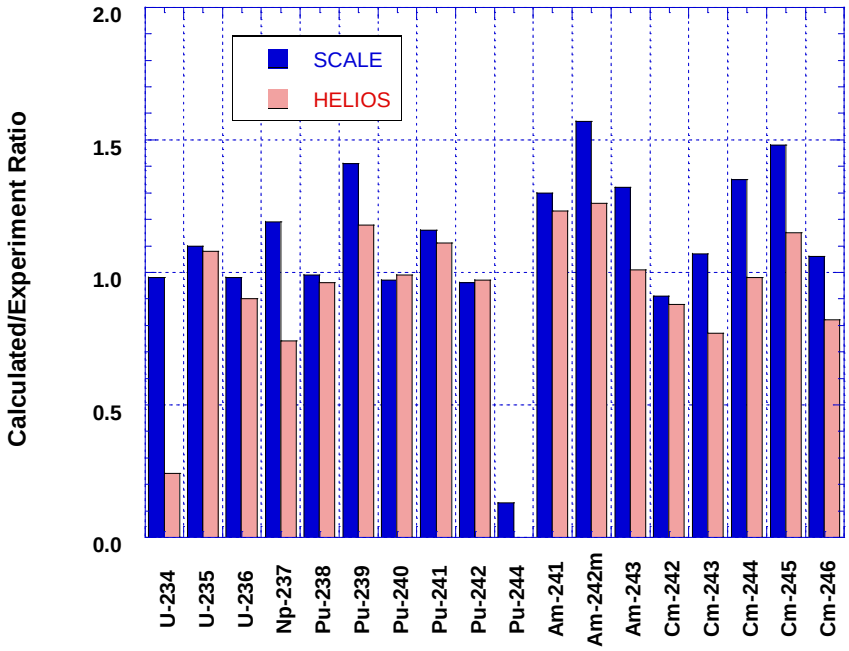
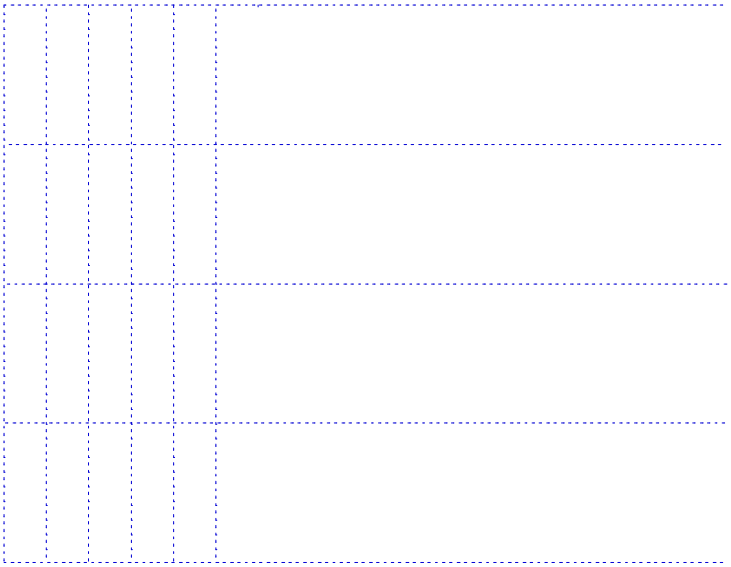


Fig. 6. C/E ratios for actinides from the Dodewaard BWR MOX sample DM2.

The burnup values for DM1 and DM2 were determined from the ^{148}Nd concentrations. As in the case of BM1 and BM3, the burnup values indicated for the SCALE and HELIOS simulations were within one percent of each other. However, these values differed from the operator-estimated sample burnups by about 12% and 14% for DM1 and DM2, respectively (operator-estimated burnups were lower). It is likely that this is a consequence of the difficulties involved in estimating burnups for samples in MOX rods that are in an assembly composed mostly of LEU.

4. BWR LEU SAMPLE RESULTS

As part of the ARIANE program, experimental UO_2 samples were exposed in both a BWR and a PWR. To date, the BWR sample measurements are more complete than the PWR measurements. Therefore, the results from the LEU sample that was exposed in the Dodewaard BWR are reported here. This sample, known as DU1, experienced a total burnup of 56.6 GWd/t over five reactor cycles. Figure 7 shows the C/E ratios for the actinides in the DU1 sample. These ratios show trends that are similar to those for the MOX samples; however, there is better agreement



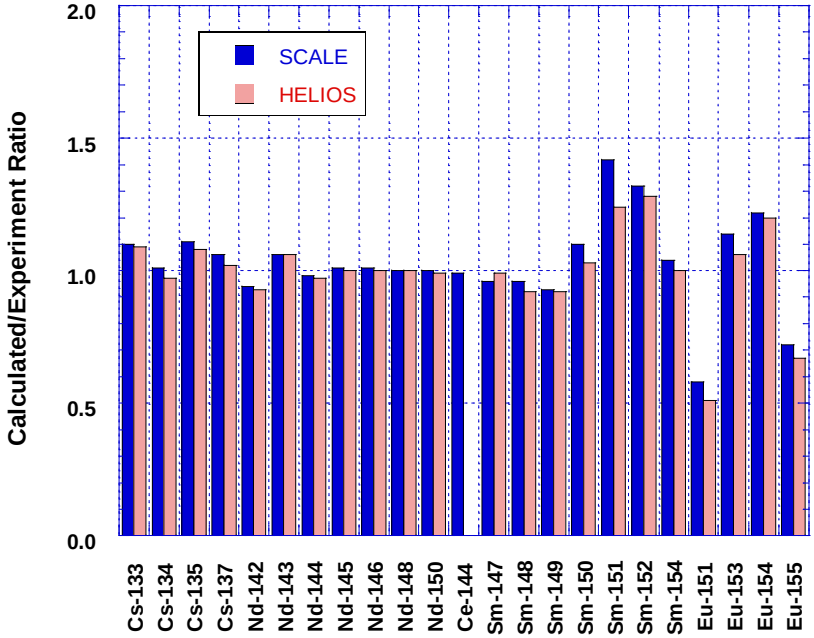


Fig. 8. C/ E ratios for fission products from the Dodewaard BWR LEU sample DU1.

5. SUMMARY AND CONCLUSIONS

The goal of this work was to investigate the ability of two burnup-simulation codes to predict spent nuclear fuel composition. The SCALE system has and will be used to predict actinide and fission-product source terms for accident analyses for U.S. reactors that will dispose of weapons-usable plutonium. The HELIOS code system will be used to verify calculated core physics parameters for MOX fuel cycles in PWRs and Russian VVERs. These data provide insight into the accuracy of source-term generation calculations.

The SAS2H sequence from the SCALE code system is a one-dimensional representation and is designed to give area-averaged values. The HELIOS code provides for a two-dimensional representation. Nevertheless, comparison of the HELIOS and SAS2H results shows that the spatial approximations necessary for preparation of the SAS2H model do not greatly impact C/E ratios. Consequently, use of SCALE/ORIGEN for radionuclide source estimation is justifiable.

The overall level of agreement seen between measurements and predictions is encouraging. However, end-of-life fissile nuclide inventory estimates do not achieve a level of accuracy

consistent with the reported uncertainty in the analytical chemistry measurements. Some fission-product poisons are also not accurately estimated.

Predictions of nuclide inventories for the MOX samples in the Dodewaard BWR were inferior to both the PWR MOX cases and the case of the LEU sample in the BWR. This situation is likely due to there being only two MOX rods in an assembly that is otherwise composed entirely of LEU.

Because this work involves small samples at various interior locations in reactor assemblies, a possible complication is the accuracy with which one can estimate the burnup at those specific locations. Even though overall assembly burnup may be well estimated, its spatial variability may not. The ^{148}Nd concentration was used to estimate sample burnup because it was believed that estimates of sample burnup provided by the reactor operators were subject to some uncertainty and, more importantly, this uncertainty may not have been adequately quantified.

To illustrate the sensitivity of calculated inventories to sample burnup estimates, we investigated the trends in the predicted concentrations for ^{235}U and ^{239}Pu as burnup is varied in the region of the value indicated by the ^{148}Nd concentration. Figures 9 and 10 refer respectively to sensitivity studies on the DU1 and DM1 samples (LEU and MOX in the Dodewaard BWR). Both figures show C/E ratios as burnup is varied over a range that is 10% above and below the ^{148}Nd-indicated value. The overprediction can clearly be seen at the burnup value used in the simulations. The ^{235}U concentration can be matched by an increase in burnup, and in the DU1 case the increase might reasonably be considered to be within its range of uncertainty. However, the overprediction of the ^{239}Pu concentration seems to be more persistent in the case of both samples.

The results of analyses performed to date lead the authors to two hypotheses **B** both based on the assumption that the analytical chemistry analyses have been performed properly. Assuming one knows the burnup accurately, the difficulty in predicting the final concentrations of ^{235}U and ^{239}Pu could be due to predictions for the production of ^{239}Pu. A large amount of ^{238}U is present in the fuel, and a small inaccuracy in its capture cross section would translate into a relatively large inaccuracy in the amount of ^{239}Pu produced, and thus the amount of fissile material in the fuel. In turn, the amount of fissile nuclei (^{235}U and ^{239}Pu) remaining at discharge would be difficult to predict. Another possibility is that the poisoning effect of the fission products that are overpredicted (Sm and Eu) compensates for the positive reactivity effect of the overpredition of the fissile isotopes. It is possible that these compensating effects could lead to an accurate estimation of burnup-dependent reactivity even though inventories of individual nuclides are not accurately calculated.

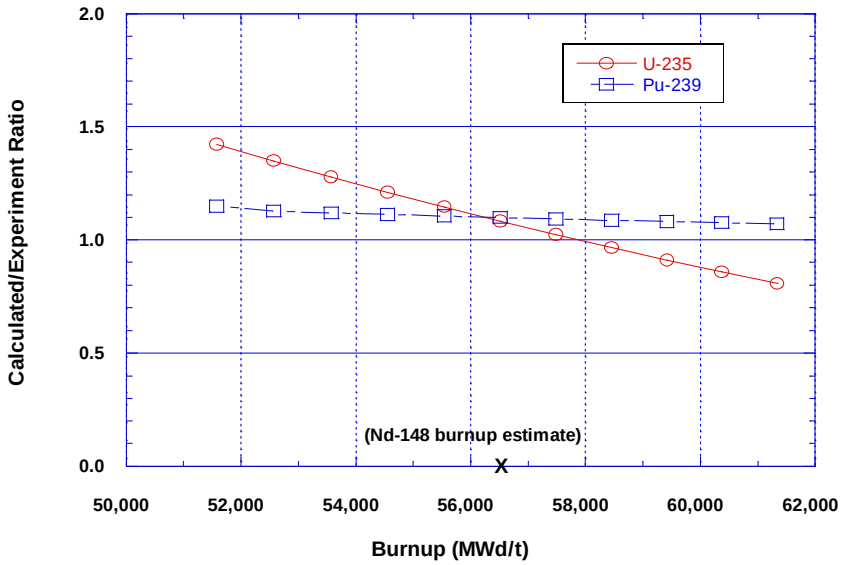


Fig. 9. The C/E ratios for two fissile species in DU1as a function of the burnup value used for simulation. The best estimate of sample burnup is marked on the abscissa.

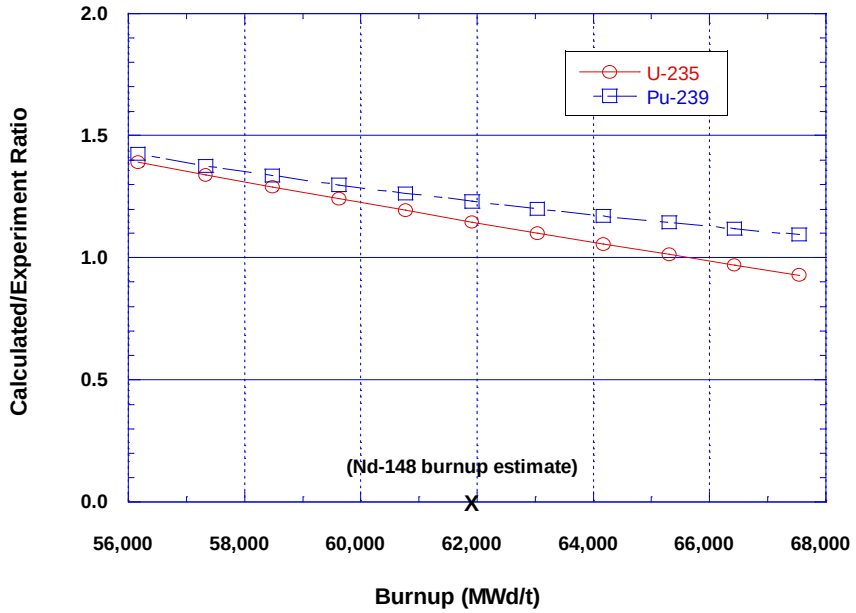


Fig. 10. The C/E ratios for two fissile species in DM1as a function of the burnup value used for simulation. The best estimate of sample burnup is marked on the abscissa.

ACKNOWLEDGMENTS

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