Comparisons of Calculated and Measured ^{241}Am and ^{243}Am Concentrations in PWR and VVER Spent Fuel

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ABSTRACT

A study was performed at Los Alamos National Laboratory to explore the accuracy of several reactor analysis codes in calculating ^{241}Am and ^{243}Am concentrations in light water reactor spent fuel. Calculated higher-actinide concentrations were compared to measured values from the literature for two reactor fuels. The fuel samples were taken from the Mihama Unit 3 pressurized water reactor and a VVER-440. The ^{241}Am and ^{243}Am concentrations were calculated using the HELIOS-1.4 lattice-physics code, the ORIGEN2 burnup code, and a linked MCNP/ORIGEN2 code named Monteburns 3.01. Comparisons were made between the calculated and measured values. It was determined that all codes performed consistently well for the Mihama Unit 3 measurements (within ±5% for ^{241}Am and ±20% for ^{243}Am); however, the ORIGEN2 pressurized water reactor libraries appear to be insufficient for the VVER-440 measurements. The HELIOS and MONTEBURNS codes both demonstrated good ability to calculate these isotopes for VVER-440 fuel (±10% for ^{241}Am and ±12% for ^{243}Am). The accuracies of these codes and the associated radiochemical measurements of these higher-actinide isotopes may be insufficient for safeguards and fuel management purposes; thus, development of new methods and modification to existing data libraries may be necessary in order to enable cost-effective safeguarding of these higher-actinide materials.

1. INTRODUCTION

The higher-actinide isotopes (specifically ^{237}Np, ^{241}Am, and ^{243}Am) contribute significantly to the long-term toxicity of high-level wastes from reprocessing plants, and these isotopes (in separated form) could be useful for the construction of nuclear explosives. Substantial quantities of these isotopes may exist in waste streams from reprocessing facilities and, due to their chemical nature, can often be separated without significant difficulty. Accurately calculating the production of these alternate nuclear materials (ANMs) may assist in decreasing the number and cost of measurements necessary to account for these materials. Because of their complicated production and decay modes, the calculations for these isotopes require sophisticated reactor physics codes with accurate cross section and nuclear data libraries. These calculations may also prove useful for waste management and burnup credit applications. The study described below

was performed using modern reactor physics codes and data libraries to determine the accuracy capable with existing methods and data to calculate the quantity of ^{241}Am and ^{243}Am.

Previous work has shown that sufficient nuclear data and methods exist in modern reactor physics codes to accurately calculate ^{237}Np quantities in spent nuclear fuel from pressurized water reactors (PWRs).[1-5] However, little effort has been made to quantify the capability of modern codes and data libraries for calculating ^{241}Am and ^{243}Am,[3-11] especially in other light water reactor (LWR) fuels such as those from Russian water cooled, water moderated energy reactors (VVERs).

2. REACTOR PHYSICS CODES

Three different code systems were chosen for evaluation in this study: HELIOS-1.4, ORIGEN2, and Monteburns 3.01. The three systems chosen were markedly different in both methods and data used. Each code is of modern design and uses nuclear data derived from the Evaluated Nuclear Data Files (ENDF). Descriptions of each code system and the source of their nuclear data are given below.

2.1. THE HELIOS-1.4 LATTICE PHYSICS CODE

The HELIOS-1.4 lattice physics code uses the method of angularly dependent, current-coupled collision probabilities to solve the neutron transport equation in nearly arbitrary geometries.[12] HELIOS-1.4 uses a set of multigroup cross sections (34, 89, or 190 neutron energy groups) derived from the ENDF/B-VI nuclear data file. It should be noted that the ^{238}U resonance parameters in these libraries have been modified to correspond to those available in ENDF/B-V. HELIOS has options available for various reactor physics capabilities, including critical spectrum calculations and input bucklings. HELIOS has been benchmarked extensively for critical experiments, assembly pin powers, and uranium, plutonium, and fission product isotopics.[13-15] However, little effort has been spent quantifying the ability of the HELIOS-1.4 code system in calculating higher-actinide quantities in spent nuclear fuel.

2.2. THE ORIGEN2 BURNUP CODE

The ORIGEN2 burnup code uses the exponential method to solve the time-dependent burnup equations.[16] One-group cross sections, flux spectra, and fission yields are acquired from predetermined, reactor-specific libraries. Current versions of ORIGEN2 contain libraries for U.S. pressurized and boiling water reactors (PWRUS and BWRUS), European pressurized and boiling water reactors (PWRE and BWRE), liquid metal fast breeder reactor (LMFBR) driver and blanket fuel, Canada Deuterium-Uranium (CANDU) reactors, as well as recycled fuel constants. Some newer libraries have been produced for different reactor designs,[17,18] but these have not had wide-scale application. The nuclear data used in producing the ORIGEN2 one-group libraries are primarily from ENDF/B-IV and ENDF/B-V.

ORIGEN2 has been used extensively for fuel management, waste analysis, shielding studies, and safeguards investigations (too many to be referenced here). Consequently, numerous benchmarking studies have been performed to illustrate the code's ability to predict uranium,

plutonium, and fission product concentrations in spent nuclear fuel (esp. for PWRs and BWRs). Significantly less effort has been spent determining the code's accuracy in predicting ^{241}Am and ^{243}Am concentrations.[3,19,20] This is primarily because of these isotopes' decreased importance in reactor operations and the difficulty associated with acquiring benchmark-quality measurements of these isotopes.

It is important to note that a significant difference exists between ORIGEN2 and the other reactor analysis codes used in this study. ORIGEN2 does not make use of a transport calculation for the specific case studied to determine collapsed cross sections and fluxes. These calculations were performed on a general basis before execution of ORIGEN2. Thus direct comparison to the other codes examined here may be inappropriate in some cases; however, the widespread use of this code mandates its inclusion in these analyses.

2.3. MONTEBURNS 3.01: A LINKED MCNP/ORIGEN2 CODE

The Monteburns 3.01 code system[21] is a newly developed code at Los Alamos National Laboratory (LANL) that links the Monte Carlo simulation code MCNP[22] to the burnup and depletion code ORIGEN2[16]. Essentially, Monteburns consists of a UNIX c-shell command file that calls a FORTRAN77 program that processes input and output files for MCNP and ORIGEN2. The program processes user input values to specify the system geometry, initial material compositions, material feed and removal, and a series of other parameters required by MCNP and ORIGEN2. The c-shell command file successively executes MCNP, the FORTRAN77 program, and ORIGEN2 to generate time-dependent results such as pin powers and spent fuel isotopics. Monteburns also outputs numerous results from ORIGEN2 and MCNP as the programs are executed. The FORTRAN77 code compiles the extensive MCNP and ORIGEN2 output files into a simpler and more compressed set of output files for use in post-processing or graphical display.

Monteburns has been applied in several space-reactor studies[23] and in some accelerator system studies[24] but has not yet been thoroughly benchmarked for conventional reactor applications.[25] There are several advantages in using a Monte Carlo code for reactor simulations, including detailed resonance self-shielding capabilities, more accurate three-dimensional (3D) transport than finite difference and nodal methods, and unparalleled versatility in handling geometric configurations and material compositions. Monteburns also has several disadvantages when compared to deterministic codes, including an inability to perform critical spectrum calculations, difficulty in capturing low probability reactions [such as (n,α) and (n,3n) reactions], and a requirement for much larger computational resources for generating flux solutions. Codes similar to Monteburns (i.e., Monte Carlo and burnup linked codes) have been used in the past,[26,27] but these have not yet gained wide acceptance because they require significant computational resources (which were not readily available at the time).

Monteburns makes use of fission product yields and decay constants from the specified ORIGEN2 library but generates one-group cross sections by collapsing reaction rates from MCNP. Thus, Monteburns can utilize any cross section data that can be processed for use with MCNP. The most common method for generating MCNP cross section data is through the use of the NJOY nuclear data processing code.[28] NJOY can be used to produce temperature-dependent,

continuous-energy cross section sets for any isotope in ENDF. The cross sections used in this study for Monteburns were processed with NJOY97 from ENDF/B-VI and ENDF/B-V data files.

3. SPENT FUEL MEASUREMENTS

Numerous spent fuel measurements exist in the literature for uranium, plutonium, and fission product isotopics; however, fewer measurements have been made for the higher-actinide isotopes, especially for more modern reactor fuels. To evaluate the capabilities of the three code systems considered here, a small set of spent fuel measurements from the literature for two reactor types were chosen. The measurements were performed on spent fuel samples from the Mihama Unit 3 PWR and a VVER-440. Included in each of these experiments are measurements of the concentration of ^{241}Am and ^{243}Am, as well as a burnup determination. The sample dissolutions were performed at laboratories located in their respective regions; thus, the experiments essentially constitute an independent set of experimental measurements. Below is a description of each reactor system and the fuel characterizations performed. A listing of the measured fuel burnup values for each sample used in the comparisons is included in Table I.

3.1. MIHAMA UNIT 3

The Mihama Unit 3 reactor is a PWR consisting of Westinghouse-type 15 × 15 assemblies with 3.20 w/o ^{235}U fuel surrounded by Zircaloy-4 cladding. The reactor operates with a nominal specific power of 34.0 W/g. A listing of the fuel parameters used in these analyses can be found in Table II. Nine spent fuel samples, with burnup ranging from 6,900 to 34,100 MWd/MTU and cut from three assemblies, were surveyed at the Tokai Research Establishment in Japan. The fuel was extensively examined using alpha, mass, and gamma-ray spectrometry. Ceramography of the fuel was also performed.[29]

The actinide concentrations were determined from radiochemical analyses (i.e., α- and γ-spectroscopy) of the fuel samples. Mass concentrations per metric ton of fuel were listed for ^{237}Np, ^{241}Am, and ^{243}Am. The percent isotopic compositions of ^{240}Pu and ^{239}Pu were also listed. All of the data reported were from the time of measurement. Burnup was determined using destructive ^{137}Cs and ^{148}Nd examinations. No measurement uncertainties were listed.

3.2. VVER-440

In the early 1980s, several fuel samples from VVER-440 spent fuel were radiochemically analyzed to determine the quantity of ^{241}Am, ^{243}Am, and curium and plutonium isotopes in the fuel.[30] The VVER-440 reactor is a PWR consisting of triangular-lattice fuel assemblies with 126 UO_2 fueled rods per assembly.[31] The 2.40 wt% fuel is clad with Zr-1% Nb, and the reactor operates with a nominal specific power of 33.0 W/g. A listing of the fuel parameters used in these analyses can be found in Table III. Seven spent fuel samples, with burnup ranging from 20,500 to 38,500 MWd/MTU and cut from the same height in one assembly, were surveyed at the V.G. Khlopin Radium Institute in Leningrad (present day St. Petersburg).

The actinide concentrations were determined from mass-spectroscopy of the fuel samples. Mass concentrations per metric ton of fuel were listed for ^{241}Am and ^{243}Am as well as for several

curium and plutonium isotopes. All of the data reported were from the time of measurement. Burnup was determined using destructive ^{148}Nd examinations. Measurement uncertainties were included and generally were on the order of ±10%.

4. CALCULATIONAL MODELS

Calculations were performed to simulate the irradiation of each of the above fuels using the three code systems of interest. In all cases, an assumed average specific power for the assembly was used (34.0 W/g for Mihama Unit 3 and 33.0 W/g for the VVER-440), and each fuel was modeled as having been irradiated from 0 to 50,000 MWd/MTU, using 1000 MWd/MTU burnup steps. Reactivity control (i.e., chemical shim concentrations and control rod movements) was neglected in this study due to the added modeling complexity and relatively small effect on higher actinide isotopics.[25] Descriptions of each of the models used for each code system are given below.

4.1. HELIOS-1.4

All HELIOS-1.4 calculations were performed using a two-dimensional (2D) pin cell model similar to that described elsewhere.[25] This model has been shown to be sufficiently accurate for PWR actinide and fission product concentrations from 0 to 50,000 MWd/MTU. A bulk coolant density of 0.703 g/cc was used for the Mihama Unit 3 simulations. A bulk coolant density of 0.749 g/cc was used for the VVER-440 simulations. The fuel-to-clad gap was ignored in all calculations. The central hole of the VVER-440 fuel was simulated using a small void region (filled with low-density oxygen gas). Critical spectrum calculations were used for all simulations. All other material and geometric properties (including the triangular VVER lattice) were simulated explicitly. The 89-group HELIOS library was used for all cross section data.

4.2. ORIGEN2

All ORIGEN2 calculations were performed using a stepwise irradiation of one metric ton of UO_2 fuel using prebuilt ORIGEN2 nuclear data libraries (essentially a zero-dimensional calculation) with constant power irradiations. The PWRUS data libraries were used for all Mihama Unit 3 calculations. The PWRE libraries were used for all the VVER-440 libraries. No specific VVER-440 library was available for use; thus, because of the lower enrichment of the VVER fuel (2.4 wt%), the European PWR libraries were employed. In the future, it would be advisable to develop a specific VVER library (as well as RBMK and BN-350 libraries).

4.3. MONTEBURNS 3.01

All Monteburns 3.01 calculations were performed using a 2D pin cell model similar to that described elsewhere[25] and used in the HELIOS calculations described above. A bulk coolant density of 0.703 g/cc was used for the Mihama Unit 3 simulations. A bulk coolant density of 0.749 g/cc was used for the VVER-440 simulations. The fuel-to-clad gap was ignored in all calculations. The central hole of the VVER-440 fuel was simulated using a small void region (filled with low-density helium gas). All other material and geometric properties were simulated explicitly. The Monteburns 3.01 calculations used all ENDF/B-VI Mod 3 data except for the ^{238}U cross sections, which were produced from ENDF/B-V. All cross section data were

processed using NJOY97 at the fuel, clad, and coolant temperatures described in Section 3 above.

5. RESULTS AND DISCUSSION

Plots of the calculated and measured ^{241}Am and ^{243}Am concentrations versus burnup for each reactor fuel are shown in Figs. 1-4. The average percent differences between the calculated and measured values for the ^{241}Am and ^{243}Am concentrations for each fuel and using each code system are given in Tables 4 and 5, respectively. Generally, reasonable agreement is found in all case except for the ORIGEN2 calculations for the VVER-440 fuel. However, upon further investigation, several trends and characteristics become apparent.

ORIGEN2 is unable to accurately calculate the americium isotopes for VVER-440 spent fuel using the PWRE library. The other PWR libraries available in ORIGEN2 were also studied to some degree, and it was determined that they yielded similarly poor results for these isotopes. It remains to be determined whether the ORIGEN2 PWR libraries will yield reasonable calculations of plutonium isotopes for VVER-440 reactors. Based on these conclusions, specific VVER libraries would need to be developed if ORIGEN2 were to be used for the calculation of ANMs in VVER fuel.

For the Mihama Unit 3 measurements, all code systems yield generally good results for the ^{241}Am concentrations; however, the calculated ^{243}Am concentrations tend to be systematically low by ~15% (especially at higher burnups). This trend has been observed in some other PWR fuel comparisons and may suggest that the ENDF/B-VI ^{243}Am absorption cross sections are overestimated.[1] This conclusion is contradicted by the VVER results for ^{243}Am (Fig. 4), where the calculations are in good agreement with the measured values. It should be noted that the VVER experiments are mass-spectrometry measurements, whereas the PWR experiments (reported here and in Ref. 1) are α-spectrometry measurements. This tends to suggest that better measurement techniques are needed for the accurate determination of ^{243}Am quantities in spent fuel and before drawing any solid conclusions about the existing ^{243}Am cross section data. It is also interesting to note that the ORIGEN2 ^{241}Am values tend to be underestimated at high burnups. This is most likely because older cross section data are used in ORIGEN2.

The results presented here suggest that both HELIOS and Monteburns can generate accurate ^{241}Am predictions for PWR and VVER spent fuel; however, ORIGEN2 should not be used for VVER reactors unless a specific VVER library is developed. Also, HELIOS tends to be more accurate than Monteburns for the calculation of these isotopes (most likely because of the addition of the critical spectrum capabilities in HELIOS). The existing ^{241}Am measurement techniques and calculations are reasonably accurate (yielding ~5% accuracy), but the ^{243}Am measurements (and perhaps calculations) are insufficient for some purposes. It is suggested that better spent fuel measurements (especially for more modern fuels) are necessary to allow proper determination of the need for alteration of the reported cross section for ^{243}Am.

CONCLUSIONS

This study explored the accuracy of three reactor analysis codes (the HELIOS-1.4 lattice-physics code, the ORIGEN2 burnup code, and a linked MCNP/ORIGEN2 code named Monteburns 3.01) in calculating ^{241}Am and ^{243}Am concentrations in light water reactor spent fuel. Calculated higher-actinide concentrations were compared to measured values from the literature for Mihama Unit 3 PWR fuel and VVER-440 fuel. It was determined that all codes performed consistently well for the Mihama Unit 3 measurements. It was determined that the ORIGEN2 pressurized water reactor libraries are insufficient for the VVER-440 measurements. The HELIOS and MONTEBURNS codes both demonstrated good ability to calculate these isotopes for VVER-440 fuel. Significant scatter existed in the measured results, suggesting that the accuracies of the radiochemical measurements of these higher-actinide isotopes (esp. ^{243}Am) may be insufficient for safeguards and fuel management purposes. Also, development of new methods and modification of existing data libraries may be necessary to enable cost-effective safeguarding of these higher-actinide materials.

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Table I. Measured Burnup Values for Each Fuel Sample Used in Comparisons

Fuel Sample	**Burnup (MWd/MTU)**
Mihama Unit 3	
86B02	*8300*
86B03	*6900*
86G05	*15300*
86G03	*21200*
86G07	*14600*
87C03	*29440*
87C04	*32300*
87C07	*33700*
87C08	*34100*
VVER-440	
1	*20500*
2	*22500*
3	*33500*
4	*34200*
5	*34600*
6	*36100*
7	*39000*

Table II. BOL Fuel Assembly Characteristics Used in the Calculations for Mihama Unit 3 Fuel

Vendor:	Mitsubishi Heavy Industries
Type:	15×15 (square)
Pin-to-Pin Pitch:	1.4920 cm[a]
Fuel Pellet Diameter:	0.932 cm
Clad Inner Diameter:	0.948 cm
Clad Outer Diameter:	1.072 cm
Fuel Density:	10.198 g/cc
Fuel Enrichment:	3.20 w/o ^{235}U
Fuel Temperature:	700 K
Active Fuel Length:	366 cm
Clad Material:	Zircaloy-4
Clad Density:	6.56 g/cc
Clad Temperature:	600 K
Coolant Material:	Light Water
Coolant Density:	0.7027 g/cc
Coolant Temperature:	561 K
Specific Power:	34.0 W/g
Decay Time:	2853 days

[a] Adjusted to produce a pin cell with the same fuel-to-moderator ratio as an assembly.

Table III. BOL Fuel Assembly Characteristics Used in the Calculations for VVER-440 Fuel

Vendor:	Ministry of Power and Electrification of the USSR
Type:	126 rods in hexagonal lattice
Pin-to-Pin Pitch:	1.2828 cm[a]
Fuel Pellet Diameter:	0.756 cm
Clad Inner Diameter:	0.780 cm
Clad Outer Diameter:	0.910 cm
Fuel Density:	10.4 g/cc
Fuel Enrichment:	2.4 w/o ^{235}U
Fuel Temperature:	700 K
Active Fuel Length:	250 cm
Clad Material:	Zr-1% Nb
Clad Density:	6.49 g/cc
Clad Temperature:	610 K
Coolant Material:	Light Water
Coolant Density:	0.749 g/cc
Coolant Temperature:	540 K
Specific Power:	33.0 W/g
Decay Time:	3900 days

[a] Adjusted to produce a pin cell with the same fuel-to-moderator ratio as an assembly.

Table IV. Comparisons of Average Percent Difference between Calculated and Measured Values of ^{241}Am for Each Reactor Fuel and Each Code System

	HELIOS	ORIGEN2	Monteburns
Mihama Unit 3	*3.7 %*	*4.6 %*	*4.1 %*
VVER-440	*7.3 %*	*32.4 %*	*10.5 %*

Table V. Average Percent Difference between Calculated and Measured Values of ^{243}Am

	HELIOS	ORIGEN2	Monteburns
Mihama Unit 3	*17.7 %*	*14.4 %*	*16.8 %*
VVER-440	*9.8 %*	*139.9 %*	*11.6 %*

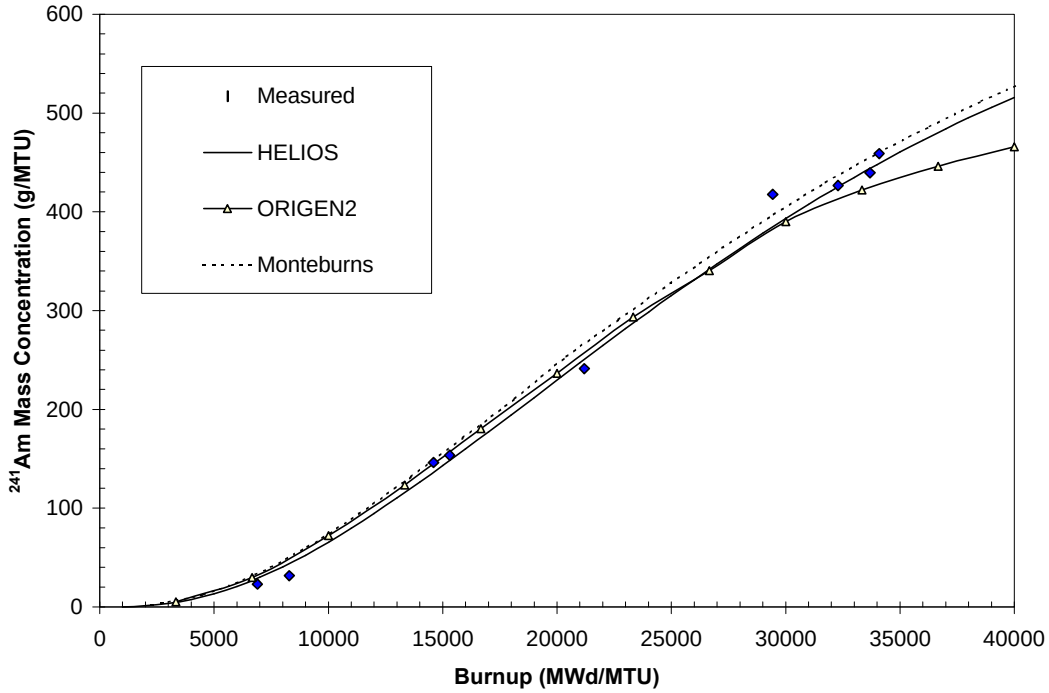

Fig. 1. ^{241}Am concentration versus burnup for a Mihama Unit 3 fuel pin (calculated and measured).

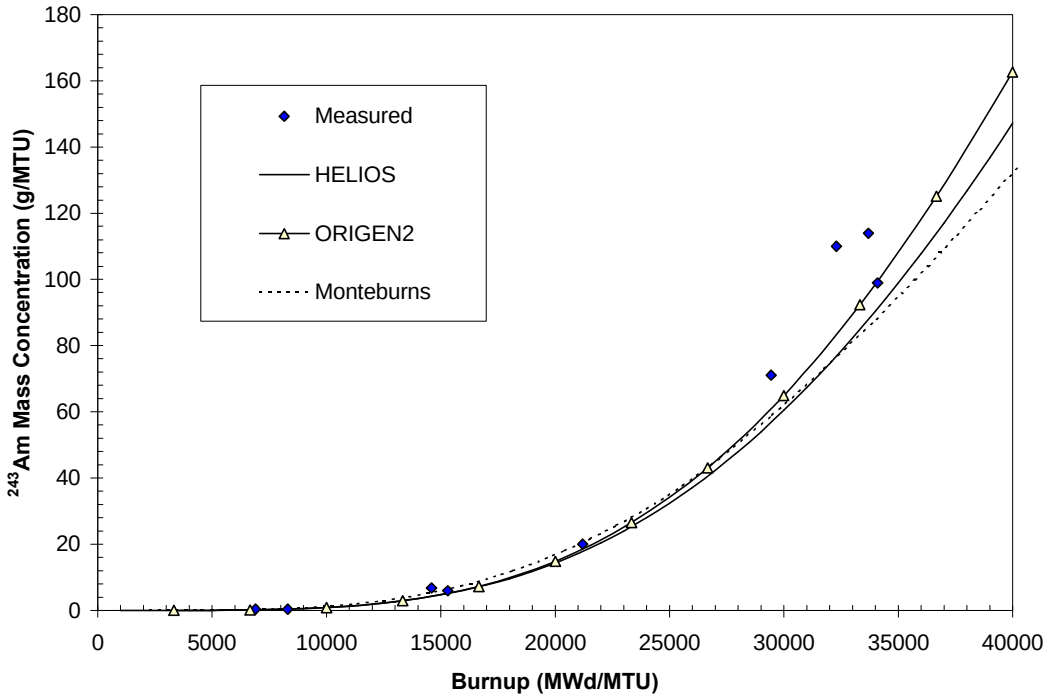

Fig. 2. ^{243}Am concentration versus burnup for a Mihama Unit 3 fuel pin (calculated and measured).

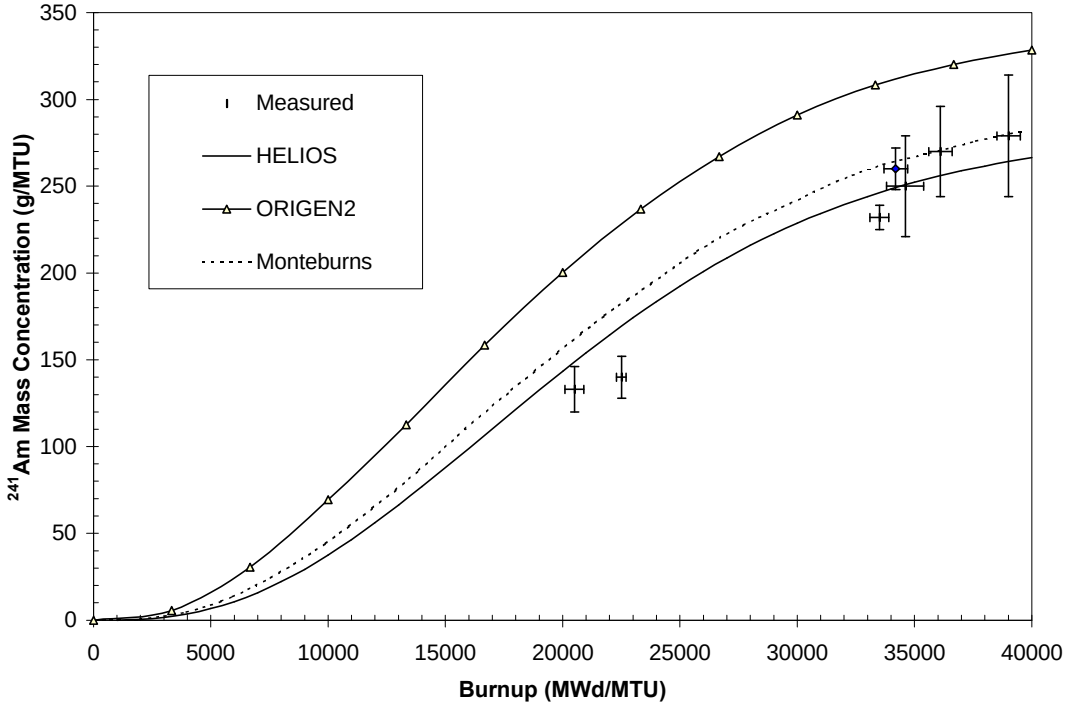

Fig. 3. ^{241}Am concentration versus burnup for a VVER-440 fuel pin (calculated and measured).

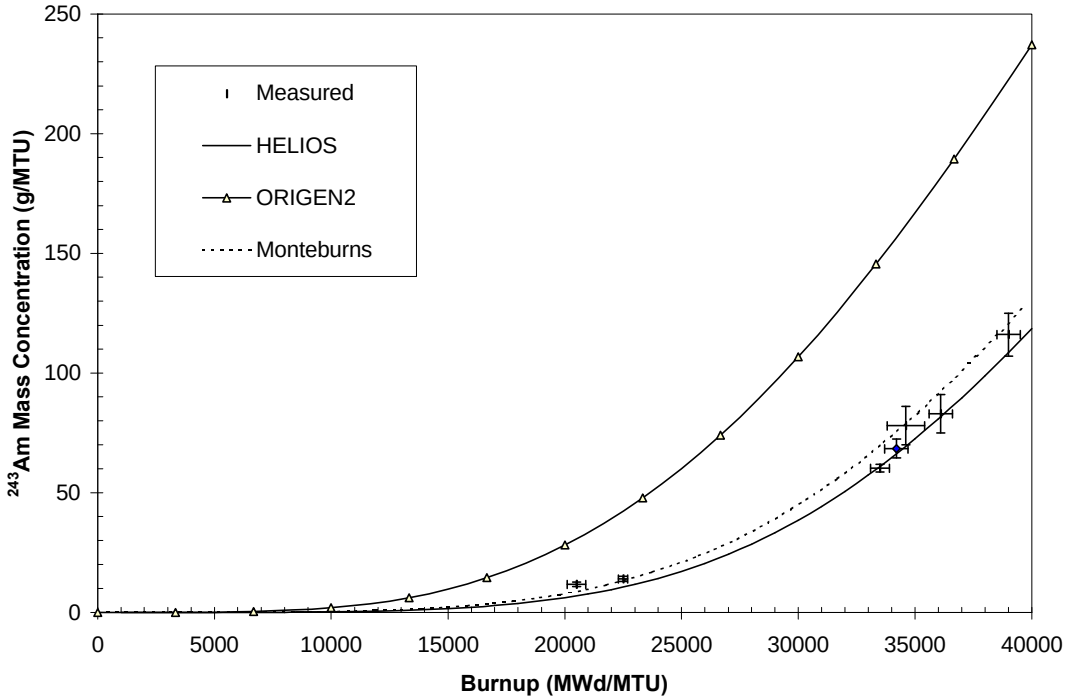

Fig. 4. ^{243}Am concentration versus burnup for a VVER-440 fuel pin (calculated and measured).