

VALIDATION OF MONTEBURNS FOR MOX FUEL USING ARIANE EXPERIMENTAL RESULTS

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ABSTRACT

The Monte Carlo burnup code *Monteburns*, which couples MCNP or MCNPX with ORIGEN2.1, was used to calculate the isotopic composition of the M308 MOX fuel assembly irradiated at the Beznau PWR in Switzerland. Two segments (BM5 and BM6) from fuel rod K7 in this assembly were analyzed radiochemically as part of the ARIANE Project. The calculation is based on a three-dimensional, radially reflected model of the single fuel assembly, which includes details of the axial reflectors and spacers. The irradiation history covers six consecutive reactor cycles (20 through 25) from 2 July 1990 to 28 June 1996. The calculation was performed with MCNPX 2.4.K. The continuous energy cross-section libraries used by MCNPX were generated with the NJOY code system for the specific fuel and coolant temperature profiles encountered during this irradiation. The burnups calculated for segments BM5 and BM6 agree with the recommended experimental values within 5%, and the corresponding isotopic content of these segments generally straddle the measured concentrations within the 2- σ error band. A detailed comparison is presented for the composition of fuel segment BM5.

Key Words: Monte Carlo, burnup, MONTEBURNS, MCNPX, ORIGEN2.1

1. INTRODUCTION

The ARIANE (Actinides Research In A Nuclear Element) Project was launched in 1994 with the aim of generating an accurate experimental database of isotopic inventories for irradiated UO₂ and MOX fuel. The purpose of such a database is to facilitate the validation of codes used for source-term prediction in reactor safety research, the investigation of high-burnup cores and of MOX fuel recycling scenarios. Irradiated fuel samples were selected from several light-water reactors, including the Dutch Dodewaard BWR (UO₂ and MOX) and the Swiss Goesgen (UO₂) and Beznau (MOX) PWRs. These samples were analyzed radiochemically by three nuclear centres—SCK·CEN (Belgium), PSI (Switzerland) and ITU (Germany)—using different analytical methods. The final report with the recommended sample compositions was issued in December 2000 [1].

Burnup calculations have been traditionally performed using deterministic lattice codes, which utilize two-dimensional (2D) models to deplete the fuel as a function of power, fuel and coolant temperatures and boron concentration. The resulting nuclide densities are used to compute few-group, macroscopic cross-sections for use in three-dimensional (3D) diffusion calculations. With the increased use of Monte Carlo methods for core design and criticality analysis, several codes have become available that couple MCNP with ORIGEN2 for the fuel depletion [2]. *Monteburns* is the latest of these Monte Carlo fuel depletion code systems [3].

Monteburns was used to analyze MOX fuel assembly M308, which was irradiated at Beznau Unit 1 during six consecutive reactor cycles (20 through 25) from 2 July 1990 to 28 June 1996. The results of the *Monteburns* analysis are presented in this report, which is organized as follows: a description of the code system and the associated cross-section libraries is provided in Section 2; the ARIANE project is described in Section 3; details of the MCNPX model appear in Section 4, and the *Monteburns* burnup calculation is outlined in Section 5. The results of the analysis are summarized in Section 6 and discussed in Section 7. Concluding remarks appear in Section 8.

2. THE CODE SYSTEM

The burnup code system used in this study to model the irradiation of the M308 fuel assembly consists of the Monte Carlo transport code MCNPX, the ORIGEN2.1 isotope generation and fuel depletion code, and the *Monteburns* program that couples these two codes. These three components of the code system, and the cross-section libraries used in this study, are described below.

2.1 MCNPX

MCNP (Monte Carlo N-Particle) is a general-purpose, Monte-Carlo transport code [4]. Its generalized geometry features and use of continuous-energy cross-sections can generate benchmark-quality results for a variety of reactor applications. The standard version of the code, MCNP4C3, is capable of transporting neutrons, photons and electrons. A high-energy version, MCNPX, is capable of extending the coupled transport capability to 36

elementary particles including protons [5]. The calculations reported here were performed using MCNPX Version 2.4.K.[†] MCNPX can be used to calculate spectrum-averaged, one-group cross-sections and fluxes for any system. Its ability to model detailed 3D geometries makes MCNPX more useful than 2D discrete-ordinates codes.

2.2 ORIGEN2

The ORIGEN2.1 isotope generation and depletion code uses either the matrix exponential method or the Bateman equations to solve a set of coupled ordinary differential equations that describe the transmutation and decay of radionuclides [6,7]. An isotope and photon decay library and a large number of one-group cross-section libraries are provided for different reactor systems. Use of the code requires specification of the initial material inventories, continuous material feed and removal rates when applicable, and the irradiation history (power or total flux and duration). The output at each time step includes detailed material inventories for actinides, fission and activation products, and various other parameters.

2.3 MONTEBURNS

Monteburns is a fully automated tool that links MCNP or MCNPX with ORIGEN2.1 [3]. It is also compatible with ORIGEN2.2 [8] and CINDER90 [9]. The code consists of a *Perl* script and the FORTRAN executable program *monteb*, which manipulate the inputs and outputs of MCNPX and ORIGEN2.1 in an automated burnup sequence. MCNPX provides one-group microscopic cross-sections and fluxes halfway through each step to ORIGEN2.1 for depletion calculations, and ORIGEN2.1 is used to calculate isotopic inventories both halfway through and at the end of each step. MCNPX calculates the effective one-group cross-sections for the system modeled, for user-specified isotopes and/or isotopes that contribute significantly to the absorption, fission, mass, or atom fraction of materials in the system. Because MCNPX cross-section libraries do not exist for all isotopes tracked in ORIGEN2.1, default cross-sections are used for the missing isotopes. Temperature-dependent calculations can be performed by changing the cross-section libraries during the burnup sequence. These libraries are generated at specific temperatures using the NJOY code system [10]. A flow chart showing the *Monteburns* calculation sequence appears in Figure 2-1.

2.4 NJOY

The NJOY99 nuclear data processing system is a modular computer code used for converting evaluated nuclear data in the Evaluated Nuclear Data File (ENDF) format into libraries useful for continuous-energy MCNP and MCNPX calculations [10]. It handles a wide variety of nuclear effects, including resonances, Doppler broadening, heating (KERMA), radiation damage, thermal scattering, gas production, neutrons and charged particles, photo-atomic interactions, self shielding, probability tables, photon production and high-energy interactions (up to 150 MeV).

[†] MCNPX is also used at PSI for high-energy physics applications, which include the modeling of the SINQ neutron spallation source.

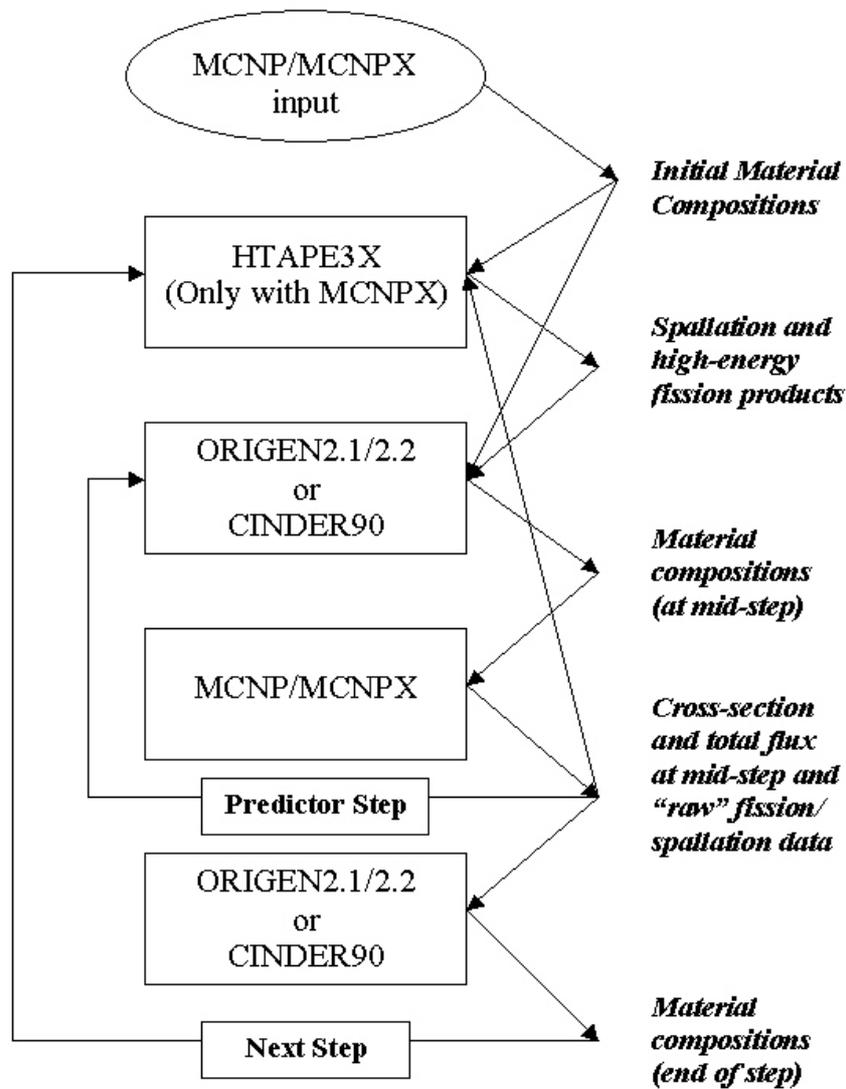


Figure 2-1. Monteburns calculation flowchart

NJOY99 was used to generate the MCNPX cross-section libraries from the ENDF/B-VI (Release 6) nuclear data file for the fuel and coolant nuclides that were modeled with *Monteburns*. Isotopes of the following elements were included: H, Be, B, O, Na, Mg, Al, Si, Cl, Ti, Cr, Mn, Fe, Co, Ni, Zr, Nb, Mo, Tc, Ru, Rh, Pd, Ag, I, Xe, Cs, Pr, Nd, Pm, Sm, Eu, Hf, Pb, Bi, Th, U, Np, Pu, Am and Cm. These cross sections were processed in a representative mesh over the full range of temperatures (285°C to 1004°C) encountered during the irradiation of the MOX fuel assembly.

3. THE ARIANE PROJECT

The ARIANE Project was initiated by Belgonucleaire to improve the accuracy of source-term prediction for highly burned UO₂ and MOX fuel [1]. The actinide and fission-product inventories of irradiated fuel are used in a variety of safety and licensing applications, including the determination of coolant activities, shielding, dose assessment, spent fuel transport and storage, safeguards, and accident analysis. Post-irradiation examinations were carried out on spent fuel from commercial PWR and BWR power plants.

3.1 The Experimental Program

The project generated an extensive experimental database of spent-fuel compositions, based on measurements of up to 53 isotopes carried out by different laboratories using a variety of radiochemical methods. The analyzed samples came from fuel irradiated in the Dodewaard BWR (Netherlands), and in the Goesgen and Beznau-1 PWRs (Switzerland).

Samples of the MOX fuel assembly irradiated in the Beznau-1 PWR were analyzed at the Belgian Nuclear Research Centre (SCK·CEN) and the Paul Scherrer Institute (PSI) in Switzerland. The reference analytical technique at SCK·CEN is Thermal Ionization Mass Spectrometry, while PSI uses High Pressure Liquid Chromatography coupled to an Inductively Coupled Plasma Mass Spectrometer. In addition, Secondary Ion Mass Spectrometry was used at PSI to determine the radial distribution of isotopes in fuel pellets.

The selected isotopes included the most relevant basic actinides, minor actinides and fission products. These were grouped into the so-called Base and Top 25 Programs. The Base group consisted of 39 isotopes, including U (atomic mass 234 to 238), Pu (238 to 244), ²³⁷Np, Am (241 to 243) and Cm (242 to 245). The fission products were chosen for their ability to determine burnup (Nd, Cs), high neutron absorption cross-sections (Sm, Eu) or long half-lives (¹²⁹I, ⁹⁹Tc). The Top 25 group extended the range of measured isotopes to a low-concentration actinide (²³²U), lanthanides (¹⁴⁷Pm and the strongly absorbing ¹⁵⁵Gd) and metal isotopes requiring special analytical methods (⁹⁰Sr, ⁹⁵Mo, ¹⁰¹Ru, ¹⁰⁶Ru, ¹⁰³Rh, ¹⁰⁹Ag and ¹²⁵Sb).

A key objective of the ARIANE program was to ensure a high level of accuracy for the isotopic measurements. This was achieved by repeating the radiochemical analyses at different laboratories using a variety of techniques. The resulting uncertainties were less than 5% for the major actinides (< 1% for the Pu vector), approximately 3% for the burnup indicators, and less than 10% for most of the other isotopes. ⁹⁹Tc, ¹⁰¹Ru, ¹⁰⁶Ru and ¹⁰³Rh were also measured, although with uncertainties reaching 20% for some samples. None of the techniques used succeeded in measuring the ²³²U content.

3.2 The M308 Fuel Assembly

The MOX fuel irradiated at the Beznau-1 PWR was fabricated by Belgonucleaire [11]. M308 is a 14×14 fuel assembly containing 179 fuel rods arranged in three zones with different plutonium contents, varying from 2.75% to 4.96% by weight, and a 1.412 cm lattice pitch. A horizontal view of the fuel assembly is shown in Figure 3-1.

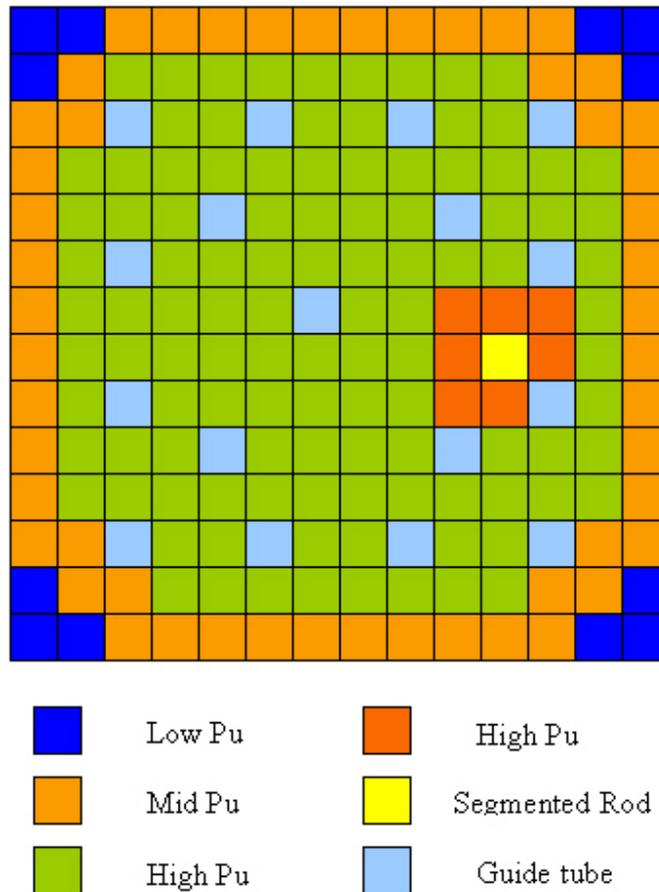


Figure 3-1. The M308 MOX fuel assembly

The active fuel height in the assembly is 302.3 cm. The fuel rods, guide thimbles and instrumentation tube are held by five spacers. Key fuel-assembly design parameters are summarized in Table 3-1.

Table 3-1. MOX fuel assembly design parameters

<i>Component</i>	<i>Value</i>
Array type	14 × 14
Cold lattice pitch (cm)	1.412
Active fuel height (cm)	302.3
Number of fuel rods	179
Number of guide thimbles	16
Number of instrument tubes	1
Number of spacers	5
Structural material	Zr-4 or Inconel-718

Assembly M308 was irradiated in symmetrically located core positions (see Figure 3-2) during cycles 20 through 25 from July 2 1990 until June 28, 1996. The average MOX core loading during this period was 33%. The main core data for the Beznau-1 PWR are summarized in Table 3-2.

Table 3-2. Beznau-1 PWR core data

<i>Parameter</i>	<i>Value</i>	<i>Units</i>
Nominal thermal power	1130	MW
Primary system pressure	155	bar
Average coolant temperature	300	°C
Fuel	UO ₂ and MOX	
Fuel density	10.3	g/cm ³
Average MOX core loading	33	%
Cladding material	Zr-4	
Number of fuel assemblies	121	
Cold assembly pitch	19.82	cm

Fuel rod K7, which achieved a relatively high burnup (~50 GWD/MTHM) because of its location next to a water hole, was selected for examination within the framework of the ARIANE program. Two segments were cut (BM5 and BM6 in Figure 3-3) that capture regions of maximum and 2/3 of maximum burnup, respectively.

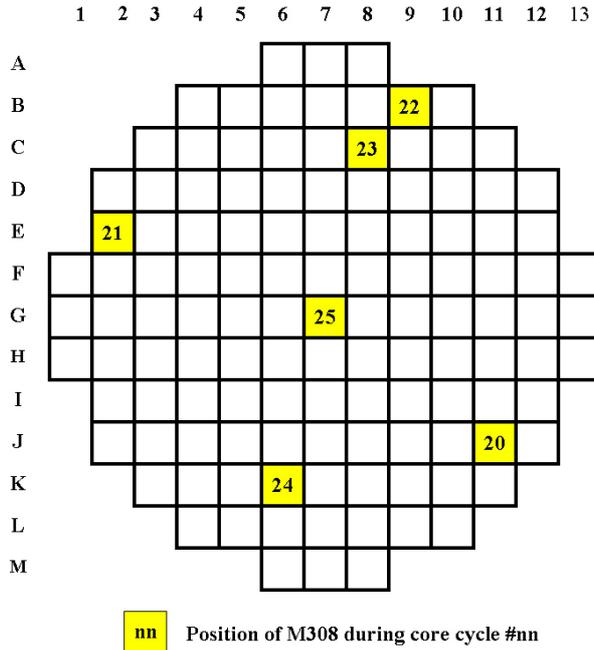


Figure 3-2. Locations of fuel assembly M308 in Beznau-1 core

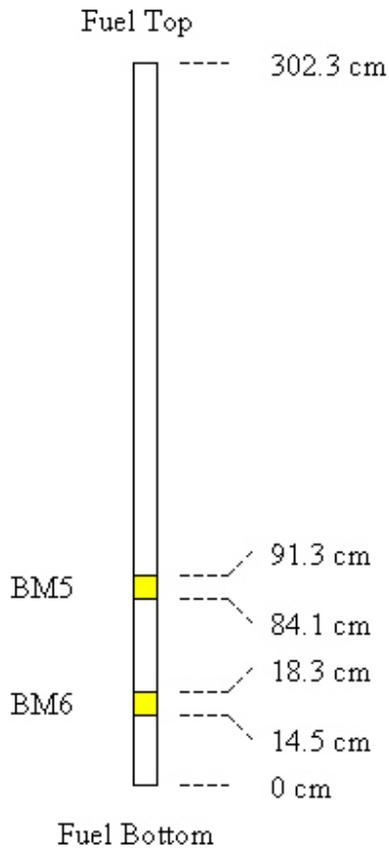


Figure 3-3. Locations of segments BM5 and BM6 in fuel rod K7

4. THE MCNPX MODEL

The detailed three-dimensional MCNPX model, which was used for the *Monteburns* calculations, was based on the CASMO-4/SIMULATE-3 code system neutronic model used for the core-follow of the Beznau-1 PWR at PSI [12,13]. The model included homogenized spacers and top and bottom reflectors. Reflective boundary conditions were used along the sides of the fuel assembly to approximate the entire core, while the axial details served to capture the correct axial buckling. Figure 4-1 shows the horizontal and vertical details of the MCNPX model. The fuel rods with high-plutonium content are shown in green, medium plutonium in gold, and low plutonium in blue. The segmented fuel rod (K7) is shown in yellow, surrounded by orange-coloured high-plutonium fuel rods (*cf.* Figure 3-1).

For the purposes of the burnup calculation, the fuel was divided into 47 radial and vertical depletion zones with the greatest detail in the immediate vicinity of the segmented fuel rod. Thus, the segmented rod was modeled with one radial and 16 axial zones that included segments BM5 and BM6, the seven adjacent pins with high plutonium content were divided into 12 axial zones, while 6 axial zones were used in each of the three radial zones with high-, medium- and low-plutonium content. The final material tracked in *Monteburns* was the borated water coolant, which was not depleted in any given burnup step because it flows continuously through the primary cooling system.

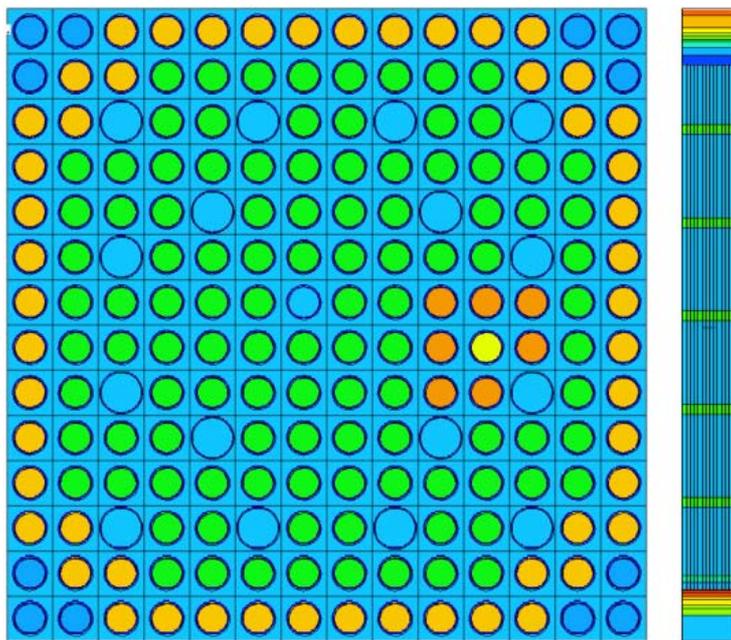


Figure 4-1. MCNPX model of the M308 MOX Fuel Assembly

5. THE MONTEBURNS SIMULATION

The *Monteburns* simulation reproduced the power history (Figure 5-1) of the M308 fuel assembly as described in the ARIANE irradiation report [11]. Adjustments to the water density and boron concentration were made discretely at the end of each burnup step (Figures 5-2 and Figure 5-3). The code was modified to facilitate changing the cross-section libraries during the irradiation to reproduce the corresponding changes in fuel and coolant temperatures. The simulation included the refuelling shutdown periods and the time delay between the irradiation end-of-life and the measurement of individual nuclide concentrations.

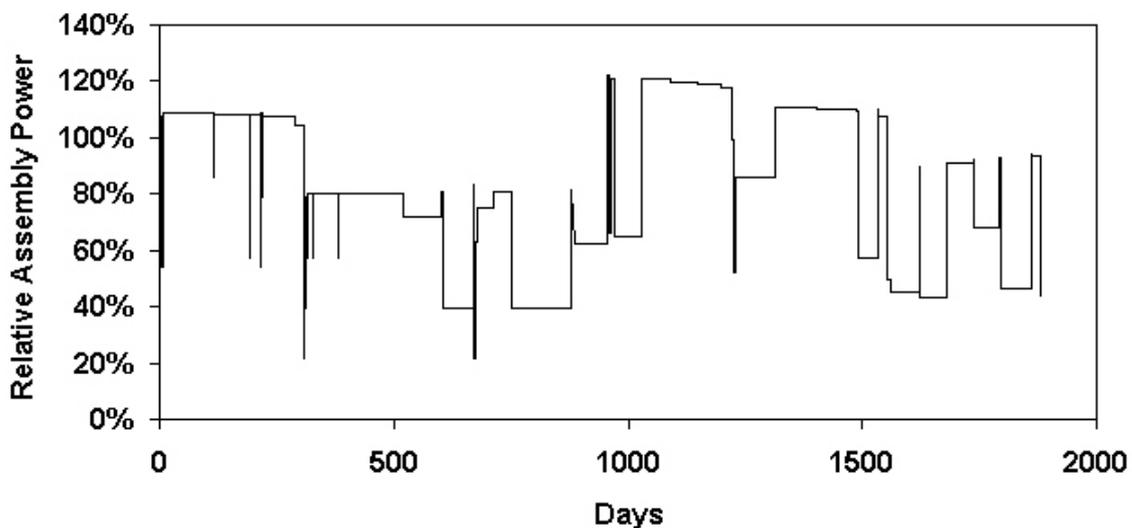


Figure 5-1. The power history of M308 fuel assembly

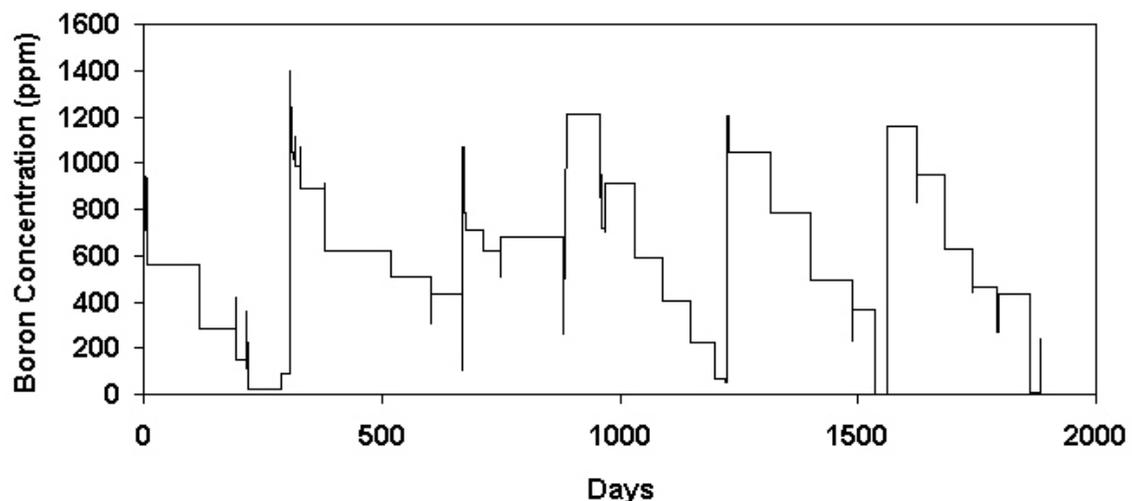


Figure 5-2. Modelled changes in boron concentration in Beznau-1

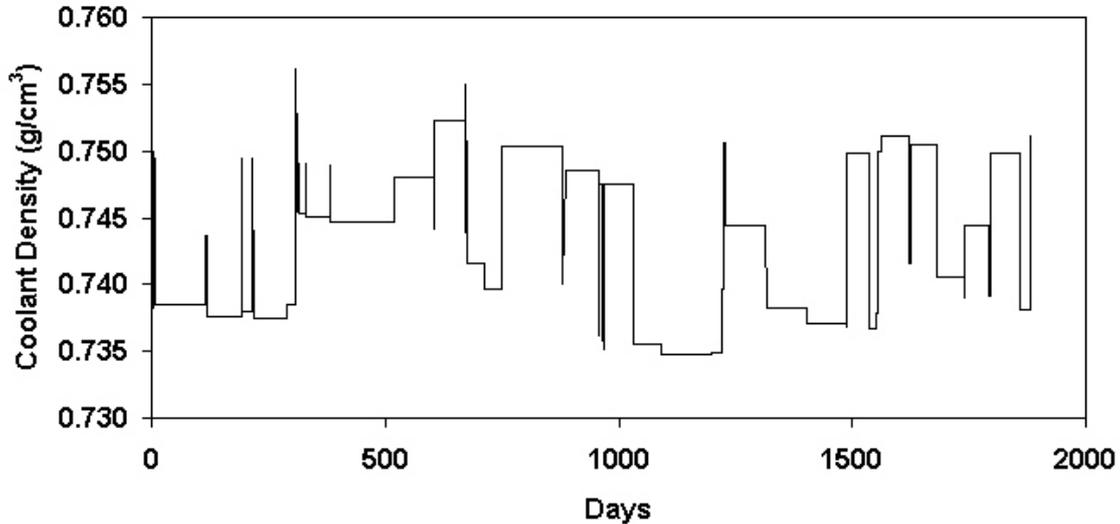


Figure 5-3. Modelled changes in coolant density in Beznau-1

The analysis was carried out using the MCNPX 2.4.K, *Monteburns* 2.0 and ORIGEN2.1 codes. The default ORIGEN2.1 library selected in *Monteburns* was PWRPUPU [6]. A total of 93 burnup steps were simulated with *Monteburns*, with an MCNPX spectrum calculation at the start of the simulation and halfway through each step. The MCNPX calculation was performed using 50 active cycles with 50,000 starting neutron histories per cycle and a continuously updated source file. To achieve reasonable execution times, the simulation was carried out in parallel on the Merlin LINUX cluster at PSI using PVM Version 3.3 [14] and fifteen 1.4 GHz AMD AthlonMP processors. A typical runtime was 1.5 hours per MCNPX case and approximately one week for the detailed fuel irradiation.

6. RESULTS

The results obtained by *Monteburns* are compared below with the measured values recommended by the ARIANE Project [1]. These include the burnup of segments BM5 and BM6, and the concentrations of the actinides and selected fission products in BM5. (The measurements for BM6 were carried out only in one laboratory and are generally considered less reliable.) The results are presented as percent differences between the calculated and measured values, along with the 2σ experimental relative errors.

The agreement between the calculated and recommended segment burnups was 2.83% for segment BM5 and 4.62% for BM6. This confirmed that the three-dimensional MCNPX model calculated the axial flux distribution correctly. The discharge burnup is the most important parameter for the accurate prediction of spent-fuel isotopic inventory.

Table 6-1 shows the results for BM5 isotopic inventory calculation. For the uranium isotopes, the error was typically within 5%. However, the depletion of ^{238}U was overestimated by 0.11 g, which resulted in higher than measured concentrations of plutonium and minor actinides (e.g., an additional 0.07 g buildup of ^{239}Pu). Uranium constitutes approximately 95% of the heavy-metal mass in the MOX fuel assembly. Therefore, the resulting 14% error for ^{239}Pu is relatively large when compared to other isotopes.

Table 6-1. Comparison of calculated BM5 isotopics with measurement

<i>Isotope</i>	$\Delta\%$	<i>Experimental. 2σ error</i>	<i>Isotope</i>	$\Delta\%$	<i>Experimental 2σ error</i>
U234	-1.71%	9.19%	Nd150	3.21%	0.58%
U235	0.87%	2.05%	Pm147	6.44%	10.46%
U236	-5.67%	4.54%	Sm147	3.50%	1.09%
U238	-0.27%	0.45%	Sm148	-3.90%	0.85%
Np237	-4.53%	0.43%	Sm149	14.34%	5.79%
Pu238	-5.36%	3.05%	Sm150	1.66%	0.91%
Pu239	13.86%	0.57%	Sm151	4.02%	1.26%
Pu240	-4.04%	0.57%	Sm152	2.47%	1.04%
Pu241	-9.33%	0.57%	Sm154	1.57%	1.27%
Pu242	-9.26%	0.57%	Eu151	-22.60%	2.10%
Am241	27.47%	2.79%	Eu153	-4.03%	1.07%
Am242m	-0.22%	4.76%	Eu154	18.01%	1.93%
Am243	10.77%	2.84%	Eu155	-32.39%	4.21%
Cm242	0.71%	3.46%	Gd155	-35.17%	3.72%
Cm243	-24.87%	10.60%	Sr90	-2.07%	9.16%
Cm244	-0.48%	2.60%	Mo95	-2.88%	3.61%
Cm245	-3.16%	3.41%	Tc99	5.33%	4.54%
Cm246	-18.12%	4.68%	Ru101	-4.12%	4.27%
Ce144	0.45%	5.15%	Rh103	12.04%	3.99%
Nd142	8.46%	7.03%	Ag109	83.78%	11.32%
Nd143	3.41%	0.56%	I129	68.51%	11.24%
Nd144	-0.63%	0.56%	Cs133	3.28%	2.14%
Nd145	1.19%	0.56%	Cs134	-6.82%	2.32%
Nd146	1.95%	0.56%	Cs135	-0.18%	2.09%
Nd148	2.99%	0.56%	Cs137	-2.34%	2.17%

The comparison of calculated and measured results for most minor actinides and fission products shows agreement within 5%. These include key burnup indicators like Nd and Cs, and most of the strongly absorbing Sm isotopes. However, significant deviations are

observed for some minor actinides (^{241}Am , ^{243}Cm and ^{246}Cm), other strong neutron absorbers (Eu and Gd), and other fission products (^{109}Ag and ^{129}I).

7. DISCUSSION

The excessive depletion of ^{238}U most likely resulted from an overprediction of the one-group capture cross-sections calculated by MCNPX, which was caused by statistical uncertainties in the cross-section calculations, the approximate representation of the energy spectrum or inaccuracies in the ENDF/B-VI cross-sections.

Inaccuracies have been largely eliminated in the latest nuclear data evaluations for well-known isotopes such as ^{238}U . Nevertheless, the current use of NJOY in generating temperature-dependent cross section libraries using ENDF/B-VI data may have led to further inaccuracies. However, these topics are beyond the scope of information presented in this paper.

Table 7-1. Uncertainties in MCNPX absorption cross-sections

<i>Isotope</i>	<i>1σ error</i>	<i>Isotope</i>	<i>1σ error</i>
U234	15.90%	Nd148	9.88%
U235	4.49%	Nd150	31.10%
U236	23.50%	Pm147	13.80%
U238	5.07%	Sm147	10.50%
Np237	13.10%	Sm148	4.62%
Pu238	5.90%	Sm149	6.65%
Pu239	5.40%	Sm150	9.37%
Pu240	7.21%	Sm151	6.40%
Pu241	5.11%	Sm152	20.80%
Pu242	18.30%	Eu154	4.76%
Am241	5.68%	Gd155	7.70%
Am242m	5.50%	Sr90	4.17%
Am243	14.70%	Mo95	16.40%
Cm242	13.20%	Tc99	15.90%
Cm243	6.07%	Ru101	15.20%
Cm244	21.70%	Rh103	9.34%
Cm245	5.63%	Ag109	14.90%
Ce144	4.62%	I129	4.46%
Nd142	5.08%	Cs133	15.10%
Nd143	5.34%	Cs134	4.85%
Nd144	10.60%	Cs135	4.54%
Nd145	11.10%	Cs137	5.60%
Nd146	8.50%	—	—

The 1σ relative errors in the spectrum-weighted absorption cross-sections, which were calculated by MCNPX in the last burnup step for all the isotopes, are shown in Table 7-1. These are approximately 5% for ^{238}U , but surpass 20% for other isotopes. The Monte Carlo statistical errors can be reduced by normalizing the tallies to a larger number of neutron histories, but at the cost of longer execution times.

The *Monteburns* analysis is based on a spectrum calculation for a single fuel assembly reflected infinitely in the radial direction. The resulting multiplication constant is shown in Figure 7-1 as a function of burnup over the six core cycles. The value of k_∞ decreases from 1.13 to 0.92 over the course of the irradiation, showing the effects of burnup and changes in the boron concentration.

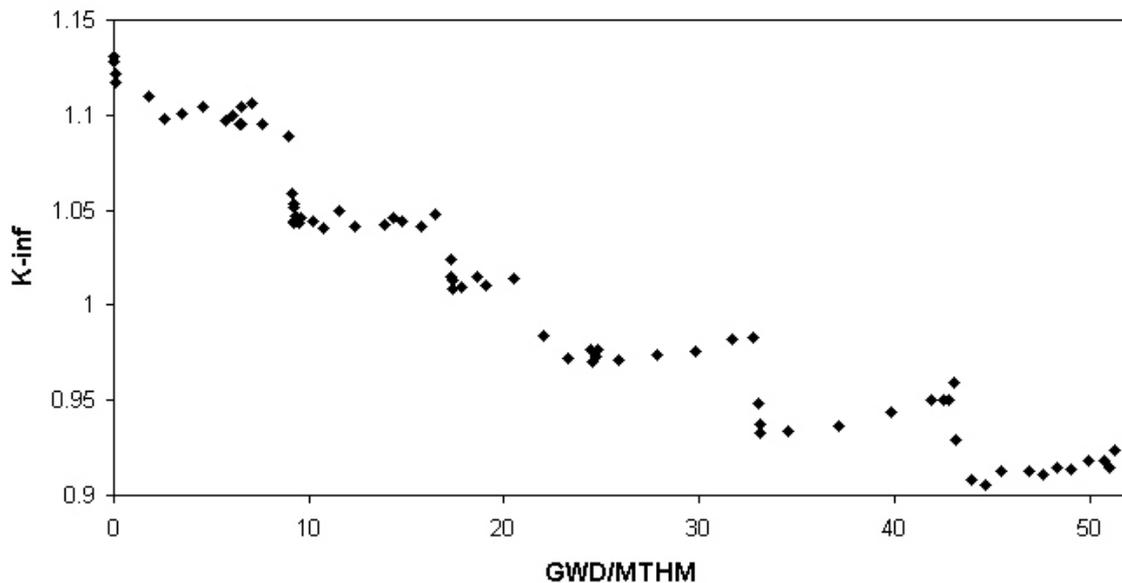


Figure 7-1. Variation of k_∞ with fuel assembly burnup

Although the MCNPX fluxes are adjusted in *Monteburns* through division by the eigenvalue at each burnup step, the resulting spectrum is not truly representative of a critical system. The harder spectrum experienced during the first half of the simulation ($k_\infty > 1$) overestimates the number of fast neutrons relative to a critical configuration, resulting in an overproduction of ^{239}Pu because of resonance captures in ^{238}U . The opposite occurs when $k_\infty < 1$.

There are several other potential spectrum effects:

- a) The accuracy of the calculated ^{239}Pu concentration was improved by subdividing the segmented fuel rod radially, but other isotope concentrations proved to be worse. This could have been caused by the coarser representation of the adjacent fuel rods, which would have affected the spectrum in the assembly.
- b) The use of reflective boundary conditions fails to account fully for the effect of the surroundings on the fuel assembly of interest.
- c) The discrete removal option used for decreasing the boron concentration in the coolant in this *Monteburns* simulation could have hardened the spectrum by ignoring the actual continuous boron depletion during the longer burnup steps. A continuous removal of boron may prove more accurate.

The larger discrepancies observed in some of the minor actinides (^{241}Am and ^{243}Cm) are possibly related to inaccuracies in the ENDF/B-VI nuclear data evaluation, which is known less well for elements above plutonium. However, it is interesting to note that the concentrations of $^{242\text{m}}\text{Am}$ and ^{244}Cm were predicted accurately. The poor prediction for some fission products may also be related to the known approximate treatment of resonance parameters for these isotopes in many nuclear data evaluations. This approximation suffices for most reactor calculations, because the relatively few neutron absorptions in these nuclides have little effect on global parameters such as reactivity and power distributions [15].

The composition of segment BM5 was calculated with ORIGEN2.1 by decaying the irradiated fuel assembly repeatedly from its discharge time to the individual measurement times of specific isotopes. This calculational method did not reproduce precisely the experimental procedure in which each measured isotope, and hence its subsequent decay products, was removed completely from the sample solution.

The above discussion focused on possible sources of error in the *Monteburns* calculation. However, considerable uncertainty is present in the measurements themselves, which is separate from the precision of the radiochemical analysis represented by the experimental 2σ errors in Table 6-1. This is evidenced by differences in some isotope concentrations measured at PSI and SCK·CEN using different methods: ^{155}Gd , 10%; ^{103}Rh , 42%; ^{109}Ag , 51%; and ^{129}I , 154%.

8. CONCLUSIONS

Monteburns was previously validated for low-burnup (~20 GWD/MTHM) MOX fuel using measurements carried out on fuel rods from the San Onofre PWR [16]. The present analysis contributes to the extension of this validation to higher burnup (~55 GWD/MTHM) and relies on a more accurate and extensive experimental database. The results generally agree with the measured isotopic inventory within 5-10%. A problem observed in both benchmarks concerns the excessive depletion of ^{238}U and the resulting

production of ^{239}Pu , which can be attributed to statistical variations in the cross section calculations and spectrum effects. Further work is planned to compare the Monte Carlo predictions with deterministic codes, and to investigate the sensitivity of spent MOX fuel isotopic inventories to (a) different cross-section libraries, (b) reactor parameters such as temperature, boron concentration and coolant density, (c) the selection of burnup regions within the fuel assembly and (d) the core environment. In conclusion, *Monteburns* is a modern technique that can be used for accurate three-dimensional depletion analyses of high-burnup MOX fuel.

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