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LITERATURE REVIEW OF UNITED STATES UTILITIES COMPUTER CODES FOR CALCULATING ACTINIDE ISOTOPE CONTENT IN IRRADIATED FUEL

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Abstract

The accuracy and precision of methods used by electric utilities to determine the actinide isotopic and element content of irradiated fuel are reviewed. The available experimental data were also used to assess the accuracy of the calculational methods. Most of the actinide isotopic content were estimated to within 5% of benchmark values, with the exception of the isotopes Pu-238, Pu-242 and Np-237. The large relative errors in Pu-238 and Pu-242 do not cause a large error in total Pu mass since their fractions in spent fuel are small.

Summary

This paper reviews the accuracy and precision of methods used by United States electric utilities to determine the actinide isotopic and element content of irradiated fuel. After an extensive literature search, three key code suites were selected for review. Two suites of computer codes, CASMO and ARMP, are used for reactor physics calculations; the ORIGEN code is used for spent fuel calculations. They are also the most widely used codes in the nuclear industry throughout the world. Although none of these codes calculate actinide isotopics as their primary variables intended for safeguards applications, accurate calculation of actinide isotopic content is necessary to fulfill their function.

These codes have been benchmarked against experimentally determined values for isotopic content of irradiated fuel and to various standard problems. The data base includes fuel rods from Vermont Yankee, Zion, H.B. Robinson, and the SAXTON nuclear plants. Comparisons have also been made to standard problems developed at the Brookhaven National Laboratory. The codes calculated the majority of the actinide isotopic content to within 5% of the benchmark values, with the exception of the isotopes Pu-238, Pu-242, and Np-237. The large relative errors in Pu-238 and Pu-242 do not cause a large error in total Pu mass since their fractions are small in power reactor spent fuel.

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1.0 Introduction

This paper reviews the accuracy and precision of methods used by United States electric utilities to determine the actinide isotopic and element content of irradiated fuel. The calculation of the actinide content of irradiated fuel requires the use of scphisticated computer codes. United States utilities have a wide variety of computer codes that do calculate actinide content. The codes used fall into two categories, depending on their primary function: 1) Reactor physics codes, whose primary purpose is to calculate neutronic parameters such as k_{eff} and Doppler coefficients, reactor power distribution, and other parameters for safe operation required by the US NRC (United State Nuclear Regulatory Commission) approved technical specifications. Accurate reactor physics information is also necessary for the optimization of fuel management through the fuel cycle. 2) Spent fuel codes, whose primary goal is to calculate such characteristics of irradiated fuel as thermal load from decay heat and radiation fields for storage and transportation safety considerations. Although neither type has as its primary calculational parameter actinide content, they both require accurate calculation of the actinide content to fulfill their functions. Therefore, as part of their validation procedure, the code predicted actinide content of irradiated fuel has been compared to the actinide content of bundles determined experimentally, usually by chemical analysis of dissolved fuel samples. The standard test method, ASTM E321-79⁽¹⁾, uses the stable fission product Nd-148 to determine burnup in irradiated uranium fuels having an initial plutonium content ranging from 0 to 50%.

Reactor physics codes are usually used to predict, before irradiation, the core neutronics parameters, local power distribution, average power level, and fuel burnup of a particular core fuel load over its expected lifetime of operation. The core neutronic parameters calculated include eigenvalues, Doppler worth, moderator worths, and control rod worths. Local power distributions and average power levels are important parameters in the technical specifications that govern the safe operation of the plant. Maximizing the fuel burnup is of extreme economic importance to the operating utility.

These parameters require an extremely accurate calculation of the neutron flux at many different spatial locations. The codes, therefore, usually perform multi-dimensional neutron physics calculations using many neutron energy groups. Sophisticated cross-section libraries are required for these calculations. The cross-sections are highly energy dependent (frequently as many as 50 groups are used), and burnup dependent. Other important neutronic parameters, such as the resonance escape probability, are computed at each stage instead of a constant value being used.

These calculations usually require the use of a suite of codes, of which the depletion code, which calculates the isotopic content, is one part. Figure 1.1 shows the process stream for one such suite of codes, MICBURN-3/CASMO-3/TABLES-3/SIMULATE-3.⁽²⁾ In this case, the calculational stream is fairly straightforward, with the MICBURN-3 code performing calculations on fuel pins containing the burnable poison gadolinium. The

Figure 1.1[•]

Process Stream of Physics Codes

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[•]Reference 7

Code Names



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Function

Provide effective cross socions for a pin as gadolinium burns out

Provide assembly homogenized two-group cross sections, kinetics data and local peaking factors. Also, flux discontinuity data, reflector cross sections and gamma detector responses.

Process cross sections, etc. into functionalized tables

Provide 3-D power, void, exposure and fission product distributions for static reactor conditions.

CASMO-3 code provides the remaining cross-sections and kinetics data necessary for the three-dimensional neutronics calculations done in SIMULATE-3. The depletion calculation is actually done in CASMO-3, although it is not shown on this chart.

However, the calculational stream is not so simple in many suites of reactor physics codes. The actual physical problem being solved is extremely complex, with much feedback between the modules. Figure 1.2 shows the calculational scheme for the EPRI (Electric Power Research Institute) suite of reactor physics codes, ARMP (Advanced Recycle Methodology Program)⁽³⁾. The ARMP suite of codes is one of the most widely used core neutronic packages in the United States and the world. As can be seen, there are many feedback loops in this arrangement, and the depletion calculations are done at several stages.

Spent fuel codes are usually run on a stand-alone basis, although they too can be part of a suite of codes. The key parameters in this calculation are usually thermal loads, photon emissions, and neutron emissions. After 3 cycles of residence in a reactor, most fuel assemblies experienced similar integrated exposure conditions. Thus, to determine the parameters of spent fuel safety, a detailed calculation of the neutron flux is not required. Because of this, the neutronics calculations are greatly simplified. Usually, the neutron flux is considered to be point-wise constant, i.e., there are no spatial changes in the flux. Since these codes are usually run after the fuel has been irradiated, and the power history and exposure known, in many cases the neutron flux is usually approximated in terms of the known average power history that the fuel was exposed to. The depletion calculation itself is greatly simplified, with the cross sections usually (with the exception of the actinides) not burnup dependent. ORIGEN2⁽⁴⁾ is probably the most widely used of the spent fuel codes.

It was decided to limit the discussion of United States utilities methods to determine actinide isotopic content to a few key computer codes, rather than conduct a full assessment of all such codes. Such a wide ranging assessment would be beyond the resources allotted to this project. In researching the reactor physics methods used by U.S. utilities, literature searches were conducted of several information data bases. A large number of reactor physics codes used by utilities or vendors were identified. Many of these codes are proprietary in nature and their documentation, therefore, extremely limited. It was decided to limit the discussion of reactor physics codes to two prototypic and widely used suite of codes, the CASMO/MICBURN/SIMULATE package and the ARMP package. CASMO is typical of the codes designed to use so-called nodal methods, which came into wide use in the 1980s. EPRI-CELL, which does the pin depletion calculation in the ARMP package, was designed to be used with PDQ-7⁽⁵⁾, a finite difference method-based code developed in the 1970s, that is still widely used. The literature searches of spent fuel codes were dominated by references to the ORIGEN family of codes. This code is the most widely used in the field. This was verified by private discussions with members of the Waste Management Division of the Department of Nuclear Energy of the Brookhaven National Laboratory.

- 3 -

Figure 1.2^{*}



Process Stream of ARMP PWR Physics Codes

- 4 -

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Section 2.0 of this paper contains summary descriptions of the three selected prototypic codes, CASMO, ORIGEN, and EPRI-CELL. Section 3.0 opens with a summary of a literature survey of the available experimental data base for validating depletion codes prepared by the Oak Ridge National Laboratory⁽⁶⁾. This is followed by descriptions of the comparisons made to experimental data of the isotopic content predictions of the three codes. Section 4.0 presents the results of two inter-code comparison studies for reactor physics codes. The section summarizes the isotopic content calculations by two utilities for a standard reactor physics problem developed at the Brookhaven National Laboratory⁽⁷⁾. A summary of the accuracy of the isotopic content calculations is then presented, along with recommendations for further research.

2.0 Summary Description of Computer Codes

2.1 CASMO-3G

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CASMO-3G⁽⁸⁾ is a two-dimensional, multi-group transport theory code for the calculation of eigenvalue, spatial reaction rate distributions, nuclide depletion of pin cells, and depletion of BWR and PWR fuel lattices. CASMO-3G is part of a larger code package⁽²⁾ that is used for reload design, steady state licensing, and plant support applications. The code is an improved version of the CASMO and CASMO-2 codes^(9,10). It can model cruciform and cluster control rods, water gaps, in-core instrumentation channels, burnable absorber rods, and fuel rods. The code can generate parameters, e.g., transport theory corrected cross sections, that are used in neutronic codes such as PDQ⁽⁵⁾ and SIMULATE-3⁽²⁾. The nuclear data library is based on ENDF/B-IV with some fission spectra data taken from ENDF/B-V⁽¹¹⁾. The library contains 40 or 70 energy group cross-sections.

CASMO-3G is divided into seven functional parts:

- 1- Nuclear data library and resonance calculation,
- 2- Calculations on unit cells to determine spatial transport and energy spectrum of neutrons in individual pin cells),
- 3- Calculation of strong absorbers (control rods, Gadolinia in fuel pins, B₄C burnable poison),
- 4- Calculation of the 2-D neutron flux within the bundle, the local bundle power distribution, and the total bundle reactivity,
- 5- Depletion calculation (i.e., isotopic content as a function of burnup),
- 6- A diffusion theory calculation for the generation of few-group diffusion theory constants for use in other reactor kinetics codes,
- 7- Gamma transport module, to accurately calculate the energy deposited by gamma rays.

A summary description of the depletion calculation from Reference 6 is given below. Descriptions of the other modules are contained in Reference 6.

In the depletion calculation, the basic burnup chains, with the isotopes linked through absorption and decay, are linearized and the differential burnup equations are solved by a fast analytical treatment. The individually treated fission products account for about 90

- 6 -

percent of the total fission product absorption. Boron and gadolinium contained in absorber rods are also depleted.

The depletion equations are solved separately for each fuel pin and each burnable absorber pin. The flux level is determined by the average power density. This level is held constant during a depletion step. The depletion calculation for each depletion step is carried out into two partial steps. First, a "predictor" step is made using previous timestep fluxes. The predictor step provides number densities at the advanced time, which are used to calculate fluxes used in a "corrector" step. The final number densities are then given by the average value of the results from the predictor and corrector steps. The size of the timesteps used in the depletion calculation is set in the program using default values, however, the user is allowed to input his own values.

This computer code has been validated against experimental data and calculations from other computer codes. The comparisons of the isotopic data are discussed below in Section 3.3. The code was also compared to experimentally determined eigenvalues, gamma scans of irradiated Quad Cities fuel bundles, and other critical experiments. The code validation was recently reviewed by the Nuclear Regulatory Commission and found acceptable for licensing purposes.

2.2 ORIGEN2

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ORIGEN2⁽⁴⁾ is the latest version of a family of computer codes used for calculating the nuclear composition and characteristics of spent fuel. The input data bases include reactor dependent cross sections, decay data, fission product yields, and neutron production rates. The current version of ORIGEN2 contains cross-section libraries for seven types of reactors, including two for PWRs and two for BWRs. The reactor type, initial material composition, irradiation history, and decay history are user input.

The ORIGEN irradiation calculation is usually done using many timesteps until the specified burnup is reached. The neutronics are based on simplified point reactor models, with an average flux calculated at each timestep based on the input power level. The point model can be considered as a neutronic calculation model collapsed (in space and energy) from the detailed reactor physics model mentioned in Sections 2.1 and 2.3. To further simplify the neutronics calculation, only the actinide nuclides have burnup dependent cross sections. A series of post-irradiation decay steps may then also be performed.

A variation on the ORIGEN code, OREST⁽¹²⁾, was developed at Gesellschaft fur Reaktorsicherheit (GRS) in Germany. This paper was limited to codes used by United States utilities. However, OREST provides an opportunity to look at the modification and use of United States developed code by a foreign user. In this variation, ORIGEN was combined with the neutron spectrum code, HAMMER, in order to overcome the limitations of the ORIGEN libraries at high burnups or with mixed oxide fuels. HAMMER is used to determine the neutron spectrum, with burnup dependent cross sections, in the actual fuel mixture at the beginning of and during burnup. HAMMER also carries out the resonance treatment for the most important uranium and transuranium isotopes.

The OREST code was benchmarked by comparisons to experimental data obtained from the Obrigheim reactor and by an intercode comparison to another ORIGEN variant, KORIGEN. The code was found to predict isotopic content within measurement uncertainties and significantly better than KORIGEN. It is not known how widely used the OREST code is.

2.3 ARMP/EPRI-CELL

EPRI-CELL⁽³⁾ is a pin depletion code. EPRI-CELL is part of the ARMP suite of codes developed by EPRI for use by utilities on various core analyses problems. The ARMP suite of codes is arguably the most widely used core physics code package by United States utilities. EPRI-CELL consists of three main subcodes: GAM, THERMOS, and CINDER. GAM performs the "fast" range neutron calculations; THERMOS the thermal range neutron calculations. THERMOS is spatially dependent, and up to 50 neutron energy groups can be modeled. The CINDER module performs the actual depletion calculation. The depletion is done at each THERMOS spatial point (up to 30 spatial points). The CINDER module contains 90 linear chains: 21 of these chains involving 116 nuclides account for the fuel, transuranic isotopes and burnable poisons which may be present in the calculation; the remaining chains determine the formation of 367 fission product nuclides.

Another code module in ARMP, EPRI-CPM, also performs depletion calculations. EPRI-CPM is a multi-group, two-dimensional collision probability code. It is used primarily to verify PDQ assembly analyses. EPRI-CPM isotopic calculations have been compared to experimental data.

3.0 Comparisons of Code Predicted Isotopic Contents to Experiments

3.1 Light Water Reactor Experimental Data Base

A literature survey of the experimental data available for determining the radiological characteristics of spent fuel was conducted at the Oak Ridge National Laboratory⁽⁶⁾. The authors state:

"most of the data available on the composition of (irradiated) reactor fuel were obtained either in fuel reprocessing studies or in destructive testing of fuel by vendors."

The authors of Reference 5 feel that the existing data base is inadequate for the validation of computer codes because of insufficient characterization of initial fuel parameters and irradiation conditions. The report lists results from 16 reactors; a total of 282 analyses of fuel samples giving experimental results for actinides. The authors stated that the H.B. Robinson data had undergone proper quality assurance. No statement was given on the level of quality assurance of the other data. The Zion data which was used to validate the CASMO code was not listed in the report. The German data from Obrigheim reactor used to validate OREST also was not listed.

3.2 Reactor Power History and Burnup

3.3 CASMO-3G Comparisons

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Comparisons were made to Vermont Yankee and Zion isotopic data as part of the validation of the CASMO-3G code⁽⁸⁾. The isotopic composition comparison was made to provide integral verification of the CASMO-3G methodology, with the depletion calculation being one part.

The Vermont Yankee plant is a boiling water reactor. The core consists on rectangular fuel bundles separated by water channels containing cruciform control rods. This arrangement affects the neutron flux locally. Therefore, the comparison of the isotopics was done using three different neutron flux spectra: perturbed, intermediate, and asymptotic. The perturbed neutron spectrum occurs in the vicinity of the water slots which surround the cruciform control rod positions and near the core reflector. The asymptotic neutron spectrum is found in those regions of the core which are well away from and unaffected by the water gaps and control assemblies. The intermediate spectrum is in those fuel regions between the perturbed and asymptotic regions. The isotopics were measured over a broad range of burnup from approximately 1,200 to 31,000 Mwd/Mtu.

The CASMO-3G calculation modeled an entire Yankee Rowe Core I assembly and zircaloy follower region. Several graphs comparing CASMO-3G calculations for various isotopes to actual plant data⁽¹³⁾ were made (Figure 3.1-3.8). The resolution of the graphs

- 9 -





*Reference 7





U-236 Atom Percent versus Burnup for Yankee Core I as Calculated by the Casmo Code

Figure 3.2[•]



U-238 Atom Percent versus Burnup for Yankee Core I as Calculated by the Casmo Code



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Pu-239/U-238 Atom Ratio versus Burnup for Yankee Core I as Calculated by the Casmo Code

Figure 3.4^{*}



Pu-239 Atom Percent versus Burnup for Yankee Core I as Calculated by the Casmo Code





Figure 3.6[•]

Reference 7



- 15 -

Reference 7

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Figure 3.8	
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is not fine enough to make a quantitative evaluation of the results, but the CASMO-3G results have a very good qualitative agreement with the Vermont Yankee Data for regions with various neutron spectra in a BWR.

CASMO-3G calculations were also compared to data obtained from the Zion Hot Cell Examination Study^(14,15). The Zion plant is a Westinghouse PWR using a 15 x 15 assembly. The data were for five different pin locations, located in two different assemblies. The pin locations represent locations in an assembly removed from a water hole (Rod 624), adjacent to a water hole (Rods 642, 616, and 614), and diagonally adjacent to a water hole (Rod 699). Therefore, these pins were exposed to different neutron spectra. The burnups ranged for 23,471 MWd/Mtu for Rod 616 to 51,754 MWd/Mtu for 614 and represent insertion in the core from one to four cycles.

The calculated versus measured results are given Table 3.1. The RMS difference for all the uranium atom ratios is less than 0.04 percent. The root mean square of the difference between the predicted and measured plutonium content for all these rods is less than 1%.

3.4 ORIGEN Comparisons

The material in this section is taken from Section 3 of Reference 6. As part of that report, the authors compiled Table 3.2 which lists the comparisons made between ORIGEN predictions and data from H.B. Robinson. They then used this data to prepare Table 3.3 which lists the standard deviations of the ratio of the predicted to measured values for several isotopes. These results were summarized Table 3.4. The plutonium actinides, with the exception of Pu-238 and Pu-242 isotopes, were all predicted within 5%. It is claimed that the predicted values are within the standard deviations of the experimental data. The authors stress, however, that because many versions of ORIGEN were used in compiling these tables that the results do not provide a full validation of the code. The authors did prepare a similar table based on only one study using a current version of ORIGEN. These results are presented in Table 3.5. Although limited in scope, the results are consistent with earlier work, with even better agreement between experiment and predictions. With Tables 3.3 and 3.5, it can be inferred that the total plutonium mass (n be predicted to within 3%) with a standard deviation less than 5%. It is also noted here that although the errors for Pu-238 and Pu-242 are large, they do not contribute much to the error for the total Pu mass since their fractions are small for spent fuel.

The Materials Characterization Center at Pacific National Laboratory is responsible for providing characterized spent fuel, call approved testing materials (ATM), for use in nuclear waster disposal studies. As part of this program, radiochemically measured values of certain isotopes, including actinides, were compared to results obtained using the ORIGEN? code⁽¹⁶⁾. The two test bundles were from the Calvert Cliffs plant, a PWR. The calculations

Table 3.1^{*}

Zion Isotopics as Calculated by the CASMO Code

*Reference 7

ZION ISOTOPICS - CASHO-3G CALCULATED VS. MEASURED

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U-238 (: MEAS	4.917 5.081 5.149 5.149 4.883		- PU-242 MEAS	6.620 6.272 8.272 10.149 11.313		CH244/PU MEAS	
PU-239/ CASMO	4.881 5.104 5.211 4.974	, ;;	CASHO 7 R.61	6.677 7.953 9.312 10.973		CASHO	.065-03 .485-03 .625-02 .715-02
DIFF	.073 .066 .031 .038 .055		DIFF	.375 .375 .790 .794 .511		IFF	76-04 1 58-04 9 76-04 1 36-04 1 76-04 2 76-04 2
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U-236 - MEAS	.370 .464 .490 .503		PU-240 MEAS	11.348 15.655 15.622 17.430 17.333		DIFF 0	56-03 1. 86-03 3. 56-02 4. 56-02 5. 86-02 5.
CASMO	. 375 . 470 . 487 . 499 . 504		CASMO	21.752 25.663 26.261 26.567 226.567 27.262 27.262 27.262		J-239	03 2.6 02 1.7 1.1 1.1 1.1
DIFF	079 070 026 038 038		DIFF	627 482 442 442 1.340 039		M-243/PU MEAS	3.335-(1.905-(2.395-(4.415-(
U-235 - MEAS	1.386 .721 .536 .370 .308		PU-239 MEAS	63.450 51.849 48.316 44.986 44.986 44.986 43.854		CA5:40	- 985-03 - 885-02 - 145-02 - 815-02
CASMO	1.307 .651 .510 .383 .260		CASMO	62.823 51.367 48.758 46.326 43.815		DIFF	86-03 5 46-03 5 36-03 4 36-03 4 56-03 5 96-03 5
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	ROD 616 642 6642 6642 614 8624 8614	- 19 -	ROD	8 8 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9	-) COR	

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Table 3.2*

Comparison of H.B. Robinson Fuel Composition Data with ORIGEN Predictions

*Reference 5

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lattops	GOODE 19806	GOODE 1978	CAMPBELL 19766	CAMPBELL	MCELROY	CAMPBELL 19766	GOODE 1980b (Oconse-)	CA	MPBELL 1977c (H. B. Robassa	CADELLI (964"	CAMPBELL 19776	CAMPBELL 1978c	NEELY 1962" (Turkey Paul-3) (H	WICHNER 1983"
Total Ps Ps/U ratio		0.94			······································			0.84	· · · · ·	0.99, 0.81	1.07	0.88, 0.93,		
ະມາ ມາບ	1.08*			1.08*	10.3. 14.9*		1.00° 0.92"	1 10"				0.92, 0.92*		
^{ມາ} ປ	1.03*			1.06*	0.98, 1.02, 0.99, 1.06,		0.94*	1.14*		0.97, 0.86*		1.07, 1.07* 1.02, 1.02, 1.16, 1.01'	1.00*	
U ^{mt}	1.01*			0. 99 *	0.96, 0.97* 1.09, 1.09, 1.05, 1.03,		1.09*	0.83*		1.08, 0.95*		0.96, 0.96, 0.95, 0.99*	0 %*	
un One	1.00*			1.00°	1.08, 1.10° 1.01, 1.01, 1.01, 1.00,		1.00*	1 00*		1.00, 1.00°		1.00, 1.00, 1.00, 1.00*	1.00*	
Total II					1.01, 1.00*					100 101	0.97			
internet and the second	1.27*			1.14, 1.33, 1.35, 1.18*	1.04, 1.15, 1.07, 1.17,	1.13*	1. 98*	0.78*		1.01, 1.53*	0.77	1.23, 1 23, 1.13, 1.20°	1.0 2 °	
19 9 1	0.96*		,	0.98*	0.90, 1.00, 0.94, 1.00, 0.94, 0.94	0.92*	1.01*	1.02"		1.03, 0.84		0.97, 0.96, 1.01, 0.97*	0.99*	
	1. 09*			1.04*	0.96, 1.01, 1.00, 1.02, 1.01, 0.97*	1.03*	1.12*	0.91*		0.87, 1.11*		1.08, 1.08, 1.02, 1.08*	1.09*	
Distanting of the				0.95, 1.16,										
²⁴¹ Pu	0.94*			1.91 0.96°	0.91, 1.00, 0.96, 1.03,	1.21*	0.77*	1.04*		1.05, 1.14ª		0.94, 0.95, 0.93, 0.95*	0.88*	
HIPs .	1.07*			1.02*	1.07, 1.08	1.53*	0.92*	0.95*		1.12, 1.73°		1.07, 1.09,	0.96"	
³⁴¹ Aan		0.62		0.60, 0.27,	0.87, 1.00					1.03, 1.03				1.05
				1540° 1.10°										0.99
MCs		1.15		1.56, 3.28*										0.95
^{III} Cm		Q.4C		0.42. 0.44, 0.47, 0.929*						1.08, 1.58				0.99
^{MC} ^{MC}				3.89" 2.98" 2.0"										1.25
HCm				5.0"										Poor statistics
, H L	0.58 0.84	1.02 0.43	0.82, 0.99				0.84 1.74	0.49	0.62	1.12, 1.12	0.82, 0.84, 0.41, 0.80, 0.84, 0.76			
* C	1.09	1.04												
™Kr ₩¢.	1.29	1.34		0.81. 1.04	0.96. 0.95		1.39						*	
Te		0.52			0.80, 0.85, 0.93, 0.90, 0.95, 0.92						,			
**Ru	0.77	0.75		1.15			0.99							
¹²⁵ 8	0.91	0.50		0,58, 0.61.			1.34				0.44			
		2.20		0.63										
""Cs	0.91	0.92		0.85, 0.91, 0.95	1.01, 1.03, 1.02, 1.00, 1.04, 1.04		1.26				J.81			
""Cs	0.81	0.8 i		0.93, 0.97, 0.98			1.29				0.64			
¹³⁴ Es: ²³⁷ Np				0.29	0.92, 0.95, 1.02, 1.03	•					0.28			
"Se ""Se					0.13, 0.13 0.23, 0.23				_					

"By mass spectrometry, "Apparest error in ³¹"U assumed in feed uranium. Used ORIGEN2 code. "Receivables of the remains reported in CAMPBELL 1976c using ORIGEN2 code.

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Table 3.3*

ORIGEN Average Measured/Predicted Ratios

Reference 5

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Isotope	Low/High	No.ª	Average	SD*	Isotope	Low/High	No.ª	Average	SD°
234UC	0.92/1.10	. 8	1.10	0.10	235U	0.86/1.16	17	1.01	0.07
236U	0.95/1.09	17	1.01	0.07	²³⁷ Np	0.92/1.03	4	0.98	0.05
238Pu	0.78/1.27	21	1.15	0.15	²³⁹ Pu	0.84/1.03	18	0.97	0.05
240pu	0.87/1.12	18	1.03	0.07	²⁴¹ Pu	0.77/1.21	18	0.97	0.10
242pu	0.94/1.73	14	1.12	0.22	²⁴¹ Am	0.27/0.946	9	0.77 ⁴	0.25
242 Am		1	0.99		²⁴³ Am	,	t	0.92	
²⁴² Cm	1.08/3.28	7	1.694	0.68	²⁴³ Cm		1	0.95	
244Cm	0.40/1.12	7	0.70^{d}	0.31	245Cm		1	1.25	
246Cm	••••	1	1.04		247Cm		1	1.00	
248Cm		1	0.954		Ъ	0.58/1.02	3	0-81	0.13
1291	0.43/1.74	11	0.80	0.32	14C	1.04/1.09	2	1.06	
85Kr	1.29/1.39	3.	1.34	0.04	⁷⁹ Se	0.13/0.13	2	0.13	
⁹⁰ Sr	0.81/1.04	4	0.94	0.08	99Tc	0.52/0.95	7	0.84	0.14
106 Ru	0.75/1.15	4	0.92	0.17	125Sb	0.50/0.91	3	0.67	0.18
125Sn	0.23/0.23	2	0.23		134Cs	0.44/1.34	7	0.67	0.28
137Cs	0.81/1.04	13	0.98	0.11	1 ⁴⁴ Ce	0.64/1.29	7	0.92	0.19
154Eu	0.28/0.29	2	0.28				·		

Average measured/predicted ratios

"Number of determinations.

 $^{b}SD =$ standard deviation.

Two very high ratio (McELROY 1985) were not used.

^dThe latest calculations using ORIGEN2 (see Table A.4) gave ratios of 1.05, 1.12, and 0.99 for ²⁴¹Am, ²⁴²Cm, and ²⁴⁴Cm, respectively. The ²⁴⁸Cm single data point had poor statistics.

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Relative Agreement of ORIGEN Predictions for Various Isotopes

Reference 5

Excellent Fair (within 10%) (within 50%) 234U 242Cm 240 2440	Poor (greater than 50%) ⁷⁹ Se ¹²⁶ Sn
²³⁴ U ²⁴² Cm	⁷⁹ Se ¹²⁶ Sn
$\begin{array}{cccc} 235 U & 247 Cm^2 \\ 236 U & 3 H \\ 237 Np & 129 I \\ 239 Pu & 99 Tc \\ 240 Pu & 125 Sb \\ 241 Pu & 134 Cs \\ 241 Am & 144 Ce \\ 242 Am^3 & 238 Pu \\ 243 Am^4 & 242 Pu \\ 244 Cm^4 & 85 Kr \\ 246 Cm^4 \\ 247 Cm^4 \\ 14 C \\ 90 Sr \\ 106 Ru \\ 112 C \\ \end{array}$	154 Eu

Relative agreement of ORIGEN predictions for various isotopes

^aOnly one experimental determination.

Table 3.5[•]

Comparison of Measured and ORIGEN Predicted Values for Irradiated H.B. Robinson Fuel

[•]Reference 5

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lsotope	Average ratio (experiment/ORIGEN2)	Standurd deviation
234U	12.6	Two determinations
235U	0.98	0.03
236U	1.07	0.02
238U	1.007	0.005
²³⁸ Pu	1.09	0.05
²³⁹ Pu	0.95	0.04
²⁴⁰ Pu	1.00	0.02
²⁴¹ Pu	0.96	0.05
²⁴² Pu	1.08	Two determinations
²³⁷ Np	0.98	0.05
241 Am	0.94	Two determinations
¹⁹ Se	0.13	Two determinations
⁹⁰ Sr	0.96	Two determinations
⁹⁹ Tc	0.89	0.05
¹²⁶ Sn	0.23	Two determinations
¹³⁷ Cs	1.02	0.0!

Comparison of measured and predicted values for irradiated fuel composition⁴

^aH. B. Robinson fuel assembly BO-5; initial loading, March 1971: discharged, May 1979; average burnup estimated as 28,050 MWd/MTU. (Source: J. O. Barner, *Characterization of LWR Spent Fuel MCC-Approved Testing Material--ATM-101*, Pacific Northwest Laboratory, PNL-5109, Rev. 1, June 1985.) were run using the ORIGEN2 PWR library, which is based on a fuel with 3.2% U-235 and a burnup of 33 MWd/kg M. The two ATMs tested had enrichments of 2.45% and 2.72% U-235 with burnups ranging from 18.7 MWd/kg M to 46.5 MWd/kg M. The results for certain actinide isotopes are shown in Table 3.6. The lower burnup fuel used in ATM-103 has better agreement between measurement and calculation, probably because its lower burnup is closer to that used in the ORIGEN2 library. However, even the calculations for the higher burnup ATM-106 is generally within 10% of the measured values.

The OREST code, a combination of HAMMER and ORIGEN, was validated against assemblies obtained from the Obrigheim reactor as part of the Isotopic Correlation Experiment (ICE)⁽¹²⁾. Five assemblies were used with isotopic content determined using radiochemical methods at four different laboratories. The assemblies were processed half at a time, so a total of 40 separate measurements were made. The comparisons for several isotopes are given in Table 3.7. The deviations between experiment and calculation are summarized in Table 3.8, along the measurement uncertainty for the isotope and results from another ORIGEN based code, KORIGEN. As can be seen in Table 3.8, the deviations agree well with measurement uncertainties except for two Pu isotopes. One of these isotopes, Pu-242, was also predicted poorly in the H.B. Robinson comparisons discussed previously.

3.5 EPRI-CELL Validation

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The EPRI-CELL depletion isotopic calculation was validated against data obtained from the Yankee Rowe, Saxton, and Robinson-2 reactors⁽³⁾. Two sets of Yankee Rowe data were used: stainless steel clad pins and zircaloy clad pins. The zircaloy clad pin data was limited. The Saxton and Robinson-2 comparisons were made with an additional code, EPRI-CPM (also part of the ARMP package), being used in conjunction with EPRI-CELL. For the Saxton data, EPRI-CPM was used to provide six of the heavy element cross sections. The Robinson-2 data were for a single pin in an assembly with burnable poison pins. EPRI-CELL is a single fuel pin depletion code that represents a fuel bundle with a single average pin. Therefore it was necessary to calculate correlation factors using EPRI-CPM, which uses a full 2-D representation of a fuel bundle and can therefore account for the presence of burnable poison rods, for use in EPRI-CELL.

The predicted to experiment comparisons were made on the basis of isotopic ratios. Comparisons on this basis reduce the data scatter at high burnups. The scatter in absolute isotopic data increases with burnup. This basis also reduces the sensitivity of the calculation to various input parameters. Therefore, these comparisons, while useful for the code validation, are of limited value for this study. The comparisons do provide insight into the overall accuracy of the code.

		ATM-103ª			ATH-106"	
Nuclida	Ratio at Giv	en Fuel Burnuo,	Ind/ka H	Ratio at G	iven Fuel Burnu	o, i id/ka M
nucrue	33.2	20.0	10./	0.0	37.0	31.0
234 _U	1.02	1.11	1.06	. HA	HA	NA
235 _U	1.06	1.04	1.04	1.37	1.23	1.20
236 _U	0.93	0.93	0.93	0.96	0.95	0.96
238 _U	0.99	0.98	0.99	1.00	0.99	0.99
238 _{2u}	0.98	0.94	0.84	1.10	1.12	1.11
²³⁹ Pu	1.06	1.02	1.02	1.20	1.18	1.18
240 _{Pu}	1.02	1.02	1.03	0.98	1.01	1.02
241 _{Pu}	1.05	1.02	1.02	1.22	1.16	1.14
242 _{Pu}	0.89	0.93	0.93	0.83	0.87	0.89
237 _{Np}	1.14	0.98	1.04	1.41	1.35	1.37
241 _{Am}	1.01	1.02	1.02	0.76	1,02	1.12
$243_{Cm} + 244_{Cm}$	0.99	0.93	0.84	1.02	1.01	1.05
⁷⁹ Se	6.67	6.25	6.25	8.33	7.14	8.33
⁹⁰ sr	0.98	0.98	0.97	1.00	1.00	0.98
99 _{Tc}	1.01	1.01	0.97	1.37	1,39	1.41
126 _{Sn}	4.35	4.17	4.34	5.26	5.55	5.00
129 ₁ °	0.85	0.91	0.89	1.08	HA	1.19
135 _{Cs}	1.02	1.00	0.99	1.08	1.11	1.08
137 _{Cs}	0.97	0.96	0.95	0.94	0.99	0.95

Ratios of Predicted-to-Measured Values of Nuclides in Fuel from ATM-103 and ATM-106 (Refs. 2 and 3)

^aEnrichment was 2.72% ²³⁵U. Examined 6.5 yr after discharge on October 18, 1990. ^bEnrichment was 2.45% ²³⁵U. Examined 6.7 yr after discharge on October 18, 1980. ^cThe ¹²⁹I was measured in other samples with slightly different burnups: 33.0 MWd/kgM, 25.5 MWd/kg M, and 15.6 MWd/kg M in ATM-103, and 46.5 MWd/kg M and 29.5 MWd/kg M in ATM-106. NA = Not available.

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Table 3.7*

Percentage Deviations of OREST Calculations from the Measured Values for Obrigheim Reactor

*Reference 12

Percentage deviations of OREST calculation results from the measured values (calculation-experiment/experiment)*100

		Batch	No.									
nent Is	sotope	э 8б	87	88	68	06	16	55	63	94	95	Mîttel
n tu	235 236	-0.5 8 -4.1	-1.6 8 -4.1	-1.2 % -4.3	-1.2 % -3.6	0.4 8 -4.4	-0.3 8 -3.9	-0.9 8 -4.5	2.4 [.] 8 -2.9	-1.38	-1.3 8-	-0.6 8 -4.1
tonium	238 239	-15.1 -0.2	-22.0 4.3	-12.3 3.9	-7.0 2.7	-10.5 3.4	-11.4 2.6	-25. 6 5.8	-9.2 4.9	-13.5 3.8	-13.4 3.0	-14 3.4
,	240 241 242	7.6 2.4 3.7	12.3 4.2 9.8	12.8 6.7 12.0	11.7 4.4 11.2	10.9 4.1 6.4	11.6 4.6 10.5	13.3 6.2 9.8	13.7 6.3 9.4	11.8 3.4 10.3	10.0 4.3 9.2	11.6 4.7 9.2
ericium	241 * 243 *	-32 200(1)	-46 141(?)	-64 56	- 63 53	-67 44	-65 49	-68 42	-67 62	-68 41	(¿)0/L	-61(?) 86(?)
ium	242 * 244 *	-11	-10 8.9	- 9 3.4	-10 1.8	-11 -3.1	-5 2.8	-16 -1.7	- 16 6.5	-16 0.0	-16 2.1	-12 2.0

* only 1 moasuring point per batch; very uncertain analytical values except for Cm-244

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Table 3.8"

Comparison Between Experiment and OREST Calculations for Obrigheim Reactor

^{*}Reference 12

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Comparison between experiment and calculation as regards nuclide concentrations in fuel assemblies 168 through 176

Isotope	Deviation *) OREST	Deviation *) KORIGEN [12]	Measurement Uncertainty [12]
U- 235	-0.6 %	-1.9 %	±2 %
236	-4.1 8	-1.9 9	±1 %
Pu- 238	-14.0 %	-23.3 %	±15 %
239	3.4 %	0.4 %	±3 %
240	11.6 %	-5.1 %	±3 %
241	4.7 %	-6.0 %	±4 %
242	9.2 %	-9.0 %	±4 %
Cm- 242	-12 %	ca ~30 %	large, up to a factor
244	2 %	ca -23 %	±20 %

*) Deviation in percent equivalent to (calculation-experiment)/experiment*100

The Yankee Rowe comparisons were presented as a set of curves showing the calculation versus experimental data. Figure 3.9 shows results for the isotopic ratio of Pu-239/Pu-240 for the stainless steel clad pins; Figure 3.10 the isotopic ratios for Pu-239/Pu-240, Pu-240/Pu-241, and Pu-241/Pu-242. The graphs do not allow for a quantitative comparison of the results, but the calculations appear to agree well with the experimental data.

The Saxton data was from Core II and consisted of 20 samples with burnups ranging from 7,000 to 22,000 MWD/MTU. The results for one pin are summarized in Table 3.9. The absolute isotopic data in the upper part of the table has been corrected for burnup. That is the comparisons were adjusted back to the end of the irradiation period, hence, the 0% uncertainly in the U-238 and Pu-239 concentrations. The atom ratios in the lower part of the table are uncorrected and correspond to a particular time after shutdown. The results are very good for all isotopes except Pu-238 (which has a large experimental uncertainty), Pu-242 (which is consistently underpredicted by EPRI-CELL), and the Np-237/U-238 ratio.

The Robinson comparison was for a single rod with an average exposure of 18,000 MWD/MTU. The rod was divided into three segments with exposures of 24,570 MWD/MTU, 27,620 MWD/MTU, and 30,920 MWD/MTU. The measurement accuracy was within $\pm 5.1\%$. The middle segment was near a spacer grid, which could not be modeled with EPRI-CELL, so the comparisons were limited to the other two sections. Table 3.10 shows the comparisons for these two exposures for three isotopic ratios. In all cases, the predictions agree well with the experiments.

*Reference 2

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COMPARISON BETWEEN ANALYSIS AND YANKEE STAINLESS ISOTOPIC PATIOS

- 29 -

Figure 3.10"

Plutonium Isotopic Ratios versus U-235 Fractional Depletion for Yankee Core V Zircalloy Assemblies as Calculated by the EPRI-Cell Code

Reference 2.

COMPARISONS BETWEEN ANALYSIS AND YANKEE SIECKARY LESTEPIC PATICS

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Nuclide or Parameter	Experiment	Experimental funcertainty %	Analysis	$\left(\frac{\text{Analysis-Exp}}{\text{Exp}}\right)^{100}$
		Ato	S mo	
U- 234	.00465	28.7	.00448	-3.7
, U−235	.574	.9	.584	. <u>+</u> +1.7 -
U-236	.0355	5.6	.0328	-7.6
U- 238	99.386	0	99.378	0
Pu-238	.109	2.2	.074	-32,1
Pu-239	73.77	0	74.25	+0.6
Pu-240	19.25	. 2	18.53	-3,5
Pu-241	6.29	. 3	6.58	+4.6
Pu-242	.579	.9	.511	-11.7
		Atom Rat	ios	aanta Angara anga ata mga kana kana kana kana na sa
Pu-236/	3.62 ⁻⁹	22	3.66 ⁻⁹	+1.1
Pu-239 Np-237/	1.14-4	15	833-4	-26.0
U-238		10	.033	** * • •
Pu-239/	.04383	.7	.0447	+2.0
Pu-238/	1.75 ⁻³	. 4	1.43 ⁻³	-17.6
Am-241/	.0123	15	1.26 ⁻²	+2.4
Am-243/	<.84 ⁻²		.86 ⁻³	
Pu-239 Cm-242/	1.05 ⁻⁴	10	1.07-4	+0.4
Pu-239 Cm-244/ Pu-239	1,09 ⁻⁴	20	1.056 ⁻⁴	-3.2
Burnup Nd-148 (20,226 MWD/MTU)	3.1	20,226	
Accumula	ted Fissigns/ N	1 ⁴ 0	_ ?	
a) ND-148	2.238 - 2	~ 2.6	2.170	-3.0
Elemen	2.200 .t	4.0	2.170	- b . 4
U-235 Fraction	.2216 al Depletion	3.3	.2062	-6.9

Table 3.9[°] Comparison Between EPRI-CELL and SAXTON Experimental Data 'Reference 3

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Table 3.10^{*}

Comparison of Isotopic Ratios Calculated by EPRI-CELL and Experimental Data for H.B. Robinson

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Reference 3

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Variation in Isotopic Ratios with Exposure

Stated Exposure (NWD/NTU)	Exposure Range due to 5.1% Uncertainty (NMD/MTU)	Isotopic Ratio	Values of EFRI-CELL Ratios over Exposure Range (without Correction Factors)		
			Measurement		
24,750	23,350-25,350	239 94 ⁷ 4/ ²⁴⁰ 74 94	2.55+.07	- 12.271-0.49	
		240 _{24/} 241 94 ^{24/} 94 ²⁴	1.83 <u>+</u> .07	1.31 - 1.33	
		241 94 ⁷ 4/242 94 ⁹ 4 ⁹ 4	3.03 <u>+</u> .27	3.04 - 3.31	
			1.		
30,020	29,340-32,500	239 ₂₄ /240 ₂₄ 94 ² 4/94 ² 4	2.15+.05	1.04 - 2.11	
		240 94 ⁷ 4/241 94 ⁹ 4	1.79+.05	1.73 - 1.75	
		241 _{24/242} 242 94 ² 4/242	2.33+.19	2.34 - 2.63	

4.0 Comparisons to BNL Standard Physics Problem

 The Brookhaven National Laboratory has developed a series of standard problems for the evaluation of reactor physics codes. The problem consists of two fuel assembly lattices are to be evaluated at several statepoint conditions and compared to a BNL computer solution. Since the problem is "artificial", no experimental data exists to compare against. The problem, though, does provide insight into the consistency of the various computer codes used.

Isotopic content results from the two computer codes, CASMO and LEOPARD, are summarized in Table 4.1. The error measurements are only approximate in order to maintain the integrity of the standard problem solution. The comparisons are quite good, with the largest deviation being 10%.

Table 4.1

Comparison Between CASMO and LEOPARD Results for Brookhaven National Laboratory

BNL STANDARD PROBLEM COMPARISONS

	CASMO	LEOPARD
U-235	5*	6
U-238	.1	2
Pu-239	5	1
Pu-240	5	-7
Pu-241	5	-1
Pu-242	3	10

-ERROR - CrBNL - Calc)/ BNL X 100%

=

5.0 Conclusions

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Table 5.1 summarizes key comparisons for the various codes. With the exception of the SAXTON data, the actinide isotopic content is calculated accurately for most isotopes regardless of the code chosen. The results for the CASMO and ORIGEN codes are all within 10% of the expected values; the majority of the isotopes are within 5%. The EPRI-CELL comparison to SAXTON is less satisfactory. In this case, the calculated actinide isotopic content for Pu-238, Pu-242, and Np-237 are larger than 10%. This may be due to the non-prototypic nature of the SAXTON fuel samples. The OREST results illustrate that even a user-modified code can be expected to give reasonably accurate results.

Table 5.1 does not show what the experimental uncertainty is in the measurement for actinide isotopic content. Typically, the measurement error ranges from $\pm 2-10\%$ depending on the isotope and the measurement technique used. Additionally, the table does not show the absolute atomic percentage of each isotope. Isotopes present in small amounts may have a larger measurement error than isotopes present in large quantities.

To illustrate the effect of isotope abundance on the total uncertainty of the calculationmeasurement error, the ORIGEN results for the plutonium isotopes were examined in greater detail. The results are summarized in Table 5.2. In this table, it can be seen that the isotopes with the largest error, Pu-238 and Pu-242, occur in very small quantities. Therefore, their contribution to the overall error is also small. As a result, the overall ORIGEN error for the plutonium isotopes is only 3%, with a standard deviation of only $\pm 3\%$.

The ORIGEN results also show that for significant burnups in fuel bundles actually exposed over their lifetimes in power reactors, no detailed neutron flux calculation, such as used in CASMO or EPRI-CELL, is needed. There are two reasons for this. First, as burnup increases, the actinides very quickly obtain equilibrium or near equilibrium values. Therefore, errors in the neutron flux shape or local power level do not lead to large errors in actinide isotopic content. Second, it is standard utility practice to place the bundles in the core so that a uniform burnup is obtained. Specifically, over the lifetime of the bundle, it is usually placed in two or three different locations in the core so that as uniform as possible a flux profile is obtained. Thus, no one bundle is placed near control rods, or other areas, where its flux profile is severely distorted for its entire lifetime.

The scope of this work was limited to a review of the current literature. This search yielded several reports that provided comparisons between computer codes and experimental data as part of validation studies. None of these studies looked in great detail at the sensitivity of the codes to various input parameters. For example, utility reported assembly burnups, a significant input to the computer codes, are calculated very accurately⁽¹⁷⁾. A

Table 5.1

Accuracy of Calculated Actinide Isotopic Contents

Code	CASMO	CASMO	LEOPARD	ORIGEN	OREST	EPRI- CELL
DATA	ZION	BNL	<u>BNL</u>	H.B Rob- inson	<u>Obergheim</u>	<u>SAXTON</u>
U-235	8	~5	~6	-2	6	1.7
U-236	0.4	x	x	7	-4.1	-7.6
U-238	0.05	~.1	~2	0.7	x	0
Pu-238	5	x	X	9	-14	-32.1
Pu-239	1	~5	~7	-5	3.4	0.6
Pu-240	1	~5	~7	0	11.6	-3.5
Pu-241	3	~5	~-1	-4	4.7	+4.6
PU-242	4	~3	~10	8	9.2	-11.7
NP-237	x	×	×	2	x	-26.9
- Error =	(Meas-Calc)/Meas x 100	% (?)			

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Т	ab	1	е	5	•	2	a
-	~~~~	-	-	•	٠	-	~

	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Isotope ^a Fraction (%)	1	58	25	9	7
Error ^a	1.15	0.97	1.03	0.97	1.12
Relative Error	1.15	56.26	25.75	8.73	7.84
Standard Deviation ^a	0.15	0.05	0.07	0.10	0.22
Variance	0.03	7.91	3.25	0.76	2.97
Mean Error for all Pu Isotopes	0.9973	With	a Standard I	Deviation of	0.04

^aFrom Table 3.3

Table 5.2b

	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Isotope ^b Fraction (%)	1	58	25	9	7
Error ^b	1.09	0.95	1	0.96	1.08
Relative Error	1.09	55.1	25	8.64	7.56
Standard Deviation ^b	0.05	0.04	0.02	0.05	0.22
Variance	0.003	4.86	0.25	0.19	2.77
Mean Error for all Pu Isotopes	0.97	With	a Standard	Deviation of	0.03

^bFrom Table 3.5, H. B. Robinson Data

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recent paper indicates that discharge burnups are probably within 2% of actual burnups for fuel with three or more cycles of irradiation. The sensitivity of the calculated actinide content to this low burnup uncertainty should be investigated. The sensitivity studies should also be done for such parameters as enrichment uncertainties, location effects, and material compositions.

Additional studies of reprocessed fuel may also be needed in order to establish a consistent and comprehensive data base for experimentally determined actinide isotopic content. Once such a data base is established, an inter-code comparison study using many different computer codes could be done to determine how individual codes perform relative to each other. Such a study would provide additional information to that obtained by having computer codes run the BNL standard problem.

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