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Radiological Characteristics of Light-Water Reactor Spent Fuel: A Literature Survey of Experimental Data

J. W. Roddy
J. C. Mailen

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RADIOLOGICAL CHARACTERISTICS OF LIGHT-WATER REACTOR SPENT FUEL: A LITERATURE SURVEY OF EXPERIMENTAL DATA

J. W. Roddy
J. C. Mailen

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CONTENTS

Page

LIST OF TABLES v

LIST OF FIGURES vii

GLOSSARY ix

ABSTRACT 1

1. INTRODUCTION 2
   - 1.1 SPECIAL DOE/OCRWM DATA NEEDS 2
   - 1.2 COMPUTER CODE SELECTION 2
   - 1.3 DESCRIPTION OF ORIGEN2 3
   - 1.4 OBJECTIVES 3

2. REACTOR DATA 5

3. RADIONUCLIDE CONTENT OF IRRADIATED FUEL 7

4. HEAT GENERATION RATES OF ASSEMBLIES 9

5. COMPARISON OF MEASURED AND PREDICTED RADIATION LEVELS 14
   - 5.1 PREDICTED AND MEASURED GAMMA-RAY COMPARISONS 14
   - 5.2 PREDICTED AND MEASURED NEUTRON DOSE-RATE COMPARISONS 21

6. CONCLUSIONS 24
   - 6.1 RADIONUCLIDE INVENTORY 24
   - 6.2 DECAY HEAT 25
   - 6.3 GAMMA EMISSION RATES 25
   - 6.4 NEUTRON PRODUCTION RATES 26
   - 6.5 SUMMARY 26

7. RECOMMENDATIONS 27

7.1 IMPORTANT VARIABLES AND RANGES 27

7.2 FUEL SELECTION 27

7.3 ANALYTICAL PROCEDURES AND ACCURACIES 27

7.4 SAMPLE SELECTION 28

7.5 CALCULATIONAL SUPPORT 29

7.6 QUALITY ASSURANCE PROGRAM 30

7.7 ADDITIONAL MEASUREMENTS 31

7.7.1 Decay Heat 31

7.7.2 Photon Emission Rates 31

7.7.3 Neutron Emission Rates 32

7.8 COST-BENEFIT ANALYSIS 32

8. REFERENCES 36

Appendix A. FUEL COMPOSITION DATA 43

LIST OF TABLES

1 Characteristics of assemblies/rods considered during this study 6

2 Comparison of measured and predicted values for irradiated fuel composition 8

3 Comparison of heat measurements with ORIGEN2 calculations 10

4 Input specifications for ORIGEN calculations 16

5 Predicted and measured exposure rates adjacent to the corners of pressurized-water reactor fuel bundles 16

6 Predicted and measured exposure rates along the sides and above pressurized-water reactor fuel bundles 17

7 Predicted and measured exposure rates near boiling-water reactor fuel bundles 18

8 Comparisons of predicted gamma dose rates with data for a partially loaded cask 19

9 Comparisons of predicted gamma dose rates with data for a fully loaded cask 20

10 Comparison of predicted neutron dose rates with data for a fully loaded cask 22

11 Accuracy and applicability of current analytical methods 29

12 Summary of total-system life-cycle cost estimates for the authorized repository system 34

13 Summary of total-system life-cycle cost estimates for the improved-performance system 35

A.1 Analyses of selected isotopes for H. B. Robinson spent fuel assembly BO-5 44

A.2 Radionuclide analyses of spent fuel from various light-water reactors 45

A.3 Currently available isotopic data for reactor fuel 46

A.4 Comparison of H. B. Robinson fuel composition data with ORIGEN predictions 48

A.5 Average measured/predicted ratios 49

A.6 Relative agreement of ORIGEN predictions for various isotopes 49

LIST OF FIGURES

1 Correlation between burnup analyses and ^{137}Cs gamma counting 5

2 Predicted and measured decay heat rates for Dresden, Cooper, and Monticello spent fuel assemblies 12

3 Location of detector tubes 15

4 Composite of ^{137}Cs gamma scans for rod N-9 from assembly BO-5, H. B. Robinson Unit 2 30

GLOSSARY

ATM	Approved Testing Materials
BWR	Boiling-water reactor
DOE	Department of Energy
EDB	Energy Data Base
EIA	Energy Information Administration
EPA	Environmental Protection Agency
EMAD	Engine, Maintenance, and Disassembly Facility
EPA	Environmental Protection Agency
FWMS	Federal Waste Management System
GE-MO	General Electric–Morris Operation
LWR	Light-water reactor
MCC	Materials Characterization Center
MRS	Monitored retrievable storage
MTU	Metric tons uranium
MWd	Megawatt-days
NWPA	National Waste Policy Act
OCRWM	Office of Civilian Radioactive Waste Management
ORNL	Oak Ridge National Laboratory
PNL	Pacific Northwest Laboratory
PWR	Pressurized-water reactor
QA	Quality assurance
RCA	Radiochemical analysis
REA	Ridihalgh, Eggers & Associates
SSMS	Spark-source mass spectrometry
TEIDMS	Thermal emission isotopic dilution mass spectrometry

RADIOLOGICAL CHARACTERISTICS OF LWR SPENT FUEL: A LITERATURE SURVEY OF EXPERIMENTAL DATA

J. W. Roddy
J. C. Mailen

ABSTRACT

This survey brings together the experimentally determined light-water reactor spent fuel data comprising radionuclide composition, decay heat, and photon and neutron generation rates as identified in a literature survey. Many citations compare these data with values calculated using a radionuclide generation and depletion computer code, ORIGEN, and these comparisons have been included. ORIGEN is a widely recognized method for estimating the actinide, fission product, and activation product contents of irradiated reactor fuel, as well as the resulting heat generation and radiation levels. These estimates are used as source terms in safety evaluations of operating reactors, for evaluation of fuel behavior and regulation of the at-reactor storage, for transportation studies, and for evaluation of the ultimate geologic storage of spent fuel.

Although substantial fuel compositional data have been generated, it is difficult to compare these values with results calculated by radionuclide generation and depletion computer codes because many of the necessary parameters are unavailable for the fuel (burnup and power history) and the analyses (accuracy and method). Also, the information was not collected under currently required quality assurance procedures.

Current data on decay heat generation rates are reasonably well characterized. Comparisons of measured values with those predicted by ORIGEN agree to within $\pm 10\%$; however, the experimental data exhibit about the same amount of scatter. Also, the data cover only a limited number of cooling times and burnups.

Both gamma and neutron production rates have been measured, and the values have been compared with ORIGEN predictions. The results obtained by using ORIGEN agree well with measured values over the length of the neutron shield. The gamma dose rate peaks are underpredicted by a factor of 2, and the neutron predictions are as much as three times greater than the actual measurements above and below the neutron shield.

The current data are very limited in scope in that they cover only a narrow range of burnups and decay times, a select number of nuclides, and emphasize pressurized-water reactors. Recommendations are given for an experimental program that would eliminate these gaps in the present knowledge for the radiological parameters considered.

1. INTRODUCTION

Under the auspices of the Department of Energy (DOE), the Office of Civilian Radioactive Waste Management (OCRWM) has been designated to site, design, and construct mined geologic repositories for the disposal of spent nuclear reactor fuel (including UO_2, cladding, and all nonfuel components) and high-level radioactive wastes (NWPA 1983). The physical, chemical, and radiological properties of these radioactive waste materials must be well characterized. Physical characteristics are relatively easy to define, but radiological properties, which continue to change with the composition and the age of the material, are much more difficult to quantify. Various computer codes have been developed to predict the radionuclide content, the associated heat, and the radiation characteristics of spent nuclear fuel as functions of irradiation history, type of reactor, and decay time (the period since fuel was discharged from a reactor). This information is vital to DOE and OCRWM because it serves as input data to many of their designers and licensing applicants in the areas of performance assessment (source terms), heat transfer (thermal generation rates), criticality (radionuclide composition), and shielding (gamma and neutron generation rates). Without accurate predictions from a well-documented and properly quality-assured radionuclide generation and depletion computer code, the licensing of any part of the Federal Waste Management System (FWMS) could experience serious delays.

1.1 SPECIAL DOE AND OCRWM DATA NEEDS

The DOE and OCRWM system (project offices, contractors, national laboratories, etc.) has special needs in the estimation of the radiological characteristics of nuclear fuels. Present requirements (DOE 1986a) state that the fuel accepted by DOE must have a minimum cooling time of 5 years; therefore, only long-lived isotopes will be of concern. In addition, the Environmental Protection Agency (EPA 1985) limits projected releases of radioactivity to the accessible environment for 10,000 years after disposal. Hence, a computer code used by DOE and OCRWM must be able to predict the radionuclide content of any radioactive waste over extended decay periods.

Present economic factors indicate that reactor fuels will be operated to their maximum feasible burnup; thus, in the future the FWMS will receive fuels with burnups (40,000 to 60,000 MWd/MTU) much higher than the ones currently available. A computer code used by the DOE and OCRWM system must be able to treat these higher burnups.

In summary, the principal requirements of a reactor physics code to meet the needs of DOE and OCRWM are that (1) it provide ample information concerning the composition of nuclear materials, (2) it be used to determine the principal characteristics of the nuclear materials, and (3) it be able to predict these parameters for any reasonable decay period.

1.2 COMPUTER CODE SELECTION

Over the years, many computer codes have been developed to estimate the composition of nuclear reactor fuels during irradiation. Several of these codes are complex and use extensive multiple-energy-group neutronics and cross sections; however, in many cases, they are restricted in application because of the limited number of nuclides that are computed. Examples of such codes are the ones developed by the vendors and used in the design, heat transfer analysis, and fuel management modes for their nuclear reactors. Additional problems with these reactor physics codes are that they are difficult to use, expensive to run, and usually proprietary. Very few computer codes

have been developed which meet all the criteria and needs of DOE and OCRWM. The code should (1) supply results in a convenient format, (2) be sufficiently accurate, (3) have adequate quality assurance (QA) for licensing applications, (4) be readily available, (5) be relatively inexpensive to run, (6) have seen extensive use, and (7) be well supported. One radionuclide generation and depletion code that meets all of these requirements and properly addresses DOE and OCRWM's needs is ORIGEN2 (BELL 1973, CROFF 1983).

1.3 DESCRIPTION OF ORIGEN2

ORIGEN2 is a versatile computer code for calculating the nuclear composition and characteristics of spent fuel. The input data bases include reactor-dependent cross sections; decay data (list of nuclides, decay half-lives and the branching fractions, internal transitions, etc.); fission product yields; photon production rates and energy; and neutron production rates and energies. The current version of ORIGEN2 contains cross-section libraries for seven types of reactors. The pertinent ones for DOE and OCRWM are the two pressurized-water reactor (PWR) models (33,000 and 50,000 MWd/MTU) and the two boiling-water reactor (BWR) models (27,500 and 40,000 MWd/MTU). The user must select the type of reactor, initial material composition, irradiation history, and decay history. ORIGEN2 contains three data bases comprising separate categories of nuclides: 130 actinides, 850 fission products, and 720 activation products. Several of the nuclides appear in more than one category. The code can predict the composition (g or g-atom), radioactivity (Ci), and the thermal power (W) for each of the nuclides. In addition, the neutron generation rates through alpha-neutron reactions and spontaneous fission and the photon production rates from gamma rays, X-rays, bremsstrahlung, and so forth can be obtained. ORIGEN2, which is written in the FORTRAN language, was developed for mainframe computers such as the IBM, UNIVAC, CDC 7000 series, CRAY, and others. Versions have also been written for minicomputers like the VAX and DEC, and a version is being tested for the IBM PC-AT microcomputer.

A typical ORIGEN2 calculation usually consists of at least three processes in addition to the input data supplied by the user. First, the code performs the irradiation calculations, almost always accomplished in a series of incremental steps, until the desired burnup is reached. A single operation would result in unacceptably large numerical errors in the algorithms employed in ORIGEN2. A typical irradiation would require five to eight steps, although more can be used if the compositions at the intermediate burnups are of interest. The second process is the postirradiation decay of the spent fuel, which is very similar to the irradiation and usually involves multiple decay steps. The final process in the calculation is to multiply the g-atoms of each nuclide (the unit ORIGEN2 uses to process and store information) by a factor, contained in one of its data bases, which converts this compositional value to that of the desired characteristic (e.g., thermal output in W) for each time step. The value for each individual radionuclide may be printed as calculated, or it can be added to other values to produce a summary table for printing.

1.4 OBJECTIVES

DOE and OCRWM must be assured that any computer code that will be used in a licensing activity has passed several tests—most importantly, that it be a correct representation of the process or system for which it is intended. The term for this activity is validation, and it is usually accomplished by determining how well a computer program simulates field or laboratory experiments. In

applying this term to ORIGEN2, one needs to demonstrate that the results of the code are in agreement, to a specified degree of accuracy, with experimentally determined values. The predictions of ORIGEN2 that are potentially measurable are the composition, thermal power, photon spectrum, and neutron emission rate of some specified nuclear material. Characterization data on fuel samples suitable for validating ORIGEN2 should include (1) a complete description of the fuel's physical parameters, including enrichment, initial impurity content, pellet density and dimensional configuration, clad material and thickness, spacer material and location, poisons, moderators, and a description of the location of the sample fuel within the reactor; and (2) a detailed power history, including power density and final discharge date.

The purposes of this report are (1) to review and assess the available experimental data that may prove useful in the validation of nuclide generation and depletion computer codes and (2) to determine how well the data agree with the prediction from one such code, ORIGEN2. This review will also be of help in defining an experimental program to eliminate the gaps in the present knowledge for the radiological parameters of spent fuel.

Computerized literature search facilities were used to access the Energy Data Base (EDB). Keywords were "LWR" plus "dissolution," "computer codes" plus "reactor physics," "ORIGEN" or "ORIGEN2," and "LWR" plus "dissolution" plus "spent fuels." The EDB system covers the period from 1976 to the present. Manual searches of the Nuclear Science Abstracts for the period 1962–1975 (information not available on the EDB computer system) were made, as well as the retrieval of data from specific studies performed at ORNL for fuel reprocessing application. Much of the latter material has had limited distribution and has not been abstracted and placed in commercial data bases. Experts in each of the fields were contacted for their input. Materials referenced in the primary retrieved reports were also examined.

This report emphasizes validation data requirements to meet DOE and OCRWM needs. It is therefore limited to data for LWR assemblies and examines the composition of spent reactor fuel plus its heat generation rates and gamma and neutron radiation levels. The values predicted by ORIGEN2 or its earlier versions are given in cases where calculations are currently available. Additional calculations were not attempted because either the needed burnup data were not quoted, the analytical precision of the experimental results was questionable, or the pedigree of the samples was suspect.

2. REACTOR DATA

Generating ORIGEN2 predictions that can be compared with experimental parameters requires knowledge of the configuration of the type of reactor under consideration, the initial isotopic composition and impurity levels of the fuel (important if activation products are of interest), and the specific power history for the fuel being examined. In situations where portions of a fuel rod are examined, either a gamma scan along the rod (using a fission or activation product that does not migrate) or an equivalent method (ASTM 1985a; ASTM 1985b; ASTM 1985c) is needed to determine the level of burnup of the portion of interest. Some of this information exists and may be available from the vendors and the reactor operators, although its retrieval may not be easy or free of charge. Barner (BARNER 1985) has reported that there is a good correlation (Fig. 1) between the measured burnup of a fuel specimen and its ^{137}Cs gamma activity, provided that no

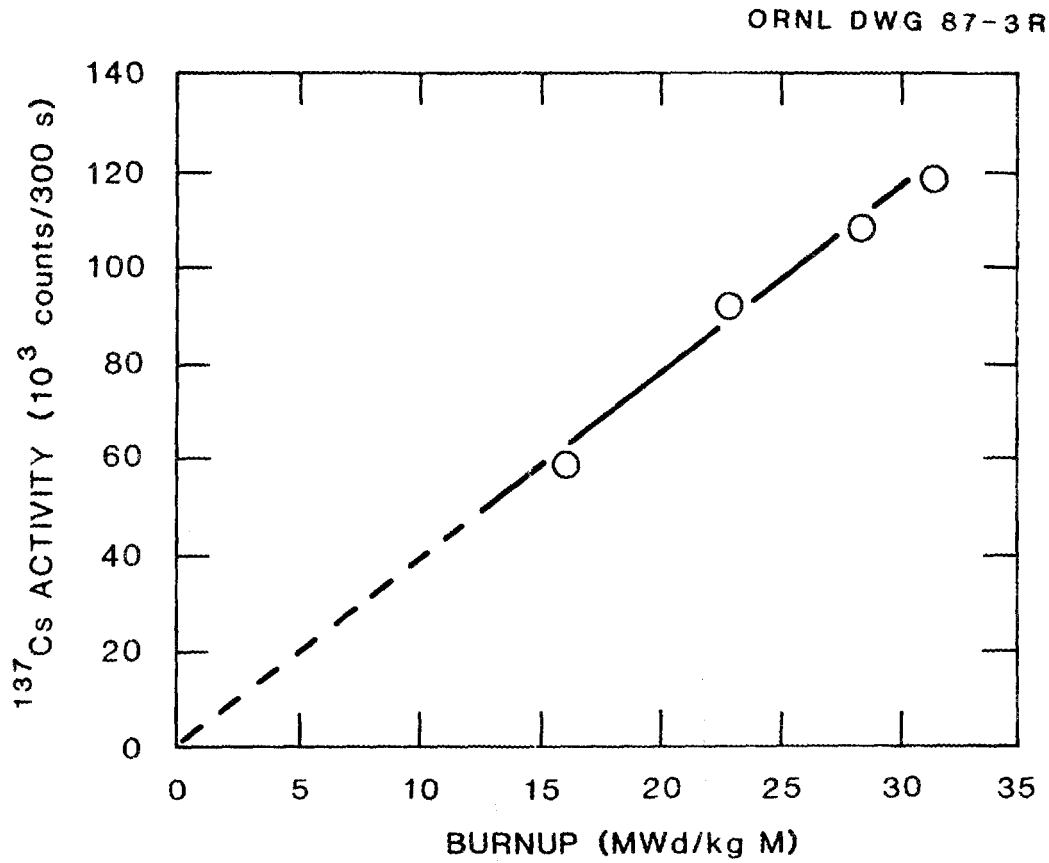

Fig. 1. Correlation between burnup analyses and ^{137}Cs gamma counting. (Source: J. O. Barner, *Characterization of LWR Spent Fuel MCC-Approved Testing Material—ATM-101*, PNL-5109, Rev. 1, June 1985.)

cesium migration has taken place within the rod from thermally driven redistribution. Frequently, data such as gamma scans may not exist and, when found, may be proprietary. A subtask is under way to obtain values for impurity levels in fuels and metallic components (NOTZ 1986). The collection of operating data for reactors (DOE 1986b) is also in progress. Table 1 outlines the information on reactor geometry, reactor operations, and initial fuel composition found during this survey. Operational data were not available for several reactors for which experimental compositions of irradiated fuel were found, so these reactors are listed without data sources (to indicate the need for information).

Table 1. Characteristics of assemblies/rods considered during this study[a]

Reactor (type)[b]	Source of information	Assembly/rod[c]	Assembly geometry	Gamma scans (assembly/rod)	Initial uranium isotopics	Linear heat rating (assembly/rod)
H. B. Robinson	MacDONALD 1975	BO-5				
(PWR)	GOODE 1978	BO-5		X[d]		X
	RYMAN 1982	X			X	
	WILSON 1983	BO-5/P8			X	BO-5/P8
	WILSON 1983	BO-5/E14			X	BO-5/E14
	BARNER 1985	BO-5	BO-5	BO-5[e]	*e*	X
Oconee-1	MONTGOMERY 1976	Cy 1	1A16		X	
(PWR)	MONTGOMERY 1978	Cy 1,2	1C56	X[d]		
	B&W 1979		1A16	X[f]	*d*	X
	BAIN 1983		X	X	X[g]	
Monticello	PASUPATHI 1982		X	*h,i*		
(BWR)	BAUMGARTNER 1984	X	X	*h,i*	*j*	X
Yankee-Rowe	NODVIK 1966	X	X	X	X	
(PWR)	RYMAN 1982				X	
Garigliano	CADELLI 1984			63/212	*j,k*	
(BWR)						
BR3	ADAMS 1984	4A, 4B		X	X	X
(PWR)	CADELLI 1984				*k,l*	
Big Rock Point						
(BWR)						
Dresden-3						
(BWR)						
Saxton						
(PWR)						
Quad Cities	WILSON 1983	GEB-161/BSG856			X	GEB-161/BSG856
(BWR)	LOWRY 1982			X	*j*	X
Obrigheim	FISCHER 1983	X			*j*	X
(PWR)						
Trino Vercellese	FISCHER 1983	509-069				
(PWR)						
SENA	FISCHER 1983				*j*	
Calvert Cliffs	WILSON 1983	BTO3/AHSO24			X	BTO3/AHSO24
Dodewaard	CADELLI 1984				*l,m*	
Turkey Point	DAVIS 1980		X			
(PWR)	ATKIN 1980		X		*j*	
	DAVIS 1981		X		*j*	X
	RYMAN 1982				X	
Surry-2	BALFOUR 1981	X	X	X		X
(PWR)						
Connecticut Yankee	LANGSTAFF 1982	X	X	X	*j*	
(PWR)						

[a]An X in a column indicates that only general data are given (i.e., not for a specific assembly or rod).
[b]PWR = pressurized-water reactor; BWR = boiling-water reactor.
[c]Power history given for designated assembly/rod in source of information.
[d]Assembly 1C56, rods 32006, 32028, and 32222.
[e]Rods C-5, D-10, G-9, G-13, J-12, L-8, N-4, N-9, and N-12.
[f]Assembly 1A16, rods 47004, 47005, and 47102.
[g]Assembly 1D13, rod 08639, piece 13; assembly 1D13, rod 08747, piece 10; assembly 1D13, rod 09603, piece 13; assembly 1D13, rod 08639; assembly 1D13, rod 08747; assembly 1D13, rod 09603.
[h]Assemblies MTB099 and MTB048 (destructive examinations); assembly MTB028 and MTBO84 (nondestructive examinations).
[i]Rods BND-304, BND-5204, BNA-0164, BND-0329, BNC-0356, BND-1688, BND-5202, BNE-0475, BNE-0583, BNG-0641, BNH-0760, and BNH-0710.
[j]Initial enrichment only.
[k]Assembly SA 56, rod 031; assembly SA 58, rod 056.
[l]Assembly ZO-100, uranium rod.
[m]Assembly B201, uranium rod.

3. RADIONUCLIDE CONTENT OF IRRADIATED FUEL

Most of the data available on the composition of reactor fuel were obtained either in fuel reprocessing studies or in destructive testing of fuel by vendors. Because of insufficient characterization of initial fuel parameters and irradiation conditions, as well as an inadequate QA basis, these data cannot be used for the validation of radionuclide generation and depletion codes. Current QA standards for critical information require that all samples to be analyzed must be completely identified and documented at each step of the handling and analytical procedures. The only studies (BARNER 1985; McELROY 1985) that meet these requirements are those by Pacific Northwest Laboratory (PNL), where a Materials Characterization Center (MCC) has been formed by DOE and OCRWM to provide spent fuel Approved Testing Materials (ATMs) to investigate nuclear waste disposal forms. In addition, the only comparisons made with the latest version of ORIGEN2 are those at PNL and the recalculation performed at ORNL (WICHNER 1983). For these reasons, most of the existing data should only be used as a guide to pinpoint areas where additional emphasis is needed. The present ORIGEN2 code should be able to predict compositions at least as well as, or better than, the older versions. Therefore, the results reported by PNL are the only currently available data that can legitimately be used for validation.

The data found in this review have been included in Appendix A, principally as a convenient resource of the existing information on fuel composition. The data of McElroy (McELROY 1985), which are presented in Table A.4 (Appendix A), were obtained under PNL's QA program, and the calculations were performed using ORIGEN2. The average ratios of measured values to values predicted by ORIGEN2, and their standard deviations, are given in Table 2.

The large ratio for ^{234}U (Table 2) is thought to be due to an error in the initial content of ^{234}U as input to ORIGEN2. The other ratios are within the standard deviations of the experimental data or the predicted precisions of the analyses for ^{235}U, ^{238}U, ^{240}Pu, ^{241}Pu [$\pm 2\%$ by thermal emission isotope dilution mass spectrometry (TEIDMS)], ^{237}Np, ^{241}Am, ^{90}Sr, and ^{137}Cs (± 5 to 10% by mass radiochemical analysis). The ORIGEN2 values appear to be low for ^{236}U, ^{238}Pu, and ^{242}Pu; slightly high for ^{239}Pu; high for ^{99}Tc; and very high for ^{79}Se and ^{126}Sn. Broad conclusions for only one fuel should not be made from these data; however, certain problems in the predictions of ^{79}Se and ^{126}Sn are evident, and some radioisotopes (^{237}Np, ^{129}I, curium isotopes, etc.) are completely lacking.

The information presented here should be considered as a guideline for the task of validating a radionuclide generation and depletion code by data with proper QA standards. However, additional data need to be generated via an experimental program.

Table 2. Comparison of measured and predicted values for irradiated fuel composition[a]

Isotope	Average ratio (experiment/ORIGEN2)	Standard deviation
^{234}U	12.6	Two determinations
^{235}U	0.98	0.03
^{236}U	1.07	0.02
^{238}U	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
^{79}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	0.01

[a]H. 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.)

4. HEAT GENERATION RATES OF ASSEMBLIES

Decay heat levels of complete spent fuel assemblies have been measured in two apparatus: (1) an in-pool calorimeter that was designed, constructed, and tested by General Electric at the Morris Operation (GE-MO) for DOE (JUDSON 1982), and (2) a water boil-off calorimeter that was designed and fabricated by PNL and tested in the engine, maintenance, and disassembly hot bay facility (EMAD) located on the Nevada Test Site and operated by Westinghouse Advanced Energy Systems Division (CREER 1981). The GE-MO test equipment is composed of two concentric pipes with an insulated annular space. The system accepts both BWR and PWR fuels and utilizes resistance temperature detectors. The PNL system consists of five major sections: (1) calorimeter vessel and support structures, (2) water supply/storage tank and fill pump, (3) steam condenser, (4) condensate collection apparatus, and (5) control and data acquisition instrumentation.

Comparisons of the results from measurements for fuel assemblies (JUDSON 1982; SCHMITTROTH 1984a,b; CREER 1984; McKINNON 1986a,b; WILES 1986) with these two systems for fuel assemblies and those predicted with ORIGEN2 are given in Table 3 and Fig. 2. The traceability of the assemblies used for the heat generation data appears to meet QA requirements.

In most cases, the agreement between the experimental and calculated thermal values is quite good. For Cooper fuel (25,000 to 28,000 MWd/MTU and cooling times of 2.3 to 4.1 years), the predictions were 2.3 W greater than the measured values, with a standard deviation of the average difference between predicted and measured values of ± 18 W, utilizing the GE-MO system (WILES 1986). The standard deviation about the average percentage difference was found to be $\pm 6.2\%$. Fourteen repeat determinations on one fuel assembly resulted in a standard deviation of ± 14 W, which indicates that the agreement between predictions and data is almost as good as the repeatability of the test data.

Measurements (McKINNON 1986a,b) made with the in-pool calorimeter on Monticello fuel (9,000 to 21,000 MWd/MTU and cooling times of 9 to 10 years) at different times dramatically demonstrate the importance of the calibration procedure. The newest predicted decay heat values were found to be 0 ± 15 W less than the values measured using a 1984 calibration method and 21 ± 2 W less than that measured when the 1985 calibration method was used. A standard deviation of ± 14.3 was calculated by the authors from a linear regression analysis of the observed vs predicted data (Fig. 2). They found that the 1985 calibration method resulted in a 20-W average difference between the data and ORIGEN2 predictions, whereas the 1984 method resulted in less than a 4-W average difference. Also, the Monticello calorimetry data obtained from the 1985 measurement method are the first BWR data in which ORIGEN2 predictions are consistently lower than the measured data. The authors concluded that ORIGEN2 predictions are accurate to within $\pm 10\%$ when used for assemblies of interest to at-reactor and interim storage facilities (200 W or greater). The results were determined to be very sensitive to the operational method used and demonstrated that the accuracy of the data is linked to the calibration method.

In earlier reports, Schmittroth (SCHMITTROTH 1984a,b) observed that the uncertainty in many of the measured values at $\pm 5\%$ gives considerable confidence to the ORIGEN2 predictions.

Table 3. Comparison of heat measurements with ORIGEN2 calculations

Reactor (type)[a]	Fuel assembly	Burnup (MWd/MTU[b])	Cooling time (d)	Decay heat (W)		Measured/predicted
				Measured	Predicted[c]	
San Onofre (PWR)	C-01	26,540	3011	359	379.4	0.95
	C-16	28,462	2012	384[d]	408.9	0.94
	C-16	28,462	2012	458[e]	408.9	1.12
	C-19	30,426	3011	418	441.4	0.95
	C-20	32,363	3011	456	469.6	0.97
	D-01	31,393	2358	499	522.2	0.96
	D-46	32,318	2360	510	541.5	0.94
	E-18	32,357	1794	635	659.4	0.96
	F-04	30,429	1078	934	963.3	0.97
WEPCo (PWR)	C-52	31,914	1635	724[d]	694.5	1.04
	C-52	31,914	1635	723[e]	694.5	1.04
	C-56	38,917	1634	921	893.1	1.03
	C-64	39,384	1633	931[d]	907.8	1.03
	C-64	39,384	1633	825[e]	907.8	0.91
	C-66	35,433	1630	846	809.2	1.04
	C-67	38,946	1629	934	896.1	1.04
	C-68	37,059	1630	874	851.4	1.03
Turkey Point (PWR)	D-04	28,430	913	1385	1555	0.89
	D-15	28,430	962	1423	1469	0.95
	D-15	28,430	1144	1126	1217	0.95
	D-15	28,430	2077	625	642	0.97
	D-22	26,485	963	1284	1357	0.95
	D-34	27,863	864	1550	1640	0.95
	B-43	25,595	1782	637	638	1.00
Dresden (BWR)	D212	5,280	4566	19	29	0.65
Cooper (BWR)	CZ102	11,667	2541	62	84	0.74
	CZ147	26,710	1294	276.7	296.4	0.93
	CZ148	26,310	1283	273.5	293.6	0.93
	CZ182	26,820	860	342.6	364.9	0.94
	CZ195	26,390	1289	255.5	289.6	0.88
	CZ205	25,340	857	324.0	331.7	0.98
	CZ205	25,340	868	368.0	328.3	1.12
	CZ205	25,340	871	343.5	327.4	1.05
	CZ205	25,340	872	352.9	327.1	1.08
	CZ205	25,340	886	331.7	322.8	1.03
	CZ205	25,340	887	338.6	322.5	1.05
	CZ205	25,340	892	327.4	321.0	1.02
	CZ205	25,340	896	313.5	320.0	0.98
	CZ205	25,340	899	311.3	319.2	0.98
	CZ205	25,340	900	314.0	313.8	1.00
	CZ205	25,340	936	331.0	315.5	1.05
	CZ205	25,340	946	317.2	318.2	1.00
	CZ205	25,340	1089	289.7	276.0	1.05
	CZ205	25,340	1103	308.0	273.6	1.13
	CZ209	25,380	891	279.5	289.3	0.97
	CZ211	26,680	1261	296.0	302.7	0.98
	CZ211	26,680	1491	240.3	266.6	0.90
	CZ222	26,690	898	355.7	346.2	1.03

Table 3 (continued)

Reactor (type)[a]	Fuel assembly	Burnup (MWd/MTU[b])	Cooling time (d)	Decay heat (W)		Measured/predicted
				Measured	Predicted[c]	
	CZ225	25,800	835	333.5	321.4	1.04
	CZ239	27,250	893	366.5	358.8	1.02
	CZ246	27,360	896	320.9	364.4	0.88
	CZ246	27,360	899	341.7	363.4	0.94
	CZ256	27,596	834	397	384	1.03
	CZ259	26,470	1288	247.6	290.9	0.85
	CZ259	26,470	1340	288.5	281.7	1.02
	CZ259	26,470	1485	254.1	260.0	0.98
	CZ264	26,500	1282	263.8	292.6	0.90
	CZ277	26,480	1287	262.7	290.9	0.90
	CZ277	26,480	1497	243.0	261.2	0.93
	CZ286	27,140	930	278.4	326.8	0.85
	CZ286	27,140	1104	284.2	290.8	0.98
	CZ296	26,390	1293	256.7	297.0	0.86
	CZ296	26,390	1492	251.9	266.0	0.95
	CZ302	26,590	1283	285.6	290.8	0.98
	CZ308	25,820	895	269.7	298.7	0.90
	CZ311	27,390	890	356.9	340.1	1.05
	CZ315	26,880	932	328.0	317.2	1.03
	CZ318	26,570	931	277.6	297.9	0.93
	CZ331	21,332	2346	167	162	1.03
	CZ337	26,720	891	347.7	346.4	1.00
	CZ337	26,720	1095	300.4	295.5	1.02
	CZ342	27,070	931	280.1	320.1	0.88
	CZ342	27,070	1101	300.0	286.1	1.05
	CZ346	28,050	890	388.7	376.5	1.03
	CZ348	27,480	894	342.8	355.5	0.96
	CZ351	25,750	931	313.8	297.1	1.06
	CZ355	25,420	891	290.5	293.0	0.99
	CZ357	27,140	932	320.3	326.3	0.98
	CZ369	26,580	858	347.7	343.3	1.01
	CZ370	26,340	1257	293.6	292.4	1.00
	CZ372	25,850	1256	294.3	286.3	1.03
	CZ379	25,930	898	287.4	296.9	0.97
	CZ398	27,480	890	372.0	361.0	1.03
	CZ415	25,860	1255	289.3	286.7	1.01
	CZ416	27,460	894	319.8	339.1	0.94
	CZ429	27,640	889	385.6	370.3	1.04
	CZ430	26,820	894	353.3	344.2	1.03
	CZ433	25,980	1255	287.4	281.6	1.02
	CZ433	25,980	1492	256.7	252.7	1.02
	CZ460	26,510	933	313.5	308.9	1.01
	CZ466	26,080	861	302.1	309.4	0.98
	CZ468	26,760	935	325.3	317.9	1.02
	CZ472	25,960	859	325.0	321.2	1.01
	CZ473	26,520	934	293.2	297.6	0.99
	CZ498	26,480	888	359.4	345.0	1.04
	CZ508	26,360	933	310.0	309.6	1.00
	CZ515	25,740	1254	294.0	285.0	1.03
	CZ515	25,740	1285	296.0	279.2	1.06
	CZ526	27,600	864	397.0	378.9	1.05
	CZ526	27,600	1097	321.8	323.1	1.00

Table 3 (continued)

Reactor (type)[a]	Fuel assembly	Burnup (MWd/MTU[b])	Cooling time (d)	Decay heat (W) Measured	Decay heat (W) Predicted[c]	Measured/predicted
	CZ528	25,720	1284	297.6	279.0	1.07
	CZ531	26,700	893	347.2	343.2	1.01
	CZ536	26,590	1256	295.2	296.1	1.00
	CZ542	26,690	932	311.9	312.4	1.00
	CZ545	26,670	935	295.2	300.6	0.98

[a]PWR = pressurized-water reactor; BWR = boiling-water reactor.
[b]MTU = metric tons of uranium.
[c]By ORIGEN2 calculations.
[d]Static test.
[e]Recirculation test.

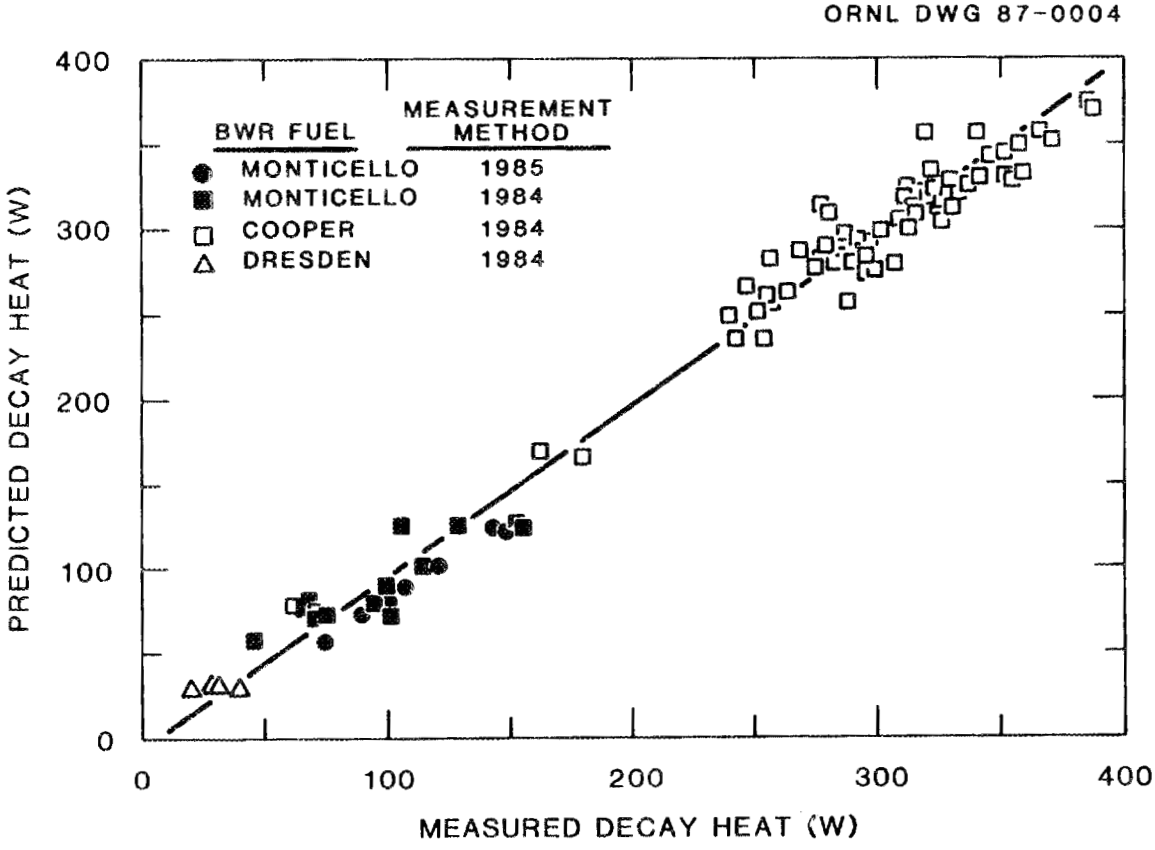

Fig. 2. Predicted and measured decay heat rates for Dresden, Cooper, and Monticello spent fuel assemblies. (Source: M. A. McKinnon et al., *Monticello BWR Spent Fuel Assembly Decay Heat Predictions and Measurements*, PNL-5799, June 1986.)

However, he cautions that the excellent agreement may apply only over a limited cooling time period during which the decay heat is dominated by contributions from a limited number of fission product isotopes; agreement over longer periods may not be as good, especially for decay times over 100 years, in which actinide decay heat becomes dominant. McKinnon et al. (McKINNON 1986a) concurred with this conclusion by stating that the results of their study are not applicable to old fuels that have very low decay heat rates.

5. COMPARISON OF MEASURED AND PREDICTED RADIATION LEVELS

The major interest in measuring radiation levels in spent fuel assemblies has been as an assay tool for the accounting and control of nuclear materials. Both gamma-ray (PHILLIPS 1985; NICHOLSON 1981; MOSS 1980; LEE 1979) and neutron (LEE 1985; PHILLIPS 1980; YOKOYAMA 1981; HSUE 1979) methods have been tested for this application. However, the data generated in these experiments are not applicable for a validation effort because the studies were designed to estimate burnup only.

5.1 PREDICTED AND MEASURED GAMMA-RAY COMPARISONS

One report of the measurement of gamma exposure rates both in air and underwater was found (STRICKLER 1981); an early version of ORIGEN was used in this study. Traceability of the assemblies used in these radiation-level experiments is probably inadequate for current QA requirements since the reactor from which the fuel was obtained was not identified. Determination of the gamma exposure depends primarily on the easily measured gamma-emitting radioisotopes (^{144}Ce, ^{137}Cs, ^{134}Cs, and ^{106}Ru) in the fuel, which are also the most easily quantified components. The early versions of ORIGEN apparently predict these important radioisotopes accurately, as confirmed by the material presented in Appendix A.

Gamma exposure rates (a measure of gamma radiation levels) near spent fuel bundles were determined in air as well as underwater. The latter determination was accomplished using a "diving bell" sized to fit over the fuel and detector in the storage pool. The apparatus was designed to hold both the fuel bundle and an ion chamber underwater in any of several fixed geometries.

Maximum exposure rates varied from 36,000 R/h adjacent to a 3.5-year-old PWR bundle with a 30,000-MWd/MTU burnup to 40 R/h for a 10.5-year-old BWR bundle with a burnup of only 180 MWd/MTU. The utilities that were the sources for the fuel were not identified.

An early version of the ORIGEN code was used, with data supplied by the utilities, to calculate the photon energies generated within the bundles (STRICKLER 1981). The geometric attenuation code, QAD-F (MALENFANT 1967), was used to calculate the dose at the chosen point from a distributed source which approximated the size and shape of a fuel bundle. Figure 3 identifies the locations of the detector tubes used in the counting measurements. Table 4 summarizes the ORIGEN input specifications for the fuels examined. Tables 5 through 7 compare the predicted and the measured exposure rates at various points on or near PWR and BWR bundles.

The agreement between the predicted and measured radiation levels for locations alongside the fuel was usually within a factor of 2. According to Strickler, this agreement demonstrates that both ORIGEN and QAD-F are adequate for estimation of radiation levels. The major differences for locations above the fuel are said to be due to deficiencies in the geometric model (QAD-F). The usefulness of this ORIGEN comparison is further limited by its dependence on another code.

Axial and radial gamma-ray dose rate measurements have been made on a partially and fully loaded Ridihalgh, Eggers & Associates REA 2023 BWR spent fuel storage cask with thermo-

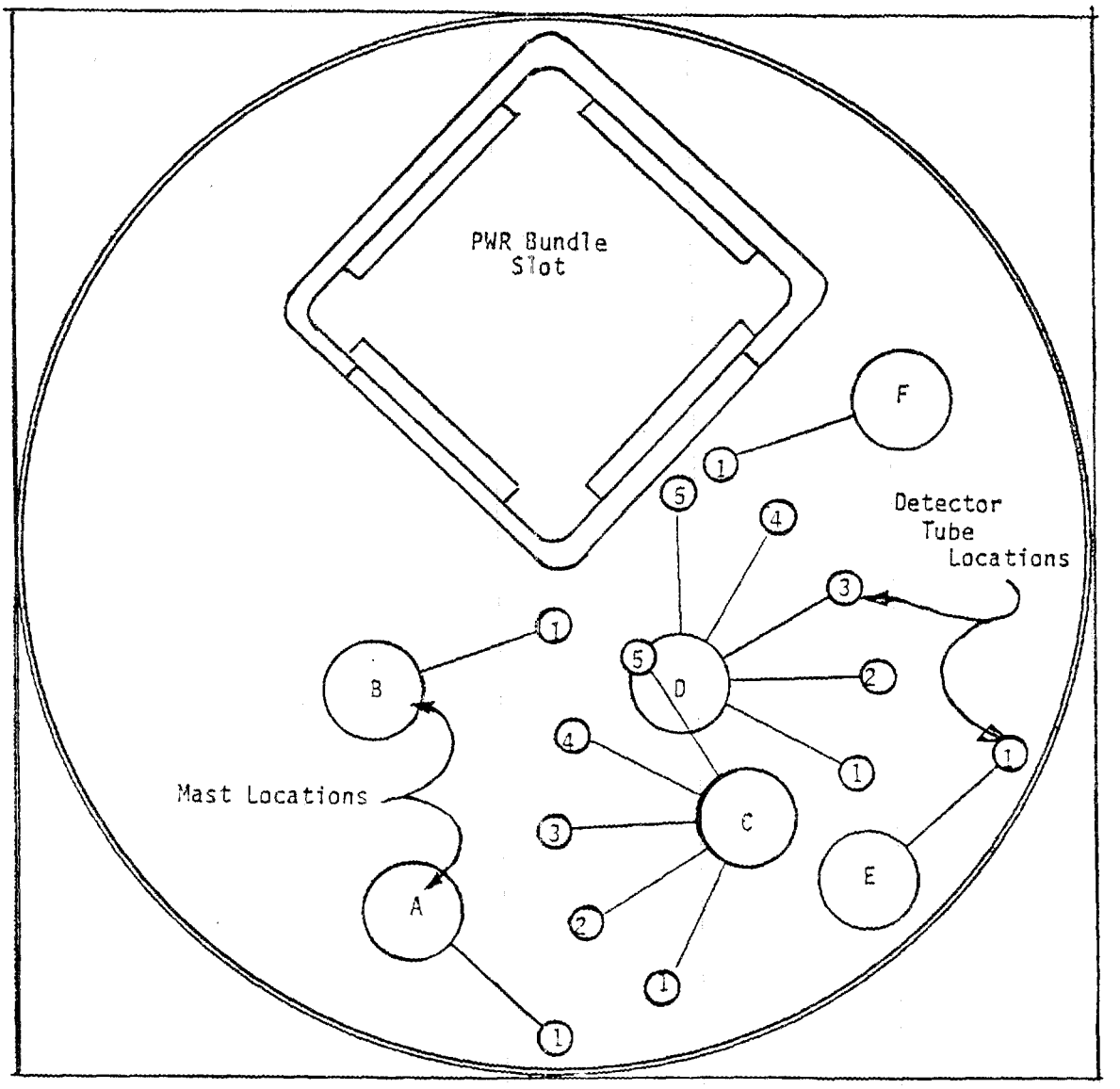

Fig. 3. Location of detector tubes. (Source: B. F. Judson, *In-Plant Test Measurements for Spent Fuel Storage at Morris Operation,* NEDG-24922-2, September 1981.)

luminescent dosimeters, an intrinsic germanium spectrometer, and portable gamma meters (McKINNON 1986a; WILES 1986). Fuel assemblies were obtained from Northern States Power Company's Monticello Nuclear Generating Plant and Nebraska Public Power District's Cooper Nuclear Station, respectively. ORIGEN2 was used to calculate the photon source terms in the axial direction for both of these cases, and the QAD-CG code (MALENFANT 1967; ORNL 1977) was then used to calculate gamma-ray dose rates on the outer surface of the cask. The QAD-CG code calculates the fast-neutron and gamma-ray penetrations through various shielding configurations using the point-kernel method. In this method, the source volume is represented by a number of point isotropic sources, and the code computes the line-of-sight distance from each of these source

Table 4. Input specifications for ORIGEN calculations[a]

Reactor type[b]	Bundle	Burnup (MWd/MTU[c])	Specific power (MW/MTU)	Initial enrichment (%)	Decay time[d] (months)
PWR	1A	40,200	31.8	3.397	48
PWR	1B	32,800	31.8	3.397	48
PWR	1C	26,400	31.8	2.264	64
PWR	2A	32,400	23.5	3.865	93
PWR	2B	26,400	23.5	3.865	93
PWR	2C	32,400	23.5	4.005	72
PWR	2D	30,400	23.5	3.996	30
PWR	2E	32,400	23.5	4.005	54
BWR	3A	5,130	10.9	2.13	107
BWR	3B	2,080	10.9	2.13	119
BWR	3C	3,070	10.9	2.13	107

[a]Parameters were chosen to match those reported by the utilities for the bundles that were measured.
[b]PWR = pressurized-water reactor; BWR = boiling-water reactor.
[c]MTU = metric tons of uranium.
[d]Time from discharge from the reactor to measurement.

Table 5. Predicted and measured exposure rates adjacent to the corners of pressurized-water reactor fuel bundles

		Exposure rate (R/h)							
		B-1 Profile				A-1 Profile			
	Vertical	Water		Air		Water		Air	
Bundle[a]	position[b]	Predicted	Measured	Predicted	Measured	Predicted	Measured	Predicted	Measured
1A	Top	6,850	7,000	5,150	10,000	1,590	1,700	4,310	6,500
	Inter[c]	29,100	28,500	20,100	33,500	4,640	5,300	10,800	18,000
	Middle	40,700	31,300	27,800	34,800	4,680	5,600	11,100	18,100
1B	Top	4,940	4,400			1,160	1,200		
	Middle	21,000	20,500			3,340	4,100		
	Bottom	29,400	24,000			3,380	4,600		
1C	Top	3,250	3,200			762	760		
	Middle	13,800	12,600			2,210	2,300		
	Bottom	19,300	13,800			2,225	2,500		
2A	Top	2,920	2,800	2,110	3,750	619	680	1,650	2,600
	Middle	11,000	12,300	7,510	16,000	1,720	2,400	3,930	7,450
	Bottom	13,600	14,000	9,310	16,800	1,730	2,600	4,050	7,650
2B	Top	2,220	2,400	1,620	3,000	471	540	1,250	2,150
	Inter[c]	8,379	8,700	5,710	12,500	1,310	1,900	2,980	6,000
	Middle	10,400	9,000	7,080	13,500	1,315	2,000	3,080	6,150
2C	Top	3,610	4,200			769	1,100		
	Inter[c]	13,600	16,900			2,140	3,000		
	Middle	16,900	16,900			2,150	3,400		
2D	Top	6,720	10,000	4,900	12,000	1,440	1,900	3,770	8,000
	Inter[c]	25,300	32,000	17,300	37,000	3,960	5,900	9,040	20,300
	Middle	31,450	35,900	21,400	40,200	4,010	6,800	9,330	21,900
2E	Top	4,650	5,600			992	1,300		
	Inter[c]	17,500	22,000			2,740	4,000		
	Middle	21,700	25,000			2,750	4,600		

[a]See Table 4.
[b]With respect to the active fuel length.
[c]Inter. = three-fourths of fuel length from bottom.

Table 6. Predicted and measured exposure rates along the sides and above pressurized-water reactor fuel bundles

Bundle[a]	Vertical position[b]	Exposure rate (R/h)							
		E-1 Profile				Above[c]			
		Water		Air		Water		Air	
		Predicted	Measured	Predicted	Measured	Predicted	Measured	Predicted	Measured
1A	Top+[d]	2,010	2,100	4,260	5,800	7	85	25	475
	Inter[e]	6,370	6,200	11,300	18,000				
	Middle	6,430	6,800	11,500	18,000				
1B	Top	1,550	1,600						
	Inter[e]	4,600	5,300						
	Middle	4,650	5,500						
1C	Top	1,070	1,000						
	Inter[e]	3,020	2,800						
	Middle	3,050	3,000						
2A	Top+	825	890	1,600	2,630	2.5	40	6.8	275
	Inter[e]	2,340	2,800	4,050	7,300				
	Middle	2,350	3,000	4,140	7,510				
2B	Top+	628	650	1,220	2,020	1.9	30	5.2	220
	Inter[e]	1,780	2,200	3,084	5,750				
	Middle	1,790	2,300	3,150	5,700				
2C	Top+	1,020	2,200			3.2			
	Inter[e]	2,900	3,600						
	Middle	2,910	4,000						
2D	Top+	1,920	2,200	3,680	9,600	6.5	145	17	800
	Inter[e]	5,420	6,500	9,280	20,800				
	Middle	5,440	7,400	9,510	22,000				
2E	Top	1,320	1,600						
	Inter[e]	3,730	4,800						
	Middle	3,750	5,400						

[a]See Table 4.
[b]Given with respect to the active fuel length.
[c]53 and 64 cm above the active length for Utilities 1 and 2, respectively.
[d]At top or above.
[e]Inter. ≈ three-fourths of fuel length from bottom.

points to a detector point. From the distances through the shielding regions and the attenuating characteristics of the shielding materials, both geometric and material attenuation are determined.

The predicted gamma-ray dose rates obtained from these measurements and calculations are approximately 6% less than the measured values for the side of a partially loaded cask. Differences between predictions and measurements average −1.2 mrem/h on the top of the cask; the standard deviation is a relatively high ±20.1 mrem/h, and the average predictions are 8% less than the measurements. The centerline dose rate calculated for the bottom of the cask is 11% higher than the measurement. Comparisons of calculated dose rates with measured values for the fully loaded cask are similar to those for the partially loaded cask; that is, near the cask centerline, the calculated dose rates are higher than the experimental data, but the periphery agreement is good. A summary of the results is given in Tables 8 and 9.

In summary, Wiles et al. (WILES 1986) found that the predictions agree well with measured data over the length of the neutron shield. Above and below the ends of the shield, the dose rate peaks were underpredicted by a factor of 2. The reasons for the discrepancy may be due to an

Table 7. Predicted and measured exposure rates near boiling-water reactor fuel bundles

Bundle[a]	Vertical position[b]	Exposure rate (R/h)			
		Water		Air	
		Predicted	Measured	Predicted	Measured
		B-1 profile (corner)			
1	Top	87	105	91	185
	Inter[c]	470	940	437	1200
	Middle	510	885	482	1400
2	Top	32	35		
	Inter[c]	172	250		
	Middle	289	568		
		A-1 profile (corner)			
1	Top	20	30	72	162
	Inter[c]	97	190	262	650
	Middle	106	190	296	730
2	Top	7	15		
	Inter[c]	35	60		
	Middle	60	122		
		E-1 profile (side)			
1	Top+[e]	20	40	59	130
	Inter[c]	103	220	232	540
	Middle	113	245	264	600
2	Top+	8	15		
	Inter[c]	39	60		
	Middle	43	90		
3	Top+	12	11		
	Inter[c]	60	132		
	Middle	65	136		
		Above[d]			
1	Top+	0.05	2	0.2	15
2	Top+	0.02			
3	Top+	0.03			

[a]See Table 4.
[b]With respect to the active fuel length.
[c]Inter = three-fourths of fuel length from bottom.
[d]74 cm above active length.
[e]At top or above.

Table 8. Comparisons of predicted gamma dose rates with data for a partially loaded cask

Radius (cm)	Angle (deg)	Elevation (cm)	Measured	Predicted	Pred.−Meas.	Pred./Meas.
			Side (45°) dose rates (mrem/h)			
		40.0	11.5	5.6	−5.8	0.49
		46.0	5.2	4.4	−0.8	0.85
		60.0	5.3	5.8	0.4	1.08
		100.0	6.7	10.1	3.4	1.51
		150.0	8.5	11.9	3.4	1.40
		223.1	8.0	11.9	3.9	1.49
		300.0	8.3	9.6	1.3	1.15
		350.0	6.6	6.9	0.3	1.04
		380.0	5.5	4.8	−0.7	0.87
		408.5	6.3	4.8	−1.5	0.76
		420.0	6.8	5.4	−1.5	0.79
		440.0	12.2	16.1	4.0	1.33
		450.0	11.3	11.5	0.3	1.02
		460.0	8.3	7.0	−1.3	0.85
		470.0	14.8	8.2	−6.5	0.56
		480.0	19.4	9.3	−10.1	0.48
		490.0	3.8	1.3	−2.5	0.35
				Average:	−0.8	0.94
				Standard deviation:	±3.7	±0.35
			Top dose rates (mrem/h)			
0.0	0		26.8	64.9	38.1	2.42
13.0	45		25.3	46.4	21.1	1.83
37.6	45		20.7	30.3	9.6	1.47
61.0	45		18.3	21.9	3.6	1.19
64.5	24.5		13.2	7.3	−5.9	0.55
80.0	26.5		31.1	4.9	−26.2	0.16
80.0	45		28.4	5.2	−23.2	0.18
90.0	26.5		16.4	3.2	−13.2	0.20
90.0	45		19.3	4.5	−14.8	0.23
				Average:	−1.2	0.92
				Standard deviation:	±20.1	±0.80
			Bottom dose rates (mrem/h)			
0.0	0		68.6	76.1	7.5	1.11
				Overall average:	−0.6	0.94
				Overall standard deviation:	±12.1	±0.58

Source: L. E. Wiles et al., *BWR Spent Fuel Storage Cask Performance Test, Volume II, Pre- and Post-Test Decay Heat, Heat Transfer, and Shielding Analyses*, Pacific Northwest Laboratory, PNL-5777, Vol. II, June 1986.

Table 9. Comparisons of predicted gamma dose rates with data for a fully loaded cask

Radius (cm)	Elevation (cm)	Angle (deg)	Measured	Predicted	Pred.−Meas.	Pred./Meas.
			Dose rates on side of cask (mrem/h)			
	100.0	26.5	11.7	15.5	3.8	1.33
	223.1	45.0	11.6	18.6	7.0	1.60
	300.0	26.5	13.4	14.6	1.3	1.10
	400.0	26.5	10.8	6.3	−4.4	0.59
	440.0	26.5	26.6	24.6	−2.1	0.92
	475.0	26.5	36.8	13.1	−23.7	0.36
				Average:	−3.0	0.98
				Standard deviation:	±10.0	±0.42
			Dose rates on top of cask (mrem/h)			
0.0		0	28.6	60.5	31.8	2.11
13.0		45	26.9	42.9	16.0	1.59
25.3		45	31.5	46.0	14.5	1.46
37.6		45	30.9	45.6	14.7	1.48
49.3		45	37.3	53.5	16.2	1.44
61.0		45	33.1	30.7	−2.4	0.93
63.5		24.5	32.3	30.8	−1.5	0.95
71.0		45	35.9	20.0	−15.9	0.56
75.0		45	47.9	13.7	−34.1	0.29
77.5		45	61.6	10.3	−51.3	0.17
80.0		45	60.7	14.6	−46.1	0.24
82.5		45	57.0	17.4	−39.6	0.31
85.0		45	49.9	15.4	−34.5	0.31
90.0		26.5	29.9	8.5	−21.4	0.28
90.0		45	26.4	7.9	−18.5	0.30
95.0		45	11.6	2.6	−9.1	0.22
				Average:	−11.3	0.79
				Standard deviation:	±24.7	±0.62
			Dose rates on bottom of cask (mrem/h)			
0.0	0	69.7	72.1	2.4	1.03	
61.0	45	83.2	37.2	−46.0	0.45	
				Average:	−21.8	0.74
				Standard deviation:	±24.2	0.29
				Overall average:	−10.1	0.83
				Overall standard deviation:	±21.9	±0.56

Source: L. E. Wiles et al., *BWR Spent Fuel Storage Cask Performance Test, Volume II, Pre- and Post-Test Decay Heat, Heat Transfer, and Shielding Analyses,* Pacific Northwest Laboratory, PNL-5777, Vol. II, June 1986.

incorrectly assumed cobalt content in the stainless steel assembly end fittings (because the authors did not know its exact concentration).

5.2 PREDICTED AND MEASURED NEUTRON DOSE-RATE COMPARISONS

Neutron dose-rate measurements were made with a ^{3}He spectrometer, tissue equivalent proportional counter, multisphere spectrometer, and several survey instruments (McKINNON 1986a; WILES 1986) on the same partially and completely filled storage cask described previously. The absolute neutron source strength was based on ORIGEN2 values calculated for each assembly, and the resulting dose rates were computed for the outer surface of the cask, utilizing the DOT computer code (RHOADES 1982). This code calculates neutron and photon particle fluxes in two dimensions using the method of discrete ordinates to solve the Boltzmann transport equation. Balance equations are solved for the flow of particles moving in a set of discrete directions in each cell of a mesh space and in each group of a multigroup energy structure. Mesh spacings and discrete directions are selected by the user. Anisotropic cross sections can be expressed in a Legendre expansion of arbitrary order.

The authors found that the neutron dose rates predicted for the ends and side of the cask where there is no neutron shield were a factor of approximately 2 to 6 higher than those determined experimentally. Calculated neutron dose rates were approximately a factor of 2 higher than the experimental data for the side of the cask where there is shielding. Reasons for the differences were given as (1) inaccurate modeling of the cask, (2) incorrect neutron cross sections, and (3) incorrect axial neutron source strength profiles. The authors believe that the last item is the major reason for the difference. Comparisons of predicted vs measured neutron dose rates are given in Table 10.

The Japanese have measured the neutron emission rates for irradiated BWR fuel assemblies and compared them with calculated values (YOKOYAMA 1981). Measurements were performed on spent fuel with burnups ranging from 11,000 to 19,000 MWd/MTU and cooling times varying from 247 to 711 d. Beta activities of neutron-activated gold-foil were determined with two calibrated 2-in.-diam plastic scintillation detectors. Although ORIGEN was not used as the primary source for neutron strength calculations, the code was used to calculate corrections to the atomic concentrations obtained for some of the nuclides, especially for ^{242}Cm. Only eight nuclides (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, and ^{244}Cm) were considered in their analysis for neutron production. They found that the calculational methods used generally underestimated the neutron emission rate for highly exposed fuel (19,000 MWd/MTU). They concluded that this underestimation occurred because the neutron absorption cross sections for the nuclides ^{241}Pu through ^{243}Am in their cross-section library were too small.

Table 10. Comparison of predicted neutron dose rates with data for a fully loaded cask

Elevation (cm)	Angle (deg)	Measured	Predicted	Pred.−Meas.	Pred./Meas.
		Side of cask (mrem/h)			
0.00	26.5	5.0	24.7	19.7	4.9
3.33	26.5	8.8	28.5	19.7	3.2
6.67	26.5	7.7	32.0	24.3	4.2
10.00	26.5	10.7	34.5	23.8	3.2
13.33	26.5	12.8	38.5	25.7	3.0
16.67	26.5	15.0	43.0	28.0	2.9
20.00	26.5	14.3	47.0	32.7	3.3
23.33	26.5	12.5	51.0	38.5	4.1
26.67	26.5	15.6	55.0	39.4	3.5
30.00	26.5	16.2	56.6	40.4	3.5
33.33	26.5	18.7	57.0	38.3	3.0
36.67	26.5	14.2	50.0	35.8	3.5
40.00	26.5	9.2	37.0	27.8	4.0
43.33	26.5	2.1	29.0	26.9	13.8
46.67	26.5	0.4	11.0	10.6	27.5
50.00	26.5	1.2	6.5	5.3	5.4
60.00	26.5	1.5	3.0	1.5	2.0
100.00	26.5	1.5	5.0	3.5	3.3
150.00	26.5	3.4	6.8	3.4	2.0
223.10	0.0	3.8	6.0	2.2	1.6
223.10	4.0	3.6	6.0	2.4	1.7
223.10	26.5	6.2	6.0	−0.2	1.0
223.10	41.0	2.8	6.0	3.2	2.1
223.10	45.0	2.6	6.0	3.4	2.3
300.00	26.5	3.2	4.1	0.9	1.3
380.00	26.5	1.7	1.2	−0.5	0.7
420.00	26.5	0.6	0.6	0.0	1.0
430.00	26.5	0.9	1.2	0.3	1.3
433.33	26.5	0.9	1.9	1.0	2.1
436.67	26.5	1.5	5.0	3.5	3.3
440.00	26.5	9.3	36.8	27.5	4.0
443.33	26.5	8.8	37.0	28.2	4.2
446.67	26.5	7.9	34.5	26.6	4.4
450.00	26.5	6.0	32.2	26.2	5.4
453.33	26.5	6.0	29.8	23.8	5.0
456.67	26.5	8.7	27.0	18.3	3.1
460.00	26.5	7.0	24.3	17.3	3.5
463.33	26.5	5.1	22.0	16.9	4.3
466.67	26.5	4.8	19.2	14.4	4.0
470.00	26.5	4.3	16.5	12.2	3.8
473.33	26.5	3.9	14.0	10.1	3.6
476.67	26.5	3.4	11.5	8.1	3.4
480.00	26.5	4.6	9.0	4.4	2.0
490.00	26.5	2.1	2.5	0.4	1.2
			Average:	+15.8	3.9
		Standard deviation:		±13.1	±4.1
		Top of cask (mrem/h)			
0.0	45	12.6	38.1	25.5	3.0
60.0	45	12.3	25.1	12.8	2.0

Table 10 (continued)

Elevation (cm)	Angle (deg)	Measured	Predicted	Pred. − Meas.	Pred./Meas.
80.0	45	5.8	12.9	7.1	2.2
95.0	45	2.7	3.7	1.0	1.4
			Average:	+11.6	2.2
			Standard deviation:	±9.0	±0.6
		Bottom of cask (mrem/h)			
0.0	45	32.8	192.0	159.2	5.9
0.0	135	29.8	192.0	162.2	6.4
0.0	225	34.2	192.0	157.8	5.6
0.0	315	23.7	192.0	168.3	8.1
80.0	45	15.5	65.0	49.5	4.2
95.0	45	4.5	23.3	18.8	5.2
97.5	45	6.0	22.6	16.6	3.8
100.0	45	4.0	24.2	20.2	6.1
105.0	45	5.3	28.0	22.7	5.3
107.5	45	3.6	29.0	25.4	8.1
110.0	45	2.6	27.5	24.9	10.6
			Average:	+75.1	6.3
			Standard deviation:	±66.2	±1.9
			Overall average:	+26.6	4.2
			Overall standard deviation:	±30.8	±3.7

Source: L. E. Wiles et al., *BWR Spent Fuel Storage Cask Performance Test, Volume II, Pre- and Post-Test Decay Heat, Heat Transfer, and Shielding Analyses,* Pacific Northwest Laboratory, PNL-5777, Vol. II, June 1986.

6. CONCLUSIONS

The goals of any validation procedure can be stated in terms of how accurately a computer code predicts one or more quantities of interest; hence, the validation effort requires the availability of suitable experimental data for detailed comparisons. Specific output quantities of importance are used as indicators for determining whether a code provides satisfactory agreement with experimental results. These indicators can serve to measure a code's level of validation. The qualification that the data be suitable implies that (1) the important variables have been studied, (2) the correct ranges for these variables have been covered, (3) the proper measurements have been made, (4) the errors in the data are small enough to permit comparisons with the predictions, and (5) the experimental data have been generated under the guidance of a QA program. These five criteria may be assumed to be qualifying standards for validation purposes. The comparison of the predicted and experimental results must consider whether the experiments will provide a direct measure of the physical processes of interest. The experiments should be selected so that the expected variables and mechanisms will be studied and the validation assessment process will become one of direct comparisons between the predictions and experimental results.

In a strict validation effort, predictions from each of the four LWR models available in ORIGEN2 need to be compared with experimentally measured values for all of the important variables and indicators. Four such indicators used in ORIGEN2 validation are the radionuclide inventory, decay heat, photon production rate, and neutron production rate. Each of these will be considered by applying the qualifying standards to the experimental data currently available to determine whether the data are sufficient for the validation of ORIGEN2 or any similar code.

6.1 RADIONUCLIDE INVENTORY

Although many citations were retrieved during the examination of the literature, only one was found to have the proper credentials for use in validating ORIGEN2-type codes. The program (BARNER 1985; McELROY 1985), initiated by the DOE/OCRWM at PNL through the MCC, is the only one identified in this survey as meeting the requirements of NQA-1 (ANSI 1983). However, the MCC's primary mission (BARNER 1984) is to provide reference and testing materials for use in development of reference nuclear waste materials and samples and for repository-related tests in the several National Waste Terminal Storage projects; it does not involve generation of data for a validation effort of radionuclide inventory computer codes. Since theirs is a new program, the information generated at PNL is limited to only a few elements (uranium, plutonium, and selected fission products) that are considered significant in terms of the FWMS.

Of the 33 radionuclides identified in the survey as having had both analyses and ORIGEN predictions, agreement was found to be within 10% for 17, within 50% for 13, and greater than 50% for 3 (Table A.6). Most of the actinides, ^{14}C, ^{90}Sr, ^{106}Ru, and ^{137}Cs fall into the first group. The middle group contains six actinides and seven fission products. The last group is comprised of ^{79}Se, ^{126}Sn, and ^{154}Eu. Many of these experimental values resulted from a single determination on fuel

that had not been properly documented, and the burnup information was questionable. In addition, the version of ORIGEN used to obtain the calculational values is not easily traceable in many instances. Virtually no measurements that were obtained under appropriate QA methods have been made for BWRs.

6.2 DECAY HEAT

The radioactive decay data base in ORIGEN2 supplies the recoverable heat per decay for each radionuclide, which is the heat that would be deposited within the nuclear material itself or in a very large surrounding shield. ORIGEN2 will output such data for each radionuclide individually and will give a total value.

Comparisons of ORIGEN2 predictions with calorimetric data indicate that the computer code approaches the precision of the experimental measurements and may exceed them in some cases. The general consensus is that ORIGEN2 predictions are accurate to within at least $\pm 10\%$. The major caveat that must be considered is the limited range of cooling times (3 to 8 years) exhibited by the fuel assemblies available for study. For these short cooling periods, the decay heat is preponderantly that generated by fission products. The actinide heat generation rates exceed those from fission products after 60 to 70 years and are the major producers (>99%) after 300 years (RODDY 1986). A significant difference (40%) is noted for the two low-burnup (5000- and 12,000-MWd/MTU) fuel assemblies, probably resulting from the difficulty encountered in experimentally measuring such low heat contents.

6.3 GAMMA EMISSION RATES

The ORIGEN2 photon data base supplies the number of photons per decay in an 18-energy-group structure. These values are used to output a table that gives the number of photons and the photon energy emission rates in these energy groups as a function of irradiation and decay time, as well as a value for the total production. Usually, additional computer codes are required before comparisons can be made between experimental and calculated values.

Two literature citations were identified that give quantitative data relating to photon production of spent fuel assemblies. The purpose of the tests was to determine the self-shielding effects of the hardware and the end pellets in LWR fuel assemblies at various locations surrounding the assemblies.

Utilizing an earlier version of ORIGEN2 to find the intensities of the gamma emissions and the geometric computer code QAD-F to determine the exposure rate, Strickler (STRICKLER 1981) found reasonable agreement (a factor of 2) between the predicted and the measured values for locations near the sides of the fuel; however, there were large discrepancies (greater than an order of magnitude) when the codes were applied to locations above the fuel. He concluded that these differences may be due to deficiencies in the shielding model.

Wiles et al. (WILES 1986) used ORIGEN2 to calculate photon source terms and QAD-CG (ORNL 1977) to calculate the gamma-ray radiation dose rates for the REA cask containing selected BWR spent fuel assemblies. They concluded that the QAD code may be utilized for shielding calculations, even though it underpredicts gamma-ray radiation dose peaks by as much as a factor of 2. Limitations of the point-kernel method are cited for these discrepancies between measured and predicted values. They found that 60% of the predicted gamma dose rate on the side of the cask was due to secondary gamma rays. The major contributor to predicted dose rates was caused by ^{60}Co. Thus, the ^{59}Co content in all of the nonfuel components must be accurately known.

6.4 NEUTRON PRODUCTION RATES

Neutron emission rates are of importance for use in transportation studies, for the design of shipping casks, for spent fuel handling and emplacement, and for use in criticality analyses. Neutrons are generated in a discharged fuel assembly by essentially two mechanisms: spontaneous fission and alpha interaction with a radionuclide. ORIGEN2 will produce a table that lists the production rate by each of these mechanisms for each radionuclide and also a total neutron production rate.

Wiles et al. (WILES 1986) used ORIGEN2 to calculate the neutron source strength and spatial distributions of selected spent fuel assemblies contained in a storage cask and used the DOT code to calculate the neutron dose rates on the outer surface of the cask. They found that the DOT predictions are a factor of 2 to 4 higher than the measured values and concluded that the neutron source strengths as predicted by ORIGEN2 may be too high. They recommend that one or two casks should be analyzed with a Monte Carlo code to determine whether (1) more accurate predictions are practical and (2) the DOT code is really as conservative as it appears.

6.5 SUMMARY

All of the information retrieved in this literature survey failed on at least one of the five qualifying standards: (1) important variables studied, (2) critical ranges covered, (3) proper measurements made, (4) precision requirements sufficient, and (5) QA procedures adequate. These standards were established to judge current experimental data's adequacy to validate ORIGEN2's use in regulatory and licensing procedures for a geologic repository. This failure is not a criticism of the previous research but indicates that the primary mission of the researcher and sponsor of the earlier studies was not to validate computer codes. The appropriate choice of measured radionuclides was limited by the availability of the analytical procedures and the equipment for handling highly radioactive spent fuel. Then, too, no one had performed a critical assessment to identify the important radionuclides that should be considered when disposing of highly radioactive nuclear waste in a geologic repository or for other parts of FWMS. Most of the previous research did not require the strict QA documentation that is now mandatory. In most cases, each researcher had to procure his own spent fuel for analysis and the proper documentation of the specific reactor power history was difficult to obtain. Finally, improvements have been made in some of the analytical procedures, increasing the sensitivity and precision of the radioisotopic analyses and reducing the error of measurement.

7. RECOMMENDATIONS

Each of the five qualifying standards above must be considered in order to adequately validate ORIGEN2-type codes for use in activities related to geologic repository systems. The recommendations presented in this section will emphasize the requirements necessary to quantify one of the more important indicators, the radionuclide inventory, as predicted by ORIGEN2. This indicator is most significant because it is the working unit used by ORIGEN2 to process and store data. Suggestions for the determination of the other three indicators will be given at the end of this section.

7.1 IMPORTANT VARIABLES AND RANGES

Since the utilities are showing increased interest in extending the burnups of their current and future fuel, a concerted effort is needed to compare ORIGEN2 predictions for the extended-burnup models with experimental data obtained from high-burnup BWR (>40,000-MWd/MTU) and PWR (60,000-MWd/MTU) fuels, the materials expected to make up the major inventory of a geologic repository. Although such data are not currently available, fuel assemblies approaching this burnup have been identified (Zion—55,000 MWd/MTU and Monticello—46,000 MWd/MTU). Additionally, radiochemical analyses of fuel at several different burnups must be made and compared with code predictions for both BWR and PWR fuels. Also, there is a critical need for the chemical determination of activation products generated in the various hardware pieces associated with spent fuel assemblies. Whether such material can be considered as low-level waste is questionable; hence, the resulting analyses will provide guidance in this area as well as yield data for the code validation study.

7.2 FUEL SELECTION

The primary requirement of any validation effort is to obtain spent fuel that has a well-characterized power history, including burnup, shutdown date, and fuel location. Both PWR and BWR fuels, as well as a variety of burnups and cladding materials (e.g., stainless steel, Zircaloy-2, and Zircaloy-4), must be considered. The original composition of the nuclear material is one of the important inputs into a radionuclide inventory code, and its availability is highly desirable. The initial impurity levels of each of the nonfuel materials should be determined if these values are not available from the vendors or suppliers. The criteria, as developed by the MCC (BARNER 1984) for the selection of approved testing materials in repository-related predisposal tests, should be used as input for the identification of LWR spent fuel to be included in an experimental program.

7.3 ANALYTICAL PROCEDURES AND ACCURACIES

The appropriate selection for the method of analysis is dependent on the specific characteristic to be determined, the accuracy desired, the quantity of material available, and the availability of

test equipment and facilities for handling nuclear material. One of the most important steps in any validation effort is the accurate determination of the radionuclide inventory.

Selection of the method to be used for analysis will also depend on the size of the sample, the radionuclide(s) to be determined, and the dissolution characteristics of the sample. Recommended methods for the analysis of LWR spent fuel include spark-source mass spectrometry (SSMS), TEIDMS, and radiochemical analyses (RCA). The SSMS will provide data for nearly all radionuclides from ^{7}Li to ^{252}Cf with an accuracy that varies with the specific radionuclide. This technique also has the advantage that it can be used on solid samples such as difficult-to-dissolve residues and hardware. The TEIDMS technique yields more precise results and has a range that covers many of the fission products and actinides. Because RCA may require some form of chemical separation as part of the procedure, this method will be used in special cases for difficult-to-analyze radioisotopes. Table 11 lists the expected accuracies and applicability of these methods for many elements and radioisotopes. Critical parameters for input to ORIGEN2 are the power history and the estimated burnup for each of the samples selected for analysis. One of the American Society for Testing and Materials' standard procedures (E 219-80, E 321-79, or E 244-79) is recommended for determining the burnup. Thus, any validation effort is not totally dependent on the utility to supply the necessary burnup data and power history.

7.4 SAMPLE SELECTION

An LWR fuel rod consists essentially of a stack of UO_2 pellets encapsulated within a metal tube. The fuel assemblies are constructed from a number of individual rods arranged in square arrays and held in place with tie plates, grid spacers, springs, and other hardware (RODDY 1986). The materials of construction are usually Zircaloy, stainless steel, and Inconel. As a minimum, a representative sample of each of the materials should be examined using SSMS. Extensive tests should be performed on the UO_2 fuel pellets and cladding, and duplicate experiments should be run for the most critical radioisotopes. Any residues remaining after dissolution from radiochemical separations should be analyzed via SSMS, followed by complete dissolution and final analysis.

Temperature excursions experienced by any fuel assembly cause serious problems in the correct determination of several of the radioisotopes. Many of the fission products (e.g., Cs, I, and Sb) are semivolatile at high temperatures and can migrate via diffusion to the cooler portions of a fuel rod. Barner (BARNER 1984) designates those fuels in which >10% of fission gas has been "released" as "high releasing" materials. He further states that fuel material restructuring, porosity agglomeration, and noble-metal fission product agglomeration occur concurrently in the central portion of high-releasing fuel material. Samples from UO_2 pellets originating from such fuel should be collected radially across the pellet and analyzed to determine whether fission product migration has occurred.

All reactor fuel rods exhibit variations in burnup along their length. This phenomenon is especially evident near the ends of a rod where the neutronic flux is measurably lower. Barner (BARNER 1985) has used ^{137}Cs to estimate the axial distribution of this important variable (Fig. 4). When selecting an appropriate sample for validation use, an analyst must be aware that the production of nuclides in a spent fuel rod is a function of the burnup, that the burnup is a function of the spatial fuel rod location in the reactor, and that the burnup experienced by a specific part of a fuel rod is a function of its axial location. Thus, a sample must be selected with considerable care.

Table 11. Accuracy and applicability of current analytical methods

Element or radioisotope	SSMS[a]	TEIDMS[b]	RCA[c]	Element or radioisotope	SSMS[a]	TEIDMS[b]	RCA[c]
Li	X			^{154}Sm	X	X	
Ti	X			^{153}Eu	X	X	
Cr	X			^{154}Eu	X	X	
Fe	X			^{155}Eu	X	X	
Ni	X			^{143}Nd	X	X	
Mo	X			^{144}Nd	X	X	
Th	X			^{145}Nd	X	X	
B	X			^{146}Nd	X	X	
V	X			^{148}Nd	X	X	
Mn	X			^{150}Nd	X	X	
Co	X			^{232}Th	X		
Nb	X			^{232}U			X
U[d]	X			^{233}U	X	X	
Pu		X		^{234}U		X	
^{79}Se	X			^{235}U		X	
^{90}Sr	X		X	^{236}U		X	
^{93}Zr	X			^{238}U		X	
^{94}Nb	X			^{237}Np	X		X
^{99}Tc	X	X		^{238}Pu		X	
^{106}Ru	X		X	^{239}Pu		X	
^{107}Pd	X		X	^{240}Pu		X	
^{125}Sb	X		X	^{241}Pu		X	
^{126}Sn	X			^{242}Pu		X	
^{129}I	X		X	^{244}Pu		X	
^{133}Cs	X			^{241}Am			X
^{134}Cs	X		X	^{242}Am			X
^{135}Cs	X			^{242m}Am			X
^{137}Cs	X		X	^{243}Am			X
^{144}Ce	X	X	X	^{242}Cm			X
^{147}Pm	X			^{243}Cm			X
^{147}Sm	X	X		^{244}Cm			X
^{148}Sm	X	X		^{245}Cm			X
^{149}Sm	X	X		^{246}Cm			X
^{150}Sm	X	X		^{247}Cm			X
^{151}Sm	X	X		^{248}Cm			X
^{152}Sm	X	X					

[a]Beta-gamma spark-source mass spectrometry; ±25%. Isotope dilution, in cases where an enriched spike is available, can be used to obtain ±10% precision. Chemical separations from elements with the same mass numbers may be necessary.

[b]Thermal-emission, isotopic dilution mass spectrometry; ±1 to 2%.

[c]Radiochemical analyses; ±5 to 10%.

[d]Uranium is often analyzed by extraction and a Davies-Gray potentiometric titration; ±1%.

7.5 CALCULATIONAL SUPPORT

Any experimental program must be closely coordinated with the results that are obtained from ORIGEN2 calculational predictions. The validation process is cyclical in nature and includes the (1) formulation of a model (ORIGEN2); (2) design of validation experiments, including establishment of validation goals; (3) collection of experimental data; (4) analysis of experimental results

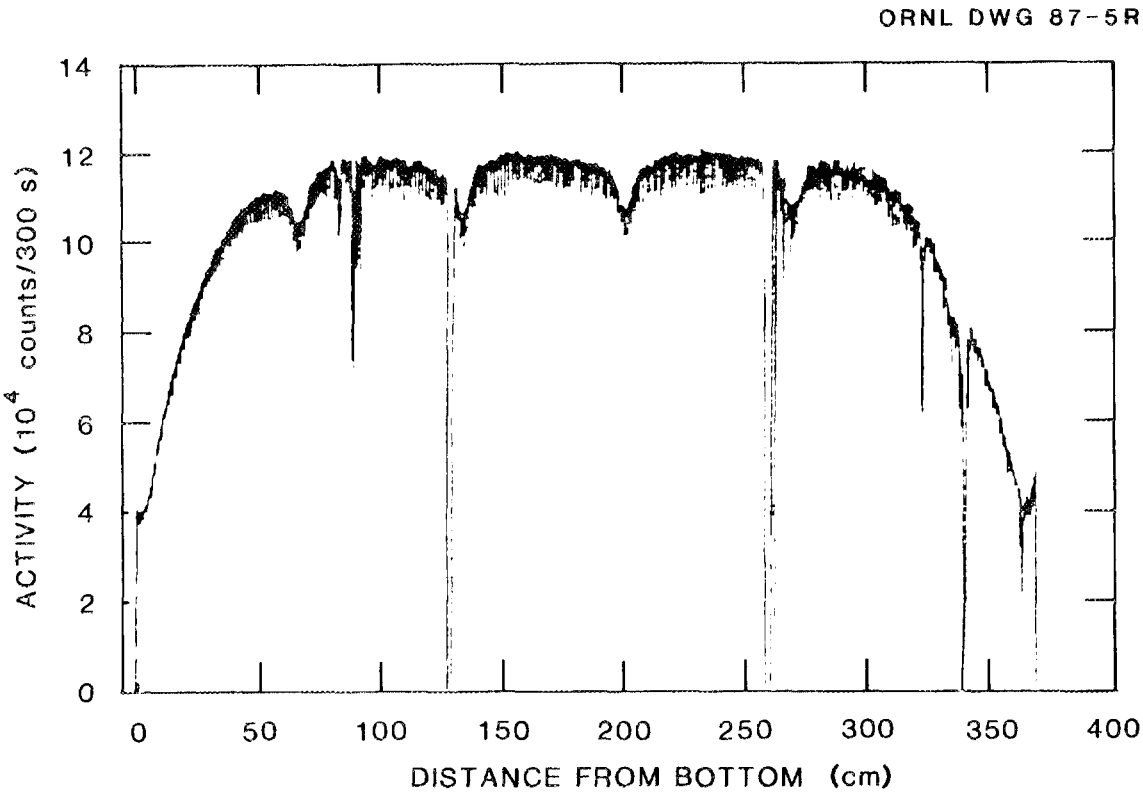

Fig. 4. Composite of ^{137}Cs gamma scans for rod N-9 from assembly BO-5, H. B. Robinson Unit 2. (Source: J. O. Barner, *Characterization of LWR Spent Fuel MCC-Approved Testing Material—ATM-101*, PNL-5109, Rev. 1, June 1985.).

compared with code predictions; and (5) revision of the code, if necessary. The need for revision and recycling will depend on whether the code is judged to have met the validation goals or criteria. These goals and criteria must be in place as early as possible in the validation process so as to permit the appropriate selection of experimental procedures with the necessary precision. If the results of the code are to be used for licensing purposes, input from the specific licensing organization should be used to assist in setting the validation criteria.

7.6 QUALITY ASSURANCE PROGRAM

All aspects of the experimental work must be performed under the guidance of a strict QA program. The basic requirements should be developed in accordance with those reported in 10 CFR 60 (NRC 1985a), applicable parts of Appendix B of 10 CFR 50 (NRC 1985b), and NQA-1 (ANSI 1983). Many elements of the sections in these three documents are not applicable because they were designed for geologic repositories, nuclear power plants, and fuel reprocessing plants, while the experimental program will deal with data generation. However, the information contained in the documents should be used as a basis for preparing a QA program plan.

7.7 ADDITIONAL MEASUREMENTS

Reliable estimates of decay heat and gamma and neutron radiation levels are important in the design of a repository, monitored retrieval storage facility (MRS), and transportation casks. As has been described previously in this review, ORIGEN2 can predict values for each of these.

7.7.1 Decay Heat

Two citations were identified that describe methods used to measure the decay heat of fuel assemblies: (1) the water boil-off spent fuel calorimeter system (CREER 1981) and (2) the water temperature rise spent-fuel system (JUDSON 1982). Results of tests indicated that the calorimeter system can be used to measure heat generation rates between 0.5 and 2.5 kW at an accuracy of about ±5%. Measurements of heat generation rates of ~0.1 kW were obtained within ±15%. The calculational accuracy estimated for the water temperature rise method was ~1.8% when used in the recirculation mode and ~1.2% when used in the static mode. Judson concluded that the total error associated with the thermal measurements was ~1% for the range of thermal outputs observed (360 to 940 W). However, large discrepancies (+12.8 and −16.2%) were noted when both calorimeter methods were used to measure two separate assemblies.

Although the accuracy stated for the water temperature rise method is more than adequate to validate ORIGEN2 for decay heat predictions, the poor agreement found for the measurements of the same assembly by the two modes casts doubt as to its accuracy. Even though ORIGEN2 predictions agree with measurements for these two methods, additional tests are required and other techniques may need to be considered to improve the precision of the experimental technique, especially if ORIGEN2 is to be validated for longer-decay fuel where the observed heat contents will be much lower.

7.7.2 Photon Emission Rates

The two studies (STRICKLER 1981, WILES 1986) that analyzed radiation emission rates (1) utilized ORIGEN2 to estimate the intensities of gamma emissions for various energy levels from spent fuel assemblies, (2) used this information as input to one of the versions of QAD to determine the exposure rates at several points, and (3) compared the calculated values with measured ones. The difficulty with this approach is the necessity of using two computer codes in series before predictions can be compared with measurements. Hence, the validity of each code is suspect if the comparisons are poor because one code may be correct while the second code is inadequate. The validity is not certifiable if the comparisons are excellent because the good agreement may be fortuitous. Also, more sophisticated general-purpose shielding codes are available than QAD. Before additional experimental measurements are generated, it is recommended that the available data be reanalyzed using one of the newer general-purpose Monte Carlo multigroup gamma-ray transport codes. The code, MORSE-CG (STRAKER 1970), would permit a direct comparison with values generated by QAD. Calculations should also be made with the shielding code MCNP, which was developed at Los Alamos National Laboratory (LANL 1979). Finally, the results obtained from MCNP should be compared with those produced by one of the versions of MORSE (EMMETT 1985).

Relatively simple gamma-ray measurements should be made using the well-characterized spent fuel that is available from the MCC. A small quantity (a few milligrams) of UO_2 powder would

serve as a point source, and a suitable detector and multichannel analyzer could be used to provide the necessary counting statistics for ^{134}Cs, ^{137}Cs, and ^{154}Eu. The results obtained from the measurement of these three isotopes would then be compared directly with ORIGEN2 predictions without the need for further calculations via another computer code. The necessary counting equipment is available at several laboratories, including ORNL.

7.7.3 Neutron Emission Rates

Although neutrons are a serious hazard in the disposal of spent fuel, only two citations (MCKINNON 1986a, WILES 1986) utilizing the measurement of dose rates were identified that attempted to quantify neutron emission rates. ORIGEN2 was used to calculate the absolute neutron source strength, and DOT was used to calculate the resulting dose rates. The authors believe that the source strengths provided by ORIGEN2 are the major reason for the differences between predicted and measured values. These measurements from a completely filled shipping cask are of maximum benefit, but the same problems that affect gamma-ray comparisons of predicted values with experimental values (i.e., multicode requirements to account for self-shielding and attenuation) also present difficulties for neutron comparisons. For fuel of similar decay time and burnup (10-year-old fuel of 33,000-MWd/MTU burnup), neutron emission rates are dramatically lower ($\sim 1 \times 10^7$) than those found for gamma rays. Hence, a much larger sample (1 g would provide 200 counts/s) and extensive shielding would be needed to perform a point-source measurement similar to that recommended for the gamma-ray studies. This experiment should be relatively easy and must be given high priority in a validation effort.

7.8 COST-BENEFIT ANALYSIS

Some differences will always be observed between a measured experimental value and one predicted by a computer code. These differences may be the result of poor analytical data, improper code methodology, or inadequate data base information. Of these three areas of uncertainty, the one that will be of long-term use to the project offices is the generation of improved analytical information. However, to properly justify the allocation of funds for an extensive experimental program that would answer this need, the accuracy requirements for each of the code's individual predictions as stated by the users of the code should be ascertained, the total costs to obtain the data should be estimated, and the total benefits that would accrue with the availability of a validated version of ORIGEN2 should be assessed.

A formal cost-benefit analysis of the validation effort for the ORIGEN2 computer code is not feasible at this time because much of the necessary data is not available. Nevertheless, it is possible to identify several benefits of a validation effort and to compare the estimated costs with those projected for the repository system.

Since ORIGEN2 provides source terms for many design studies, an enhancement of the code's predictions via validation with reliable experimental data may result in a substantial savings in constructional and operational costs. Although the actual amounts cannot be quantified, the incremental costs to the repository system will be discussed.

DOE (DOE 1986c) has performed an analysis of the life cycle cost of the total system for the civilian radioactivity waste management program for 32 cases. The cases are distinguished by the quantity of spent fuel generated [126,600 MTU for the 1985 Energy Information Administration

(EIA) midcase through the year 2020 vs 87,400 MTU for the EIA "no new orders" case]; repository startup dates (on time, 5-year delay, and 10-year delay); repository host rocks and location (basalt, tuff, and salt in Deaf Smith County, Texas, for the first repository, and tuff and salt, a generic "high-cost" crystalline-rock site, and a generic "low-cost" crystalline-rock site for the secondary repository); and the authorized vs the improved-performance system. All cases included the costs of accepting 16,000 canisters of defense high-level waste. The principal findings of their analysis were as follows: (1) the total-system cost for the authorized system in the reference case ranged from $23.6 billion to $32.6 billion (in constant 1985 dollars), depending on the repository-site combination; (2) the total-system cost for the improved-performance system in the reference case ranged from $26.2 billion to $34.0 billion, depending on the repository-site combination; (3) the total-system costs for the "no new orders" case are 8 to 12% ($2.0 billion to $4.9 billion) lower than the total-system cost for the midcase projections; and (4) the maximum total-system cost is estimated to be $40.5 billion. These results are summarized in Tables 12 and 13 for the authorized and improved-performance systems, respectively. Note that the second system includes an MRS facility. The development and evaluation cost columns cover all the siting, design development, testing, regulatory, and institutional activities associated with the repositories, MRS, and transportation.

If a completely validated version of ORIGEN2 results in an expenditure of $1 million, the incremental cost would be less than 0.005% of the total cost for the authorized system in the reference case. If the validation cost were applied only to the transportation cost estimates ($2 billion), the incremental increase would be just 0.1%. The resulting benefits in greater ORIGEN2 accuracy would be reduced design margins and lower weights (and thus lower construction and hauling costs) for all shipping casks.

Other benefits of a successful validation effort, which cannot be quantified, would be to ensure that the methodology is the most current, the data bases are up to date, and the computer program contains no major flaws. Any of these deficiencies would have a serious impact on the utilization of the code in a licensing hearing or review. ORIGEN2 output is also used for the generation of source terms for shielding analysis, criticality analysis, decay heat predictions, and dose-to-man estimation; each of these areas would benefit directly from a proper validation of the code.

Table 12. Summary of total-system life-cycle cost estimates for the authorized repository system

(Billions of 1985 dollars)

Repository-site combination	D&E[a]	Transportation	Repository First	Repository Second	Repository Subtotal	At-reactor storage	Total[b]
Reference Cases (EIA Midcase)							
Basalt/crystalline (high)	9.4	2.1	11.3	9.5	20.8		32.3
Basalt/salt	9.4	2.2	11.3	7.1	18.4		29.9
Basalt/tuff	9.4	2.5	11.3	5.6	16.9		28.8
Basalt/crystalline (low)	9.4	1.8	11.3	6.5	17.8		29.0
Salt/crystalline (high)	9.4	1.7	7.9	9.5	17.4		28.5
Salt/salt	9.0	1.9	7.9	7.1	15.0		26.0
Salt/crystalline (low)	9.4	1.5	7.9	6.5	14.4		25.3
Tuff/crystalline (high)	9.4	2.0	6.0	9.5	15.5		26.9
Tuff/salt	9.0	2.1	6.0	7.1	13.1		24.3
Tuff/crystalline (low)	9.4	1.8	6.0	6.5	12.5		23.6
Sensitivity Cases: (1) EIA "no new orders" case							
Basalt/crystalline (high)	9.3	1.6	9.9	7.7	17.6		28.4
Tuff/crystalline (low)	9.3	1.4	5.3	5.3	10.6		21.3
(2) 5-year repository delay (EIA midcase)							
Basalt/crystalline (high)	10.1	2.1	11.3	9.5	20.8	2.7	35.7
Tuff/crystalline (low)	10.1	1.8	6.0	6.5	12.5	2.7	27.0
(3) 10-year repository delay (EIA midcase)							
Basalt/crystalline (high)	10.8	2.0	11.3	9.9	21.2	4.9	38.9
Tuff/crystalline (low)	10.8	1.8	6.1	6.8	12.9	4.9	30.4

[a]D&E = development and evaluation.

[b]Cost categories may not add to total because of independent rounding.

Source: U.S. Department of Energy, *Analysis of the Total System Life Cycle Cost for the Civilian Radioactive Waste Management Program, Volume 1: The Analysis and Its Results,* DOE/RW-0047, April 1986.

Table 13. Summary of total-system life-cycle cost estimates for the improved-performance system[a]

(Billions of 1985 dollars)

Repository-site combination	D&E[a]	Transportation	Repository First	Repository Second	Repository Subtotal	MRS	At-reactor storage	Total[b]
Reference cases (EIA midcase)								
Basalt/crystalline (high)	9.5	1.9	10.2	9.5	19.7	2.8		34.0
Basalt/salt	9.5	2.1	10.2	7.1	17.3	2.8		31.6
Basalt/tuff	9.5	2.3	10.2	5.6	15.8	2.8		30.4
Basalt/crystalline (low)	9.5	1.8	10.2	6.5	16.7	2.8		30.8
Salt/crystalline (high)	9.5	1.9	6.9	9.5	16.4	2.8		30.6
Salt/salt	9.2	2.0	6.9	7.1	14.0	2.8		27.9
Salt/crystalline (low)	9.5	1.7	6.9	6.5	13.4	2.8		27.4
Tuff/crystalline (high)	9.5	2.2	5.4	9.5	14.9	2.9		29.4
Tuff/salt	9.2	2.3	5.4	7.1	12.5	2.9		26.7
Tuff/crystalline (low)	9.5	2.0	5.4	6.5	11.9	2.9		26.2
Sensitivity cases: (1) EIA "no new orders" cases								
Basalt/crystalline (high)	9.4	1.6	10.2	6.8	17.0	2.8		30.9
Tuff/crystalline (low)	9.4	1.7	5.3	4.8	10.1	2.9		24.2
(2) 5-year repository delay (EIA midcase)								
Basalt/crystalline (high)	10.3	2.0	10.2	9.5	19.7	3.1	2.1	37.2
Tuff/crystalline (low)	10.3	2.0	5.4	6.5	11.9	3.1	2.1	29.4
(3) 10-year repository delay (EIA midcase)								
Basalt/crystalline (high)	10.9	2.0	10.5	9.9	20.4	3.3	3.9	40.5
Tuff/crystalline (low)	10.9	2.1	5.5	6.8	12.3	3.3	3.9	32.6

[a]D&E = development and evaluation.

[b]Cost categories may not add to total because of independent rounding.

Source: U.S. Department of Energy, *Analysis of the Total System Life Cycle Cost for the Civilian Radioactive Waste Management Program, Volume 1: The Analysis and Its Results,* DOE/RW-0047, April 1986.

8. REFERENCES

ADAMS 1984. J. P. Adams and B. R. Dabell, *Characteristics of UO_2-Zr Fuel Rods Irradiated in the BR3 Reactor; Draft Preliminary Report for Comment,* EG&G Idaho, Inc., November 1984.

ANSI 1983. The American Society of Mechanical Engineers, *Quality Assurance Program Requirements for Nuclear Facilities,* ANSI/ASME NQA-1, 1983 Ed.

ASTM 1985a. American Society for Testing and Materials, *Atom Percent Fission in Uranium Fuel (Radiochemical Method),* E 219-80 (Reapproved 1985).

ASTM 1985b. American Society for Testing and Materials, *Atomic Percent Fission in Uranium and Plutonium Fuel (Neodymium-148 Method),* E 321-79 (Reapproved 1985).

ASTM 1985c. American Society for Testing and Materials, *Atomic Percent Fission in Uranium and Plutonium Fuel (Mass Spectrometric Method),* E 244-79 (Reapproved 1985).

ATKIN 1980. S. D. Atkin, *Destructive Examination of 3-Cycle LWR Rods from Turkey Point Unit 3 for the Climax-Spent Fuel Test,* Hanford Engineering Development Laboratory, TC-1867, December 1980.

ATKIN 1981. S. D. Atkin, *Destructive Examination of 3-Cycle LWR Fuel Rods from Turkey Point Unit 3 for the Climax-Spent Fuel Test,* Hanford Engineering Development Laboratory, HEDL-TME 80-89, June 1981.

B&W 1979. Babcock & Wilcox and Duke Power Company, *Post-Irradiation Examination of Oconee 1 Fuel; Cycle 1 Destructive Test Phase,* COD-4711-23 (BAW-1533), 1979.

BAIN 1983. G. M. Bain and W. A. McInteer, *Data Package for High Burnup Fuel Samples,* Babcock & Wilcox Company, LRC 4063, January 1983.

BALFOUR 1981. M. G. Balfour, G. R. Schmidt, M. F. Muenks, P. G. Isaac, and G. H. Eng, *Surry Unit 2 End of Cycle 4 Onsite Fuel Examination—Reduced Data and Operating History,* Westinghouse Electric Corporation, WCAP-9839, February 1981.

BALFOUR 1982. M. G. Balfour, W. C. Chubb, and R. F. Boyle, *BR-3 High Burnup Fuel Rod Hot Cell Program, Volume 1 and 2: Data Summary,* Westinghouse Electric Corporation, WCAP-10238, Volume 1 (DOE/ET 34073-1), November 1982.

BARNER 1984. J. O. Barner, *LWR Spent Fuel Approved Testing Materials for Radionuclide Release Studies,* Pacific Northwest Laboratory, PNL-4686, January 1984.

BARNER 1985. J. O. Barner, *Characterization of LWR Spent Fuel MCC-Approved Testing Material—ATM-101,* Pacific Northwest Laboratory, PNL-5109, Rev. 1, June 1985.

BAUMGARTNER 1984. J. A. Baumgartner, *BWR Fuel Bundle Extended Burnup Program; Final Report,* General Electric Company and Northern States Power Company, DOE/ET/34031-18, December 1984.

BELL 1973. M. J. Bell, *ORIGEN—The ORNL Isotope Generation and Depletion Code,* Oak Ridge National Laboratory, ORNL-4628, 1973.

CADELLI 1984. N. Cadelli and M. Lippens, "Post-Irradiation Examination of Plutonium-Bearing Fuel Elements Irradiated in LWR Power Plants—A European Community Campaign," pp. 182-207 in *Improved Utilization of Water Reactor Fuel with Special Emphasis on Extended Burnups and Plutonium Recycling* (Proceedings of IAEA Specialists' Meeting, Mol, Belgium, May 7-11, 1984); CONF-8405285.

CAMPBELL 1976a. D. O. Campbell et al., pp. 6-16 in *LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period January 1 to March 31, 1976,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-5447, May 1976.

CAMPBELL 1976b. D. O. Campbell et al., pp. 10-31 in *LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period April 1 to June 30, 1976,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-5547, July 1976.

CAMPBELL 1976c. D. O. Campbell, pp. 23-38 in *LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period July 1 to September 30, 1976,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-5660, November 1976.

CAMPBELL 1977a. D. O. Campbell et al., pp. 13-40 in *LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period October 1 to December 31, 1976,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-5760, February 1977.

CAMPBELL 1977b. D. O. Campbell et al., pp. 2-6 through 2-25 in *LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period January 1 to March 31, 1977,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-5864, May 1977.

CAMPBELL 1977c. D. O. Campbell et al., pp. 2-15 through 2-32 in *LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period April 1 to June 30, 1977,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-5987, August 1977.

CAMPBELL 1977d. D. O. Campbell et al., pp. 2-13 through 2-41 in *Alternate Fuel Cycle Technologies Program Quarterly Report for Period July 1 to September 30, 1977,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-6076, November 1977.

CAMPBELL 1978a. D. O. Campbell et al., pp. 2-8 through 2-16 in *Alternate Fuel Cycle Technologies/Thorium Fuel Cycle Technology Programs Quarterly Report for Period October 1 to December 31, 1977,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-6206, February 1978.

CAMPBELL 1978b. D. O. Campbell et al., pp. 2-6 through 2-9 in *Alternate Fuel Cycle Technologies/Thorium Fuel Cycle Technology Programs Quarterly Report for Period January 1 to March 31, 1978,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-6307, May 1978.

CAMPBELL 1978c. D. O. Campbell et al., pp. 2-6 through 2-9 in *Alternate Fuel Cycle Technologies/Thorium Fuel Cycle Technology Programs Quarterly Report for Period April 1 to June 30, 1978,* B. L. Vondra, Program Manager, Oak Ridge National Laboratory, ORNL/TM-6429, August 1978.

CAMPBELL 1981. D. O. Campbell and W. L. Pattison, *The Effect of Heat Treatments on Tritium Volatility and the Behavior of Other Radioactive Constituents in Irradiated Fuel,* Oak Ridge National Laboratory, ORNL/TM-7326, April 1981.

CAMPBELL 1986. D. O. Campbell, Oak Ridge National Laboratory, personal communication of unpublished data to J. C. Mailen, Oak Ridge National Laboratory, March 1986.

COLEMAN 1980. T. A. Coleman, *Qualification of the B&W Mark B Fuel Assembly for High Burnup, Fourth Semi-Annual Progress Report: January-June 1980,* Babcock and Wilcox Company, DOE/ET/34212-28 (BAW-1546-4), September 1980.

COLEMAN 1981a. T. A. Coleman, *Qualification of the B&W Mark B Fuel Assembly for High Burnup, Fifth Semi-Annual Progress Report: July-December 1980,* Babcock and Wilcox Company, DOE/ET/34212-31 (BAW-1546-5), April 1981.

COLEMAN 1981b. T. A. Coleman, *Qualification of the B&W Mark B Fuel Assembly for High Burnup, Sixth Semi-Annual Progress Report: January-June 1981,* Babcock and Wilcox Company, DOE/ET/34212-32 (BAW-1546-6), October 1981.

COLEMAN 1982. T. A. Coleman, *Qualification of the B&W Mark B Fuel Assembly for High Burnup, Seventh Semi-Annual Progress Report: July-December 1981,* Babcock and Wilcox Company, DOE/ET/34212-33 (BAW-1546-7), May 1982.

CREER 1981. J. M. Creer and J. W. Shupe, Jr., *Development of a Water Boil-Off Spent Fuel Calorimeter System,* Pacific Northwest Laboratory, PNL-3434, May 1981.

CREER 1984. J. M. Creer, *Decay Heat and Heat Transfer Predictions for Spent Fuel Storage Systems,* Pacific Northwest Laboratory, PNL-SA-12658, October 1984.

CROFF 1983. A. G. Croff, "ORIGEN2: A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials," *Nucl. Technol. 62,* 335 (1983).

DAVIS 1980. R. B. Davis, *Data Report for the Nondestructive Examination of Turkey Point Spent Fuel Assemblies B02, B03, B17, B41, and B43,* Hanford Engineering Development Laboratory, HEDL-TME 79-68, April 1980.

DAVIS 1981. R. B. Davis and V. Pasupathi, *Data Summary Report for the Destructive Examination of Rods G7, G9, J8, I9, and H6 from Turkey Point Fuel Assembly B17,* Hanford Engineering Development Laboratory, HEDL-TME 80-85, April 1981.

DOE 1986a. U.S. Department of Energy, Code of Federal Regulations, Standard Contract for Disposal of Spent Nuclear Fuel and/or High-Level Radioactive Waste, 10 CFR 961, pp. 539–56 (January 1986).

DOE 1986b. U.S. Department of Energy, *Integrated Data Base for 1986: Spent Fuel and Radioactive Waste Inventories, Projections, and Characteristics,* DOE/RW-0006, Rev. 2, September 1986.

DOE 1986c. U.S. Department of Energy, *Analysis of the Total System Life Cycle Cost for the Civilian Radioactive Waste Management Program, Volume I: The Analysis and Its Results,* DOE/RW-0047, April 1986.

EMMETT 1985. M. B. Emmett, *MORSE-CGA: A Monte Carlo Radiation Transport Code with Array Geometry Capability,* ORNL-6174, April 1985.

EPA 1985. U.S. Environmental Protection Agency, *"Environmental Standards for the Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Wastes: Final Rule," 40 CFR 191 (Code of Federal Regulations)*; Fed. Regist. *50* (No. 182), Sept. 19, 1985.

FISCHER 1983. U. Fischer and H. W. Wiese, *Improved and Consistent Determination of the Nuclear Inventory of Spent PWR Fuel on the Basis of Cell-Burnup Methods Using KORIGEN,* Karlsruhe Nuclear Research Center, ORNL-tr-5043; translation of KfK-3014, January 1983.

GOODE 1978. J. H. Goode and R. G. Stacy, *Head-End Reprocessing Studies with H. B. Robinson-2 Fuel,* Oak Ridge National Laboratory, ORNL/TM-6037, June 1978.

GOODE 1980a. J. H. Goode, R. G. Stacy, and V. C. A. Vaughen, *Head-End Studies of H. B. Robinson-2 Fuel: II. Parametric Voloxidation Studies,* Oak Ridge National Laboratory, ORNL/TM-6888, May 1980.

GOODE 1980b. J. H. Goode, R. G. Stacy, and V. C. A. Vaughen, *Comparison Studies of Head-End Reprocessing Using Three LWR Fuels,* Oak Ridge National Laboratory, ORNL/TM-7103, June 1980.

HEEB 1979. C. M. Heeb et al., *Analysis of Alternative Light Water Reactor (LWR) Fuel Cycles,* Pacific Northwest Laboratory, PNL-2792, December 1979.

HSUE 1979. S. T. Hsue et al., *Passive Neutron Assay of Irradiated Nuclear Fuels,* Los Alamos Scientific Laboratory, LA-7645-MS (ISPO-43) (February 1979).

JOHNSON 1978. "Light Water Reactor Fuel Reprocessing: Dissolution Studies of Voloxidized Fuel," paper presented at the ANS Topical Meeting on the Back End of the LWR Fuel Cycle, Savannah, Ga., Mar. 19-23, 1978; Savannah River Laboratory, DP-MS-77-77, March 1978.

JOHNSON 1980. D. R. Johnson and J. A. Stone, *Light Water Reactor Fuel Reprocessing: Dissolution Studies of Voloxidized and Nonvoloxidized Fuel,* Savannah River Laboratory, DP-1520, April 1980.

JUDSON 1982. *In-Plant Test Measurements for Spent Fuel Storage at Morris Operation. Volume 3: Fuel Bundle Heat Generation Rates,* General Electric Co., NEDG-24922-3, February 1982.

LANGSTAFF 1982. D. C. Langstaff, W. J. Bailery, A. B. Johnson, Jr., M. P. Landow, V. Pasupathi, and R. W. Klingensmith, *Examination of Stainless Steel-Clad Connecticut Yankee Fuel Assembly S004 After Storage in Borated Water,* Pacific Northwest Laboratory, PNL-3828, September 1982.

LANL 1979. Los Alamos National Laboratory, *MCNP: A General Monte Carlo Code for Neutron and Photon Transport,* LA-7396-M (Revised), November 1979.

LEE 1979. D. M. Lee et al., *A New Approach to the Examination of LWR Irradiated Fuel Assemblies Using Simple Gas Sample Chamber Techniques,* Los Alamos Scientific Laboratory, LA-7655-MS (ISPO-48) (March 1979).

LEE 1985. D. M. Lee, "Passive and Active Neutron Methods for Assay/Verification of Spent Fuel," pp. 14-1 through 14-21 in *Proceedings: International Training Course on Implementation of State Systems of Accounting for and Control of Nuclear Materials, Santa Fe, New Mexico and San Clemente, California,* June 3-21, 1985, LA-10672-C, June 1986.

LOWRY 1982. L. M. Lowry and V. Pasupathi, *Postirradiation Examination of Fuel Rods from the Quad Cities-1 Reactor,* Battelle Columbus Laboratories, BCL-585-82-3, August 1982.

MacDONALD 1975. P. E. MacDonald, Aerojet Nuclear Company, letter to A. P. Malinauskas, Holifield (Oak Ridge) National Laboratory, July 30, 1975.

MALENFANT 1967. R. E. Malenfant, *QAD: A Series of Point-Kernel General-Purpose Shielding Programs,* Los Alamos Scientific Laboratory, LA-3573, April 1967.

McELROY 1985. J. L. McElroy, Pacific Northwest Laboratory, letter report to Jerry D. White, Richland Operations Office, Richland, Wash., Nov. 7, 1985. McKINNON 1986a. M. A. McKinnon et al., *Monticello BWR Spent Fuel Assembly Decay Heat Predictions and Measurements,* Pacific Northwest Laboratory, PNL-5799, June 1986.

McKINNON 1986b. M. A. McKinnon et al., *BWR Spent Fuel Storage Cask Performance Test, Volume I, Pre- and Post-Test Decay Heat, Heat Transfer, and Shielding Analyses,* Pacific Northwest Laboratory, PNL-5777, Vol I, June 1986.

MONTGOMERY 1976. M. H. Montgomery, Babcock & Wilcox, letter to B. L. Vondra, Oak Ridge National Laboratory, Oct. 13, 1976.

MONTGOMERY 1978. M. H. Montgomery, Babcock & Wilcox, letter report to B. L. Vondra, Oak Ridge National Laboratory, Apr. 5, 1978.

MOSS 1980. C. E. Moss et al., *Gross Gamma-Ray Measurements of Light Water Reactor Spent-Fuel Assemblies in Underwater Storage Arrays,* Los Alamos Scientific Laboratory, LA-8447 (ISPO-132) (December 1980).

MOSS 1982. H. D. Moss, *Extended Fuel Burnup Demonstration Program Semiannual Technical Progress Report for the Period from July 1981 through December 1981,* Westinghouse Electric Corporation and Virginia Electric and Power Company, WCAP-10199 (DOE/ET 34014-5), December 1982.

NEELY 1982. G. J. Neely and F. Schmittroth, *Decay-Heat and Gamma-Dose Rate Prediction Capability in Spent LWR Fuel,* Hanford Engineering Development Laboratory, HEDL-TC-1787, August 1982.

NICHOLSON 1981. N. Nicholson et al., *Irradiated Fuel Examination Using the Cerenkov Technique,* Los Alamos National Laboratory, LA-8767-MS (March 1981).

NODVIK 1966. R. J. Nodvik, *Evaluation of Mass Spectrometric and Radiochemical Analyses of Yankee Core I Spent Fuel,* Westinghouse Electric Corporation, WCAP-6068, March 1966.

NOTZ 1986. K. J. Notz, Oak Ridge National Laboratory, personal communication to J. W. Roddy, Oak Ridge National Laboratory, 1986.

NRC 1985a. U.S. Nuclear Regulatory Commission, "Disposal of High-Level Radioactive Wastes in Geologic Repositories," 10 CFR 60 (*Code of Federal Regulations*), pp. 563-617, January 1985.

NRC 1985b. U.S. Nuclear Regulatory Commission, "Quality Assurance Criteria for Nuclear Power Plants and Fuel Reprocessing Plants," 10 CFR 50, Appendix B (*Code of Federal Regulations*), pp. 475-78, January 1985.

NWPA 1983. Nuclear Waste Policy Act of 1982 (NWPA), Pub L. 87-425, 42 U.S.C. 10101 (Jan. 7, 1983).

ORNL 1977. Oak Ridge National Laboratory, *QAD-CG: A Combinatorial Geometry Version of QAD-P5A, a Point-Kernel Code for Neutron and Gamma-Ray Shielding Calculations,* CC-307, Radiation Shielding Information Center, Oak Ridge National Laboratory.

PASUPATHI 1982. V. Pasupathi, L. M. Lowry, and M. C. Matthews, *Final Report on Postirradiation Examination of Fuel Rods from the Monticello Reactor to General Electric Company,* Battelle Columbus Laboratories, BCL-585-82-2, June 1982.

PHILLIPS 1980. J. R. Phillips et al., *Application of Nondestructive Gamma-Ray and Neutron Techniques for the Safeguarding of Irradiated Fuel Materials,* Los Alamos Scientific Laboratory, LA-8212 (ISPO-77) (May 1980).

PHILLIPS 1985. J. R. Phillips, "Passive Gamma-Ray Methods for Assay/Verification of Spent Fuel," pp. 13-1 through 13-14 in *Proceedings: International Training Course on Implementation of State Systems of Accounting for and Control of Nuclear Materials, Santa Fe, New Mexico and San Clemente, California,* June 3-21, 1985, LA-10672-C, June 1986.

RHOADES 1982. W. A. Rhoades and R. L. Childs, *An Updated Version of the DOT-4 One- and Two-Dimensional Neutron/Photon Transport Code,* Oak Ridge National Laboratory, ORNL-5851, July 1982.

RODDY 1986. J. W. Roddy, H. C. Claiborne, R. C. Ashline, P. J. Johnson, and B. T. Rhyne, *Physical and Decay Characteristics of Commercial LWR Spent Fuel,* Oak Ridge National Laboratory, ORNL/TM-9591/V1&R1, January 1986.

RYMAN 1982. J. C. Ryman, O. W. Hermann, C. C. Webster, and C. V. Parks, *Fuel Inventory and Afterheat Power Studies of Uranium-Fueled Pressurized Water Reactor Fuel Assemblies Using the SAS2 and ORIGEN-S Modules of Scale with an ENDF/B-V Updated Cross Section Library,* Oak Ridge National Laboratory, NUREG/CR-2397; ORNL/CSD-90, September 1982.

SCHMITTROTH 1980. F. Schmittroth, G. J. Neely, and J. C. Krogness, *A Comparison of Measured and Calculated Decay Heat for Spent Fuel Near 2.5 Years Cooling Time,* Hanford Engineering Development Laboratory, TC-1759, August 1980.

SCHMITTROTH 1984a. F. Schmittroth, *ORIGEN2 Calculations of PWR Spent Fuel Decay Heat Compared with Calorimeter Data,* Hanford Engineering Development Laboratory, HEDL-TME 83-32, January 1984.

SCHMITTROTH 1984b. F. Schmittroth, "ORIGEN2 Calculations of PWR Spent Fuel Decay Heat Compared with Calorimeter Measurements," pp. 2-69 through 2-74 in *Proceedings of Fuel Reprocessing and Waste Management, Jackson, Wyoming,* August 1984.

STRAKER 1970. E. A. Straker, P. N. Stevens, D. C. Irving, and V. R. Cain, *MORSE Code: A Multigroup Neutron and Gamma-Ray Monte Carlo Transport Code, Oak Ridge National Laboratory,* ORNL-4585, September 1970.

STRICKLER 1981. H. R. Strickler and K. J. Eger, *In-Plant Test Measurements for Spent Fuel Storage at Morris Operation,* General Electric Co., NEDG-24922-2, September 1981.

WICHNER 1983. R. P. Wichner, Oak Ridge National Laboratory, memorandum to T. S. Kress, Oak Ridge National Laboratory, and L. Chan, U.S. Nuclear Regulatory Commission, "ORIGEN2 Description and Verification Status," July 25, 1983.

WILES 1986. L. E. Wiles et al., *BWR Spent Fuel Storage Cask Performance Test, Volume II, Pre- and Post-Test Decay Heat, Heat Transfer, and Shielding Analyses,* Pacific Northwest Laboratory, PNL-5777, Vol. II, June 1986.

WILSON 1983. W. B. Wilson, R. J. LaBauve, and T. R. England, "Calculations of Spent Thermal Reactor Fuel Nuclide Inventories and Comparisons with Measurements," pp. 8-1 through 8-27 in *Proceedings: Thermal Reactor Benchmark Calculations, Techniques, Results, and Applications, Upton, New York, May 17-18, 1982,* NP-2855, February 1983.

YOKOYAMA 1981. T. Yokoyama et al., "Measurement and Analysis of Neutron Emission Rate for Irradiated BWR Fuel," *J. Nucl. Sci. Technol.* 18(4), 249 (1981).

Appendix A. FUEL COMPOSITION DATA

The fuel composition data accumulated during this review are assembled here. Some of these data are scattered throughout documents that are not easily obtainable; the results of one series of studies available only piecemeal in progress reports (CAMPBELL 1976a,b,c; 1977a,b,c,d; 1978a,b,c) have been summarized in Tables A.1 and A.2. The analytical methods used in generating the data in this table were as follows:

for ^{129}I—volatilization from the fuel solution, trapping in sodium hydroxide solution, purification by extraction, and neutron activation;

for ^{14}C—volatilization from the fuel solution, trapping in sodium hydroxide solution, and beta counting;

for ^{3}H—sampling of fuel solution by distillation and counting;

for $^{238,239,240}Pu$—thenoyltrifluoroacetone extraction, alpha counting, and alpha pulse;

for $^{238}Pu + {}^{241}Am$, $^{239,240}Pu$, ^{242}Cm, ^{244}Cm—gross alpha and alpha pulse;

for ^{106}Ru, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{125}Sb, ^{154}Eu, ^{95}Zr—gamma spectrometry;

for $^{90}Sr/^{90}Y$—chemical separation and beta counting;

for total uranium—Davies-Gray, chemical separation and mass analyses, colorimetric, and isotopic dilution mass analyses;

for $^{234,235,236,237,238}U$, $^{238,239,240,241,242}Pu$—mass spectrometry; and

for $^{242,243}Am$, $^{245,246,247,248}Cm$—ion-exchange separation and mass spectrometry.

The requirements for validation of ORIGEN were not the primary concern of the fuel reprocessing studies. In some cases, the source of the fuel studied is not completely specified. In at least one case, the date for the radiochemical counting of the isotopes was not given (this was retrieved from the original notebooks). Analyses of several isotopes, where quoted, are of doubtful use. About 50% of the tritium in the fuel migrates into the Zircaloy cladding (GOODE 1978, 1980a,b; CAMPBELL 1986); in most studies, the quantity of tritium in the cladding was not determined. A significant fraction of the noble metals (ruthenium, palladium, etc.) does not dissolve in nitric acid, and determination of the fraction of the heavy metals in the residue is quite difficult. The finely divided residue is difficult to recover and must be accurately weighed. Data for ^{14}C are often not available and, where given without information on the nitrogen content of the fuel, are of marginal value.

Experimental data in which the reported tritium and ruthenium values do not take into account the quantities of material in the cladding (tritium) or in the residue (ruthenium) are excluded from this appendix.

Table A.3 lists the literature sources for information on the various elements. The uranium and plutonium isotopes were generally determined by TEIDMS; the results, which are usually given as isotopic ratios, should have accuracies of ± 1 to 2%. Americium and curium can be determined either by mass spectrometry or by RCA. The remaining isotopes are usually determined by radiochemical means and, with some exceptions, should have accuracies of between 5 and 10%. Exceptions are ^{129}I, tritium (^{3}H), ^{106}Ru, and ^{14}C, where recovery and/or the radiochemical methods are complex. The isotopic concentrations of uranium and plutonium have been determined in a

Table A.1. Analyses of selected isotopes for H. B. Robinson spent fuel assembly BO-5[a]

Radioisotope	Concentration (Ci/MTU)[b]		
	Loose fuel fragments	Center of rod A-1	Center of rod O-14
^{3}H	2.57E+02	3.08E+02	
^{14}C	2.70E−01	2.40E−01	
^{90}Sr	5.31E+04		6.81E+04
^{95}Zr	≤2.05E+03		
^{106}Ru	1.22E+05	1.34E+05	
^{129}I	2.77E−02	2.63E−02	2.98E−02
^{125}Sb	≤3.46E+03	≤5.02E+03	≤3.16E+03
^{134}Cs	6.58E+04	6.90E+04	7.17E+04
^{137}Cs	8.52E+04	9.05E+04	9.48E+04
^{144}Ce	1.75E+05	1.83E+05	1.86E+05
^{154}Eu	≤3.62E+03		4.04E+03
^{234}U	1.86E−04		
^{235}U	2.99E−06		
^{236}U	4.76E−05		
^{238}U	7.06E−05		
^{238}Pu	2.41E+03	2.80E+03	2.84E+03
^{239}Pu	3.14E+02	} 9.73E+02	} 8.49E+02
^{240}Pu	5.14E+02		
^{241}Pu	1.15E+05		
^{242}Pu	1.88E+00		
^{241}Am	3.41E+02	1.54E+02	3.04E+02
^{242}Am	4.02E+05		
^{243}Am	1.18E+01		
^{242}Cm	2.67E+03	2.38E+03	2.49E+03
^{243}Cm	1.68E+01		
^{244}Cm	1.97E+03	2.09E+03	2.22E+03
^{245}Cm	2.04E−01		
^{246}Cm	3.74E−02		
^{247}Cm	1.00E−07		
^{248}Cm	1.14E−06		

[a]H. B. Robinson fuel assembly BO-5; initial loading, March 1971; discharged, May 1979; average burnup estimated at 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.)

[b]MTU = metric tons of uranium.

Source: CAMPBELL 1976a,b,c; 1977a,b,c,d; 1978a,b,c.

large number of cases to allow burnup calculations; in most cases, however, the traceability of the samples does not meet current QA standards. Data for americium and curium are much more sparse, and information on the higher isotopes of americium and curium is limited to a single paper (CAMPBELL 1976c). Isotopes for which only a few data are available include those of Nd, Eu, Sr, Sb, Zr, Ru, Np, Tc, Se, Sn, and Xe. Many other isotopes have never been determined, or at least the results were not located for this review.

Table A.2. Radionuclide analyses of spent fuel from various light-water reactors

Reactor[a]	Rod	Radioisotope (Ci/MTU)[b]			
		^{129}I	^{14}C	^{244}Cm	^{3}H
H. B. Robinson	P-5	7.95E−03	1.50E−01	2.11E+02	
H. B. Robinson	H-10, P-4, P-8	2.49E−02	4.20E−01		
H. B. Robinson	J-6	2.12E−02	3.70E−01		
H. B. Robinson	0-14	2.33E−02	3.20E−01		
H. B. Robinson	Unknown	2.07E−02	3.62E−01		
H. B. Robinson	J-6	1.92E−02	4.00E−01		
H. B. Robinson	J-6	3.60E−02	3.60E−01		
H. B. Robinson	J-10				3.80E+02
Oconee-1[c]	Unknown	6.80E−03	1.06E−01		
Oconee-1[d]	32028	1.73E−02	2.00E−01		
Big Rock Point	CM-0042	1.13E−02	1.31E−01		

[a]H. B. Robinson fuel assembly BO-5; initial loading, March 1971; discharged, May 1979; average burnup estimated at 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.)

[b]MTU = metric tons of uranium.

[c]Cycle 1.

[d]Cycle 2.

Source: CAMPBELL 1976a,b,c; 1977a,b,c,d; 1978a,b,c.

A.1. COMPARISONS OF AVAILABLE FUEL COMPOSITIONS WITH ORIGEN

The available comparisons of fuel compositions with ORIGEN predictions are summarized in Table A.4. The actinides are listed first, followed successively by volatile fission products and the other fission products, in order of atomic weight. Early versions of ORIGEN were used in most cases; however, ORIGEN2 was used in the studies made after 1980. Current QA methods were used in only one study (McELROY 1985).

The high and low ratios of the measured to the predicted values, the number of comparisons found, the average ratios, and the standard deviations are given in Table A.5 for all isotopes for which data were found. The values in Table A.5 are indicative of ORIGEN's ability to predict various nuclide concentrations in irradiated fuel, but they must be used with caution since (1) most studies did not use appropriate QA standards, (2) certain nuclides are difficult to analyze, and (3) the more recent ORIGEN2 code gives better agreement in some areas. For instance, ORIGEN2 permits much closer agreement with the limited data for higher actinides (WICHNER 1983) (Table A.4). It would be expected that uncertainties in the chemical analyses would be reflected in a large range of ratios and standard deviations such as those found for ^{242}Cm, ^{244}Cm, ^{129}I, ^{106}Ru, ^{241}Am, ^{3}H, ^{125}Sb, ^{134}Cs, and ^{144}Ce.

The data in Table A.5 can be divided into isotopes which are relatively well predicted by ORIGEN (within ±10%), those which are moderately under- or overpredicted (within ±50%), and those which are grossly under- or overpredicted, as given in Table A.6. The extent to which the disagreements are due to inaccuracies in ORIGEN or from experimental errors cannot be easily determined.

Table A.3. Currently available isotopic data for reactor fuel

Reactor[a]	Source	Burnup (MWd/kg)	Isotopes determined in studies									
			U	Pu	Am	Cm	T	C	Kr	I	Nd	Cs
H. B. Robinson	WILSON 1983	25	X	X							X	
	WILSON 1983	31	X	X							X	
	WILSON 1983	30	X	X	X	X						X
	JOHNSON 1980	27	X	X						X		X
	JOHNSON 1980	27	X	X								X
	JOHNSON 1980		X	X								X
	JOHNSON 1980	19	X	X			X	X	X		X	
	JOHNSON 1980											X
	JOHNSON 1980	28										X
	GOODE[b]	31	X	X	X	X	X	X	X	X		X
	CAMPBELL[c]	31	X	X	X	X	X	X		X		X
	CAMPBELL[c]	31				X		X		X		
	CAMPBELL[c]	31		X	X	X	X			X		X
	CAMPBELL[c]	31		X	X	X				X		X
	CAMPBELL[c]	31	X	X				X		X		
	CAMPBELL[c]	31						X		X		
	CAMPBELL[c]	31						X		X		
	CAMPBELL[c]	31	X	X				X		X		
	CAMPBELL[c]	31						X		X		
	CAMPBELL[c]	31						X		X		
	CAMPBELL 1981	31	X	X		X					X	
	BARNER 1985	31	X	X							X	X
	McELROY 1985	31	X	X	X		X				X	
	RYMAN 1982	31	X	X	X	X				X		X
	HEEB 1979[d]	25-31	X	X								
	CAMPBELL 1986	31					X					
Oconee-1	GOODE 1980b[e]	24	X	X			X		X	X		X
	CAMPBELL[c]	31	X	X				X		X		
	CAMPBELL[c]	31						X		X		
	MONTGOMERY 1978		X	X								
	B&W 1979	11	X	X								
		11	X	X								
		11	X	X								
	JOHNSON 1980	10	X	X						X		X
	JOHNSON 1980	11	X	X						X		X
	JOHNSON 1980	11	X	X				X	X			X
	JOHNSON 1980		X	X				X	X	X		X
	JOHNSON 1980	12	X	X				X	X	X		X
	JOHNSON 1980	9						X	X			X
	JOHNSON 1980							X	X			X

Table A.3. (continued)

Reactor[a]	Source	Burnup (MWd/kg)	Isotopes determined in studies									
			U	Pu	Am	Cm	T	C	Kr	I	Nd	Cs
Monticello	BAUMGARTNER 1984	44	X	X		X						
Yankee-Rowe	NODVIK 1966[f]	1-31	X	X								X
	RYMAN 1982	14	X	X							X	
	HEEB 1979[g]	14,24	X	X								
Garigliano	CADELLI 1984	18	X	X								
BR3	ADAMS 1984	35	X	X								X
	CADELLI 1984	44	X	X	X	X						
	CADELLI 1984	40	X	X	X	X						
Big Rock Point	CAMPBELL[b]	20						X		X		
Dresden-3	GOODE 1980b	14		X			X	X	X	X		X
Saxon	JOHNSON 1980	4	X	X			X	X	X			X
	JOHNSON 1980	4					X	X	X			X
	JOHNSON 1980	8	X	X			X	X	X			X
Quad Cities	WILSON 1983	12	X	X	X	X						
Obriqheim	FISCHER 1983	30	X	X	X	X			X		X	X
Trino Vercellese	FISCHER 1983		X	X	X	X						
SENA	FISCHER 1983	20	X	X								
Turkey Point-3	ATKIN 1981	28	X	X							X	
	ATKIN 1980	31	X	X					X			
	DAVIS 1981	27	X	X								
	NEELY 1982	27	X	X								
Connecticut Yankee	LANGSTAFF 1982	40	X	X								
Point Beach	JOHNSON 1980[h]	12-28	X	X	X	X					X	

[a]BWR—Monticello, Garigliano, Big Rock Point, Dresden-3, and Quad Cities. PWR—remainder.
[b]GOODE 1978, 1980a,b.
[c]CAMPBELL 1976a,b,c; CAMPBELL 1977a,b,c,d; CAMPBELL 1978a,b,c.
[d]Three samples.
[e]Data for H. B. Robinson-2 as of 4/1/77; for Oconee-1 as of 2/1/79; for Dresden-3 as of 3/23/79.
[f]Analyses of 219 pellet-sized samples.
[g]Two samples.
[h]Twelve samples.

Table A.4. Comparison of H. B. Robinson fuel composition data with ORIGEN predictions

Isotope	Measured/predicted ratio												
	GOODE 1980b	GOODE 1978	CAMPBELL 1976b	CAMPBELL 1976c	McELROY 1985	CAMPBELL 1976b	GOODE 1980b (Oconee-1)	CAMPBELL 1977c (Oconee-1)	CAMPBELL 1977c (H. B. Robinson)	CADELLI 1984[a] (BR3)	CAMPBELL 1977a	CAMPBELL 1978c	NEELY 1982[c] (Turkey Point-3)	WICHNER 1983[d] (H. B. Robinson-2)
Total Pu		0.94								0.99, 0.81	1.07			
Pu/U ratio								0.86				0.88, 0.93, 0.92, 0.92[a]		
^{233}U							1.00[a]							
^{234}U	1.08[a]			1.08[a]	10.3, 14.9[b]		0.92[a]	1.10[a]				1.27, 1.20, 1.07, 1.07[a]		
^{235}U	1.03[a]			1.06[a]	0.98, 1.02, 0.99, 1.06, 0.96, 0.97[a]		0.94[a]	1.14[a]		0.97, 0.86[a]		1.02, 1.02, 1.16, 1.01[a]	1.00[a]	
^{236}U	1.01[a]			0.99[a]	1.09, 1.09, 1.05, 1.03, 1.08, 1.10[a]		1.09[a]	0.83[a]		1.08, 0.95[a]		0.98, 0.98, 0.95, 0.99[a]	0.96[a]	
^{238}U	1.00[a]			1.00[a]	1.01, 1.01, 1.01, 1.00, 1.01, 1.00[a]		1.00[a]	1.00[a]		1.00, 1.00[a]		1.00, 1.00, 1.00, 1.00[a]	1.00[a]	
Total U										1.00, 1.01	0.97			
^{238}Pu	1.27[a]			1.14, 1.33, 1.35, 1.18[a]	1.04, 1.15, 1.07, 1.17, 1.05, 1.05[a]	1.13[a]	1.08[a]	0.78[a]		1.01, 1.53[a]		1.23, 1.23, 1.13, 1.20[a]	1.02[a]	
^{239}Pu	0.96[a]			0.98[a]	0.90, 1.00, 0.94, 1.00, 0.94, 0.94[a]	0.92[a]	1.01[a]	1.02[a]		1.03, 0.84[a]		0.97, 0.96, 1.01, 0.97[a]	0.99[a]	
^{240}Pu	1.09[a]			1.04[a]	0.98, 1.01, 1.00, 1.02, 1.01, 0.97[a]	1.03[a]	1.12[a]	0.91[a]		0.87, 1.11[a]		1.08, 1.08, 1.02, 1.08[a]	1.09[a]	
$^{239,240}Pu$				0.95, 1.16, 1.01										
^{241}Pu	0.94[a]			0.96[a]	0.91, 1.00, 0.96, 1.03, 0.92, 0.91[a]	1.21[a]	0.77[a]	1.04[a]		1.05, 1.14[a]		0.94, 0.95, 0.93, 0.95[a]	0.88[a]	
^{242}Pu	1.07[a]			1.02[a]	1.07, 1.08	1.53[a]	0.92[a]	0.95[a]		1.12, 1.73[a]		1.07, 1.09, 0.94, 1.09[a]	0.96[a]	
^{241}Am		0.62		0.60, 0.27, 0.54, 0.946[a]	0.87, 1.00					1.03, 1.03				1.05
^{242}Am				1560[a]										0.99
^{243}Am				1.10[a]										0.92
^{242}Cm		1.15		1.67, 1.49, 1.56, 3.28[a]										1.12
^{243}Cm				8.64[a]										0.95
^{244}Cm		0.40		0.42, 0.44, 0.47, 0.929[a]						1.08, 1.58				0.99
^{245}Cm				3.89[a]										1.25
^{246}Cm				2.98[a]										1.04
^{247}Cm				2.0[a]										1.00
^{248}Cm				5.0[a]										Poor statistics
^{3}H	0.58	1.02					0.84			1.12, 1.12				
^{129}I	0.84	0.43	~0.82, 0.99				1.74	0.49	0.62			0.82, 0.84, 0.41, 0.80, 0.84, 0.76		
^{14}C	1.09	1.04												
^{85}Kr	1.29	1.34					1.39							
^{90}Sr				0.81, 1.04	0.96, 0.95									
^{99}Tc		0.52			0.80, 0.85, 0.93, 0.90, 0.95, 0.92									
^{106}Ru	0.77	0.75		1.15			0.99							
^{125}Sb	0.91	0.50					0.59							
^{134}Cs	0.57	0.55		0.58, 0.61, 0.63			1.34					0.44		
^{137}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					0.81		
^{144}Ce	0.81	0.81		0.93, 0.97, 0.98			1.29					0.64		
^{154}Eu				0.29								0.28		
^{237}Np					0.92, 0.95, 1.02, 1.03									
^{79}Se					0.13, 0.13									
^{126}Sn					0.23, 0.23									

[a]By mass spectrometry.
[b]Apparent error in ^{234}U assumed in feed uranium.
[c]Used ORIGEN2 code.
[d]Recalculation of the results reported in CAMPBELL 1976c using ORIGEN2 code.

Table A.5. Average measured/predicted ratios

Isotope	Low/High	No.[a]	Average	SD[b]	Isotope	Low/High	No.[a]	Average	SD[b]
^{234}U[c]	0.92/1.10	8	1.10	0.10	^{235}U	0.86/1.16	17	1.01	0.07
^{236}U	0.95/1.09	17	1.01	0.07	^{237}Np	0.92/1.03	4	0.98	0.05
^{238}Pu	0.78/1.27	21	1.15	0.15	^{239}Pu	0.84/1.03	18	0.97	0.05
^{240}Pu	0.87/1.12	18	1.03	0.07	^{241}Pu	0.77/1.21	18	0.97	0.10
^{242}Pu	0.94/1.73	14	1.12	0.22	^{241}Am	0.27/0.946	9	0.77[d]	0.25
^{242}Am		1	0.99		^{243}Am		1	0.92	
^{242}Cm	1.08/3.28	7	1.69[d]	0.68	^{243}Cm		1	0.95	
^{244}Cm	0.40/1.12	7	0.70[d]	0.31	^{245}Cm		1	1.25	
^{246}Cm		1	1.04		^{247}Cm		1	1.00	
^{248}Cm		1	0.95[d]		^{3}H	0.58/1.02	3	0.81	0.18
^{129}I	0.43/1.74	11	0.80	0.32	^{14}C	1.04/1.09	2	1.06	
^{85}Kr	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	^{99}Tc	0.52/0.95	7	0.84	0.14
^{106}Ru	0.75/1.15	4	0.92	0.17	^{125}Sb	0.50/0.91	3	0.67	0.18
^{125}Sn	0.23/0.23	2	0.23		^{134}Cs	0.44/1.34	7	0.67	0.28
^{137}Cs	0.81/1.04	13	0.98	0.11	^{144}Ce	0.64/1.29	7	0.92	0.19
^{154}Eu	0.28/0.29	2	0.28						

[a]Number of determinations.
[b]SD = standard deviation.
[c]Two very high ratio (McELROY 1985) were not used.
[d]The latest calculations using ORIGEN2 (see Table A.4) gave ratios of 1.05, 1.12, and 0.99 for ^{241}Am, ^{242}Cm, and ^{244}Cm, respectively. The ^{248}Cm single data point had poor statistics.

Table A.6. Relative agreement of ORIGEN predictions for various isotopes

Excellent (within 10%)	Fair (within 50%)	Poor (greater than 50%)
^{234}U	^{242}Cm	^{79}Se
^{235}U	^{245}Cm[a]	^{126}Sn
^{236}U	^{3}H	^{154}Eu
^{237}Np	^{129}I	
^{239}Pu	^{99}Tc	
^{240}Pu	^{125}Sb	
^{241}Pu	^{134}Cs	
^{241}Am	^{144}Ce	
^{242}Am[a]	^{238}Pu	
^{243}Am[a]	^{242}Pu	
^{244}Cm[a]	^{85}Kr	
^{246}Cm[a]		
^{247}Cm[a]		
^{14}C		
^{90}Sr		
^{106}Ru		
^{137}Cs		

[a]Only one experimental determination.

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