HOW WELL DOES ORIGEN PREDICT SPENT LWR FUEL CHARACTERISTICS?

J. C. Mailen and J. W. Roddy
Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831

ABSTRACT

The ORIGEN computer code is widely used to estimate the radionuclide content (actinides, activation and fission products) of irradiated reactor fuel and the resultant heat generation and radiation levels associated with such fuel. 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 the fuel.

Calculated values determined using several variations of ORIGEN have been compared with experimentally determined values for actual fuel for many, but not all, of the parameters desired. In most cases, the comparisons did not use the most recent ORIGEN2 program, the most recent data libraries, or currently required quality assurance (QA) procedures. Comparisons of fuel composition data with ORIGEN2 are very limited, and the only data with proper QA are currently being acquired by Battelle Pacific Northwest Laboratory.

This survey summarizes the fuel data available in the open literature and, where given, the calculated values by ORIGEN. Plans for additional analyses of well-characterized reactor fuel samples to improve the validation of ORIGEN2 are discussed.

INTRODUCTION

ORIGEN and derivative programs are used throughout the world to estimate the actinide, fission product, and activation product contents of irradiated reactor fuels and the resultant heat generation and radiation levels associated with such fuels. These estimates are used as source terms in safety evaluations of operating reactors, for assessment of fuel behavior and regulation of the storage at the reactor, for transportation studies, and for evaluation of the ultimate geologic storage of the fuel. ORIGEN2 (1,2), a revised and updated version of ORIGEN, is an important code for formulating these predictions since it is currently used by many groups, including utilities, the U.S. Department of Energy (DOE), and the U.S. Nuclear Regulatory Commission (NRC), for such calculations.

The predictions of ORIGEN2 have not been adequately compared with experimental data for all fuel characteristics. This paper briefly summarizes the existing comparisons of experimental data with ORIGEN, with particular emphasis on the composition of irradiated fuel.

DESCRIPTION OF ORIGEN2

ORIGEN2 is a versatile computer code for calculating the nuclear composition and characteristics of fission reactor fuel. The input data bases include reactor-dependent cross sections, decay data (decay half-lives and branching fractions, internal transitions, etc.), fission product yields, photon production rates and energies, and neutron production rates. The current version of ORIGEN2 contains cross-section libraries for seven types of reactors. The user must specify 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 from alpha-neutron reactions and spontaneous fission and the photon production rates from gamma rays, X-rays, bremsstrahlung, etc., can be obtained. The results are presented in a tabular form. ORIGEN2 is written in the FORTRAN language and was developed for mainframe computers, such as the IBM, UNIVAC, CDC 7000 series, and CRAY and has also been used on "minicomputers," such as the VAX and DEC. A version is being tested for the IBM PC-AT microcomputer. The ORIGEN2 program is readily available to all users through the Oak Ridge National Laboratory/Radiation Shielding Information Center (ORNL/RSIC).

WHY VALIDATE ORIGEN?

Better estimates of the radioactive composition of the fuel, through improvements in the predictive capability of ORIGEN2, would be important in waste packaging, transportation, and fuel handling, where substantial savings in fabrication and operation costs could be possible. Although the actual amounts cannot be quantified, the incremental costs to one portion of the repository system will be discussed as an example.

The U.S. Department of Energy (DOE) (3) has performed an analysis of the waste management costs for the Davis Canyon site. The estimates for shipping spent fuel are ~$2 billion, while the number of casks required for shipment is 156 ± 22 by truck and 96 ± 15 by rail. Capital costs for each truck and rail cask are estimated to be $1.5 million and $2.5 million, respectively. If a completely validated version of ORIGEN2 results in an expenditure of $1 million, the incremental cost would be <0.1% of the total capital cost for the casks and would have a negligible effect

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on the total shipping costs. The benefits of greater ORIGEN2 accuracy would be reduced design margins and lower cask weights, which would result in lower construction and hauling costs. An identical scenario can be made for the waste repository and monitored retrievable storage facilities, in which shielding thicknesses and heat-removal requirements can be reduced.

Another benefit of a successful validation effort that cannot be quantified is the assurance that the methodology is the most current, the data bases are up to date, and the program contains no major flaws. Deficiencies in any of these areas would have a serious impact on the acceptability of the code results for design and licensing, resulting in expensive delays or facility modifications.

COMPARISON OF ORIGEN PREDICTIONS WITH EXPERIMENTAL RESULTS

Values determined using several variations of ORIGEN have been compared with those determined for actual fuel for many, but not all, of the parameters desired. In most cases, the comparison was not made with the most recent ORIGEN2 program (2), the most up-to-date nuclear data libraries, or currently required quality assurance (QA) procedures. An earlier summary has been published (2) in which heat generation rates, actinide contents, and krypton and xenon isotope contents of irradiated fuels are compared with values calculated using ORIGEN2.

In this report, the main effort has been to assemble the existing comparisons of experimental reactor fuel compositions with those predicted by ORIGEN, since predictions of composition are required as input to calculations of heat and radiation generation. This effort has additional value, since the fuel composition data had not been completely surveyed in the past. The available comparisons of heat and radiation generation by fuel assemblies with ORIGEN predictions will be summarized first, followed by the fuel composition comparisons.

Heat Generation

The experimental heat-generation data for fuel assemblies have been compared with those predicted using ORIGEN2 (4-11). Comparisons of measured values with those predicted by ORIGEN agree to within ±10%; the experimental data scatter is also ~±10%. The data cover only a limited number of cooling times and burnups.

Gamma Rates

Gamma exposure rates have been measured near full fuel bundles in a pool experiment (12) and surrounding a partially and fully loaded spent fuel storage cask (8,11). In both cases, a combination of an ORIGEN code for prediction of the photon source and a radiation transport code were required; this makes it impossible to completely separate experimental errors from inaccuracies introduced by the codes. Further, inaccuracies introduced by the individual codes cannot be determined. Strickler (12) found reasonable agreement (a factor of 2) between the predicted [ORIGEN and QAD-F (3)] 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. Wiles (11) found the predictions [ORIGEN and QAD-CG (14)] agreed well with measured data over the length of the neutron shield, but, above and below the ends of the shield, dose rate peaks were underpredicted by a factor of 2. The discrepancies between the predictions and measurements could have been due to the predicted quantities of radioisotopes by ORIGEN (or ORIGEN2), from approximations within the geometric codes (QAD-F or QAD-CG), from errors in the experiment, or from inaccuracies in the assumed fuel attributes.

Neutron Rates

Neutron dose rate measurements were made on the same partially and completely filled storage cask as was used for gamma measurements (8,11). The absolute neutron source strength was based on ORIGEN2 values calculated for each assembly, and the resulting dose rates were computed for the cask outer surface utilizing the DOT computer code (15). The predicted neutron dose rates on the ends and side of the cask on which there is no neutron shield were a factor of ~2 to 6 higher than experimental data. Calculated neutron dose rates were a factor of ~2 higher than the experimental data on the side of the cask having shielding.

Composition Data

Fuel composition data were found for 16 reactors; a total of 295 analyses of fuel samples were identified, with 282 of these giving experimental contents of actinides and 265 giving experimental contents of fission products (16-48). It should be noted that 219 of the analyses were of pellet-sized samples from Yankee-Rowe (38), which used stainless-steel clad fuel. A complete report presenting data sources and surveying the currently available data for all the parameters (90 references) is in preparation (49).

Data comparing experimental fuel compositions with ORIGEN predictions were found only for the H. B. Robinson (18,19,22,23,26,30,47,48), Oconee-1 (20,26), BR3 (39), and Turkey Point-3 (45) reactors. Table I gives the high and low ratios of the experimentally determined radioactive concentration in irradiated fuel to the values calculated by ORIGEN, the number of comparisons found, the average ratios, and the standard deviations for all isotopes for which usable data were found. The values in this table are only indicative of the ability of ORIGEN to predict various nuclide concentrations in irradiated fuel and must be used with caution, since most studies did not use current QA standards, some nuclides are difficult to analyze, and the more recent ORIGEN2 code yields better agreement in some areas. Isotopes that are difficult to determine in irradiated light-water reactor (LWR) fuel include: (1) tritium, of which ~50% is in the Zircaloy cladding, which is often not analyzed; (2) ^{106}Ru, a portion of which is not readily soluble in the nitric acid commonly used for dissolving irradiated fuel; (3) ^{129}I, which must be volatilized from the fuel solution, trapped, and determined by neutron activation; and (4) ^{14}C, which must be volatilized from the fuel solution and trapped. Data from experiments in which the experimental procedures did not ensure nearly complete recovery of these isotopes were not included in the compiled data. Experimental errors can also arise from errors in sampling or equipment calibration. ORIGEN2 predictions that are an improvement over earlier versions of the code include much better agreement with the limited data for higher actinides (2). Uncertainties in the chemical analyses and experimental difficulties would be expected to result 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, and ^{125}Sb. The large scatter found for ^{134}Cs and ^{144}Ce is difficult to explain, since these isotopes should be relatively easy to measure.

TABLE I

Average Ratios[a] (Experiment/ORIGEN)

Isotope	High/Low	No.[b]	Average	SD[c]
^{3}H	0.58/0.81	3	0.81	0.18
^{14}C	1.14/1.09	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
^{126}Sn	0.23/0.23	2	0.23	
^{129}I	0.43/1.74	11	0.80	0.32
^{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	
^{234}U	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]	

[a]Ratios of absolute concentrations.
[b]Number of determinations.
[c]Standard deviation.
[d]The latest calculations using ORIGEN2 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.

The data in Table I can be divided into nuclides which have relatively good agreement (within ±10%), moderately good agreement (within ±50%), and poor agreement between the experimental data and the ORIGEN predictions, as shown in Table II. The extent to which the disagreements are due to inaccuracies in ORIGEN calculations (possibly from nuclear data libraries or reactor models), experimental limitations, or analytical inaccuracies cannot be easily determined unless replicate measurements are made using well-characterized fuel samples and approved QA procedures (e.g., analytical techniques, sample traceability).

PROBLEMS WITH EXPERIMENTAL DATA

The experimental data on heat and radiation-generation rates from irradiated LWR fuel are presumed adequate. Agreement between experimental and predicted heat-generation rates is excellent for early time periods. Additional efforts in radiation (gamma and neutron) rates should address the radiation transport

TABLE I.

Relative Agreement of Experimental Data and ORIGEN Predictions for Various Isotopes

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

[a]Only one experimental determination.

calculations, since the codes used in the reported studies were relatively inaccurate.

Comparisons of fuel composition data with ORIGEN2 are very limited, and the only data with proper QA are being currently acquired by Batelle Pacific Northwest Laboratory (32,50). In most cases, the other comparisons did not use the most recent ORIGEN2 program, the most recent data libraries, or currently required QA procedures. Many of the earlier data were used in the development of the ORIGEN codes and cannot be used for validation efforts. A significant additional effort is needed to adequately validate ORIGEN2 for the prediction of fuel compositions.

FUTURE STUDIES

Work is currently under way at PNL in the Materials Characterization Center (33,50) to obtain well-characterized LWR fuel for use in studies related to fuel storage in geologic repositories. The availability of portions of these well-characterized fuel samples from PNL makes this an ideal time to validate the ORIGEN2 code. A spark-source mass spectrometer (SSMS) at ORNL, which is capable of analyzing fully irradiated fuel samples, will make it possible to examine isotopes which are difficult to determine using standard radiochemical techniques.

The identification of the important isotopes that need to be predicted in spent fuel is being undertaken by assembling the recommendations of personnel in the waste project repository offices, those performing criticality calculations, and experts in the use of the ORIGEN2 code. These recommendations, along with information on analytical capabilities and funding availability, will be used to define a program to improve the validation of ORIGEN-type programs. New analyses resulting from such a program will allow the validation of the ORIGEN2 code required for its use in future years. The data base generated will also be available for development or validation of other similar codes.

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