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AN AUTOMATED PROCEDURE FOR CALCULATING TIME-DEPENDENT SENSITIVITIES IN ORIGEN2

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I. INTRODUCTION

The determination of spent fuel and waste characteristics forms the basis for the study and design of fuel reprocessing plants, spent fuel shipping casks, waste treatment and disposal facilities, and waste shipping casks. The principal quantities of interest are the composition and amounts of the spent fuel and the associated radioactivity, decay heat, and toxicity. The neutronics calculation need only be sophisticated enough to determine the nuclide compositions of interest both during reactor operation and during subsequent storage and disposal. In practice, these quantities are calculated based on the average flux and fuel composition in the reactor (referred to as "point depletion" since the spatial characteristics of the flux and fuel are not considered).

ORIGEN2[1] is a widely used point-depletion and radioactive-decay computer code for use in simulating nuclear fuel cycles and/or spent fuel characteristics. The data used by the code are of two types: generic data that are part of the code package and are read from a series of library files describing the nuclide chains, cross sections, fission-product yields, decay data and decay photon data; and problem-specific data, including the composition of the materials in the reactor and a series of commands describing, step by step, the nuclear fuel cycle scenarios considered in the problem. The code calculates the amount of each nuclide being considered in the problem at a specified number of times, and upon request, a database of conversion factors relating mass compositions to specific nuclear material characteristics is used to calculate and print the total and nuclide-dependent radioactivity, thermal power, and toxicity, as well

as absorption, fission, neutron emission, and photon emission rates. The calculated masses, radioactivities, etc. will be referred to as output responses.

The flexibility in use of the ORIGEN2 code, along with its extensive database, allows complex problems to be solved with a minimal burden on the user. However, as is the case with any code with a large database and numerous user input options, studies seeking to quantify data sensitivities (the change in a given response with respect to the change in data) often require an impractical number of recalculations. The objective of this paper is to present an automated procedure for calculating partial first derivatives, d(response)/d(data), and the application of this procedure to the ORIGEN2 code for performing a sensitivity analysis of a high-level waste disposal problem.

Section II describes the automated procedure for calculating the partial derivatives. Section III presents a sample problem typical of an application of ORIGEN2 in analysis of high-level waste disposal. Section IV is a discussion of the key sensitivity results for this study and Section V details computer code performance. Section VI presents some conclusions about the use of the automated procedure for calculating partial derivatives.

II. AUTOMATED CALCULATION OF SENSITIVITY COEFFICIENTS

An automated procedure based on the use of computer calculus was developed[2] to perform large-scale sensitivity analyses. The procedure was embodied in a FORTRAN pre-compiler called GRESS,[3] which automatically processes computer codes and adds derivative-taking capabilities to the

normal calculational scheme. Thus, in a single run, all sensitivities of interest can be obtained from these calculated derivatives. The procedure allows derivatives of any real variable used in the code to be calculated with respect to another variable or an input parameter. The independent variables (generic or problem-specific data), with respect to which GRESS will generate partial derivatives in the enhanced FORTRAN source code, may be freely selected by the user. The only theoretical restriction to this choice is that the sequence of computations in the source code must be continuous in differentiable form for the derivatives generated to have any practical meaning. In the enhanced code the derivatives are calculated according to the chain rule of differential calculus and are propagated from operation to operation analytically by implicit differentiation rules. Inherently, therefore, no numerical difference scheme is used and the derivative results are as accurate as the analytical computations in the reference code. Specific details explaining how GRESS enhances conventional FORTRAN programs with analytic differentiation of arithmetic statements can be found in Reference 4.

The use of GRESS is illustrated in Fig. 1. In the preliminary processing step the input program is separated into two subsets. The first, which contains all main sequence computations, is hand modified for submission to the GRESS pre-compiler. The amount and nature of these modifications depends upon the particular application at hand and the limitations of the current version of GRESS. The second subset (possibly null) is composed of subroutines whose only communication with the first subset is through the arguments in their calling sequence. These subroutines are usually associated with input, output, and peripheral program

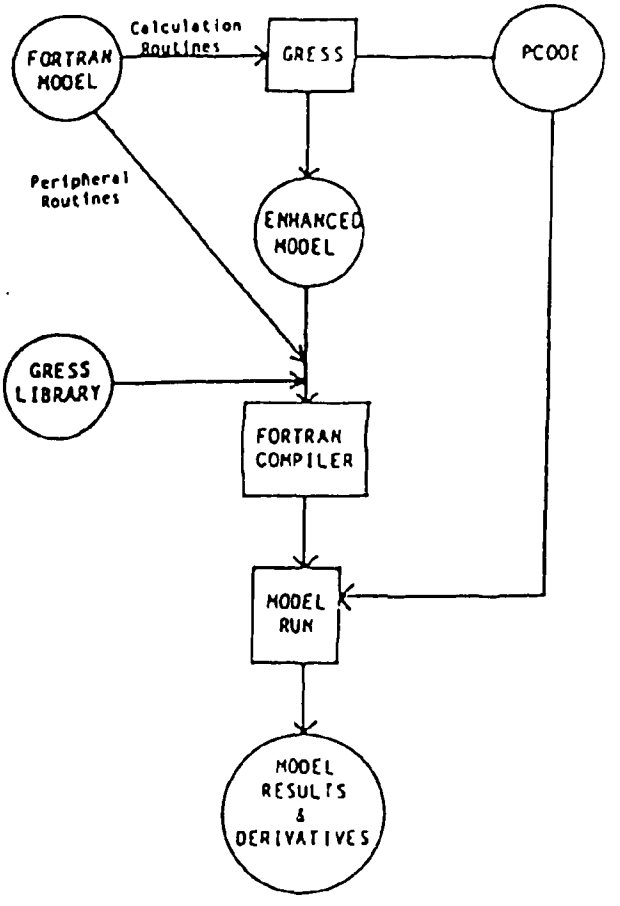

Fig. 1. Flowchart of Procedures for the Use of GRESS

analysis functions. They do not require GRESS compilation and may usually be submitted unchanged to the FORTRAN compiler.

The GRESS pre-compilation step is the one in which the additional code translation necessary to compute derivatives is performed using automated computer calculus. A rearrangement of the program data structure and a substitution of calls to GRESS interpretive software are performed automatically by GRESS for all arithmetic lines of coding. The modified program is then compiled into pseudo-machine code (the GRESS P-CODE) for use during program execution. The two output files of this step are, therefore, the set of transformed FORTRAN subroutines and the binary P-CODE file.

In the final stage of the GRESS procedure, the transformed FORTRAN subroutines are combined with the untranslated subset of subroutines and both are submitted to the normal FORTRAN compiler. The relocatable object modules which result from actual compilation are then input to the system link-edit loader, which combines them with appropriate portions of the GRESS interpretive library and the P-CODE file to form the complete executable program. The library contains the P-CODE interpreter, a series of support routines, and a set of utility subroutines which may be directly referenced by the translated program to display or manipulate derivatives. The calls to these utility routines are added, as needed, in the original source code before the pre-compilation step. The resulting translated program will execute identically to the original source program with the option of also calculating derivatives.

The application of GRESS to the ORIGEN2 code required about 2 man months effort. First, the ORIGEN2 FORTRAN was modified to make the code

compatible with the GRESS precompiler and the modified code was tested to ensure that the changes had no effect on calculated results. Second, after a GRESS version of the code was obtained, the computed derivatives were compared with derivatives estimated from reruns. We feel that this initial investment in programming effort is extremely efficient considering that 1) ORIGEN2 is a large code of approximately 10,000 lines with an extensive data base for the 1300 unique nuclides and 2) the capability now exists for calculating partial derivatives of all independent variables with respect to all data and input parameters.

III. SAMPLE PROBLEM

The sample problem calculates the decay heat and radioactivity of the high-level waste resulting from the reprocessing of one metric ton of initial heavy metal of 33 GWd/ton spent PWR fuel for decay times up to 1000 years following reprocessing. The fuel is irradiated for 880 full-power days with two decay intervals of 110 days each. After the fuel has been removed from the reactor and a 150 day cooling period has been allowed, the volatiles and U/Pu are separated from the waste. The fractional recovery rates for the uranium and plutonium isotopes are 0.999 and 0.9999, respectively. The resulting high-level waste (HLW) nuclide compositions are calculated at 0.0, 0.5, 1.0, 5.0, 10.0, 50.0, 100.0, 200.0, 300.0, and 1000.0 years following reprocessing.

The initial fuel loading, which represents an average for the reactor equilibrium core, is 3.2% enriched uranium plus various fuel impurities. The ORIGEN2 input is shown in Table I.

Initially, the HLW is made up of 28 kg of fission products and 5.4 kg of actinides. Approximately 85% of the mass of the fission products is accounted for by the 10 elements in Table II. Of these 40 fission-product nuclides, 34 are either stable or have half-lives longer than a million years. The ^{238}U comprises 87% of the actinides not recovered in the reprocessing.

The output responses are the radioactivity and decay heat by nuclide at each of the ten time steps. For this sample problem there are 129 actinides and 879 fission products of interest. Thus the total number of output responses is $(129 + 879) \times 2 \times 10 = 20,160$. However, as can be seen from Tables III and IV, 12 actinides and 17 fission products constitute the bulk of the decay heat and radioactivity over the 1000-year period.

A sensitivity analysis of this problem entails consideration of the effect of the generic and problem-specific data upon the responses of interest - the nuclide-dependent radioactivity and decay heat. The generic data are the nuclide cross sections, half-lives, decay branching fractions, fission-product yields, and photon decay rates. Examples of the problem-specific data are the irradiation times, fuel enrichment, shutdown time periods, cooling periods, specific power, etc. Conventionally, the sensitivity analysis would begin by first identifying the data that would most effect the response of interest and/or would have a relatively high uncertainty. The selection of these data would be based either upon some prior experience or auxiliary calculations. Then, for N data selected, the code would be rerun N times to access the effect of each datum change upon the response of interest. Likewise, the effect of input parameters would be calculated by rerunning the code. For a code

with a large database and numerous input parameters such as ORIGEN2, a prior knowledge of the most critical and/or uncertain data is imperative to keep the reruns to an affordable number. In the next two sections we discuss how the GRESS automated procedure was used to calculate sensitivity data for the sample problem; how the sensitivity data might be used; and how the use of GRESS circumvents the necessity to limit the selection of data to be analyzed.

IV. SAMPLE PROBLEM SENSITIVITY COEFFICIENTS

Decay Heat Sensitivity Coefficients

The ORIGEN2-GRESS (ORIGEN2G) version of ORIGEN2 produces first derivatives and calculates sensitivity coefficients of output responses to generic and problem-specific data (hereinafter referred to as parameters). The sensitivity coefficient is defined as the percent change in an output response per percent change in a parameter. The user specifies both the output responses (i.e., calculated variables or functions of variables) and the parameters for which sensitivity coefficients are to be calculated. For the HLW sample problem, the sensitivity of the decay heat and the radioactivity of the 129 actinides and 879 fission products at each of the ten times steps was calculated for seven selected parameters, producing a total of 141,120 sensitivity coefficients. The parameters chosen for this problem are the fission cross sections of ^{235}U, ^{239}Pu, and ^{241}Pu (generic data) and the specific power, initial ^{235}U loading, irradiation time, and shutdown time during irradiation (problem-specific data).

The sensitivity coefficients were stored on disk for subsequent selection retrieval. Note from Table III that for the time period 0-1 year

and 5-50 years the fission products account for most of the high-level waste decay heat, whereas Pu and Am account for a comparable fraction of the decay heat between 100-1000 years. Tables V-XI summarize the sensitivities of decay heat to each of the seven selected parameters for the nuclides with relatively high fractions of total decay heat in each of these three time periods. Table XII shows the sensitivity of total decay heat versus decay time. The sensitivities to radioactivity are not shown but are identical to the decay heat sensitivities since radioactivity and decay heat are proportional.

With the exception of ^{95}Nb, the fission-product sensitivities shown in Tables V-XI do not change as a function of decay time. In addition, the ^{90}Sr and ^{90}Y sensitivities are identical, as are the ^{137}Cs and ^{137m}Ba sensitivities, because the relatively long decay time of ^{90}Sr and ^{137}Cs effectively control the rate of decay heat released by ^{90}Y and ^{137m}Ba (relatively short decay times). Table XII reveals that the sensitivity of total decay heat to irradiation time is larger than the sensitivity to the other six selected parameters for all decay times. In particular, of the nuclides producing the greater fraction of heat in the first 0-1 and 5-50 year time spans, the decay heats of ^{134}Cs and ^{106}Rh are the most sensitive to irradiation time. The largest nuclide-dependent sensitivities to irradiation time, however, are for the decay heat of ^{240}Pu and ^{243}Am, for which a one percent change in irradiation time changes the decay heat by over eight percent. A thousand years after reprocessing, these two nuclides account for over 30 percent of the total decay heat. After the irradiation time, the relative ranking of the sensitivity of the total decay heat to the remaining six parameters are: downtime, specific power,

^{239}Pu fission cross section, ^{235}U loading, ^{235}U fission cross section, and the ^{241}Pu fission cross section.

The decay heat sensitivities change as a function of decay time as exemplified by the selected data shown in Table XIII. From the time of reprocessing to a thousand years later, the ^{238}Pu sensitivities to irradiation time and downtime decreases by 63% and 79% respectively, and the relative rankings of the sensitivities change. On the other hand, the ^{241}Am decay heat sensitivities change only slightly with time and the relative ranking of the sensitivity to each of the parameters remains unchanged.

Definition of Decay Heat Coefficients

A relatively high nuclide-dependent sensitivity coefficient is not necessarily important if that nuclide response contributes negligibly to the total response of interest. For example, the ^{244}Cm sensitivity coefficient of decay heat to irradiation time is 11.3 at 0 years, but the ^{244}Cm decay heat is only 0.337% of the total. However, at 10 years the ^{244}Cm decay heat is almost 5% of the total and the high sensitivity of decay heat to irradiation time is significant. Thus a much more informative number is the change in decay heat per percent change in the parameter of interest. This number is simply the sensitivity coefficient times the decay heat for the nuclide divided by 100.0. For the sample problem this quantity was calculated for all nuclides for the seven selected parameters. Table XIV lists the nuclides and input parameters that result in the 20 highest changes in the nuclide-dependent values of decay heat at time of reprocessing per one percent change in the data or input parameter. Thus, a one percent increase in irradiation time

results in an increase in decay heat of 123.1 watts in ^{106}Rh at the time of reprocessing. Note that irradiation time, downtime, and specific power are the most contributing parameters as expected. Although fission products produce most of the decay heat at the time of reprocessing, the actinide ^{242}Cm, because of its high sensitivity coefficient, appears fairly high in the list. We shall refer to the quantities listed in Table XIV as the decay heat coefficients to distinguish them from the decay heat sensitivity coefficients.

An important feature of saving the decay heat coefficients is that uncertainty analyses can be performed for given relative uncertainties in the data and input parameters. These relative uncertainties, normalized and folded with the decay heat coefficients, would give the total uncertainty (and nuclide-dependent uncertainties) in decay heat as a function of decay time.

Because the decay heat coefficient depends upon the decay heat of a nuclide at a reference state, the decay heat coefficient of a nuclide is not equal to its radioactivity coefficient even though its decay heat sensitivity coefficient is equal to its radioactivity sensitivity coefficient. Table XV lists the 20 largest radioactivity coefficients at time of reprocessing. The radioactivity coefficient of ^{106}Rh to a change in the irradiation time is the largest, as was the decay heat coefficient of ^{106}Rh to irradiation time (Table XIV). But note that ^{106}Ru and ^{144}Ce contribute significantly to the change in radioactivity for a given change in irradiation time, but not to the change in decay heat.

A thousand years after reprocessing, the 20 largest radioactivity coefficients, shown in Table XVI, are smaller by about four orders of

magnitude. This is slightly less than the fractional decrease in total radioactivity (Table IV) because the nuclide sensitivities of the important nuclides 1000 years after reprocessing are greater than the sensitivities of the important nuclides at the time of reprocessing. Interestingly, the fission product ^{99}Tc is fourteenth in ranking. Note also that generic data parameters, the U and Pu fission cross sections, appear in this list. The significance of ranking the radioactivity coefficients is the ease in recognizing that if the uncertainty in the ^{239}Pu fission cross section is six times greater than the uncertainty in the other parameters listed, then the largest contribution to the total uncertainty in radioactivity at 1000 years will be the uncertainty in the ^{239}Pu fission cross section.

Associated Sensitivity Coefficients

Given the downtime, irradiation time, specific power, and ^{235}U loading sensitivity data, other secondary sensitivities can be calculated. For example, suppose the capacity factor is changed X_C percent by changing the downtime by X_D percent. Then

$$X_D = - \frac{X_C(1 + I/D)}{1 + 0.01\ X_C} ,$$

where I = full power irradiation time and D = downtime. The sensitivity of response i to capacity factor, α_{iC}, is given by

$$\alpha_{iC} = - \frac{\alpha_{iD}(1 + I/D)}{1 + 0.01\ X_C} ,$$

where α_{iD} is the sensitivity of response i to downtime. Likewise, if the irradiation time is changed by X_I percent to change the capacity factor by X_C percent,

$$X_I = \frac{X_C}{(1 - C_f) - 0.01\ C_f\ X_C} \quad ,$$

where C_f is the unperturbed capacity factor. The effective sensitivity is then

$$\alpha_{ic} = \frac{\alpha_{iI}}{(1 - C_f) - 0.01\ C_f\ X_C} \quad ,$$

where α_{iI} is the sensitivity of response i to irradiation time.

The sensitivity to burnup can also be calculated from the basic sensitivity data if an estimate of the relationship between ^{235}U loading and burnup is available from auxiliary calculations. For a known function f such that $X_{U5} = f(X_B)$, where X_{U5} and X_B are the percent changes in ^{235}U loading and burnup respectively, the sensitivity of response i to burnup, α_{iB}, is given by

$$\alpha_{iB} = \alpha_{iX} + \left(\frac{X_{U5}}{X_B}\right)\alpha_{iU5} \quad ,$$

where α_{iX} is the sensitivity of response i to specific power or irradiation time and α_{iU5} is the sensitivity to ^{235}U loading.

As an example, the ratio X_{U5}/X_B for a typical light-water reactor varies between 0.4 and 0.7. If it is assumed that $(X_{U5}/X_B) = 0.57$ and that the burnup is changed by changing the specific power and if the data in Table VIII are used, then the sensitivities to burnup for ^{106}Rh, ^{134}Cs,

^{137}Cs, and ^{243}Am would be as shown in Table XVII. This table reveals that if the burnup is changed by changing the specific power while keeping the irradiation time fixed, the sensitivities to burnup can be dramatically different from the sensitivities to specific power because of the effect of the change in the ^{235}U loading.

V. ORIGEN2 PERFORMANCE

The execution time of ORIGEN2G is approximately 30 times that of the ORIGEN2 standalone version for the sample problem. However, because all partial derivatives are stored and used in calculating the total derivative for the selected responses, the additional computation required for determining sensitivities to other parameters is negligible. For example, instead of identifying only 7 parameters as we did in this study, we could have selected any number of parameters and still used approximately 30 times the execution time of the standalone version. Thus, for 30 or more parameters the time required to run the reference calculation and calculate sensitivities would be less than or equal to the time required to perform the reference calculation and perform reruns as would be done in the conventional sensitivity study.

Two important points should be made with regard to GRESS and conventional sensitivity studies in general. First, even with reruns the sensitivities (i.e., derivatives) would have to be hand calculated or computed in an auxiliary calculation. While this calculation is trivial, the accurate calculation of derivatives based on reruns is not necessarily straightforward. If the change in the data or input parameter is too small, the change in the response may not be significant

enough to accurately calculate the first derivative. If the parameter change is too large the change in response may include higher order effects such that the local first derivative about the reference model solution is not accurately calculated. Depending upon the application, a true response change may be or may not be more desired than the exact local first derivative. On the other hand, the GRESS version of a code calculates derivatives using simple arithmetic operations resulting from the rules of differentiation. The local first derivatives are accurate to within the computational accuracy of the reference computer model.

Second, the availability of selected or complete sensitivity data as an option to a reference calculation is a valuable analysis tool. The limited selection of sensitivities shown for the sample problem reveals relationships not easily recognized even with reruns.

VI. CONCLUSIONS

The automated GRESS precompiler was used to produce the ORIGEN2G version of the ORIGEN2 code. This new version has the added capability of calculating derivatives of any user-specified real variable with respect to another variable or input parameter. By using the GRESS precompiler, we were able to automate almost all of the effort of producing ORIGEN2G and thus dramatically reduce the initial analytical effort that would have been required in programming a code of comparable capability. Furthermore, the ability to calculate all partial derivatives on demand in ORIGEN2G opens new avenues of study that would have been out of the scope of other conventional sensitivity techniques given the same time frame in which to perform the analysis.

The calculated sensitivity results are of particular interest because the long time ranges make many experimental tests impractical. Sensitivity studies such as this one will be used to understand overall uncertainties and will be an important factor in the eventual design of a high-level waste disposal site. The calculation tools presented in this paper are a first step towards achieving this understanding.

Table I. ORIGEN2 Input for Sample Problem

```
-1
-1
-1
 CUT  -1
 RDA     SPECIFY WHICH DATA BASES ARE TO BE PRINTED
 LIP     0 0 0
 RDA     READ DECAY AND CROSS SECTION DATA BASES
 LIB     0  1 2 3  204  205  206   9   3   0  1   1
 RDA     READ PHOTON DATA BASE
 PHO     101  102  103  30
 RDA     SET BASIS FOR CALCULATION
 BAS     ONE METRIC TON INITIAL HEAVY METAL
 RDA     READ INITIAL FUEL COMPOSITION
 INP     1   1   -1   -1   1   1
 HED     1   CHARGE
 RDA     BUP COMMANDS SURROUND BASIS IRRADIATION STEPS
 BUP
 RDA     IRRADIATE FUEL
 IRP     100.0  37.5  1  2  4  2
 IRP     300.0  37.5  2  3  4  0
 DEC     410.0         3  1  4  0     DECAY FOR 110 DAYS
 IRP     550.0  37.5  1  2  4  0
 IRP     690.0  37.5  2  3  4  0
 DEC     800.0         3  4  4  0     DECAY FOR 110 DAYS
 IRP     950.0  37.5  4  5  4  0
 IRP    1100.0  37.5  5  6  4  0
 BUP
 RDA     DECAY OF FUEL OVER SHORT-TERM
 DEC     60.0  6  7  4  1
 DEC     90.0  7  8  4  0
 DEC     120.0  8  9  4  0
 DEC     150.0  9  10  4  0
 RDA     ***  FUEL REPROCESSING
 RDA      REMOVE VOLATILES FROM 150-DAY-OLD FUEL
 PRO     10   1   2   2
 RDA      SEPARATE U/PU FROM HLW
 PRO      1   3   4   1
 RDA      SEPARATE U AND PU
 PRO      3   5   6   8
 HED     1   '  FUEL
 HED     2   '  VOL
 HED     3   '  U-PU
 HED     4   '  HLW
 HED     5   '  U
 HED     6   '  PU
 RDA     PRINT REPROCESSING RESULTS
 TIT      REPROCESSING VECTORS
 OPTL      6*8     8 8 8   15*8
 OPTA      6*8     8 8 8   15*8
 OPTF      6*8     8 8 8   15*8
 MOV     4  1  0  1.0
 HED     1   '  HLW
 RDA     DECAY HLW FOR ONE THOUSAND YEARS
 DEC     0.5  1  2  5  1
 DEC     1.0  2  3  5  0
 DEC     5.0  3  4  5  0
 DEC     10.0  4  5  5  0
 DEC     50.0  5  6  5  0
 DEC     100.0  6  7  5  0
 DEC     200.0  7  8  5  0
 DEC     300.0  8  9  5  0
 DEC     1.0   9  10  7  0
 RDA     PRINT HLW DECAY RESULTS
 TIT      DECAY OF PWR-U HLW
 OPTL      6*8     8 8 8   15*8
 OPTA      6*8     5 8 5   15*8
 OPTF      6*8     5 8 5   15*8
 OUT      10  1  -1  0
 END
 2   922340  290.0  922350  32000. 922380   967710.     0    0.0   FUEL 3.2%
 4   030000  1.0    050000  1.0    060000   89.4   070000  25.0   FUEL IMPU
 4   080000 134454. 090000  10.7   110000   15.0   120000   2.0   FUEL IMPU
 4   130000  16.7   140000  12.1   150000   35.0   170000   5.3   FUEL IMPU
 4   200000  2.0    220000  1.0    230000   3.0    240000   4.0   FUEL IMPU
 4   250000  1.7    260000  18.0   270000   1.0    280000  24.0   FUEL IMPU
 4   290000  1.0    300000  40.3   420000   10.0   470000   0.1   FUEL IMPU
 4   480000  25.0   490000  2.0    500000   4.0    640000   2.5   FUEL IMPU
 4   740000  2.0    820000  1.0    830000   0.4    0        0.0   FUEL IMPU
 0
```

Table II. Selected Fission-Product Concentrations at Time of Reprocessing

	Nuclides	Mass (g)
	Light Mass Nuclides	
40	Zr-90, 91, 92, 93, 94, 96	4229
42	Mo-95, 96, 97, 98, 100	3302
44	Ru-100, 101, 102, 104	2173
46	Pd-104, 105, 106, 107, 108	1202
		10906
	Heavy Mass Nuclides	
55	Cs-133, 134, 135, 137	2703
56	Ba-134, 136, 137, 138	1397
57	La-139	1214
58	Ce-140, 142, 144	2589
59	Pr-141	1112
60	Nd-142, 143, 144, 145, 146, 148, 150	3791
		12806

Table III. Fraction of Total Decay Heat as a Function of Time Following Reprocessing for Selected Nuclides[a]

Nuclide	Fraction of Total Decay Heat at									
	0.0 yr	0.5 yr	1.0 yr	5.0 yr	10.0 yr	50.0 yr	100.0 yr	200.0 yr	300.0 yr	1000.0 yr
^{238}Pu	2.02-5[b]	2.70-4	5.58-4	3.17-3	4.82-3	1.02-2	2.18-2	6.66-2	7.77-2	2.84-3
^{240}Pu	4.17-6	8.28-6	1.27-5	7.51-5	1.44-4	6.77-4	2.29-3	1.47-2	3.50-2	9.55-2
^{241}Am	3.30-4	6.32-4	9.39-4	4.47-3	7.13-3	2.02-2	5.83-2	3.19-1	6.53-1	6.26-1
^{243}Am	2.79-5	5.33-5	7.91-5	3.72-4	5.87-4	1.67-3	5.17-3	3.28-2	7.82-2	2.15-1
^{242}Cm	4.91-2	4.33-2	2.96-2	4.29-4	2.27-4	5.41-4	1.34-3	5.43-3	8.27-3	9.99-4
^{243}Cm	3.97-5	7.50-5	1.10-4	4.70-4	6.57-4	7.10-4	6.53-4	3.68-4	7.77-5	0
^{244}Cm	3.37-3	6.33-3	9.22-3	3.73-2	4.86-2	3.00-2	1.38-2	1.92-3	1.00-4	0
^{89}Sr	1.92-2	3.00-3	3.63-4	0	0	0	0	0	0	0
^{90}Sr	4.25-3	8.04-3	1.18-2	5.05-2	7.08-2	7.81-2	7.38-2	4.37-2	9.73-3	1.66-9
^{90}Y	2.03-2	3.84-2	5.63-2	2.41-1	3.38-1	3.73-1	3.52-1	2.09-1	4.65-2	7.93-9
^{91}Y	3.47-2	7.63-3	1.30-3	0	0	0	0	0	0	0
^{95}Zr	8.09-2	2.14-2	4.39-3	2.76-9	0	0	0	0	0	0
^{95}Nb	1.45-1	4.36-2	9.17-3	5.81-9	0	0	0	0	0	0
^{103}Ru	1.96-2	1.50-3	8.86-5	0	0	0	0	0	0	0
^{106}Rh	1.96-1	2.66-1	2.80-1	8.44-2	4.28-3	0	0	0	0	0
^{134}Cs	6.81-2	1.10-1	1.38-1	1.70-1	4.98-2	2.06-7	0	0	0	0
^{137}Cs	5.79-3	1.09-2	1.60-2	6.89-2	9.69-2	1.10-1	1.08-1	6.84-2	1.63-2	4.53-9
^{137m}Ba	1.94-2	3.67-2	5.39-2	2.32-1	3.26-1	3.69-1	3.61-1	2.30-1	5.48-2	1.52-8
^{141}Ce	5.01-3	1.95-4	5.90-6	0	0	0	0	0	0	0
^{144}Ce	2.57-2	3.14-2	2.99-2	4.00-3	7.34-5	0	0	0	0	0
^{144}Pr	2.84-1	3.48-1	3.31-1	4.43-2	8.14-4	0	0	0	0	0
^{154}Eu	4.57-3	8.40-3	1.20-2	4.08-2	4.31-2	4.90-3	2.71-4	5.48-7	0	0
Total Decay Heat, watts	19,577	10,237	6,899	1,464	928	324	104	16.3	6.78	2.31

[a]Nuclides with decay heat >0.1 watt; 0 is shown for values < 1.0×10^{-9}.

[b]Read 2.02×10^{-5}.

Table IV. Fraction of Total Radioactivity as a Function of Time Following Reprocessing for Selected Nuclides[a]

Nuclide	Fraction of Total Radioactivity at 0.0 yr	0.5 yr	1.0 yr	5.0 yr	10.0 yr	50.0 yr	100.0 yr	200.0 yr	300.0 yr	1000.0 yr
^{239}Np	3.77-6	7.29-6	1.05-5	4.02-5	5.74-5	1.57-4	5.00-4	4.73-3	2.96-2	1.48-1
^{238}Pu	2.66-6	3.59-5	7.21-5	3.32-4	4.57-4	9.27-4	2.04-3	9.30-3	2.85-2	1.90-3
^{239}Pu	3.48-7	6.73-7	9.73-7	3.72-6	5.34-6	1.49-5	4.86-5	4.76-4	3.09-3	1.93-2
^{240}Pu	5.83-7	1.17-6	1.75-6	8.38-6	1.45-5	6.56-5	7.28-4	2.19-3	1.37-2	6.80-2
^{241}Pu	1.37-4	2.59-4	3.65-4	1.15-3	1.29-3	5.18-4	1.53-4	5.22-5	2.55-4	1.29-3
^{241}Am	4.32-5	8.37-5	1.21-4	4.67-4	6.74-4	1.83-3	5.46-3	4.44-2	2.39-1	4.18-1
^{242m}Am	1.62-6	3.12-6	4.50-6	1.68-5	2.35-5	5.38-5	1.37-4	8.29-4	3.32-3	7.30-4
^{242}Am	1.61-6	3.10-6	4.47-6	1.68-5	2.34-5	5.35-5	1.36-4	8.25-4	3.30-3	7.27-4
^{243}Am	3.77-6	7.29-6	1.05-5	4.02-5	5.74-5	1.57-4	5.00-4	4.73-3	2.96-2	1.48-1
^{242}Cm	5.81-3	5.17-3	3.44-3	4.04-5	1.94-5	4.43-5	1.13-4	6.82-4	2.73-3	6.01-4
^{243}Cm	4.71-6	8.99-6	1.28-5	4.45-5	5.63-5	5.83-5	5.54-5	4.64-5	2.58-5	0
^{244}Cm	4.20-4	7.96-4	1.13-3	3.70-3	4.37-3	2.59-3	1.22-3	2.54-4	3.49-5	0
^{89}Sr	2.42-2	3.82-3	4.50-4	0	0	0	0	0	0	0
^{90}Sr	1.60-2	3.05-2	4.35-2	1.51-1	1.92-1	2.03-1	1.98-1	1.75-1	1.02-1	3.17-8
^{90}Y	1.60-2	3.05-2	4.36-2	1.51-1	1.92-1	2.03-1	1.98-1	1.75-1	1.02-1	3.17-8
^{91}Y	4.21-2	9.35-3	1.55-3	0	0	0	0	0	0	0
^{95}Zr	6.95-2	1.86-2	3.71-3	1.89-9	0	0	0	0	0	0
^{95}Nb	1.32-1	4.00-2	8.20-3	4.21-9	0	0	0	0	0	0
^{103}Ru	2.56-2	1.97-3	1.13-4	0	0	0	0	0	0	0
^{103m}Rh	2.31-2	1.78-3	1.02-4	0	0	0	0	0	0	0
^{106}Ru	8.92-2	1.22-1	1.25-1	3.05-2	1.40-3	0	0	0	0	0
^{106}Rh	8.92-2	1.22-1	1.25-1	3.05-2	1.40-3	0	0	0	0	0
^{134}Cs	2.91-2	4.76-2	5.81-2	5.78-2	1.54-2	6.11-8	0	0	0	0
^{137}Cs	2.28-2	4.35-2	6.22-2	2.16-1	2.75-1	3.00-1	3.03-1	2.86-1	1.79-1	9.08-8
^{137m}Ba	2.16-2	4.12-2	5.88-2	2.05-1	2.61-1	2.84-1	2.86-1	2.71-1	1.70-1	8.59-8
^{141}Ce	1.49-2	5.87-4	1.73-5	0	0	0	0	0	0	0
^{144}Ce	1.69-1	2.09-1	1.93-1	2.09-2	3.48-4	0	0	0	0	0
^{144}Pr	1.69-1	2.09-1	1.93-1	2.09-2	3.48-4	0	0	0	0	0
^{147}Pm	2.64-2	4.48-2	5.67-2	7.52-2	2.87-2	2.02-6	0	0	0	0
Total Radioactivity, Curies	4.494+6	2.327+6	1.609+6	4.217+5	2.951+5	1.076+5	3.359+4	3.522+3	5.574+2	1.041+2

[a]Nuclides with radioactivity > 1 Curie; 0 is shown for values < 1.0×10^{-9}.

[b]Read 3.77×10^{-6}.

Table V. Sensitivity of the Decay Heat of Selected Nuclides to the ^{235}U Fission Cross Section Following Reprocessing

Nuclide	Sensitivity Coefficient		
	0.0 yr	0.5 yr	1.0 yr
^{95}Zr	4.29 - 2	4.29 - 2	4.29 - 2
^{95}Nb	4.50 - 2	4.33 - 2	4.30 - 2
^{106}Rh	-3.48 - 1	-3.48 - 1	-3.48 - 1
^{134}Cs	-1.58 - 1	-1.58 - 1	-1.58 - 1
^{144}Pr	6.49 - 2	6.49 - 2	6.49 - 2
	5.0 yr	10.0 yr	50.0 yr
^{90}Sr	1.90 - 1	1.90 - 1	1.90 - 1
^{90}Y	1.90 - 1	1.90 - 1	1.90 - 1
^{134}Cs	-1.58 - 1	-1.58 - 1	-1.58 - 1
^{137}Cs	-7.96 - 3	-7.96 - 3	-7.96 - 3
^{137m}Ba	-7.96 - 3	-7.96 - 3	-7.96 - 3
	100.0 yr	300.0 yr	1000.0 yr
^{238}Pu	-7.75 - 1	-7.48 - 1	-6.00 - 1
^{240}Pu	-1.19 + 0	-1.20 + 0	-1.20 + 0
^{241}Am	-4.00 - 1	-4.00 - 1	-4.04 - 1
^{243}Am	-1.24 + 0	-1.24 + 0	-1.24 + 0

Table VI. Sensitivity of the Decay Heat of Selected Nuclides to the ^{239}Pu Fission Cross Section Following Reprocessing

Nuclide	Sensitivity Coefficient		
	0.0 yr	0.5 yr	1.0 yr
^{95}Zr	1.53 - 2	1.53 - 2	1.53 - 2
^{95}Nb	1.51 - 2	1.52 - 2	1.53 - 2
^{106}Rh	9.85 - 3	9.85 - 3	9.85 - 3
^{134}Cs	-1.33 - 1	-1.33 - 1	-1.33 - 1
^{144}Pr	-1.15 - 2	-1.15 - 2	-1.15 - 2
	5.0 yr	10.0 yr	50.0 yr
^{90}Sr	-3.54 - 2	-3.54 - 2	-3.54 - 2
^{90}Y	-3.54 - 2	-3.54 - 2	-3.54 - 2
^{134}Cs	-1.33 - 1	-1.33 - 1	-1.33 - 1
^{137}Cs	7.08 - 3	7.08 - 3	7.08 - 3
^{137m}Ba	7.08 - 3	7.08 - 3	7.08 - 3
	100.0 yr	300.0 yr	1000.0 yr
^{238}Pu	-6.15 - 1	-5.99 - 1	-5.09 - 1
^{240}Pu	-8.78 - 1	-8.80 - 1	-8.80 - 1
^{241}Am	-5.41 - 1	-5.41 - 1	-5.43 - 1
^{243}Am	-8.53 - 1	-8.53 - 1	-8.53 - 1

Table VII. Sensitivity of the Decay Heat of Selected Nuclides to the ^{241}Pu Fission Cross Section Following Reprocessing

Nuclide	Sensitivity Coefficient		
	0.0 yr	0.5 yr	1.0 yr
^{95}Zr	-8.68 - 3	-8.68 - 3	-8.68 - 3
^{95}Nb	-8.83 - 3	-8.71 - 3	-8.67 - 3
^{106}Rh	4.35 - 2	4.35 - 2	4.35 - 2
^{134}Cs	-3.82 - 2	-3.82 - 2	-3.82 - 2
^{144}Pr	-1.45 - 3	-1.45 - 3	-1.45 - 3
	5.0 yr	10.0 yr	50.0 yr
^{90}Sr	-1.17 - 2	-1.17 - 2	-1.17 - 2
^{90}Y	-1.17 - 2	-1.17 - 2	-1.17 - 2
^{134}Cs	-3.82 - 2	-3.82 - 2	-3.82 - 2
^{137}Cs	5.33 - 4	5.33 - 4	5.33 - 4
^{137m}Ba	5.33 - 4	5.33 - 4	5.33 - 4
	100.0 yr	300.0 yr	1000.0 yr
^{238}Pu	-4.40 - 1	-4.41 - 1	-4.42 - 1
^{240}Pu	-3.17 - 1	-3.19 - 1	-3.19 - 1
^{241}Am	-5.19 - 1	-5.19 - 1	-5.19 - 1
^{243}Am	-4.84 - 1	-4.84 - 1	-4.84 - 1

Table VIII. Sensitivity of the Decay Heat of Selected Nuclides to the Specific Power Following Reprocessing

Nuclide	Sensitivity Coefficient		
	0.0 yr	0.5 yr	1.0 yr
^{95}Zr	8.38 - 1	8.38 - 1	8.38 - 1
^{95}Nb	8.39 - 1	8.38 - 1	8.38 - 1
^{106}Rh	1.55 + 0	1.55 + 0	1.55 + 0
^{134}Cs	1.96 + 0	1.96 + 0	1.96 + 0
^{144}Pr	8.74 - 1	8.74 - 1	8.74 - 1
	5.0 yr	10.0 yr	50.0 yr
^{90}Sr	7.64 - 1	7.64 - 1	7.64 - 1
^{90}Y	7.64 - 1	7.64 - 1	7.64 - 1
^{134}Cs	1.96 + 0	1.96 + 0	1.96 + 0
^{137}Cs	1.00 + 0	1.00 + 0	1.00 + 0
^{137m}Ba	1.00 + 0	1.00 + 0	1.00 + 0
	100.0 yr	300.0 yr	1000.0 yr
^{238}Pu	1.86 + 0	1.71 + 0	8.64 - 1
^{240}Pu	3.43 + 0	3.45 + 0	3.45 + 0
^{241}Am	6.54 - 1	6.56 - 1	6.66 - 1
^{243}Am	3.41 + 0	3.41 + 0	3.41 + 0

Table IX. Sensitivity of the Decay Heat of Selected Nuclides to the Initial ^{235}U Loading Following Reprocessing

Nuclide	Sensitivity Coefficient		
	0.0 yr	0.5 yr	1.0 yr
^{95}Zr	1.64 - 1	1.64 - 1	1.64 - 1
^{95}Nb	1.65 - 1	1.64 - 1	1.64 - 1
^{106}Rh	-7.35 - 1	-7.35 - 1	-7.35 - 1
^{134}Cs	-4.52 - 1	-4.52 - 1	-4.52 - 1
^{144}Pr	1.61 - 1	1.61 - 1	1.61 - 1
	5.0 yr	10.0 yr	50.0 yr
^{90}Sr	3.46 - 1	3.46 - 1	3.46 - 1
^{90}Y	3.46 - 1	3.46 - 1	3.46 - 1
^{134}Cs	-4.52 - 1	-4.52 - 1	-4.52 - 1
^{137}Cs	-6.80 - 3	-6.80 - 3	-6.80 - 3
^{137m}Ba	-6.80 - 3	-6.80 - 3	-6.80 - 3
	100.0 yr	300.0 yr	1000.0 yr
^{238}Pu	-1.18 + 0	-1.11 + 0	-7.49 - 1
^{240}Pu	-2.06 + 0	-2.07 + 0	-2.07 + 0
^{241}Am	-5.26 - 1	-5.27 - 1	-5.33 - 1
^{243}Am	-2.12 + 0	-2.12 + 0	-2.12 + 0

Table X. Sensitivity of the Decay Heat of Selected Nuclides to the Irradiation Time Following Reprocessing

Nuclide	Sensitivity Coefficient		
	0.0 yr	0.5 yr	1.0 yr
^{95}Zr	1.09 - 2	1.09 - 2	1.09 - 2
^{95}Nb	7.48 - 2	2.20 - 2	1.30 - 2
^{106}Rh	3.20 + 0	3.20 + 0	3.20 + 0
^{134}Cs	4.42 + 0	4.42 + 0	4.42 + 0
^{144}Pr	1.39 + 0	1.39 + 0	1.39 + 0
	5.0 yr	10.0 yr	50.0 yr
^{90}Sr	1.78 + 0	1.78 + 0	1.78 + 0
^{90}Y	1.78 + 0	1.78 + 0	1.78 + 0
^{134}Cs	4.42 + 0	4.42 + 0	4.42 + 0
^{137}Cs	2.35 + 0	2.35 + 0	2.35 + 0
^{137m}Ba	2.35 + 0	2.35 + 0	2.35 + 0
	100.0 yr	300.0 yr	1000.0 yr
^{238}Pu	3.76 + 0	3.53 + 0	2.32 + 0
^{240}Pu	8.08 + 0	8.13 + 0	8.13 + 0
^{241}Am	2.10 + 0	2.10 + 0	2.12 + 0
^{243}Am	8.06 + 0	8.06 + 0	8.06 + 0

Table XI. Sensitivity of the Decay Heat of Selected Nuclides to the Downtime Following Reprocessing

Nuclide	Sensitivity Coefficient		
	0.0 yr	0.5 yr	1.0 yr
^{95}Zr	-1.15 - 1	-1.15 - 1	-1.15 - 1
^{95}Nb	-1.66 - 1	-1.23 - 1	-1.16 - 1
^{106}Rh	-2.17 + 0	-2.17 + 0	-2.17 + 0
^{134}Cs	-2.69 + 0	-2.69 + 0	-2.69 + 0
^{144}Pr	-1.28 + 0	-1.28 + 0	-1.28 + 0
	5.0 yr	10.0 yr	50.0 yr
^{90}Sr	-1.05 + 0	-1.05 + 0	-1.05 + 0
^{90}Y	-1.05 + 0	-1.05 + 0	-1.05 + 0
^{134}Cs	-2.69 + 0	-2.69 + 0	-2.69 + 0
^{137}Cs	-1.38 + 0	-1.38 + 0	-1.38 + 0
^{137m}Ba	-1.38 + 0	-1.38 + 0	-1.38 + 0
	100.0 yr	300.0 yr	1000.0 yr
^{238}Pu	-1.48 + 0	-1.35 + 0	-6.45 - 1
^{240}Pu	-4.66 + 0	-4.69 + 0	-4.69 + 0
^{241}Am	-9.47 - 1	-9.49 - 1	-9.63 - 1
^{243}Am	-4.67 + 0	-4.67 + 0	-4.67 + 0

Table XII. Sensitivity of Total Decay Heat to Seven Selected Parameters as a Function of Decay Time Following Reprocessing

Parameter	Sensitivity Coefficient at 0.0 yr	0.5 yr	1.0 yr	5.0 yr	10.0 yr	50.0 yr	100.0 yr	200.0 yr	300.0 yr	1000.0 yr
Irradiation Time	1.71	2.36	2.60	2.97	2.75	2.40	2.29	2.53	2.93	4.11
Downtime	-1.21	-1.63	-1.77	-1.80	-1.61	-1.39	-1.32	-1.34	-1.46	-2.20
Specific Power	1.14	1.25	1.30	1.31	1.18	1.02	0.97	1.02	1.10	1.59
Initial ^{235}U Loading	-0.14	-0.24	-0.26	-0.18	-0.07	-0.03	-0.03	-0.30	-0.67	-1.05
^{235}U Fission Cross Section	-0.09	-0.12	-0.13	-0.08	-0.04	0.01	0	-0.22	-0.46	-0.67
^{239}Pu Fission Cross Section	-0.04	-0.05	-0.05	-0.08	-0.08	-0.06	-0.08	-0.28	-0.51	-0.64
^{241}Pu Fission Cross Section	-0.02	-0.02	-0.01	-0.03	-0.04	-0.04	-0.06	-0.23	-0.43	-0.47

Table XIII. Selected Actinide-Dependent Sensitivities at 0 and 1000 Years After Reprocessing

Parameter	Sensitivity Coefficient			
	0 yr		1000 yr	
	^{238}Pu	^{241}Am	^{238}Pu	^{241}Am
Irradiation Time	5.54 + 0	2.06 + 0	2.32 + 0	2.12 + 0
Downtime	-3.06 + 0	-8.93 - 1	-6.45 - 1	-9.63 - 1
Specific Power	2.29 + 0	6.11 - 1	8.64 - 1	6.66 - 1
^{239}Pu Fission Cross Section	-3.82 - 1	-5.32 - 1	-5.09 - 1	-5.43 - 1
^{235}U Loading	-7.27 - 1	-5.18 - 1	-7.49 - 1	-5.33 - 1
^{235}U Fission Cross Section	-9.67 - 1	-4.07 - 1	-6.00 - 1	-4.04 - 1
^{241}Pu Fission Cross Section	-1.17 - 1	-5.12 - 1	-4.42 - 1	-5.19 - 1

Table XIV. Twenty Largest Absolute Changes in Decay Heat at Time of Reprocessing Per Percent Change in the Selected Data Input Parameters

Change in Decay Heat (watts)	Nuclide	Parameter
123.1	^{106}Rh	Irradiation Time
-83.6	^{106}Rh	Downtime
77.2	^{144}Pr	Irradiation Time
-71.5	^{144}Pr	Downtime
59.8	^{106}Rh	Specific Power
58.9	^{134}Cs	Irradiation Time
48.6	^{144}Pr	Specific Power
-35.8	^{134}Cs	Downtime
35.4	^{242}Cm	Irradiation Time
-28.3	^{106}Rh	Initial ^{235}U Loading
26.1	^{134}Cs	Specific Power
23.8	^{95}Nb	Specific Power
18.1	^{242}Cm	Specific Power
-13.4	^{106}Rh	^{235}U Fission Cross Section
-13.3	^{242}Cm	Downtime
13.3	^{95}Zr	Specific Power
-12.0	^{242}Cm	Initial ^{235}U Loading
9.0	^{144}Pr	Initial ^{235}U Loading
8.9	^{137m}Ba	Irradiation Time
7.5	^{244}Cm	Irradiation Time

Table XV. Twenty Largest Absolute Changes in Radioactivity at Time of Reprocessing Per Percent Change in the Selected Data Input Parameters

Change in Radioactivity (curies)	Nuclide	Parameter
12,835	^{106}Rh	Irradiation Time
12,835	^{106}Ru	Irradiation Time
10,502	^{144}Pr	Irradiation Time
10,502	^{144}Ce	Irradiation Time
- 9,729	^{144}Pr	Downtime
- 9,729	^{144}Ce	Downtime
- 8,716	^{106}Rh	Downtime
- 8,716	^{106}Ru	Downtime
6,619	^{144}Pr	Specific Power
6,619	^{144}Ce	Specific Power
6,618	^{106}Rh	Specific Power
6,232	^{106}Ru	Specific Power
5,788	^{134}Cs	Irradiation Time
4,967	^{95}Nb	Specific Power
-3,521	^{134}Cs	Downtime
-2,948	^{106}Rh	Initial ^{235}U Loading
-2,948	^{106}Ru	Initial ^{235}U Loading
2,620	^{95}Zr	Specific Power
2,562	^{134}Cs	Specific Power
2,406	^{137}Cs	Irradiation Time

Table XVI. Twenty Largest Absolute Changes in Radioactivity a Thousand Years After Reprocessing Per Percent Change in the Selected Data Input Parameters

Change in Radioactivity (curies)	Nuclide	Parameter
1.25	^{239}Np	Irradiation Time
1.25	^{243}Am	Irradiation Time
0.92	^{241}Am	Irradiation Time
-0.72	^{239}Np	Downtime
-0.72	^{243}Am	Downtime
0.58	^{240}Pu	Irradiation Time
0.53	^{243}Am	Specific Power
0.53	^{239}Np	Specific Power
-0.42	^{241}Am	Downtime
-0.33	^{240}Pu	Downtime
-0.33	^{239}Np	Initial ^{235}U Loading
-0.33	^{243}Am	Initial ^{235}U Loading
0.29	^{241}Am	Specific Power
0.26	^{99}Tc	Irradiation Time
0.24	^{240}Pu	Specific Power
-0.24	^{241}Am	^{239}Pu Fission Cross Section
-0.23	^{241}Am	Initial ^{235}U Loading
-0.23	^{241}Am	^{241}Pu Fission Cross Section
-0.19	^{239}Np	^{235}U Fission Cross Section
-0.19	^{243}Am	^{235}U Fission Cross Section

Table XVII. Sensitivities of the Decay Heat of Selected Nuclides to Specific Power and Burnup

Nuclide	Sensitivity Coefficient		
	Specific Power	Burnup	Percent Difference
^{106}Rh	1.55	1.13	-27
^{134}Cs	1.96	1.70	-13
^{137}Cs	1.00	1.00	0
^{243}Am	3.41	2.20	-35

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ABSTRACT

ORIGEN2 is a widely used point-depletion and radioactive-decay computer code for use in simulating nuclear fuel cycles and/or spent fuel characteristics. This paper presents the application of the GRESS procedure to the ORIGEN2 code for performing a sensitivity analysis of a high-level waste disposal problem. The GRESS procedure uses computer calculus and the GRESS precompiler to automate the generation and calculation of gradients in a computer code. The GRESS version of ORIGEN2 is used to calculate the nuclide-dependent sensitivities of the decay heat and radioactivity of 1,008 nuclides comprising reprocessed high-level waste to changes in data and input parameters. The sensitivities are calculated in a single execution of the revised code as compared to the conventional method of rerunning the code numerous times. The availability of sensitivity data as an option in ORIGEN2 reveals relationships not easily recognized even with reruns.