**DECAY-HEAT AND GAMMA-DOSE-RATE
PREDICTION CAPABILITY IN SPENT LWR FUEL**

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August 1982

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DECAY HEAT AND GAMMA DOSE RATE PREDICTION CAPABILITY
IN SPENT LWR FUEL

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I. SUMMARY

The ORIGEN2⁽¹⁾ code was established as a valid means to predict decay heat from LWR spent fuel assemblies for decay times up to 10,000 years. Calculational uncertainties ranged from 8.6% to a maximum of 16% at 2.5 years and 300 years cooling time respectively. The calculational uncertainties at 2.5 years cooling time are supported by experiment. Major sources of uncertainty at the 2.5 year cooling time were identified as irradiation history (5.7%) and nuclear data together with calculational methods (6.3%). These uncertainties were added quadratically to give an overall one-sigma uncertainty of 8.6%. Although limited information is available concerning experimental/calculational actinide inventory comparisons in spent LWR fuel, a comparison given within this document gives good agreement and supports decay heat calculations at long cooling times.

The QAD⁽²⁾ shielding code was established as a valid means to predict interior and exterior gamma dose rates of spent LWR fuel assemblies. A calculational/measurement comparison was done on two assemblies with different irradiation histories and supports a 35% calculational uncertainty at the 1.8 and 3.0 year decay times studied. Uncertainties at longer times are expected to increase, but not significantly, due to an increased contribution from the actinides whose inventories are assigned a higher uncertainty. The uncertainty in decay heat rises to a maximum of 16% due to actinide uncertainties. Adding the uncertainty in actinide concentrations quadratically to those already assigned in the gamma dose rate calculation would not significantly increase the overall gamma dose rate uncertainty. It should be noted that these uncertainties are stated for a properly modeled assembly geometry.

A previous study was made of the neutron emission rate from a typical Turkey Point Unit 3, Region 4 spent fuel assembly at 5 years decay time. A conservative estimate of the neutron dose rate at the assembly surface was less than 0.5 rem/hr. This is negligible with respect to the unshielded gamma dose rate. The gamma dose rate decreases much faster with time than

the neutron dose rate. However, at shorter times the dose rate due to neutrons would not be expected to increase significantly since their source is (α ,n) reactions and spontaneous fission in the actinides. It should be noted, however, that conventional gamma shielding materials such as lead and steel are not effective in attenuating neutrons.

II. INTRODUCTION

This report documents a computer code package to calculate decay heat and gamma dose rates for spent LWR fuel assemblies. Computational/experimental decay heat and gamma dose rate comparisons were made to provide additional confidence for this study. This activity is being performed by the Westinghouse Hanford Company (WHC) and is in support of programs managed by the Office of Nuclear Waste Isolation (ONWI) for the Department of Energy to demonstrate the safe disposal of commercial spent fuel. The capability to characterize spent fuel at various times throughout the disposal cycle is necessary for understanding spent fuel performance and provide information for the design of spent fuel repositories.

Although improvements in depletion codes such as ORIGEN⁽³⁾ have been made in recent years, experimental validation is desirable. This is due to uncertainties in nuclear data, reactor operating histories, and modeling approximations.

Decay heat measurements⁽⁴⁾ performed with a boiling water calorimeter on four Turkey Point Unit 3 spent fuel assemblies at the Engine Maintenance Assembly and Disassembly (EMAD) facility located within the Nevada Test Site was the first opportunity for a comparison between calculated and measured decay heat for an entire spent fuel assembly. Gamma dose rate measurements made at the Battelle Columbus Laboratories on two assemblies together with the calorimeter measurements provide a direct validation of calculations of decay heat and gamma dose rates for complete spent fuel assemblies.

III. DECAY HEAT PREDICTION CAPABILITY

Decay heat assessments can be roughly divided into two categories: short cooling times when the fission products dominate, and long cooling times when actinides are the predominate contributors.

A. RESULTS.

Analytical and measured decay heat values were compared for four Turkey Point spent fuel assemblies D34*, D04, D15, and D22⁽⁴⁾. Except for assembly D04, the agreement is excellent (6%) with respect to the 8.6% calculational uncertainty and provides strong confirmation of these decay heat calculations. A discrepancy for assembly D04 is attributed to measurement problems (see Reference 4).

B. UNCERTAINTIES ASSOCIATED WITH DECAY HEAT CALCULATIONS.

With relatively large uncertainties of 6% assigned to the assembly power and its irradiation time, the expected calculational decay heat uncertainty is nearly 13% (one sigma)⁽⁵⁾. Over half of this uncertainty arose from the power and operating history. Further study showed that a decay heat uncertainty of 5.7% can be attributed to operating parameters⁽⁴⁾. Further discussion of these uncertainties can be found in Reference 4.

If the power history of a given assembly is well known, specifically burnup, irradiation time, and capacity factor together with the uranium mass, then the uncertainties given here will apply. However, if the assembly power history is not known, new uncertainty assessments must be made. This could be done by assigning new uncertainties to the power, irradiation time, and burnup and applying Eq. (A.4) from Reference 4. These uncertainty assessments apply not only to the Turkey Point assemblies studied here but to any LWR assembly.

*Turkey Point fuel assembly serial numbers.

Actinides are formed for the most part through a series of spectrum dependent neutron captures, and subsequent decays, making uncertainty calculations difficult⁽⁵⁾. Also, few data are available on calculational/experimental comparisons of actinide inventories in spent fuel. However, a recent comparison shown in Table 1 indicates that smaller uncertainties may be assigned to actinide inventories than the conservative estimates used in Reference 5. Table 1 gives a comparison between measured and calculated uranium and plutonium isotope inventories. The measurement is described in Reference 6, and calculations were performed using the ORIGEN2⁽¹⁾ code. Figure 1 shows a plot of decay heat uncertainty vs cooling time for cooling times to 10,000 years. The largest uncertainties are for cooling times around three hundred years and arise from uncertainties in the ^{241}Pu inventory which decays to ^{241}Am with a 14.7 year half-life. For large cooling times, the uncertainties decrease to around 12% as ^{239}Pu and ^{240}Pu dominate the decay heat. Table 2 shows uncertainties assigned to the important actinides at the confidence level of one standard deviation. The ^{239}Pu inventory is assumed to be known within 10% and an additional 5% uncertainty is assigned to each neutron capture along the plutonium chain.

At discharge the ^{241}Pu inventory is some 36 times greater than that of ^{241}Am . Around 500 years cooling time ^{241}Am becomes an important contributor to the overall decay heat, well after all the ^{241}Pu has decayed to ^{241}Am . For this reason uncertainty in the neutron capture cross section of ^{241}Am is considered unimportant in assigning an uncertainty to the ^{241}Am inventory (20%).

It should be mentioned that the new ORIGEN2 code has a nuclear data library that has been processed to account for self shielding and other effects. Using another nuclear data set such as the Evaluated Nuclear Data File (ENDF/B-V), after the appropriate reactor physics processing, one would expect to obtain results as valid as those reported here with respect to the given uncertainties.

Table 1.

Comparison of Calculated and Measured U and Pu Isotopic Concentrations from Turkey Point Unit 3 Assembly B17.

U-Isotope/U Total (Atom Percent)

<u>Isotope</u>	<u>C[†]</u>	<u>E^{††}</u>	<u>(C-E)/E x100</u>
²³⁵ _U	0.725	0.728	-0.412
²³⁶ _U	0.320	0.335	-4.478
²³⁸ _U	98.955	98.923	0.034

Pu-Isotope/Pu Total (Atom Percent)

<u>Isotope</u>	<u>C</u>	<u>E</u>	<u>(C-E)/E x100</u>
²³⁸ _{Pu}	1.346	1.377	-2.251
²³⁹ _{Pu}	57.777	57.415	0.630
²⁴⁰ _{Pu}	23.147	25.193	-8.121
²⁴¹ _{Pu}	12.573	11.055	13.731
²⁴² _{Pu}	5.146	4.960	3.750
<u>Atom ratios</u>	<u>C</u>	<u>E</u>	<u>(C-E)/E x100</u>
¹⁴⁸ _{Nd} / ²³⁸ _U (X10 ⁴)	5.25	5.09	3.14
²³⁹ _{Pu} / ²³⁸ _U (X10 ³)	5.17	5.06	2.17

† Calculated value from ORIGEN2

†† Experimental value from sample H6-B. See Ref. 6, pp. 60-61.

Figure 1. Uncertainties in Decay Heat Calculations as a Function of Cooling Time.

Table 2.

Uncertainty Assignments to Important Actinide Inventories for Decay
Heat at Long Cooling Times.

<u>Isotope</u>	<u>Assigned Inventory Uncertainty</u>
^{238}Pu	10%
^{239}Pu	10%
^{240}Pu	15%
^{241}Pu	20%
^{242}Pu	25%
^{241}Am	20%

An important difference between the new ORIGEN2 code and the older version ORIGEN is the use of burnup-dependent cross sections in ORIGEN2. Also, many of the effective cross sections in ORIGEN2 are based on detailed multigroup spectrum calculations specific to the desired reactor type. The older ORIGEN code required user supplied input parameters to approximately model the neutron spectrum.

IV. GAMMA DOSE RATE PREDICTION CAPABILITY

Gamma dose rate calculation for spent fuel assemblies from Turkey Point Unit 3, a Westinghouse PWR, were made and compared with measurements made by WHC at Battelle Columbus Laboratories. Measurements were made with thermal luminescent dosimeters (TLD's) and calculations were performed with the QAD point kernel shielding code.

A. RESULTS.

A description of the QAD code and the input data needed to model these spent fuel assemblies are described fully in Reference 7.

Two assemblies were measured, a two-cycle assembly (B03) at three years decay time, and a three-cycle assembly (D04) at 1.8 years decay time. Gamma dose rates for these assemblies were measured by exposing TLD's at the locations shown in Figure 2. For the two cycle assembly (B03) thirteen-foot tubes containing TLD's were exposed in the central instrument tube, in a guide thimble tube, along the assembly surface, and one-foot above the assembly. A seven-foot tube was also exposed perpendicular to the assembly at the axial midplane. Identical measurements were made on the three cycle assembly (D04) except for the two tubes exposed within the assembly.

In Figure 3 (a) the circled measurements are part of measurements taken perpendicular to the assembly at the axial midplane (tube 5, Figure 2) at the assembly surface and at a height of one-foot. These circled values are compared with axial measurements made at the assembly surface and one-foot above (tubes 2 and 1 respectively). Agreement between the circled midplane and axial measurements is very good at the assembly surface but there is a 25% difference in the same set of measurements at a height of one-foot. The discrepancies could be due to experimental problems such as positioning of the TLD's or errors in the exposure times. Agreement between measurement and calculation is best near the assembly midplane. The measurements, however, indicate a much higher dose rate from the top half of the assembly than from the bottom half. Axial measurements inside the assembly shown

1. 12 INCHES ABOVE ASSEMBLY FLAT
2. CONTACT IN MIDDLE OF FLAT
3. GUIDE THIMBLE TUBE
4. CENTRAL INSTRUMENT TUBE
5. PERPENDICULAR TO FUEL ASSEMBLY

FIGURE 2. Locations of Gamma Dosimetry Measurements. (7)

Figure 3 (a). Axial Dose Rates for Two-Cycle Assembly after 3 Years Decay⁽⁷⁾.
The curves are calculated values. The symbols are measured values.

Figure 3 (b). Axial Dose Rates Within the Two-Cycle Assembly after 3 Years Decay⁽⁷⁾.
Except as noted all values are measured. The curves are eye guides.

in Figure 3 (b) did not show this type of distribution and were even slightly peaked toward the bottom of the assembly. The higher dose rates near the bottom can be attributed to measurement technique. The tubes containing the TLD's were inserted from the top of the assembly causing the TLD's meant for the bottom of the assembly to receive more exposure during loading and unloading. Also shown in Figure 3 (b) is the calculated axial midplane dose rate at the guide tube location. The 15% agreement between the measured and calculated dose rates indicated that QAD can be used for dose rate calculations inside the source region.

A previous study of these two-cycle assemblies used ISOSHL⁽⁸⁾ to calculate axial midplane gamma dose rates. The main difference in input between the two codes was the amount of ^{60}Co assumed to be present as an impurity. Reference 7 gives a comparison between axial midplane gamma dose rates calculated by QAD and ISOSHL. The ISOSHL calculation with no ^{60}Co contribution was only about 15% higher than measurements indicating that ISOSHL can also be used for these gamma dose rate calculations.

Gamma dose rate measurements and corresponding calculations for the three-cycle assembly D04 were made perpendicular to the assembly (tube 5, Figure 2). Agreement for the three-cycle assembly between measurement and calculation perpendicular to the assembly is not as close as for the two-cycle assembly, B03. At three feet or more from the surface of assembly D04, the measured and calculated dose rates were within 20%, however, at distances closer than three feet the calculated dose rates were about 40% higher than measured values.

Plots of measured and calculated axial gamma dose rates along the surface and at a height of one-foot above the three-cycle assembly are shown in Reference 7. The measured midplane dose rates at the assembly surface and at a height of one-foot are also included.

The TLD at the surface of the assembly in the group extending out radially at the axial midplane indicated a 33% lower dose rate than the corresponding axial measurement (see Figure 2). At a height of one-foot the TLD in the same group extending radially was about 10% higher than the corresponding axial measurement. A 10% agreement was seen between calculation and experiment axially along the assembly surface except for the upper three-feet.

B. GAMMA DOSE RATES AT LONGER COOLING TIMES.

A plot of the calculated axial midplane gamma dose rates as a function of time for the two- and three-cycle assemblies are shown in Reference 7. Only after 30 years do the actinides begin to make a small contribution to the gamma dose rate. Between 1 to 50 years, gamma dose rates are closely approximated by a straight line on a logarithmic graph and after 50 years are only 3.5% of their value at one year.

C. DISCUSSION OF UNCERTAINTIES.

The major source of uncertainty in these gamma dose rate calculations is the choice of the buildup factor. Other sources of uncertainty include the point kernel method used by QAD, the ORIGEN calculation of the gamma spectrum and intensity, attenuation coefficients, axial and radial source distributions and flux to dose rate conversion factors.

The point kernel method used by QAD is also a significant source of uncertainty. Representing the source as a discrete number of points, integrating the contributions from all these point sources at the detector location, attenuating between the source and detector and correcting these attenuated values for scattering through the use of infinite medium buildup factors, all contribute to the uncertainty in the calculated gamma dose rates.

The gamma source intensity and spectrum calculated by the ORIGEN code also contributes to the uncertainty. This is directly related to uncertainties in the amount of fission and activation products calculated by ORIGEN at the desired decay time. The ORIGEN calculation in turn depends on input values such as power, irradiation time, initial inventory, and also on the nuclear data library built into the ORIGEN code.

Combining all uncertainty sources indicates that the calculated gamma dose rate has an overall uncertainty of about 35% at cooling times around 1.8 to 3 years. At longer cooling times calculational uncertainties are expected to increase, but not more than a few percent. This is due to greater uncertainties in actinide inventories where contribution to the gamma dose rate increases with time.

V. CONCLUSIONS

The ORIGEN2 code was experimentally established as a valid means to predict decay heat from LWR spent fuel assemblies for decay times around 2.5 years. Estimated calculational uncertainties are 8.6% at this cooling time. At longer cooling times the actinides are the predominate decay heat contributors. Decay heat uncertainties rise to about 16% near 300 years cooling time and level off to about 12% at longer times as shown in Figure 1.

Uncertainties associated with an ORIGEN2 calculation can be attributed to operating characteristics as well as uncertainties in nuclear data and computational methods. If the irradiation history of a given assembly is well known, specifically burnup, irradiation time, power history, as well as uranium mass, then the uncertainties given here will apply. However, if the assembly irradiation history is not known, new uncertainty assessments must be made. This can be done by assigning new uncertainties to the power, irradiation time and burnup and applying Eq.(A.4) from Reference 4. Experimental comparisons at 2.5 years cooling time show that a better than 6% agreement with calculations can be obtained.

Decay heat assessments at long decay times are almost completely determined by the actinides⁽⁵⁾. Since the actinides arise, for the most part, from a series of spectrum-dependent neutron captures, uncertainty assignments are difficult. Comparisons of measured and calculated actinide inventories are scarce; however, a recent comparison shows good agreement and provides some confidence in decay heat predictions at long cooling times.

Due to the large uncertainty contributions from the buildup factor choice, gamma source spectrum calculated for ORIGEN, and the point kernel method used by QAD itself, an overall uncertainty of 35% is assigned to gamma dose rate calculations at cooling times around three years. At longer cooling times uncertainties would be expected to increase slightly due to uncertainties in the actinide inventories.

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