A4165

PDR-1
LPDR-WM-10 (2)
WM-11 (2)
WM-16 (2)

WM Rec File
A4165
A35D

WM Project 10, 11, 16
Docket No.
PDR
LPDR (B, N, S)

Distribution:
Chang

(Return to WM, 623-SS)

FEB 10 A11 :43

To: Kien Chang

From: Loren Zaremba

Subject: Activity Report, January 26, 1987 - February 8, 1987

In the two week period from January 26, through February 8, 1987 (Weeks 16 and 17 of my contract) I continued to search the technical literature for reports and papers relating to the subject of decay heat (also called afterheat). The Energy Research Abstracts, which are available in hard copy in the library at the Phillips building were very helpful in this regard.

The purpose of this literature search was to find studies which identified the nuclei which are the principal contributors to the decay heat during the time period of interest in the disposal of high level radioactive waste, e.g. 10 to 10,000 years after removal from the reactor in the case of spent fuel. Initially, I was only able to find studies which related to early periods, such as those of concern in reactor loss of coolant accidents. I also found several studies, such as those by Schmittroth at Hanford, which were concerned with later periods, but these were not very specific regarding the decay heat contributions of individual nuclei. I discussed the material in Shmittroth's, as well as several other studies, in my last Activity Report for January 5 through January 25, 1987. However, early in week 16 I located a report entitled "Physical and Decay Characteristics of Commercial LWR Spent Fuel", by J.W. Roddy, H.C. Claiborne, R.C. Ashline, P.J. Johnson and B.T. Rhyne of Oak Ridge National Laboratory (ORNL/TM-9591,January 1986). This report provides the information which enabled me to identify the important contibuting nuclei to the decay heat in the period in which we are interested and identify the most significant decay rates. Consequently, I was able to develop a physical basis for choosing the set of decay rates which I use in my computer program QFIT2. This is the program I use to obtain a least squares fit to a decay heat curve using a set of exponentially decaying functions. This set of functions is used in the codes which I use to predict the temperature history at various locations in the repository.

The Oak Ridge report describes a data base comprised of output from 24 ORIGEN2 runs which provides detailed information on the decay of spent fuel from PWR and BWR reactors with burnups from 33,000 to 60,000 MWd/MTU. Table 3.14 of that report shows the individual decay heat contributions from the significant nuclei at times from 1 to 10,000 years after removal from a reactor for the specific case of PWR spent fuel and a 33,000 MWd/MTU burnup. This case will be used here as an example of how the most significant decay rates can be extracted from such information.

Table 3.14 shows that at 1 year after removal, the largest contributions to the decay heat are made by Rh-106 (26%), Cs-134 (11%) and Pr-144 (34%), which have half-lives of 29.9 sec, 2.06 yr, and 17.28 min, respectively. At first it would appear that the short lived isotopes Rh-106 and Pr-144 whould not make a significant contribution at 1 year after removal. However examination of the decay schemes provided in an appendix of the Oak Ridge report reveals that Rh-106 is produced by the decay of Ru-106, which has a half-life of 368.2 days and Pr-144 is produced by the decay of Ce-144 with a half-life of 284.3 days. Consequently, these appear to be cases of secular equilibrium.

If a parent nucleus decays with a decay rate of $\lambda_1$ to an unstable daughter which has a decay rate $\lambda_2$, the general expression for the total number of daughter nuclei, $N_2$, at time t is:

$$N_2 = \frac{\lambda_1}{(\lambda_2 - \lambda_1)} N_1^0 \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right) + N_2^0 e^{-\lambda_2 t} \qquad (1)$$

where $N_1^0$, $N_2^0$ = number of parent and daughter nuclei at t = 0

(see for example Nuclear and Radiochemistry by G. Friedlander, J.W. Kennedy and J.M. Miller, 2nd ed., 1964, John Wiley and Sons, New York).

For times long in comparison to the daughter's half-life, i.e. secular equilibrium, the factor:

$$e^{-\lambda_2 t}$$

becomes negligable and the above equation reduces to:

$$N_2 = \frac{\lambda_1}{(\lambda_2 - \lambda_1)} N_1^0 e^{-\lambda_1 t} \qquad (2)$$

Consequently, the daughter activity, $\lambda_2 N_2$, and the associated decay heat will decline with the parent's half-life. This conclusion is reinforced by using the data in Table 3.14 to determine the half-life of the decay heat contributions from Rh-106 and Pr-144, which are found to be 1.0 yr and 0.8 yr, respectively.

At 10 years after removal, the largest contributors to the decay heat are Y-90 (28%), Ba-137 (27%) and Cs-137 (8%), which have half-lives of 64.1 hr, 2.52 min and 30.17 yr, respectively. However, Y-90 is produced by the decay of Sr-90, which has a half-life of 28.6 yr, and Ba-137 is produced by Cs-137. Consequently, the presence of large decay heat contributions from Y-90 and Ba-137 at ten years again appears to result from secular equilibrium. This is supported by the data in Table 3.14 which indicates half-lives of 29 yrs and 30 yrs, respectively, for the decay heat contributions from Y-90 and Ba-137.

At 100-10,000 yrs after removal, the principal contributors to the decay heat are Pu-239, Pu-240 and Am-241, with half-lives of 24,131 yrs, 6569 yrs and 432.2 yrs, respectively. At 100 yrs, Am-241 alone contributes 43% of the heat, and at 1,000 yrs the three nuclei a total of 99%. At 10,000 yrs Pu-239 and Pu-240 alone contribute 97% of the total heat.

The decline of the decay heat contributions from Pu-239 and Pu-240 are consistent with the half-lives of these nuclei. The contribution from Am-241 is interesting because it reaches a maximum between 10 and 1,000 yrs. The presence of a maximum can be explained by the fact that Am-241 arises from the decay of Pu-241 with a half-life of 14.7 yrs. By differentiation of Equation 1 above it can be shown that the maximum daughter activity will occur at a time $t_m$, given by the expression:

$$t_m = \frac{(\ln\lambda_2 - \ln\lambda_1)}{(\lambda_2 - \lambda_1)} \qquad (3)$$

In this case the equation above predicts that the maximum decay heat from the decay of Am-241 will occur after 74.2 yrs, which is consistent with Table 3.14.

The preceding analysis suggests that the initial set of decay constants, or rates, used to fit the decay heat curve should include a rate of approximately 0.693/(1yr) for the contributions from the short-lived nuclei Rh-106, Cs-134 and Pr-144, a rate of about 0.693/(30yr) for the intermediate-lived nuclei Y-90, Ba-137 and Cs-137, and rates of 0.693/(432yr), 0.693/(6569yr) and 0.693/(24,131yr) for Am-241, Pu-240 and Pu-239, respectively. The percent of the total decay heat these rates could provide would be approximately as follows:

1 yr --- 80% - 90%
10 yrs --- 75% - 85%
100 yrs --- 85%
1,000 yrs --- 99%
10,000 yrs --- 97%

Further adjustment of these five rates by the search routine in QFIT2 should result in a considerable increase in these percentages.

To check the accuracy of the decay rates derived in the preceding analysis they were used as the initial rates in a test problem using QFIT2. The test data were obtained from a decay heat curve for a Westinghouse PWR assembly (15x15) from Turkey Point with a burnup of 32,700 MWd/MTU (see "Waste Package Preliminary Reliability Analysis Report" by S.C. Yung et al.). The output from this run is attached. For convenience, the inverse of the initial decay rates, which have the more natural units of years, are specified in the input data and output rather than the decay rates, which have units of inverse years. When the five best fit decay rates are converted they correspond to half-lives of 0.5 yrs, 29.9 yrs, 386.7 yrs, 6501 yrs and 24,168 yrs, which are relatively close to the dominant values identified in the analysis of the ORIGEN2 output.

In week 17 I returned to working on the user's manual for all of my thermal analysis codes, which is now about 75% completed. I will be on vacation during week 18. I will give the completed portion of the manual to a typist so it can be started while I am gone. I hope to finish the first draft during week 19 and begin the manual for the program CONVO ,which is used to predict the failure probability of waste packages and release rates using the convolution method, in week 20.

PROGRAM QFIT TO DETERMINE A LEAST SQUARES FIT TO A TABLE OF WASTEFORM RELATIVE DECAY HEAT VS TIME USING A SET OF EXPONENTIAL DECAY FUNCTIONS. THIS VERSION (2) INCLUDES A SEARCH ROUTINE TO DETERMINE A SET OF DECAY RATES WHICH MINIMIZES THE SQUARE ERROR. THE MAIN PROGRAM DIRECTS THE SEARCH. THE SUBROUTINE QFIT CALLS THE IMSL SUBROUTINE IFLSQ TO DO THE LEAST SQUARES FIT. IT THEN COMPUTES THE SQUARE ERROR FOR THAT SET OF DECAY RATES. THE EXTERNAL FUNCTION F COMPUTES THE VALUES OF THE EXPONENTIAL DECAY FUNCTIONS AT THE TABLE TIMES.

TABLE OF TIMES AND RELATIVE DECAY HEATS

| TIME(YRS) | REL. HEAT |
|---|---|
| 0.10000E+02 | 0.10000E+01 |
| 0.15000E+02 | 0.85100E+00 |
| 0.20000E+02 | 0.76100E+00 |
| 0.25000E+02 | 0.69100E+00 |
| 0.30000E+02 | 0.63300E+00 |
| 0.40000E+02 | 0.53800E+00 |
| 0.60000E+02 | 0.40300E+00 |
| 0.80000E+02 | 0.31500E+00 |
| 0.10000E+03 | 0.25700E+00 |
| 0.20000E+03 | 0.14700E+00 |
| 0.30000E+03 | 0.11600E+00 |
| 0.40000E+03 | 0.99000E-01 |
| 0.50000E+03 | 0.86000E-01 |
| 0.70000E+03 | 0.67000E-01 |
| 0.10000E+04 | 0.49000E-01 |
| 0.20000E+04 | 0.23000E-01 |
| 0.60000E+04 | 0.13000E-01 |
| 0.10000E+05 | 0.11000E-01 |

INITIAL DECAY RATES AND SEARCH INCREMENTS

| RATE(YRS) | INCREMENT(YRS) | NO. OF INCREMENTS |
|---|---|---|
| 0.10000E+00 | 0.20000E+00 | 20 |
| 0.42000E+02 | 0.20000E+00 | 20 |
| 0.55000E+03 | 0.20000E+01 | 25 |
| 0.93500E+04 | 0.50000E+01 | 20 |
| 0.34800E+05 | 0.10000E+02 | 20 |

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BEST OVERALL FIT

RATES(YRS)=

0.500000E+00 0.432000E+02 0.558000E+03 0.938000E+04 0.348700E+05

COEFFICIENTS= 0.395917E+08 0.924664E+00 0.167698E+00 0.105004E-01
0.960099E-02

ERR= 0.89397E-02

| TIME(YRS) | REL.HEAT | FIT | PER. ERR. |
|---|---|---|---|
| 10.0 | 1.00000 | 1.00000 | 0.00001 |
| 15.0 | 0.85100 | 0.83674 | 1.67540 |
| 20.0 | 0.76100 | 0.76387 | -0.37650 |
| 25.0 | 0.69100 | 0.69881 | -1.12968 |
| 30.0 | 0.63300 | 0.64071 | -1.21830 |
| 40.0 | 0.53800 | 0.54246 | -0.82931 |
| 60.0 | 0.40300 | 0.40119 | 0.45004 |
| 80.0 | 0.31500 | 0.31041 | 1.45659 |
| 100.0 | 0.25700 | 0.25149 | 2.14463 |
| 200.0 | 0.14700 | 0.14603 | 0.65885 |
| 300.0 | 0.11600 | 0.11854 | -2.18698 |
| 400.0 | 0.09900 | 0.10153 | -2.55210 |
| 500.0 | 0.08600 | 0.08788 | -2.18409 |
| 700.0 | 0.06700 | 0.06699 | 0.01930 |
| 1000.0 | 0.04900 | 0.04671 | 4.67789 |
| 2000.0 | 0.02300 | 0.02220 | 3.45705 |
| 6000.0 | 0.01300 | 0.01363 | -4.81184 |
| 10000.0 | 0.01100 | 0.01082 | 1.60858 |