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DECAY HEAT ANALYSIS FOR AN LMFBR FUEL ASSEMBLY USING ENDF/B-IV DATA*

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Recently evaluated ENDF/B-IV fission product data have been used in decay heat calculations for typical LMFBR fuel assemblies exposed to 100,000 MWd/MT burnup. The decay heat and radioactivity of the fuel assemblies have been calculated as a function of time from discharge. Important contributors to the decay have been identified.

(Decay Heat; LMFBR; ENDF/B-IV Fission Product; CRBR; ORIGEN)

Introduction

Accurate calculation of nuclear transmutation and decay is required in a number of areas in the fast reactor fuel cycle. These include:

Sodium Void Calculations and Safety Requirements

Tyror[1] has estimated that the fission product capture rate in a fast reactor approaches 7% of the total captures in the fuel near the end of core life. This poisoning effect is energy dependent, and drastic changes in the spectrum (due for example to sodium voiding) leads to significant changes in the fission product capture rate and a positive reactivity coefficient. The uncertainties introduced in the analysis of a loss-of-coolant accident (LOCA) due to uncertainties in the basic data must also be assessed.

Waste Projections, Transportation, and Disposal

The projected accumulation of high level wastes cladding hulls, etc., at the repositories through the year 2000 have been estimated by Blomeke[2,3] to have some 18,500 (MCi) of radioactivity and a volume of 366,000 ft^3. Projected annual shipments of strategic nuclear materials, spent fuel, and wastes will number 112,000. The planning and design of methods and facilities that will be needed for handling and storage of the spent fuel and reprocessing wastes require accurate analysis for the composition, radioactivity, thermal power, and gamma and neutron release rates of the material. Claiborne[4] and Croff[5] have investigated the effect on the nuclear fuel cycle of recycling the actinide wastes. Croff indicates that including recycled actinides in the nuclear fuel cycle would require substantial changes in the nuclear fuel fabrication and shipping sectors. Concrete shield thicknesses in a fuel fabrication plant may have to be tripled because of the highly active neutron sources in the fuel.

The production of these heavy, radioactive isotopes involves a long series of neutron captures; accurate calculation requires knowledge of the spectrum and the energy dependence of the reaction cross sections of all competing reactions as well as branching ratios, half lives, and yields.

Nuclear Safeguards and Accountability

In a mature nuclear industry, large amounts of irradiated fuel from several reactors will be shipped to the reprocessing plant. In order to close the material balance and to deter diversion at the reprocessing stage, attention is being given to a number of proposals for nondestructive assay of the irradiated fuel. Direct measurement at the spent fuel pins or bundles requires accurate assessment of the background in which the measurement is to be made[6].

Fuel Cycle Optimization

As fuel is discharged from the reactor, it is desirable to begin reprocessing as soon as possible[7]. Short cooling times can, however, lead to "hostile" environments for chemical processing. Decay heat calculations are required to minimize "fissile holdup" while permitting safe fuel handling and transportation design.

In this paper, we focus our attention on the calculation of composition, radioactivity, and thermal power of irradiated CRBR fuel subassemblies using the recently available ENDF/B-IV data library. Extensive calculations[8-9] have been made previously of the properties of irradiated fuel from power reactors. However, these calculations did not include the comprehensive data library now available. Different nuclear data sets can produce significant differences in fast reactor static and dynamic parameters. Kee et al.,[10] have reported 20% differences in the decay heat one second after irradiation. Ilberg[11] reports 30% differences in the sodium void coefficient when using different product cross section sets. The results and experience obtained using the new ENDF/B-IV library for these problems are compared to similar studies made using the older ORIGEN library.

Data Base

Calculations of nuclear transmutation and decay require basic cross section and decay information for light element, fission-product, and actinide nuclides. An extensive fission-product data library in the ENDF/B-IV format has resulted from a two-year effort by the CSWEG Fission Product Task Force. The formulation and content of this library is discussed in more detail in another paper[12] of this conference.

Table I presents a comparison of the new fission-product library with the ORIGEN[13] fission-product library. The original fission-product cross-section formats in ORIGEN allowed only (n,γ) reactions, but the code can treat other reactions. Similar characteristics are provided in Table II for the ORIGEN light element (LMFBR) and heavy metal libraries. The paucity of data for nuclear transmutation of light and heavy nuclides for ENDF/B-IV is such that inclusion in the table was deemed unnecessary.

The LMFBR cross-sections in the ORIGEN libraries, until the present work, were based on a fixed reference spectrum[14]. We have processed the ENDF/B-IV fission products' capture cross section into 124 groups using the MINX[15] code. The ORIGEN program was then modified to derive group collapsed reaction rates "on-line" with a user specified reference spectrum[16].

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Such capability permits more credible problem dependent analysis (e.g., foil and/or capsule measurements designed for dosimetry purposes.)

Table I.
Comparison of ORIGEN and ENDF/B Fission Product Data Library for the LMFBR

	ENDF/B-IV	ORIGEN
No. of nuclides	825	461
Radioactive	712	338
1st excited state	117	83
2nd excited state	7	-
Delayed neutron precursors	57	-
Alpha decay	6	0
Positron decay	17	11
Cross sections	181	423*

*1 group (n,γ) cross sections.

Table II.
ORIGEN Light Element and Heavy Metal Data Libraries for the LMFBR

	Light Elements	Heavy Metal
No. of nuclides	162	101
Naturally occurring	80	-
Radioactive	81	96
Nuclides with 1 group cross sections	142	53
Excited state	5	3
Alpha decay	1	58
Positron decay	14	2
Spontaneous fission	-	10

Methods and Data Testing

In a cooperative effort with HEDL under the auspices of the CSWEG Fission Product Task Force, the fission product decay heat following thermal neutron fission of ^{235}U was calculated[16] with the ORIGEN and RIBD[17] codes using the same ENDF/B-IV data base. This problem was picked because of its importance to the analysis of the loss-of-coolant accident (LOCA) in light water reactors, because the "zero flux", infinite irradiation benchmark case is "clean" in the sense that cross section and spectral complications are unimportant, and finally because it was thought that the myriad of new data, particularly those of importance at early times, could shed some light on the present credibility of summation calculation results at early times (> 100 seconds after shutdown).

Calculations using the ENDF/B-IV data were performed for several different irradiation times and flux levels. The total (β + γ) integral afterheat for a 60,000 day irradiation (∿ infinite) at a flux level of 10^6 n/cm^2-sec (low enough to neglect capture) is given in Table III along with ORIGEN calculations using earlier data,[18] the Perry et al.[19] evaluation, and the ANS-5 standard.[20] The calculations of ORIGEN and RIBD, using the ENDF/B-IV data base, agree to within the three figures printed on output. The largest difference between these calculations and the Perry et al., evaluation is 11% for times less than one second following discharge. At ten seconds, the difference is less than 4%. The calculated results will increase if transitions to second excited states are properly included.

Table III.
Fission Product Afterheat, (MeV/sec)/(fission/sec), for Thermal Neutron Fission of ^{235}U Following "Infinite" Operation at Constant Power

Time after Shutdown t (sec)	Total Afterheat (β + γ) ORIGEN (Earlier Data)	ANS-5	Perry et al.	ORIGEN/RIBD
1×10^0	10.0	12.50	13.52	11.72
1×10^1	9.05	10.00	10.05	9.44
1×10^2	6.82	6.62	6.54	6.35
1×10^3	4.00	3.70	3.941	3.93
1×10^4	2.01	1.93	2.001	1.93
1×10^5	1.05	0.95	1.012	.962
1×10^6	.583	0.534	0.561	.545

We conclude that for decay heat calculations of this type, ORIGEN and RIBD give essentially the same results (provided they start from the identical data base) even though the calculations use different numerical methods. The data represented by the ENDF/B-IV evaluated decay file represent a substantial improvement over previous libraries which are associated with codes of this type. Isotope generation and depletion calculations based upon such extensive libraries are feasible for determining beta and gamma energy release rates. The uncertainties associated with the proposed ANS-5[20] standard appear to be conservative.

Calculational Method - ORIGEN

The present calculations have been performed using the ORIGEN isotope generation and depletion code which uses the matrix exponential method to solve the linear differential equations for nuclear transmutation and decay which can be expressed as:

$$\dot{X} = AX(t) \tag{1}$$

where:

$$A = (B-I)\Lambda + (T-I)\Sigma\phi \tag{2}$$

and:

B = Decay transition matrix with b_{ij} = fraction of decays of nuclide j which produce nuclide i
I = Identity matrix
T = Transmutation matrix with t_{ij} = fraction of neutron captures with j to produce i
Λ and Σ = Diagonal matrices whose ith elements are λ_i and σ_i, respectively where
λ_i = decay constant (sec^{-1}) and
σ_i = capture cross sections (cm^2/atom)
$\dot{X}(t) = dX/dt$ (3)
ϕ = Scalar flux (n/cm^2-sec)

The solution is given by:

$$X(t) = X(0)\exp(-At) \tag{4}$$

A discussion of the numerical technique in ORIGEN is presented by Bell.[13]

Application to Fuel Storage Facility Problem

The fuel storage and test facility is designed to store irradiated fuel from the CRBR and FFTF as it becomes available. In order to properly design a fuel storage facility, a knowledge of the decay heat and radioactivity associated with the irradiated fuel is required input information. This study was undertaken to provide that information for equilibrium core

CRBR fuel assemblies irradiated to 100,000 MWD/MT burn-up.

The design model of the CRBR core[2] employed in this work contains 198 fuel assemblies arranged in two zones with the outer zone of 2 rows of assemblies having a higher enrichment than the inner zone to improve power flattening. Both radial and axial reflectors surround the core. The core itself is 36" in height with the upper and lower axial reflector each of 14" in height.

A fuel assembly is a hexagonal structure of 316 stainless steel about 4 1/2" flat to flat containing 217 fuel rods of Uranium-Plutonium dioxide pellets. Each rod is 0.23" in diameter and has a cladding thickness of 15 mills. Each fuel assembly consists of three distinct regions, an upper axial blanket, a core region, and the lower axial blanket. The fuel assemblies were specified as three regions for the ORIGEN calculations and the results were blended by the code to obtain an effective fuel assembly.

The power history used for the calculations was based on the equilibrium fuel management plan for the CRBR which has a goal of 274 full power days of operation (.75 capacity factor) per fuel cycle. Each subassembly will remain in the core for three cycles.

Conclusions

The results of the ORIGEN decay heat calculations for the CRBR core fuel assemblies are given in Table IV and the total activities are given in Table V. Also shown are results calculated from previous ORIGEN data. From Table IV, it can be seen that fission products are the major contributors to decay heating in CRBR fuel. The largest changes due to the use of ENDF/B-IV data occurs at early times (i.e., the first 1000 seconds following shutdown). At longer times, there is approximately 9% difference between results obtained using the previous ORIGEN fission product library and results obtained by using ENDF/B-IV fission product data. In Table V, the total activities calculated from ENDF/B-IV data are approximately the same as the total activities calculated from the previous ORIGEN library.

A list of major contributors to fission-product decay heating is given in Table VI. It should be noted that for all times listed these 17 nuclides collectively contribute more than 90% of the total. Furthermore, nuclides from three mass chains (95, 106, and 144) contribute 56.7% at 30 days, 78.2% at 90 days, 85.2% at 180 days, and 76.8% at 720 days. Thus, only a small set of nuclides are crucial for decay heat calculations in a fuel storage facility.

Table IV
Thermal Power (kW) of a CRBR Core Fuel Assembly as a Function of Time from Discharge

Time (days)	Light Elements and Actinides	Fission Products	
		ENDF/B-IV Library	Previous ORIGEN Library
0	14.74	241.00	175.00
10	1.38	9.32	9.95
20	1.00	6.95	7.54
30	0.99	5.72	6.24
60	0.80	3.99	4.36
90	0.66	3.19	3.49
180	0.39	2.02	2.20
360	0.19	1.15	1.25
720	0.10	0.57	0.63

Table V
Activity (MCi) of a CRBR Fuel Core Assembly as a Function of Time from Discharge

Time (days)	Light Elements and Actinides	Fission Products	
		ENDF/B-IV Library	Previous ORIGEN Library
0	6.560	20.90	17.70
10	0.288	2.29	2.23
20	0.120	1.77	1.75
30	0.104	1.49	1.49
60	0.089	1.05	1.07
90	0.078	0.82	0.84
180	0.059	0.49	0.51
360	0.045	0.29	0.30
720	0.036	0.15	0.16

An extensive analysis of errors was not undertaken in this study. However, based on the calculated decay heats, the data parameters which most influenced the total fission-product decay heat at the times given in Table IV were identified as:

1. Yields for 95, 106, and 144 mass chains.
2. Decay constants for ^{95}Zr, ^{95}Nb, ^{106}Rh, and ^{144}Ce.
3. Q-values for ^{95}Zr, ^{95}Nb, ^{106}Rh, and ^{144}Pr.

Captures in the fission products lead to both destruction and production of the important nuclides. However, the most important fission-product cross-sections for this problem are the total capture cross-sections of Zr-95, Nb-95, Ru-106, and Ce-144; and the n-gamma cross-sections of Zr-95, Ru-105, and Ce-143.

Table VI
Percentage Contribution to Decay Heat of Significant* Fission-Product Isotopes Discharged CRBR Core Fuel

Time since discharge from reactor (days)

	30	60	90	180	720
Sr-89	2.9	2.8	2.8	1.1	0.0
Y-90	0.4	0.6	0.8	1.2	4.2
Y-91	4.3	4.3	3.8	2.1	0.0
Zr-95	11.1	11.6	3.8	6.4	0.1
Nb-95	13.1	16.1	10.5	11.7	0.2
Ru-103	8.7	7.4	5.5	1.8	0.0
Rh-106	18.0	24.5	28.9	38.6	49.5
Cs-134	0.9	1.2	1.5	2.2	4.6
Cs-137	0.3	0.3	0.4	0.6	2.1
Ba-137m	0.8	1.1	1.4	2.2	7.6
Ba-140	2.0	0.6	0.1	0.0	0.0
La-140	12.6	3.6	0.9	0.0	0.0
Ce-141	2.6	2.0	1.3	0.3	0.0
Ce-144	1.2	1.5	1.8	2.3	2.2
Pr-143	1.4	0.4	0.1	0.0	0.0
Pr-144	13.3	17.8	20.7	26.2	24.8
Pm-147	0.2	0.3	0.3	0.5	1.2
Others	7.2	4.0	3.4	2.7	3.6

*Greater than 1% contribution to total heat generation at any listed time.

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