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COOLING-TIME DETERMINATION OF SPENT FUEL

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ABSTRACT

Two methods to determine the cooling time using data from high-resolution gamma-ray measurements are discussed; one is useful when the irradiation history information is available, the other when the irradiation history is not available. We have applied both methods and found that the cooling time can be determined to within an average of 3% and 4.1%, respectively.

I. INTRODUCTION

To inspect the irradiated fuels discharged from a reactor, both the burnup and cooling time of an assembly should be verified. The burnup measures the number of fissions that has occurred in the fuel residing in the core; the cooling time indicates how long the fuel has been discharged from the reactor core. This report discusses the methods for cooling-time determination in general and describes two methods using data from high-resolution gamma-ray measurements to determine the cooling time; one is useful when the irradiation history information <u>is</u> available, the other when the irradiation history information <u>is not</u> available. We have applied both methods to Omega West Reactor (OWR) spent-fuel elements, which are 93 per cent enriched initially, and found that the cooling time can be determined to within an average of 3% and 4.1% respectively.

II. METHOD OF COOLING-TIME DETERMINATION

To determine the cooling time nondestructively it is necessary to measure the activity ratio of two radioactive fission product nuclides. Activity ratios can be measured easily without knowing the absolute detector efficiency.

Consider a fission product nuclide that is a burnup monitor after a cooling time of t_c. These nuclides are produced directly from fission (or have very short-lived precursors) and have small absorption cross sections. The detector-efficiency-corrected activity (A_j) of a particular gamma-ray branch emitted from the nuclide having an absolute branching ratio of B_j is given by

$$A_j = \lambda_j N_j B_j e^{-\lambda_j t_c} \tag{1}$$

where λ_j is the decay constant of the nuclide and N_j the number of atoms of the nuclide at the time of discharge. For a different burnup monitor nuclide, we have

$$A_i = \lambda_i N_i B_i e^{-\lambda_i t_c} \tag{2}$$

If the decay constants of the two monitors are different, we can divide Eq. (1) by (2) and solve for t_c to obtain an equation for the cooling time

$$t_c = \frac{1}{\lambda_j - \lambda_i} \ln \frac{A_i}{A_j} \frac{\lambda_j N_j}{\lambda_i N_i} \frac{B_j}{B_i} \tag{3}$$

This equation is valid for a $^{95}Zr/^{137}Cs$ isotopic ratio. The ^{106}Rh and ^{144}Pr are short-lived isotopes that are in equilibrium with longer-lived parents. For the $^{106}Rh/^{137}Cs$ and $^{144}Pr/^{137}Cs$ activity ratios, the cooling-time equation becomes

$$t_c = \frac{1}{\lambda_j - \lambda_{ip}} \ln \frac{A_i}{A_j} \frac{\lambda_j}{\dfrac{\lambda_{ip}}{\lambda_i - \lambda_{ip}} \cdot \lambda_i} \frac{N_j}{N_{ip}} \frac{B_j}{B_i} \tag{4}$$

where N_{ip} and λ_{ip} are the number and decay constant of the parent of ith nuclide (^{106}Ru or ^{144}Ce). Notice that it is the $^{106}Ru/^{137}Cs$ or $^{144}Ce/^{137}Cs$ nuclide ratio that enters Eq. (4). When $\lambda_i \gg \lambda_{ip}$, Eq. (4) becomes the same as Eq. (3) except the number of nuclides and the decay constants of their respective parents should be used. The approximation of Eq. (3) is valid to better than 0.01% for both ^{106}Ru and ^{144}Ce.

Another isotopic ratio of interest that can be used for cooling-time determination is the $^{95}Zr/^{95}Nb$ ratio. ^{95}Nb is not a direct fission product and arises only from the decay of ^{95}Zr. ^{95}Nb decays with a 34.97-day half-life and emits a gamma ray at 765.84 keV that can easily be measured. When the reactor is in operation, the $^{95}Zr/^{95}Nb$ approaches a constant ratio; when the fuel is removed from the core, the $^{95}Zr/^{95}Nb$ ratio approaches a different constant because of the ceased production of ^{95}Zr. The cooling-time equation for this ratio is

$$t_c = \frac{1}{\lambda_{jp} - \lambda_i} \ln \frac{\dfrac{A_i}{A_{jp}} \dfrac{B_{jp}}{B_i} + \dfrac{\lambda_i}{\lambda_{jp} - \lambda_i}}{\dfrac{\lambda_i}{\lambda_{jp}} \dfrac{N_i}{N_{jp}} + \dfrac{\lambda_i}{\lambda_{jp} - \lambda_i}} \tag{5}$$

Equations (3-5) can all be used for cooling-time determination.

III. CALCULATION OF NUCLIDE RATIOS AT DISCHARGE

To determine the cooling time of the spent fuel, it is necessary to know the ratio at the time of discharge of two burnup monitor nuclides having substantially different half-lives. This nuclide ratio can be calculated, in some cases easily calculated. Assume that the reactor is operated at constant power over an irradiated period of t_i and assume that only one fissioning nuclide (^{235}U, for example) is dominant in the fuel. The number N_j of jth nuclide at the end of irradiation is proportional to

$$N_j \; \alpha \; \phi\sigma_f Y_j (1 - e^{-\lambda_j t_k}) \frac{1}{\lambda_j} \tag{6}$$

where σ_f is the fission cross section and Y_j is fission yield of the nuclide. The ratio of the jth and ith nuclides is proportional to

$$\frac{N_j}{N_i} \; \alpha \; \frac{\lambda_i Y_j}{\lambda_j Y_i} \; \frac{(1 - e^{-\lambda_j t_k})}{(1 - e^{-\lambda_i t_k}} \tag{7}$$

For multiple irradiation cycles as shown in Fig. 1, the ratio is proportional to

$$\frac{N_j}{N_i} \; \alpha \; \frac{Y_j \lambda_i \Sigma_k (\phi_k/\bar{\phi}) (1 - e^{-\lambda_j t_k}) \; e^{-\lambda_j \theta_k}}{Y_i \lambda_j \Sigma_k \; \phi_k/\bar{\phi}(1 - e^{-\lambda_i t_k}) \; e^{-\lambda_i \theta_k}} \tag{8}$$

where $\bar{\phi}$ is the averaged neutron flux in the reactor and θ_k is the time from the end of the kth irradiation cycle to the time of discharge.[1] For constant power, single cycle irradiation, the nuclide ratio is independent of the reactor flux and fission cross section but depends on fission yields and the irradiation history. For multiple cycles, the nuclide ratio is dependent on the flux or power level ratios between cycles, not the absolute flux in the reactor. Therefore if the irradiation history of the reactor is known (cycle length, relative power level, and shutdown time), the nuclide ratio at the time of discharge can be calculated and the cooling time of the fuel can then be determined by Eq. (3-5).

When there is more than one dominant fissioning nuclide in the fuel, the calculation of the N_j/N_i nuclide ratio is more involved, and a computer program COLDET has been written to perform the calculation. The program was patterned after a code developed at the Battelle Pactific Northwest Laboratories.[2] The program takes into account the fissioning of ^{235}U, ^{238}U, and ^{239}Pu and the absorption of the neutrons by the fission products during the irradiation. (The neutron flux is assumed to be constant during each irradiation cycle.) But the salient feature of the nuclide ratio remains; it is rather independent of the absolute flux of the reactor and the fission cross sections.

To investigate the sensitivity of the nuclide ratios to the flux and to the operational history of the reactor, we have performed the following calculations using the COLDET code. Thermal cross sections were used. First, for a fixed irradiation history, we calculated the nuclide ratios at discharge by varying the flux from (2.4 to 6.0) x 10^{13} n/cm$^2\cdot$s. The ^{95}Zr/^{137}Cs, ^{144}Ce/^{137}Cs, ^{106}Ru/^{137}Cs, and ^{95}Zr/^{95}Nb ratios vary by less than 4%, even though the absolute number of each nuclide has increased substantially. This calculation demonstrates that the nuclide ratios at discharge are not very sensitive to the flux or power level of the reactor.

To investigate the sensitivity of the nuclide ratios at discharge to irradiation history, we have performed the following calculation. OWR fuel is allowed to burn up to 29% of the initial fissile content according to a continuous and uniform irradiation cycle (irradiation history A) or according to intermittent irradiation cycles (irradiation histories B, C, D) as shown in Fig. 2. The COLDET code was used to calculate the nuclide ratios at the time of discharge for each of the four irradiation histories, and the results are tabulated in Table I. The ^{144}Ce/^{137}Cs and ^{106}Ru/^{137}Cs ratios vary by no more than 2% indicating they are rather insensitive to the detailed irradiation history, whereas the ^{95}Zr/^{137}Cs and ^{95}Zr/^{95}Nb ratios vary substantially indicating these ratios require accurate detailed irradiation history to determine the cooling time.

TABLE I

NUCLIDE RATIOS AT DISCHARGE
BASED ON VARIOUS IRRADIATION HISTORY

Irradiation History	Nuclide Ratio			
	^{95}Zr/^{137}Cs	^{144}Ce/^{137}Cs	^{106}Ru/^{137}Cs	^{95}Zr/^{95}Nb
A	0.237	0.559	0.045	1.814
B	0.261	0.568	0.045	2.808
C	0.240	0.552	0.044	3.077
D	0.264	0.568	0.045	2.407

We should point out that if the calculation were performed on LWR spent fuels, the same general trend should prevail, with the exception that the ^{106}Ru/^{137}Cs nuclide ratio would be more irradiation-history-dependent because of the drastically different fission yield from ^{235}U and ^{239}Pu.

That the ^{144}Ce/^{137}Cs nuclide ratios are rather insensitive to the detailed irradiation history suggests an approximate method for the cooling time determination. The following discussion describes such an approximate method. For nuclear power reactors, the shutdown time is normally one to two months per year for refueling and maintenance purposes. If the half-life of the fission product is long compared with the shutdown time of the reactor, the decay of the fission product during the shutdown is relatively small and is considered negligible in the approximation. The approximation is valid for ^{144}Pr (half-life of parent ^{144}Ce, 284.4 days) and ^{137}Cs (half-life, 30.12 yr) except for extremely long shutdowns. Therefore in calculating the nuclide ratios involving these two fission products, the reactor can be considered as operating at constant power from the time the fuel is inserted into the reactor until the time of discharge. Under this approximation, the cooling time of the nuclear fuel can be obtained simply by inserting Eq. (7) into (3)

$$t_c = \frac{1}{\lambda_j - \lambda_i} \ln \frac{A_i}{A_j} \frac{Y_j B_j}{Y_i B_i} \frac{(1 - e^{-\lambda_j t_k})}{(1 - e^{-\lambda_i t_k})} \qquad (9)$$

where t_k is now defined as the irradiation time as shown in Fig. 3. The cooling time, the irradiation time, and the activity ratio are simply related, as indicated in Fig. 4. For a measured ^{137}Cs/^{144}Pr activity ratio (deduced from the 662- to 696-keV gamma peak ratios, corrected for detector relative efficiency), the irradiation time uniquely determines the cooling time of the spent fuel.

IV. APPLICATION TO OWR SPENT FUEL.

A. Cooling Time if Complete Irradiation History is Known

We have calculated the cooling time of OWR spent fuels using the operator-declared irradiation history and the COLDET program; the results are summarized in Table II. The relative detector efficiency was determined by the various gamma rays of ^{134}Cs and ^{144}Pr using the established branching ratios. For each of the September and January exercises, data from three fuel elements were averaged to obtain the relative detector efficiency curve. The efficiency curve was then used to correct the peak areas corresponding to the gamma rays of interest. For the ^{95}Zr/^{137}Cs, ^{144}Pr/^{137}Cs, ^{106}Rh/^{137}Cs activity ratios the efficiency-corrected peak-area ratios of 756 keV/662 keV, 696 keV/662 keV, and 622 keV/662 keV were used respectively. The ^{95}Zr/^{95}Nb ratio was not used because of the relatively long cooling time encountered (500-1500 days). From the table we observe that the ^{144}Pr/^{137}Cs activity ratio gives the best cooling-time determination with an averaged difference of 3.0%. This is the best activity ratio to use for cooling times between 400 and 1500 days. It should be observed that for shorter cooling time the ^{95}Zr/^{137}Cs activity ratio can also be used to determine the cooling time of spent fuel with good precision. The activity of ^{95}Zr drops off rapidly because of its 65-day half-life and after 800-1000 days, there is hardly any ^{95}Zr activity to be measured, resulting in large percentage differences of cooling time determined with the ^{95}Zr/^{137}Cs activity ratio.

TABLE II

COOLING TIME BASED ON ACTIVITY RATIOS MEASURED WITH Ge(Li) DETECTOR

Element	Cooling Time	Cooling Time Based on Activity Ratio					
	(Day)	^{95}Zr/^{137}Cs		^{144}Pr/^{137}Cs		^{106}Rh/^{137}Cs	
		Day	% Diff.	Day	% Diff.	Day	% Diff.
356	1350	902	-33.2	1368	1.3	1314	-2.7
359	1268	888	-30.0	1296	2.2	1481	16.8
361	1202	898	-25.3	1149	-4.4	1232	2 5
363	1204	879	-27.0	1225	1.7	1208	0.3
364	1091	936	-14.2	1133	3.8	1513	38.7
368	947	1075	13.5	940	-0.7	941	-0.6
370	891	902	1.2	849	-4.7	1025	15.0
371	890	854	-4.0	865	-2.8	1407	58.1
372	750	790	5.3	758	1.1	797	6.3
378	554	565	2.0	526	-5.1	666	20.2
357	1456	950	-34.8	1506	3.4	1479	1.6
359	1374	976	-29.0	1397	1.7	1351	-1.7
368	1053	1871	77.7	1057	0.4	1014	-3.7
370	998	904	-9.4	1021	2.3	1024	2.6
373	858	834	-2.8	918	7.0	915	6.6
374	438	438	0.0	432	-1.4	394	-10.0
375	683	687	0.6	728	6.6	785	14.9
379	662	678	2.4	689	4.1	629	-5.0
Averaged Percent Difference			17.4		3.0		11.5

B. Cooling Time if Complete Irradiation History is Not Known

The procedure outlined above is useful if the detailed irradiation history is known. However, in various situations the irradiation history may not be available. In such a case the approximate method described by Eq. (9) that requires minimal irradiation history information and yet has reasonable precision is useful for the cooling-time determination. This equation is simple to apply because it does not require data on either the shutdown or the power level of the reactor. One must know only the total time interval from the time the fuel is inserted into the reactor core until the time of discharge, namely the irradiation time. It is interesting to see how well this simple approximate method works.

Table III lists the results of applying this simple method to the OWR data. In general the agreement with the declared cooling times is quite good. The main reason that the percentage difference is large for fuel element 374 is that this element had been removed from the core for 314 days before reirradiation, making the approximation no longer valid. It should be noted that for this element the cooling time determined by including the shutdown decay correction differs from the declared value by only 1.4% (Table III).

TABLE III

COOLING TIME BASED ON APPROXIMATE METHOD AND ^{144}Pr/^{137}Cs ACTIVITY RATIO

Element	Irradiated Time (Day)	Measured $\frac{A(^{137}Cs)}{A(^{144}Pr)}$	Declared Cooling Time (Day)	Measured Cooling Time (Day)	Percent Difference
356	501	79.606	1350	1350	0.0
359	541	74.528	1268	1306	3.0
361	587	55.311	1202	1164	-3.2
363	553	64.895	1204	1244	3.3
364	636	57.145	1091	1159	6.2
368	751	40.317	947	970	2.4
370	764	32.129	891	870	-2.4
371	764	33.370	890	886	-0.4
372	903	28.212	750	769	2.5
378	995	16.003	554	500	-9.7
357	501	112.086	1456	1495	2.7
359	541	94.767	1374	1408	2.5
368	751	53.256	1053	1088	3.3
370	764	48.240	998	1041	4.3
373	903	40.805	858	924	7.7
374	1266	18.878	438	492	12.3
375	1023	27.960	683	727	6.4
379	995	22.912	662	652	-1.5
			Averaged Percent Difference		4.1

V. CONCLUSION

If the detail irradiation history is available, then either $^{95}Zr/^{137}Cs$ or $^{144}Pr/^{137}Cs$ activity ratios can be measured to determine the cooling time, the former applicable to a cooling time range of 30 to 800 days and the latter suitable for 150 to 1500 days (probably longer) cooling. If the detail irradiation history is not available, then the $^{144}Pr/^{137}Cs$ activity ratio still can be applied provided the shutdown time is less than half of the half-life of ^{144}Pr (284.4 days). We feel that this simple method of cooling-time determination, even though it is slightly less accurate than the more exact method, is of value to inspectors because it requires less information from the operator. The method can be easily applied durng the inspection exercise to obtain a preliminary cooling-time determination. In the case of incomplete irradiation history information, the approximate method may be the only method that can be used.

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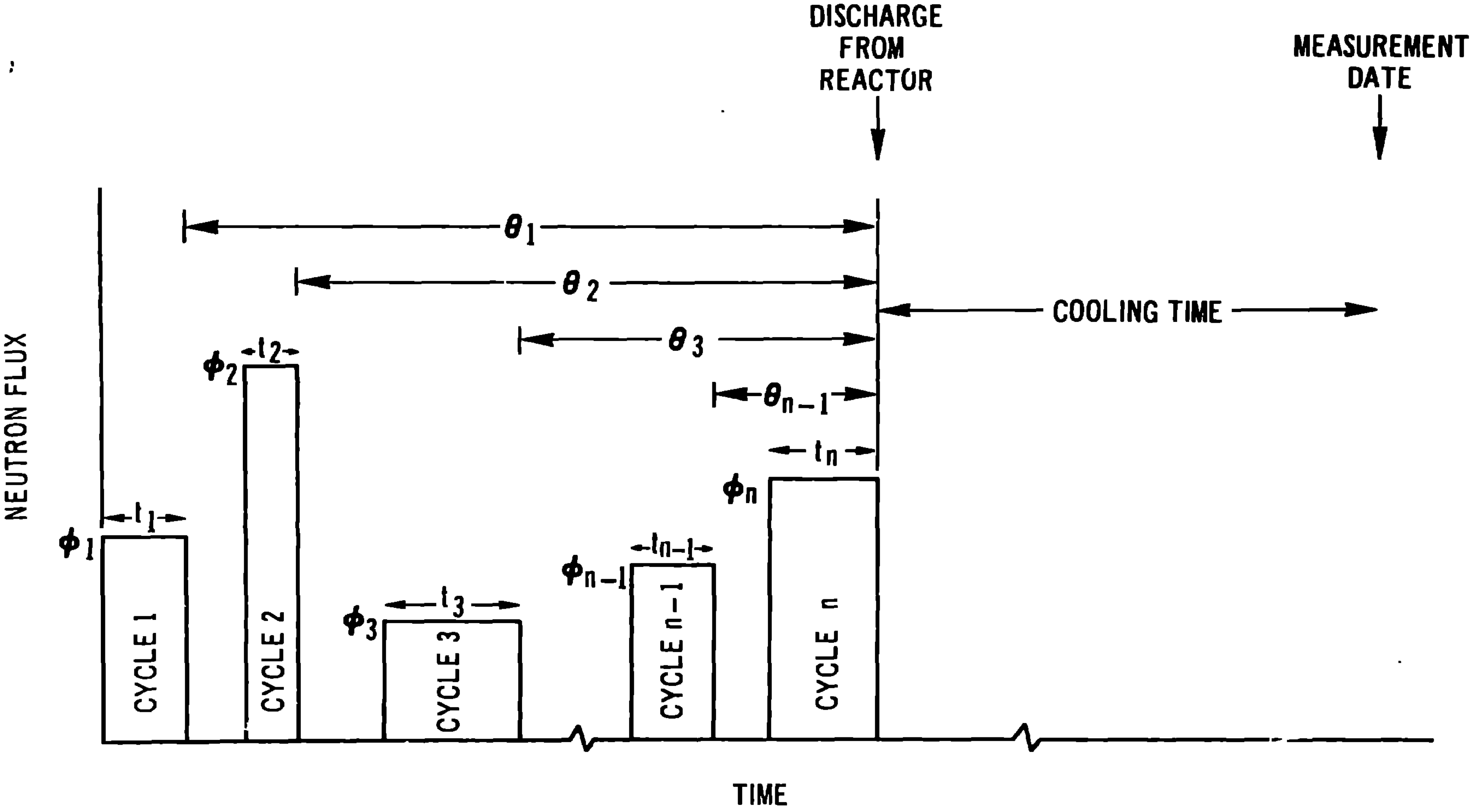
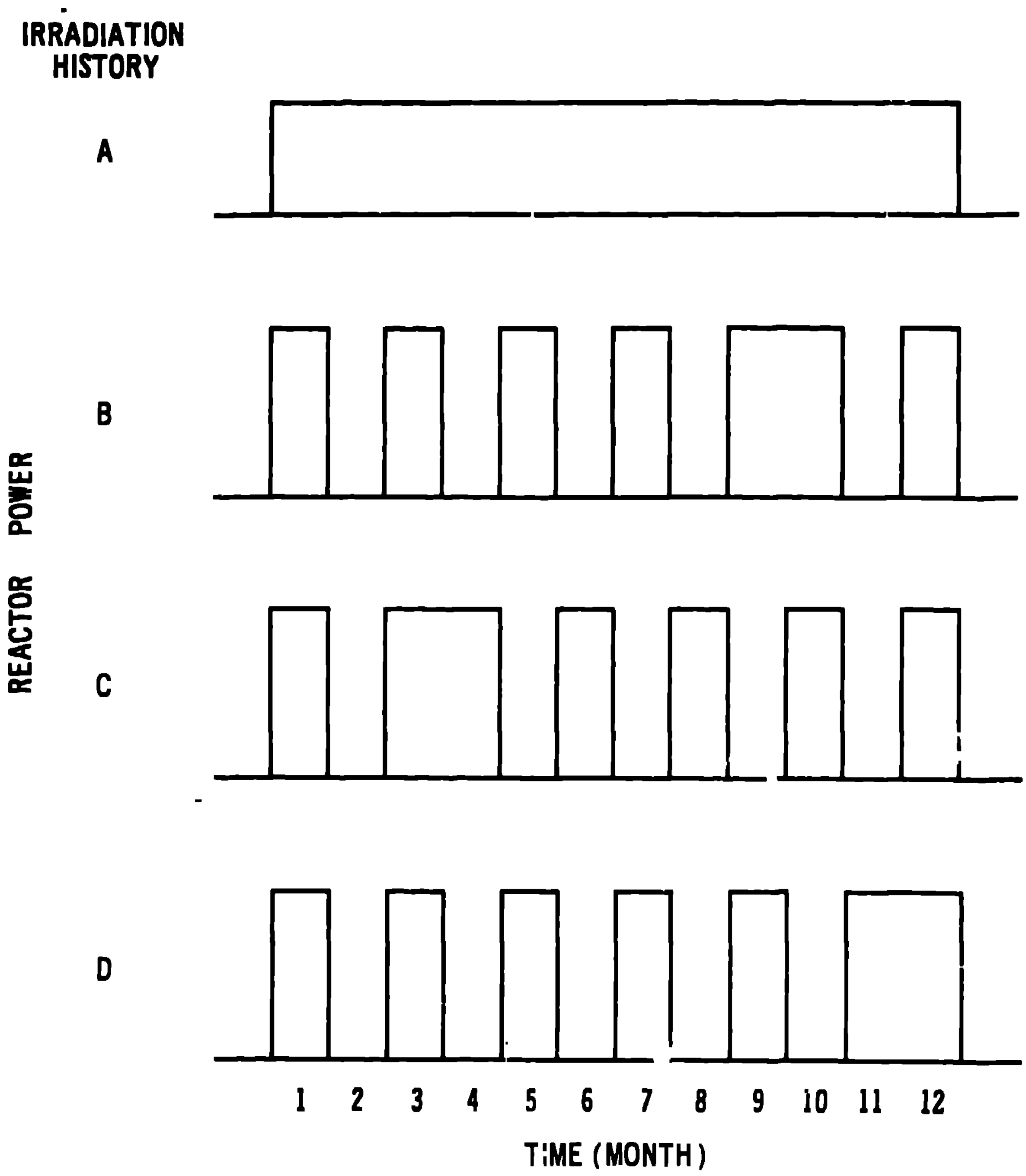
REFERENCES

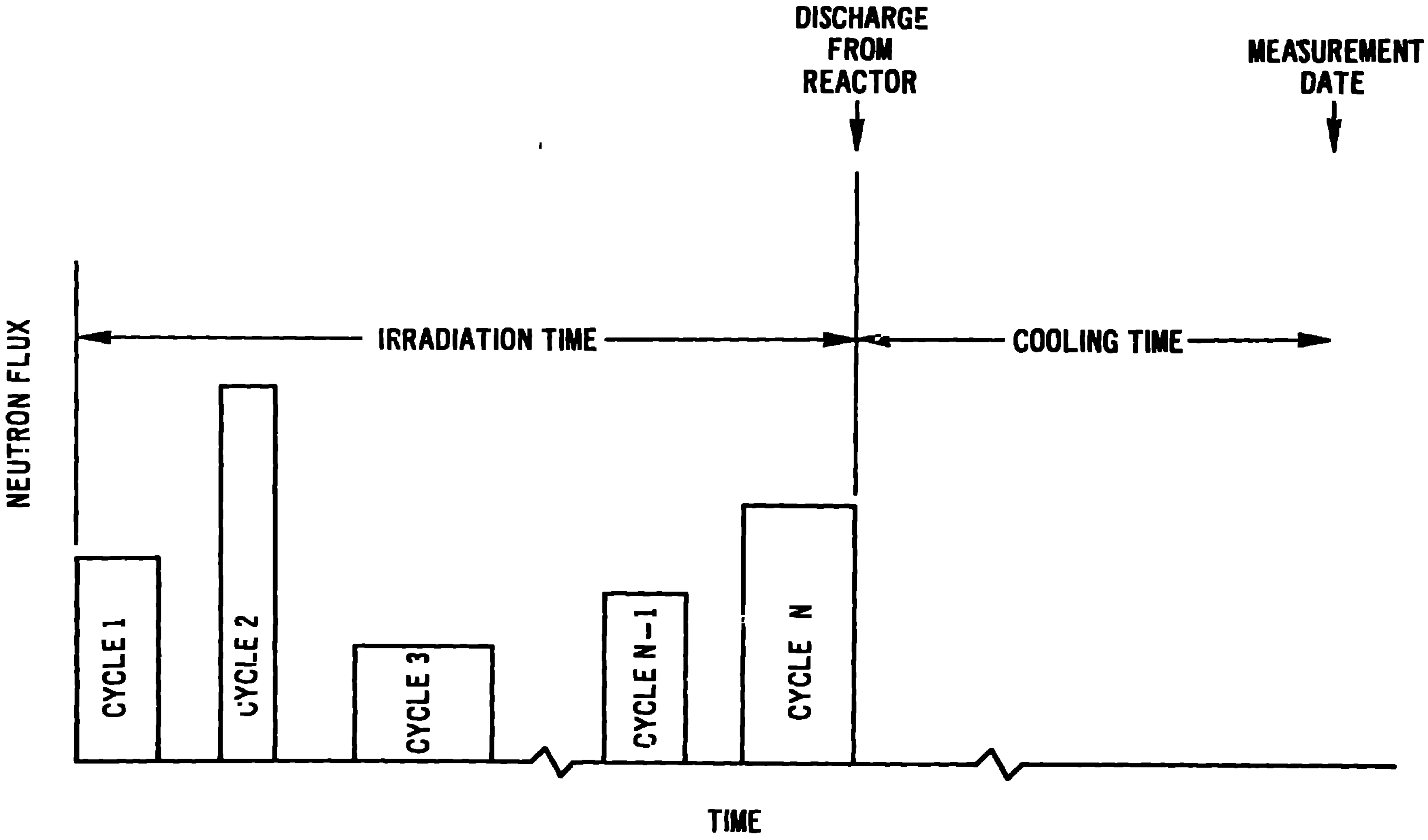
1. P. Brand and A. Cricchio, "Experimental Determination of Burnup and Heavy Isotopes at LWR Power Stations," Trans. Am. Nucl. Soc. 17, 473-474 (1973).
2. D. R. Oden and G. D. Seybold, "DRAFT - A Computer Code for the Calculation of Fission Product Activity Ratios," Battelle Pactific Northwest Laboratory report BNWL-1607 (June 1971).

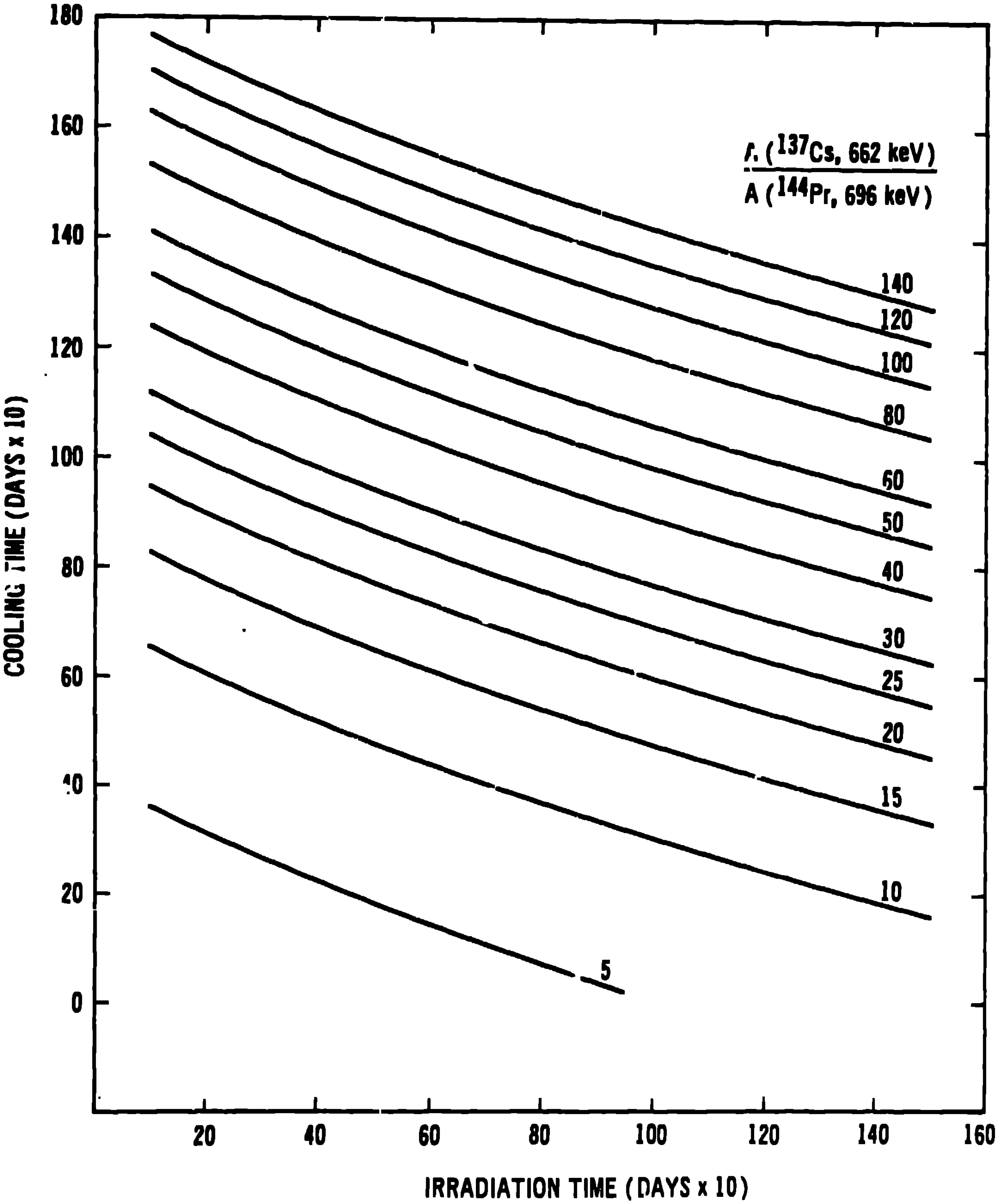
FIGURE CAPTIONS

1. Irradiation history for a typical spent fuel assembly.
2. Several hypothetical irradiation histories.
3. The irradiation time is the time the fuel starts to be irradiated until it is discharged from the reactor.
4. The cooling time versus the irradiation time for various $^{137}Cs/^{144}Pr$ activity ratios, for the simple model.


DISCHARGE FROM REACTOR
MEASUREMENT DATE
θ1
θ2
θ3
θn−1
COOLING TIME
NEUTRON FLUX
Φ1
t1
Φ2
t2
Φ3
t3
Φn−1
tn−1
Φn
tn
CYCLE 1
CYCLE 2
CYCLE 3
CYCLE n−1
CYCLE n
TIME


IRRADIATION
HISTORY
A
B
C
D
REACTOR POWER
1
2
3
4
5
6
7
8
9
10
11
12
TIME (MONTH)


DISCHARGE FROM REACTOR
MEASUREMENT DATE
NEUTRON FLUX
IRRADIATION TIME
COOLING TIME
CYCLE 1
CYCLE 2
CYCLE 3
CYCLE N−1
CYCLE N
TIME



180
160
140
120
100
80
60
40
20
0
COOLING TIME (DAYS x 10)
20
40
60
80
100
120
140
160
IRRADIATION TIME (DAYS x 10)
A (^{137}Cs, 662 keV) / A (^{144}Pr, 696 keV)
140
120
100
80
60
50
40
30
25
20
15
10
5