A BURNUP CREDIT CALCULATION METHODOLOGY FOR PWR SPENT FUEL TRANSPORTATION

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SUMMARY

A burnup credit calculation methodology for PWR spent fuel transportation has been developed and validated in CEA/Saclay. To perform the calculation, the spent fuel compositions are first determined by the PEPIN-2 depletion analysis. Secondly the most important actinides and fission product poisons are automatically selected in PEPIN-2 according to the reactivity worth and the burnup for criticality consideration. Then the 3D Monte Carlo criticality code TRIMARAN-2 is used to examine the subcriticality. All the resonance self-shielded cross sections used in this calculation system are prepared with the APOLLO-2 lattice cell code. The burnup credit calculation methodology and related PWR spent fuel transportation benchmark results are reported and discussed.

INTRODUCTION

Burnup credit is an ongoing technical concern for the criticality analysis of light-water-reactor (LWR) spent fuel transportation, storage and reprocessing (Zachar, 1994 ; Brady et al., 1996). In practice, determination of subcriticality using a burnup credit approach requires first to determinate the spent fuel compositions by the depletion analysis. Secondly the calculation of the effective multiplication factor Keff is realized for the spent fuel system based on the predicted actinides and eventually fission products (FPs) poisons. This paper presents a recently developed burnup credit calculation methodology and its validation benchmark results for PWR spent fuel transportation.

CALCULATION METHODOLOGY

The burnup credit calculation system is composed of three different computer codes developed on the reactor physics in CEA/Saclay. The major function of each code is :

. for APOLLO-2 code (Sanchez, 1989), to generate the resonance self-shielded cross sections as a function of burnup,
. for PEPIN-2 code (Tsilanizara et al., 1997), to calculate the burnup/depletion and to select the actinides and the FPs involved in burnup credit calculation, and
. for TRIMARAN-2 code (Lee et al., 1995), to estimate the Keff in a realistic 3D configuration.

Depletion calculations

The PEPIN depletion code (Nimal et al., 1990) is originally designed to predict decay heat and radiation source terms for shielding. The accurate prediction of the specific nuclide quantities, including actinides, fission and activation products, becomes important in PEPIN-2 for fuel cycle back end studies, radioactive waste evaluations, decommissioning, and hybrid spallation system research.

Two approaches, analytical and numerical, are available in PEPIN-2 to solve the Bateman equations. In order to obtain an accurate result, in burnup calculation the numerical Runge-Kutta method is recommended. For burnup credit studies two data libraries are necessary. The first one is composed of basic data such as decay chains, decay constants, branching ratios, fission yields, neutron absorption cross sections, etc.. The second concerns the resonance self-shielded cross sections.

To prepare macroscopic multigroup self-shielded cross sections and neutron spectra as a function of burnup, a constant power has been used in APOLLO-2 lattice cell burnup calculations. The nuclides processed by the self-shielding calculation are : ^{238}U, ^{235}U, ^{239}Pu, ^{236}U, ^{240}Pu, ^{241}Pu, ^{237}Np, ^{241}Am, ^{242}Pu, ^{243}Am and Zr. The established self-shielded cross sections data library includes fission, capture and (n,2n) reactions.

According to the PWR fuel type (moderation ratio), uranium enrichment (UOX fuel) and plutonium concentration (MOX fuel), the condensed and burnup dependent one-group cross sections have been built in the data library of PEPIN-2. It allows to carry out the depletion calculations of an axially-specified fuel zone according to the operation history which is composed of real power variation and different cooling intervals.

Selection criteria for actinides and fission products

The spent fuel composition calculated by PEPIN-2 code includes more than 90 actinides and heavy nuclides, and 700 FPs. The most important actinides and FP poisons for burnup credit analyses can be selected according to the reactivity worth of poisons, the burnup, and the cooling time of the spent fuel.

Actually, to prepare the burnup credit criticality calculation, besides the 5 basic fissionable nuclides ^{234}U, ^{235}U, ^{238}U, ^{239}Pu, ^{241}Pu, only 7 minor actinides (^{240}Pu, ^{241}Am, ^{236}U, ^{242}Pu, ^{237}Np, ^{238}Pu, ^{243}Am) and 16 most absorbing FPs (^{149}Sm, ^{103}Rh, ^{143}Nd, ^{133}Cs, ^{155}Gd, ^{151}Sm, ^{152}Sm, ^{99}Tc, ^{145}Nd, ^{153}Eu, ^{147}Sm, ^{95}Mo, ^{150}Sm, ^{109}Ag, ^{101}Ru, ^{135}Cs) are selected. With the exception of the ^{151}Sm the other FPs are stable and non volatile.

The selection criteria is based on the French experimental results (Santamarina, 1995). The volatile nuclide ^{131}Xe and short half-life nuclide ^{147}Pm are excluded. The reactivity worth of each nuclide selected is a function of fuel type, burnup, enrichment of uranium and cooling time. The selected 16 FPs represent more than 70% of the total fission products poisoning for a typical PWR spent fuel.

Criticality calculations

TRIMARAN-2 is a 3D-multigroup Monte Carlo code dedicated to criticality safety studies. To perform the burnup credit criticality calculation, the macroscopic multigroup resonance self-shielded cross sections and neutron spectra in the spent fuel, containing

only the above selected 12 actinides and 16 FPs, are again calculated by the lattice cell code APOLLO-2. Then TRIMARAN-2 code is executed with these cross sections to examine the subcriticality of the transportation flask.

The geometry package of TRIMARAN-2 allows the description of the transportation flask in combinatorial and/or analytical ways. A graphical display function is available to help the definition of geometry for neutron transport simulation.

For the study of axial distribution of burnup, the relative fission rates in each burnup zone can be evaluated by TRIMARAN-2. The correlation matrix Kij calculated by TRIMARAN-2 is used to study the Keff of each fission zone and the correlation between fission zones i and j. This matrix gives a clear reactivity analysis of the interference effects between axially distributed burnup zones of the spent fuel.

VALIDATION BENCHMARKS AND RESULTS

Depletion calculations benchmark

The first benchmark (DeHart et al., 1996) was performed to check the accuracy of the depletion code PEPIN-2 and its associated data libraries used to predict the isotopic concentration of the PWR fuel as a function of burnup. The Combustion Engineering 14 x 14 assembly designated as ATM-104 was used in this benchmark. Three cases, A, B, C, with corresponding cumulative burnup of 27.35, 37.12 and 44.34 GWd/MTU were investigated. Four complete operating cycles with different irradiation and cooling intervals were evaluated. The final cooling down time is about 5 years (1870 days).

The actual pin dimension and the modified fuel pin pitch were taken from the benchmark specification. The fuel-to-moderator ratio of the actual two-dimensional assembly is considered to generate the modified fuel pin pitch. The effective fuel temperature is 841 K and the water temperature 558 K. The averaged specific power (case A : 16.94 W/g U, case B : 22.99 W/g U and case C : 27.46 W/g U) and boron concentration (450 ppm) were calculated by the author to simplify the preparation of the cross sections data base.

In Table 1, the selected isotopic concentrations of 12 actinides and 16 FPs from PEPIN-2 calculation for three burnup values are presented. The calculated isotopic activities of 5 highly active nuclides are also given. The differences in percentage between PEPIN-2 calculations and measurements are designated as 'M'. The differences in percentage between PEPIN-2 calculations and averaged 21 international calculation results taken from benchmark report are designated as 'A'. PEPIN-2 calculations are generally in agreement with measurements and with averaged calculation results.

Table 2 presents the standard deviation of isotopic calculation among the 21 participant results. It is interesting to note that, except for nuclides ^{236}U and ^{153}Eu, the values 'A' ((PEPIN2 - Average) x 100 / Average) in Table 1 are always lower than the standard deviation given in Table 2.

For most of the actinides in Table 1, the 'M' and 'A' values are lower than 3%. The 'A' values of ^{238}Pu, ^{243}Am and ^{237}Np nuclides in Table 1 are about 5%. This difference may result from the thermal capture cross sections used and/or from the simplified decay

Table 1. Results of burnup/depletion analysis for actinides and fission products

Burnup (GWd/MTU)	Case A 27.35			Case B 37.12			Case C 44.34		
Nucl.	PEPIN2 (mg/g UO_2)	M (%)	A (%)	PEPIN2	M (%)	A (%)	PEPIN2	M (%)	A (%)
^{234}U	1.555E-1	- 2.8	- 2.2	1.330E-1	- 5.0	- 2.4	1.186E-1	- 2.8	- 2.2
^{235}U	8.205E+0	- 3.1	+0.2	4.883E+0	- 5.6	+0.1	3.205E+0	- 9.5	+0.1
^{236}U	3.105E+0	- 1.1	- 3.7	3.513E+0	- 0.5	- 3.3	3.642E+0	- 1.3	- 3.4
^{238}U	8.370E+2	- 0.7	- 0.1	8.296E+2	- 0.4	- 0.1	8.239E+2	- 0.1	- 0.1
^{238}Pu	9.460E-2	- 6.5	- 0.5	1.751E-1	- 7.5	- 4.6	2.444E-1	- 9.1	- 5.0
^{239}Pu	4.301E+0	+0.9	+1.7	4.357E+0	+0.0	+1.0	4.330E+0	- 6.2	+0.6
^{240}Pu	1.743E+0	+1.4	+1.9	2.231E+0	- 0.4	+1.9	2.488E+0	- 2.2	+2.1
^{241}Pu	6.906E-1	+1.3	+3.1	9.055E-1	+0.3	+2.2	1.006E-0	- 1.4	+1.7
^{242}Pu	2.842E-1	- 1.5	+2.9	5.720E-1	- 0.7	+2.3	8.197E-1	- 2.4	+2.7
^{241}Am	2.396E-1	n.a.	- 1.2	3.055E-1	n.a.	- 2.1	3.320E-1	n.a.	- 2.4
^{243}Am	3.982E-2	n.a.	- 2.9	1.085E-1	n.a.	- 4.6	1.826E-1	n.a.	- 4.7
^{237}Np	2.906E-1	+8.4	- 0.2	3.924E-1	+10.2	- 6.2	4.768E-1	+1.9	- 4.7
^{95}Mo	5.596E-1	n.a.	+1.2	7.262E-1	n.a.	- 1.0	8.391E-1	n.a.	- 0.6
^{99}Tc	5.913E-1	n.a.	- 1.2	7.743E-1	n.a.	+0.0	8.994E-1	n.a.	+0.4
^{101}Ru	5.704E-1	n.a.	+1.2	7.701E-1	n.a.	+1.2	9.149E-1	n.a.	+1.4
^{103}Rh	3.607E-1	n.a.	+3.3	4.580E-1	n.a.	+3.2	5.165E-1	n.a.	+3.5
^{109}Ag	5.680E-2	n.a.	- 3.6	8.499E-2	n.a.	- 0.8	1.058E-1	n.a.	+0.5
^{133}Cs	8.497E-1	-0.04	+1.0	1.097E+0	+0.6	+1.1	1.261E+0	+1.7	+1.4
^{135}Cs	3.797E-1	+5.5	- 0.6	4.147E-1	+3.7	- 0.02	4.340E-1	+0.9	+0.5
^{143}Nd	6.121E-1	- 0.2	- 1.8	7.154E-1	- 0.1	- 1.9	7.599E-1	- 0.4	- 1.9
^{145}Nd	5.050E-1	- 1.0	- 0.3	6.456E-1	- 1.1	+0.03	7.372E-1	- 0.9	+0.4
^{147}Sm	1.864E-1	n.a.	+3.1	2.102E-1	n.a.	+4.6	2.184E-1	n.a.	+5.5
^{149}Sm	2.039E-3	- 3.0	- 0.7	2.301E-3	-23.3	+4.2	2.511E-3	- 46.5	+7.5
^{150}Sm	1.949E-1	- 5.8	- 0.2	2.731E-1	+0.8	- 0.3	3.305E-1	- 8.4	- 0.6
^{151}Sm	8.583E-3	n.a.	-11.9	9.621E-3	n.a.	-11.9	1.039E-2	n.a.	-11.0
^{152}Sm	8.999E-2	+3.4	- 4.2	1.128E-1	+8.5	- 5.6	1.275E-1	+5.4	- 5.9
^{153}Eu	8.550E-2	+8.2	+11.7	1.276E-1	+17.1	+12.0	1.574E-1	+6.4	+12.7
^{155}Gd	2.319E-3	n.a.	- 20.0	3.868E-3	n.a.	- 23.0	5.101E-3	n.a.	-23.1
	(mCi / g UO_2)								
^{237}Np	2.048E-4	+8.3	+1.6	2.808E-4	+11.9	- 4.4	3.360E-4	+1.5	- 4.8
^{241}Am	8.068E-1	- 5.7	- 0.2	1.029E+0	-12.8	- 2.2	1.118E+0	-14.7	- 2.9
^{243}Am	7.937E-3	n.a.	- 2.1	2.163E-2	n.a.	- 5.0	3.639E-2	n.a.	- 5.7
^{99}Tc	1.004E-2	+4.7	- 1.1	1.315E-2	+8.7	- 1.6	1.527E-2	+13.1	- 1.3
^{135}Cs	4.378E-4	+6.2	+5.2	4.782E-4	+4.2	+7.1	5.004E-4	- 6.1	+7.7

M : (PEPIN2 - Measurement) x 100 / Measurement A : (PEPIN2 - Average) x 100 / Average

Average : Average calculation results from 21 international participants. (DeHart et al., 1996)

n.a. : data not available.

Table 2. Standard deviation of isotopic calculation among the 21 participant results (DeHart et al. 1996)

Burnup (GWd/MTU)	Case A 27.35	Case B 37.12	Case C 44.34	Average
Nuclide				
^{234}U	5.19	7.08	8.99	7.09
^{235}U	2.98	6.01	8.12	5.70 #
^{236}U	2.91	2.72	2.60	2.74
^{238}U	0.12	0.17	0.21	0.16
^{238}Pu	8.52	7.46	6.58	7.52
^{239}Pu	5.16	6.08	7.12	6.12 #
^{240}Pu	3.95	4.27	5.27	4.49 #
^{241}Pu	6.45	5.97	6.86	6.43 #
^{242}Pu	8.69	8.28	8.39	8.45
^{241}Am	4.22	4.35	5.29	4.62
^{243}Am	11.31	10.41	10.40	10.71
^{237}Np	8.61	8.86	9.42	8.96
^{95}Mo	1.17	1.30	1.85	1.44
^{99}Tc	5.17	3.57	4.21	4.32
^{101}Ru	1.03	1.05	1.76	1.28
^{103}Rh	4.57	5.15	5.40	5.04
$^{109}Ag+$	7.05	6.98	7.61	7.21
^{133}Cs	4.87	4.90	5.60	5.12
^{135}Cs	2.49	2.98	3.63	3.03
^{143}Nd	2.76	3.93	4.51	3.73
^{145}Nd	1.02	1.25	1.46	1.25
^{147}Sm	6.03	7.95	9.12	7.70
^{149}Sm	14.14	15.01	15.61	14.92 #
^{150}Sm	5.30	7.07	8.50	6.96
^{151}Sm	22.41	21.72	22.31	22.15 #
^{152}Sm	7.20	9.01	9.68	8.63
^{153}Eu	7.90	8.19	8.52	8.21
^{155}Gd	33.45	33.28	32.97	33.23 #
^{237}Np *	10.62	9.93	10.14	10.23
^{241}Am *	7.58	8.84	9.55	8.66
^{243}Am *	10.04	10.78	11.91	10.91
^{99}Tc *	2.52	5.81	6.49	4.94
^{135}Cs *	13.11	13.36	13.73	13.40

* Concentrations for these isotopes were calculated in units of mCi/g UO_2 .

+ Revised standard deviations for ^{109}Ag whose fission yield is higher in plutonium.

These nuclides have larger integral effect on reactivity due to their higher dispersion of calculation results. (Brady et al., 1996)

chains. The correct calculations of the modified fuel-pin-pitch and of the neutron spectrum are also important to predict Pu, Am and Np isotopes.

For most of the FPs poisons, the 'M' and 'A' values are lower than 5%. For ^{149}Sm, the most absorbing FP in spent fuel with burnup less than 30 GWd/MTU and cooling time 5 years (Santamarina, 1995), the new measurements are significantly different from measurements reported in Table 1 and more closely in agreement with the calculations (Gulliford, 1997). For ^{103}Rh another leading isotope for reducing reactivity in spent fuel, the existence of difficulties in measurements and calculations is declared. (Gulliford, 1997)

For ^{155}Gd the most absorbing FP in spent fuel with burnup higher than 40 GWd/MTU and cooling time 5 years (Santamarina, 1995), the standard deviation of 21 calculation results in Table 2 is more than 30%. Because the independent ^{155}Gd measurement is not possible, a detailed study of the resonance data of ^{155}Gd and ^{155}Eu would be helpful to explain the important dispersion of results. (DeHart et al. 1996)

For ^{238}Pu and ^{109}Ag, the revised averages and standard deviations are included in Tables 1 and 2 (Tables 16 and 18 of Ref.: DeHart et al. 1996). The inclusion of the Cm nuclides in the reaction/decay chains will improve the ^{238}Pu prediction. The direct/cumulative fission yields of ^{109}Ag from ^{239}Pu and ^{241}Pu are larger than those from ^{235}U so the ^{109}Ag for burnup credit study would be more important when burnup value of UOX fuel is high or MOX fuel is involved.

Burnup credit criticality calculations benchmark

The second benchmark (Brady et al, 1996) was utilized to check the criticality neutron transport simulation of TRIMARAN-2 Monte Carlo code. The prediction of the Keff for a conceptual PWR spent fuel burnup credit transportation container was realized. Several cases with axial distributed burnup were considered.

The realistic configuration of 21 PWR UOX spent fuel assemblies in a stainless steel transport flask was taken from benchmark specification. A borated stainless steel basket centered in the flask separates the 21 assemblies. The thickness of this basket is 1 cm. The basket (5x5 array with the 4 corner positions removed) was fully flooded with water.

The main characteristics of the fuel assembly are : 17x17 array (289 fuel rods, no guide tubes), water moderated cells with pitch equal to 1.25984 cm ; initial fuel enrichment equal to 4.5 wt % ; fuel radius equal to 0.4096 cm, fuel rod ID= 0.41785 cm and OD= 0.475 cm which lead to a moderation ratio Vmod / Vuox =1.67.

The cooling time of the spent fuel is 5 years. The spent fuel was divided axially into 9 symmetrical zones corresponding to 5 predicted fuel composition sets. Each zone is composed of only 12 actinides or 28 nuclides (12 actinides and 16 FPs described in Table 1). The axially distributed burnup is shown by the higher ^{235}U concentration at end zones and the higher ^{240}Pu, ^{236}U and FPs concentrations at central zone.

In this benchmark problem, it is interesting to evaluate the capability of TRIMARAN-2 code to treat the neutron transport by Monte Carlo method in large size (height : 477 cm, radius: 98 cm) and in relative-complicated 3D geometry. With discrete neutron poisons

(borated stainless steel basket), reflector (flask) and axial distributed burnup (21 fuel assemblies x 9 axial zones) the statistical results of Keff should be correctly calculated.

The calculation investigation includes: the burnup effect (fresh fuel, 30 and 50 GWd/MTU) on criticality, the reactivity worth of actinides and FPs, the axial burnup profile effect and the effect of accidental movement of the assemblies in the flask on Keff.

The 11 cases of TRIMARAN-2 calculation results are presented in Table 3. Comparing with the averaged Keff of the international studies (Brady et al, 1996), the TRIMARAN-2 calculations over-predict or under-predict the Keff by about $\pm 0.35\%$. It should be noted that the 3σ of TRIMARAN-2 results are already about 0.3%.

The outcome of the burnup credit criticality consideration can be found in Table 3 when we compare the Keff in case (1) (fresh fuel) with those of cases (2) to (9). With different degree of application burnup effect, the cases (2) to (9) correspond to the more realistic Keff of the transportation flask. With increasing burnup, the reduction of Keff is more evident. Although the FP's contribution to reduction Keff is less important than the actinides, the inclusion of only 16 leading FPs in the calculation increases the reduction of the Keff in the spent fuel.

With higher burnup (cases (6) to (9)), the contribution of the axial burnup distribution becomes important to obtain an higher and conservative Keff, and it is more significant in the case including FPs. This well-known end effect in burnup credit study becomes distinctive for spent fuel with load following operation history. In that case the important axial asymmetry of burnup exists and the less burnt zone in the top end will be dominant in reactivity. This end effect is also non negligible in the accidental movement of the spent fuel assemblies in the flask when moderation ratio locally increases (cases X1 and X2).

Table 3. Multiplication factors of a conceptual burnup credit transportation benchmark

Burnup (GWd/t)	Burnup profile (9 fuel zones)	Spent fuel compositions: Actinides and FPs			Actinides only		
			$Keff^{*}_{Trimaran}$	$Keff^{+}_{Average}$		$Keff^{*}_{Trimaran}$	$Keff^{+}_{Average}$
0	No	Case			Case (1)	1.1297	1.1256
30	No	(2)	0.8932	0.8934	(3)	0.9714	0.9714
	Yes	(4)	0.8916	0.8949	(5)	0.9636	0.9640
50	No	(6)	0.7613	0.7641	(7)	0.8749	0.8735
	Yes	(8)	0.7906	0.7929	(9)	0.8791	0.8781
Accidental movement of the fuel assemblies in the flask (Mennerdahl, 1995)							
30	No	X1	0.9216	(case (2) + 20 cm water at top end of fuel.)			
	Yes	X2	1.0026	(case (4) + 20 cm water at top end of fuel.)			

* one standard deviation < 0.001.

\+ 14 results from 7 countries, codes used : MCNP, SCALE, KENO, MONK...etc. (Brady et al., 1996)

CONCLUSIONS

The traditional criticality calculation of the spent fuel transportation did not consider the burnup effect because the burnup value was relatively low ten years ago and the Keff analysis, using the fresh fuel containing lower uranium enrichment, is simple and conservative.

With increasing uranium enrichment and burnup of fuel to reduce the nuclear power generation cost and to diminish spent fuel accumulation, the burnup credit criticality calculation becomes interesting. It allows us to use old transportation basket and flask design to deliver new higher burnup spent fuel if the decay heat and radiation shielding problems are solved. For storage ponds and reprocessing process it is also profitable to take the burnup effect on criticality safety consideration.

The burnup credit calculation methodology presented in this paper is a basic tool to consider the burnup effect of spent fuel. The degree of its application should be determined by the real measurements of the burnup and the caution of the user. It is possible to perform burnup credit calculation considering only limited poisons (for example, above selected actinides) in the spent fuel and/or taking axial uniform burnup with limited burnup value.

The validation benchmark results presented above give an exemplary exercise for burnup/depletion computation and criticality evaluation. The validation work is in progress and the data base covering BWR, RBMK spent fuels will be prepared.

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