Inventory Assessment of Pin-wise Isotopic Composition in the Spent Fuel

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1. Introduction

The knowledge of the detailed information of isotopic composition in the spent nuclear fuel could be beneficial in multiple ways, like; for more efficient fuel utilization by imparting highest possible burnup to each fuel pin and consequently reducing amount of nuclear waste production, improved spent nuclear fuel (SNF) handling, precise calculation of the linear heat generation ratio, better assessment of the power peaking factors. Since, there is no direct method to determine the pin-wise composition in a reasonable time frame, so, different multifaceted attempts have been made to calculate isotopic distribution of the SNF under special circumstances.

There are numerous factors that influence the final SNF composition; initial fuel composition (especially enrichment), operating conditions of the reactor and final burnup imparted to the fuel, to name a few [1]. Previously, a neutronic analysis code is used [2], which can keep account of those isotopes that are important from the neutronic perspective only. An assumption, usually employed implicitly, is that the SNF composition is a function of the burnup and the initial enrichment only. Current methodology can be utilized by making use of the three code strategy i.e. (HELIOS 1.5 [3], MASTER 2.2 [4], and ORIGEN2 [5]) to assess the amount of the isotopes that are hazardous for being radioactive as well as those ones that have some role (positive or negative) in reactivity variations inside the core. Effect of the operating power i.e. dominant neutron flux is also incorporated. The minor spectrum variation from BOC to the EOC could be neglected because (1) PWR assemblies are adorned in a way to make the neutron spectrum uniform over the core; (2) minor variation of the neutron spectrum has very little effect on the SNF composition [6].

Few isotopes like Cs-135, Pu-241, etc. show remarkable dependence on the reactor operating conditions (e.g. operating power level) while many important isotopes like U-235, I-129, Cs-137, etc. depend on the net burnup imparted to the fuel only.

2. Methods and Results

A C-1 type fuel assembly from the OPR-1000 reactor is treated as a reference case for the composition assessment methodology. Fuel assemblies in OPR-1000 reactor are classified into six types depending on their composition and layout; i.e. whether a fuel pin has burnable absorber in it or not, UO_2 comprising fuel contains natural uranium or enriched one, how are these special composition pins distributed in the assembly etc. The C-1 type fuel assembly (Fig. 1) considered to calculate the pin-wise isotopic composition is a 16 X 16 fuel assembly consisting of 236 fuel pins and five guide tubes.



Fig. 1. Pin layout of the C-1 type fuel assembly

An octant of this assembly is modeled in HELIOS 1.5 to generate cross sections for the homogenized fuel assembly. Later, MASTER 2.2 is used to generate pinwise burnup distribution by invoking its pin-by-pininformation (PPI) file generation option.

Dimensions (Table 1) and composition (Table 2) of the fuel containing pins and the guide tubes are used in HELIOS 1.5. Assembly layout and the OPR-1000 reactor operating conditions were studied earlier with MASTER 2.2 code [2]. Guide tubes are assumed to be filled with water, all control rods are treated as out of core, fission products are treated in equilibrium, boron concentration is taken as "sufficient enough to keep the reactor critical".

Table 1. Dimensions of the C-1 type fuel assembly

Item Name	OD (cm)
Fuel Pin	0.826
Clad ID	0.843
Clad OD	0.970
Pin Pitch	1.285
Guide Tube ID	1.145
Guide Tube OD	1.245
Assembly Pitch	20.78
Active Fuel Height	381

Material	Density (g/cc)	Comments
Normal fuel pin	10.97	U-235 - 3.43%
		U-238 - 96.57%
		Form: UO ₂
Lower enriched fuel pin	10.97	U-235 - 2.93%
		U-238 - 97.07%
		Form: UO ₂
Gadolinia bearing fuel pin	10.96	U-Nat: 98%
		Gd ₂ O ₃ : 2%
		Form: Gd ₂ O ₃ -UO ₂ mixed uniformaly
Zirlo (Clad)	6.55	Fe: 0.1%
		Zr: 97.9%
		Nb: 1.0%
		Tin: 1.0%
		Form: Zirlo Alloy

 Table 2. Composition of the materials used in C-1

 fuel assembly

2.1 Pin-wise Burnup Calculation

The lattice code HELIOS 1.5 gives the required necessary cross sections and the assembly heterogeneity measuring normalized flux and power profiles called respective "form functions". Then, like other nodal codes, a diffusion theory code MASTER 2.2 models the entire core and provides assembly homogenized two group neutron fluxes, called nodal homogeneous fluxes.

We make use of the reconstruction schemes distributed in the MASTER 2.2 code. To determine the continuous distribution of the neutron flux all over the fuel assembly, these node average discrete values are interpolated by a second order polynomial and hence a continuous intra-nodal neutron flux variation is obtained. This global or intra-nodal neutron flux profile is used to calculate corresponding global power profile by Equation (1).

$$[P_g^{(p)}(x,y)]^{lntra} = [\kappa \Sigma_{fg}^{(p)}(x,y)]^{lntra} [\phi_g^{(p)}(x,y)]^{lntra}$$
(1)

Where, symbols kappa and sigma connote their standard meanings of the number of neutrons per fission reaction, and macroscopic fission cross section respectively.

This intra-nodal (or global) neutron flux and power distributions in the homogenized fuel assembly are translated into their corresponding pin by pin distributions (i.e. heterogeneous distributions) by merely superimposing this continuous solution with their "single assembly" profiles (i.e. form functions obtained from HELIOS), as given in equation (2).

$$P^{(p)}(x,y) = \sum_{g=1}^{2} [P_g^{(p)}(x,y)]^{Intra} [f_g^{p,het}(x,y)] \dots (2)$$

Finally, pin wise burnup is obtained directly from the power produced within that $pin(\overline{P^{(p)}}(x, y))$, assembly average power (\overline{P}) and the total burnup imparted to the assembly $(\Delta \overline{B})$ i.e.

$$\Delta B^{(p)}(x,y) = \frac{\overline{P}^{(p)}(x,y)}{\overline{P}} \Delta \overline{B} \qquad (3)$$

This pin-wise burnup distribution is used in the next section 2.2 to calculate isotopic composition.

2.2 Composition Table Preparation

The variation of isotopic composition of the fuel with burnup and specific power level named as the composition table, which is prepared twice; once with the lattice code HELIOS 1.5 and then with the dedicated depletion analysis code ORIGEN2. Both gave a bit different results for the isotopes common to their respective libraries. However, ORIGEN2 has much larger inventory of the isotopes. The isotopic masses used in a typical fuel pin (Table 2 above) are burned at multiple power levels reaching to identical burnup levels in different time durations. Being the exclusively depletion code, ORIGEN2 gives content for roughly 1500 isotopes out of which only 143 isotopes are selected for being radioactive and having half-life more than 30 hours [5].

ORIGEN2 results are validated with NEA Phase VII publications [7]. Multiplication factor variation pattern is in good agreement and decently mimics the quoted values over the entire one million year time span (Fig. 2).



Fig. 2. Validation of the ORIGEN2 results

2.3 Parameters affecting composition

A fictitious benchmark problem is established (Fig. 3) in MCNPX 2.6.0 [8] to ascertain the effect of the different parameters like flux level (i.e. power level), neutron spectrum, location of the fuel pin etc. All the fuel pins are identical in dimensions and material composition (normal fuel pins of table 1 and table 2). The sole difference between the pins 'A', 'B' and 'C' is their location, and hence different neighborhood, consequently different neutron spectrum and flux levels.

Pin 'A', surrounded by a very strong neutron absorber B_4C faces the smallest neutron flux. Pin 'B' being adjacent to a water hole faces the softest neutron spectrum. The last pin 'C' being at the far end of the fuel assembly and also farthest from the absorber region has the highest and the hardest neutron spectrum.



Fig. 3. A benchmark problem: An OPR-1000 fuel assembly with same fuel pins exposed to steeply varying neutron flux, spectrum and neighbourhood.

Regardless of the very obvious geometrical and other, prima facie, spectral differences, we do not find any difference in the change in the composition at the EOC (Fig. 4) other than what we expect from the corresponding burnup and power levels. This may be due to the fact that other minor effects are already incorporated into these two parameters.



Fig. 4. Isotopes like I-129 depend on burnup only

However, there are some isotopes like Cs-135 (Fig. 5) that clearly depend on the operating power level also.



Fig. 5. Isotopes like Cs-135 show a proportional dependence on the neutron flux

2.4 Results and Discussion

The burnup pattern within a C-1 type fuel assembly located at B-7 position in the reference core, as expected, is biased towards the core center. Magnitude of the burnup (Fig. 6) for a certain fuel pin is obtained from the MASTER 2.2 code. To run the MASTER 2.2 code, cross sections were generated for all fuel assemblies; a full core with all rods out was modeled. However, to show the methodology of the composition calculation, only single fuel pin in a single fuel assembly is considered.



Fig. 6. Burnup distribution in the selected C-1 fuel assembly, located at B-7 position in the reference core.

Above calculated burnup is translated into the amount of a certain nuclide by interpolation from Fig. 7. For instance, a specific case of a normal fuel located at the position (14, 14) in Fig. 6, having the highest burnup in this fuel assembly (8.01 MWd/kg) is considered. From the Fig. 7, the atomic number densities of the different isotopes are the desired number densities.



Fig. 7. Composition table obtained from ORIGEN2 at specific power 36.874 W/g

 Table 3. Change in the number density of selected isotopes during 370 EFPD

Isotope	No. Density at BOC (atom/b-cm)	No. Density at EOC (atom/b-cm)
U-235	7.91E-04	5E-4
Pu-239	0.00E+00	6E-5
Pu-241	0.00E+00	7E-7
Am-241	0.00E+00	6E-9
Am-243	0.00E+00	4E-10

For the validation purposes, a composition table is also generated from HELIOS 1.5. Due to wide variation in the corresponding calculation methodologies and use of different libraries there is a reasonable difference between the two (Fig. 8). The most profound reason of non-conformity between the two codes is consideration of the different parameters while performing calculations. HELIOS 1.5, for instance, uses 45-group neutron energy spectrum while ORIGEN2 uses a single energy group neutron flux.



Fig. 8. Composition table comparison from ORIGEN2 and HELIOS 1.5

3. Conclusions

Detailed pin-wise isotopic composition of the spent nuclear fuel from PWRs could be ascertained if influence of all of the concerned parameters is considered precisely. Here, we considered effect of the burnup on the composition of a selected fuel pin from the assembly B-7. Effect of the operating power level (i.e. flux) is accounted implicitly. This technique of composition reconstruction utilizes the pin-wise burnup calculation capability of the core analysis code MASTER 2.2. The relative magnitudes of the from two different codes considered isotopes (ORIGEN2 and HELIOS 1.5) match reasonably well. However, incorporation of the more isotopes will make the difference clearer, while calculation of the composition at other pin locations will improve accuracy in the results.

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