Evaluation of the Pin-wise Isotope Inventory for the Used PWR Fuels

Zafar Iqbal ZAFAR and Myung Hyun KIM*
Department of Nuclear Engineering, Kyung Hee University
1732 Deogyeong-daero, Giheung-gu, Yongin-si, Gyeonggi-do, Korea, 446-701
**mhkim@khu.ac.kr*

1. Introduction

Accurate determination of the used nuclear fuel (UNF) composition is essential for its reliable long time storage. However, discharge composition determining factors are not identical for each fuel pin. The initial composition, discharge burnup, neutron energy spectrum, power density, presence of water holes nearby, critical boron concentration levels and other similar parameters lead to uneven fuel burning. The active life events i.e. the irradiation history of each fuel pin in each fuel assembly is different from the other. Moreover, the current fleet of the depletion codes is incapable of analyzing each fuel pin with true neutron spectrum due to impractically large required computational time. Consequently, in these codes, entire lot of the fuel pins in the fuel assemblies supposedly burn identically. This is represented by a single core average value, or in some cases of the 'detailed analysis' fuel assembly average values of the burnup only.

Fuel assembly average inventory has already been calculated and pointed out by many researchers as inadequate. For instance, Caruso et.al. found a significant effect of the neutron spectrum variation on the activation characteristics for the UO_2 fuel [3]. They also showed that neglecting or concerning tiny fractions of the various isotopes in fuel, at the beginning of life, have significant effect on the fuel composition at the end of life. Wagner et.al had pointed out that in order to take credit for the depleted fuel for criticality analysis the axial variation of the burnup could not be ignored [4].

In one way or the other, some limited but workable pin-wise composition retrieving techniques have been studied previously. They either concerned limited number of isotopes or assumed single cycle depletion [1], [2]. This manuscript deals with the fine inventory assaying option i.e. pin by pin composition assessment. Fine composition information has numerous potential applications: it could lead to reutilization of the selected fuel pins so reducing the spent fuel inventory, could help in segregating fuel pins and improving the DUPIC fuel cycle, could be useful in increasing efficiency of the specific isotope production facilities. Moreover, random placement of the fuel assemblies vs. their ordered placement in transportation or a permanent storage cask could make a big difference because the decay heat, infinite multiplication factor (k_∞), radioactivity etc. depend on the fuel geometry and pin wise composition.

Therefore, current work comprises of the nuclide wise pin by pin composition, concerning three cycle fuel depletion, and also by paying regard to the depletion rate (aka specific power) in the UNF. The resulting amounts of some of the important isotopes (like Am-241) differ from the single cycle depletion by more than 30%.

2. Methods and Results

2.1. Reactor Model

The optimum power reactor of 1000 MW electrical power capacity (OPR-1000) unit 5 located at Ulchin (UC5), is a pressurized water reactor with its core consisting of 177 fuel assemblies. Each fuel assembly has 16x16 pin configuration with five guide tubes (GT) to be used as instrumentation thimble or control rod guides (Fig. 1). In most of the fuel assemblies, some fuel pins are replaced with burnable poison (BP) pins to partially compensate for the excess reactivity and to flatten the power distribution. Guide tube inner-diameter, outer diameter, fuel pellet diameter, cladding inner and outer diameter are also given in Fig 1.

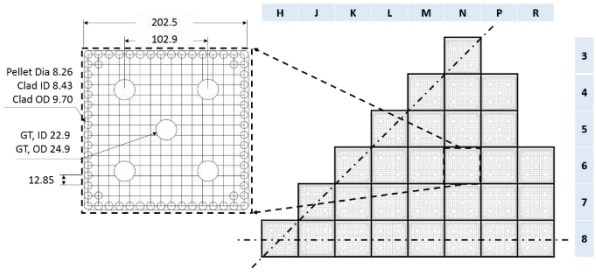

Fig. 1. Right: Octant cut from the OPR-1000 unit 5 core with FA to FA water gap of 5.28 mm. Left: Dimensions of a fuel assembly given in millimeters [5]

In the UC5 core most of the fuel assemblies consist of two types of fuel pins other than BP pins. These two differently enriched fuel pin types help in flattening power distribution and maximizing discharge burnup. The fuel assemblies equivalent to the equilibrium cycle fuel assemblies (like E type and higher) have about 4.0% and 4.5 % enriched ZIRLO clad fuel pins because these fuel assemblies are expected to stay for full three cycles and also undergo maximum possible burnup (above 45 MWD/kg). Like most of the other PWRs, this reactor also uses UO_2 fuel with smear density 10.172 g/cc.

2.2. Pin-wise Burnup from two step method

The fuel assembly average burnup has been calculated using two step method. In the first step, called lattice analysis, fuel assembly averaged cross sections (Equation 1) and form functions (Equation 2) are calculated using lattice code HELIOS 1.8 [6]. Although

both flux form functions and power form functions are calculated but only latter are used in the analysis. An octant of a fuel assembly, with all sides reflected, gives fully symmetric distribution of the neutron flux, and hence, power production and burnup. This saved the required memory and computational time should the full fuel assembly has been modeled.

$$\bar{\sigma} = \frac{\int_0^\infty \sigma(E)\phi(E)dE}{\int_0^\infty \phi(E)dE} \quad (1)$$

$$Flux\ Form\ Function = FF_\phi = \frac{\phi_p}{\phi_{FA}} \quad (2a)$$

$$Power\ Form\ Function = FF_P = \frac{P_p}{P_{FA}} \quad (2b)$$

The second step consists of nodal analysis i.e. neutron fluxes, power profile, burnup etc. are calculated at each node (here, node means a homogenized fuel assembly region in the core). That is, the neutron cross sections generated for a fuel assembly present among a lattice of the identical assemblies were used to calculate quantities like neutron flux and power over the fuel assembly region present in the real environment of the reactor core.

The burnup obtained from the nodal analysis is a fuel assembly average burnup. These node average values of the burnup need two additional operations to get pin by pin burnup. In the first operation, continuous burnup profile over the entire core region is produced by linear interpolation between the node average values. In the second operation, power form functions obtained from the lattice code at the beginning are superimposed to the continuous burnup profile. This procedure gives identical results if pin by pin power is reconstructed and then that pin wise power is used to calculate corresponding burnup.

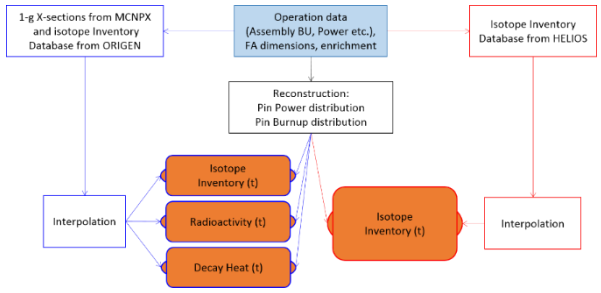

Fig. 2. Main steps of the inventory assay method

2.3. Considerations for the reference composition database

After preparing a pin by pin burnup profile for each fuel assembly, the next task is to translate this burnup into composition. One way to do this translation is by using reference composition database (RCD) method. Here, two databases are used; one prepared with HELIOS 1.8 and the other from ORIGEN2. The latter database needed additional task of one group cross sections preparation from MCNPX 2.6.0 (Fig. 2).

2.3.1. Selection of Important isotopes

At the time of UNF discharge, the content of those nuclides is provided that are either experimentally measureable or deducible from the given correlations. These isotopes are not sufficient enough to explain the characteristics of the used nuclear fuel (UNF) over extended period of time. So, long term storage facilities (repositories) require information about more isotopes, especially the ones that are produced from the radioactive decay of the other isotopes. [3]. Hence, instead of carrying entire lot of the roughly one thousand nuclides all the time, a set of the seven important nuclides (U-235, Pu-239, Am-241, Nb-94, I-129, I-131, and Cs-135) is selected to show comparative effects of different sets of the inventory more clearly. The first three nuclides are fissile actinides and have strong influence on the infinite multiplication factor of the material (k_∞). Two nuclides of Iodine are selected firstly because they are important for their medicinal characteristics and secondly to show effect of a long lived isotope I-129 ($T_{1/2}$~$15.7 \times 10^6 y$) and a relatively short-lived isotope I-131 ($T_{1/2}$~$8\ days$). Am-241 and Cs-135 are specially included to represent the neutron spectrum and flux dependent effects. Nb-94 is used to represent the most robust set of nuclides that are not affected by any factor other than the fuel burnup, irrespective of being radioactive and produced in small amount.

2.3.2. Concerning database memory requirements

Since each fuel assembly contains 236 fuel pins (including absorber pins) and 177 fuel assemblies reside in the core during each depletion cycle so, it is impractical (probably unnecessary too) to calculate pin wise composition. Hence, fuel pins were divided into different groups. All the fuel pins of any fuel assembly having identical initial composition, identical nearest four neighbors, and burned at similar specific powers are grouped together [1]. Indeed any grouping scheme could be used based on the available memory and affordable number of groups for the resultant data

2.3.3. Single cycle vs. multicycle depletion

The conventional method of the RCD preparation is based on a single spell continuous fuel burning. In that method, it is also assumed that a reactor is always operating at full power, time evolution of the composition in a fuel pin depends solely on its initial composition and neutron flux level or spectrum effects could be ignored. The assumption of depletion at constant power is a reasonable one because nuclear reactors always serve as base loads, however, the assumption becomes overly simplistic if extended to the entire life time of the reactor. Because exact power

production profile of a fuel pin varies from pin-to-pin and from cycle to cycle, so, it is assumed that the power production history of any given fuel pin could be divided into three cycles. During any given cycle, it burns at a unique rate. This assumption is reasonably valid because the main difference in power profile of a fuel pin arrives from location of its fuel assembly, which does not change during any given cycle (Fig. 3).

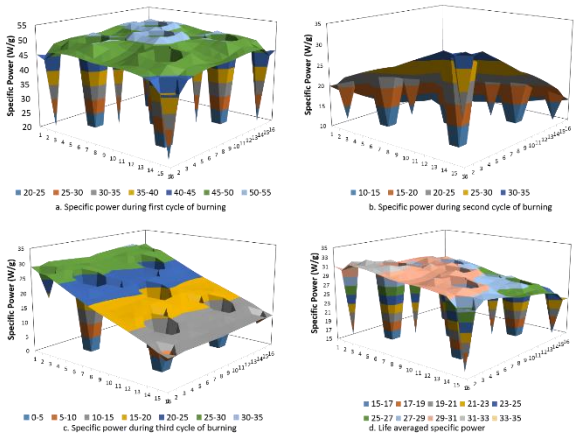

Fig. 3. Pin by pin specific power profile for the E1 type fuel assembly burned at core locations M04, D14, and R08 during reactor cycle 3 to cycle 5, respectively.

Some patterns of the specific power effect on different nuclides could entice to develop mathematical correlations to concern deviation of the depletion rates from some standard value. However, due to non-identical response of the different isotopes to the specific power, development of the mathematical correlations as correction factors to account for the difference from average value is not a practical option. So, instead of using a single database assisted with a plethora of the correlations, a simpler approach has been used here, and nine databases for depletion at different specific power levels (i.e. 10, 15, 20, 25, 30, 35, 40, 45, and 50 watts per gram) has been prepared. For a three cycle, annually recycled core these specific power values correspond to discharge burnup of 11 MWD/kg to 55 MWD/kg. While higher enriched fuel pins, especially if they happen to lie near guide tubes, are burnt more rapidly (i.e. at higher specific power). Therefore, this range of variation is wide enough to accommodate all types of fuel pins reasonably. Since number of the specific power concerning databases is arbitrary and could be refined as per requirement from the discharged fuel pins.

In case the used nuclear fuel lacked data about the cycle-wise burnup (which represents cycle average specific power too), then a single fuel assembly average value, obtained by dividing the fuel assembly average burnup with the assembly residence time could be used. This provision in the methodology is for the sake of completeness.

2.4. Reference composition database preparation

Although any depletion code can be used to construct an appropriate reference composition database. In this study, RCDs are primarily prepared using ORIGEN2. However, cases of reduced number of isotopes and fewer burnup steps have been constructed with HELIOS 1.8 and CINDER90 also. Using ORIGEN2 is simple and quick. The only input parameters provided were one group cross sections generated from MCNPX 2.6.0, initial composition, specific power, and appropriate depletion time steps. ORIGEN2 converts power into instantaneous flux using equation 1, where denominator is a summation over time dependent isotope, i, specific quantities.

$$\phi = \frac{1.0365\times10^{19}\times P}{\Sigma_i\left(X_i^f \times \sigma_i^f \times \kappa_i\right)} \qquad (3)$$

In equation 3, P is the power in Mega Watts, X_i^f and σ_i^f are the amount (in gram-atoms) and the microscopic fission cross section (barn) of the isotope i. Moreover, instead of using a conventional constant value of 200 MeV/fission, the isotope dependent value of (κ_i) represents the composition effect more accurately. κ_i is calculated by using equation 2.

$$\kappa_i = 1.29927 \times 10^{-3} \times Z^2 \times A^{0.5} + 33.12$$

Equation 3 represents the neutron flux at the beginning of a depletion time step. Time variation of the neutron flux is taken into account by making a Taylor series expansion of the equation 3, with X_i^f replaced with $X_i^f(t)$, and then integrating it over the entire time step to calculate true time step average neutron flux (Φ).

To prepare RCD with CINDER90, we need 63 group true fluxes which could be obtained from MCNPX 2.6.0 is then normalized to this one group flux (Φ). Resultantly, it becomes a tri-code (MCNPX-ORIGEN2-CINDER90) based RCD preparation. Now, CINDER90 being a Monte Carlo depletion code takes a lot more time than the deterministic point depletion code ORIGEN2. Hence, it is used for the comparison and verification purposes only.

2.5. Assaying pin by pin inventory from RCD

After preparation of the reference composition databases the process of assaying inventory comprises of reading measured or calculated pin by pin burnup BU_{pin}, fuel assembly average burnup BU_{FA}, number of cycles stayed in core (one or three), and duration of each cycle. Then specific power during depletion of a fuel pin is obtained by dividing its cycle average burnup with cycle length.

Selection of the appropriate RCD depends on introspection of the pin itself. First, its initial composition, then its location in the fuel assembly and

lastly its depletion rate (via specific power) will lead to the appropriate database.

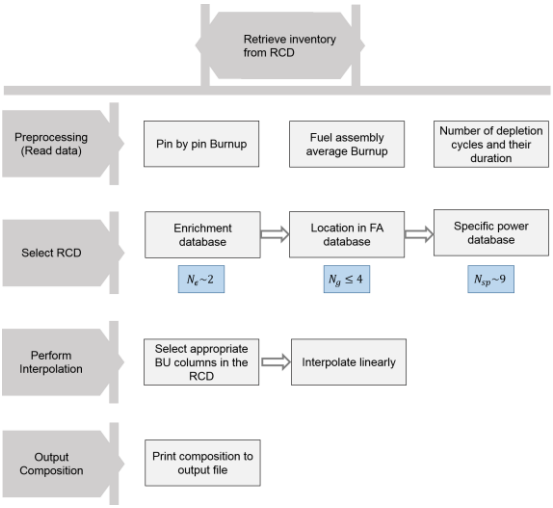

Fig. 4 Process of assaying pin by pin from given pin-by-pin Burnup and RCD.

The specific power effect on the content of different isotopes in the discharged fuel is said to be not important for most of the isotopes [7]. However, for some crucial isotopes it cannot be ignored either. For instance, Nb-94 is the only potent isotope to be used for experimental BU determination for long term storage purposes [3]. Due to small amounts of the Nb-94 isotope involved (few micrograms per ton of initial fuel), the observed little effect (about 0.3% decrease in the amount produced could lead to considerable error in the estimated BU (Table I). Benefit of concerning multicycle fuel depletion is more obvious for the short lived nuclides or from the nuclides that are produced from the short lived nuclides. For instance, Am-241 is produced by the following chain. Here, both U^{239} and NP^{239} are short lived nuclides with half lives 23 minutes and 2.36 days respectively.

$$U^{238} \rightarrow U^{239} \rightarrow NP^{239} \rightarrow Pu^{239} \rightarrow Pu^{241} \rightarrow Am^{241}.$$

Table I. Amount of the selected isotopes per ton of the discharged fuel from E1 fuel assembly of Fig. 3.

Isotope	U^{235}	Pu^{239}	Am^{241}	Nb^{94}	I^{129}	I^{131}	Cs^{135}
Single cycle	1.03E+04	5.19E+03	4.99E+01	8.90E-04	2.14E+02	8.84E+00	3.69E+02
Three cycle depletion	1.03E+04	5.18E+03	3.73E+01	8.87E-04	2.13E+02	1.18E+01	2.93E+02
Diff. (%)	0.03	0.16	34	0.30	0.70	25	26

There is additional difference of up to ± 15 % from the traditional fuel assembly average method of inventory assaying (Fig. 5). This difference could only be addressed by tracking pin by pin inventory variation. This difference is highest on the isotopes that have different initial amounts like U^{235}. Least difference happens for the isotopes with small quantities like Nb^{94} or those that depend on some nuclide which is identical among all fuel pins like Pu^{239} produced from U^{238}.

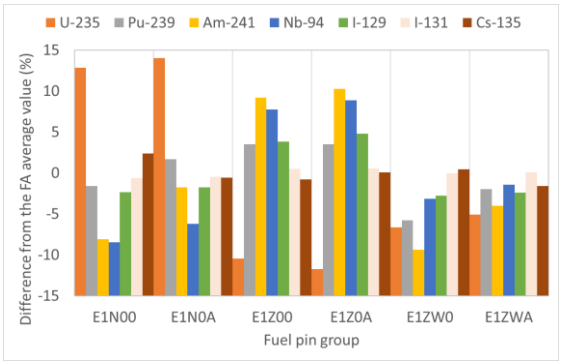

Fig. 5 Difference of the inventory of each group of fuel pins from the fuel assembly average value.

Lastly, the number of time steps is directly proportional to size of an RCD. Depletion calculations over shorter periods of time and hence larger number of time steps means it would take longer to run these depletion calculations thus increasing the database preparation time while decreasing the error that would later arise from the linear interpolation between any two consecutive tabulated inventory values.

3. Conclusions

Reference composition database technique has been used to calculate pin by pin inventory for difference fuel fuel assemblies discharged from UC5 reactor. It is concluded that treating fuel depletion as a three cycle depletion process is of paramount importance, especially if some short lived nuclide was important for the problem. Some nuclides like U^{235} are of course practically independent of the depletion rate.

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