# **Recycling Study of PWR Spent Fuel TRU in the PWR Fuel Assembly**

Ye Seul Cho, Dae Hee Hwang, and Ser Gi Hong\*

Department of Nuclear Engineering, Kyung Hee University 1732 Deogyeong-daero, Giheung-gu, Yongin, Gyeonggi-do, 446-701, Korea \*Corresponding author: sergihong@khu.ac.kr

## 1. Introduction

The safe management of spent fuel generated from PWRs is one of the most important issues in nuclear industry because the accumulated inventories of nuclear spent fuel are expected to exceed the capacities of the spent fuel storage pools inside NPP (Nuclear Power Plant)s in the near future in spite of the use of the dense racks and inter-transportation between different NPP units. For example, it is expected that the spent fuel storage pools of the Kori, Hanbit, and Hanwol units will be saturated in 2028, 2024, and 2026, respectively. Recently, the national Public Engagement Commission on Spent Nuclear Fuel Management has recommended that the geological repository is operated in 2051 after new storage facilities are used to store the spent fuels. Also, the government has supported the research and construction of sodium cooled fast reactor as a longterm solution for reduction of the spent fuel by re-using them.

In this work, an alternative option for reducing the spent fuel is studied, which uses the recycling of TRU (Transuranics) nuclides with MOX fuels (actually TRUO<sub>2</sub>-UO<sub>2</sub>) [1-3] in PWRs. In particular, this study was in the fuel assembly level, and the mass flows of actinides and the neutronic characteristics with TRU recycling are analyzed.

#### 2. Methods and Results

#### 2.1 Computational methods

The DeCART2D code [4] which was developed at KAERI is used to perform the depletion calculations in the fuel assembly level and to analyze the reactivity coefficient and mass flow coupled with ORIGEN-2 [5] for the radioactive decays. DeCART2D solves the multi-group neutron transport equation using MOC (Method of Characteristics) and it uses the subgroup method for resonance self-shielding treatment. The multi-group cross section library is 47 group cross section (DML-E71N047G018-PV01-cr08) which was generated based on ENDF/B-VII.r1. The feed TRU composition corresponds to the one of the PWR spent fuel which is discharged with 50 MWD/kg followed by 10 years cooling (4.5 % initial uranium enrichment). This TRU composition was evaluated with ORIGEN-2.

### 2.2 Assembly Design and Recycling Methods

The  $17 \times 17$  standard fuel assembly is utilized and its dimensions are summarized in Table I. The fuel rod outer diameter is 0.95 cm and the cladding material is zircaloy-4. Three different assembly configurations are investigated to shows the effects of the number of MOX fuel pins on the neutronic performances including mass flow. The configurations of three fuel assemblies are compared in Fig. 1. In the CASE 1 configuration, the MOX fuel pins are arranged only in the peripheral positions (64 MOX fuel pins) and the additional MOX pins are located in the inner and middle rings for the CASE 2 configuration (108 MOX fuel pins) while the CASE 3 configuration is loaded with all the MOX fuel pins.

Table I: Comparison of fuel assembly design parameters.

<u>1</u>		<u> </u>		
Design parameter	CASE 1 (Partial MOX 1)	CASE 2 (Partial MOX 2)	CASE 3 (Full MOX)	
Assembly array	17×17	17×17	17×17	
Number of UO2 rods	200	156	0	
Number of MOX rods (UO <sub>2</sub> -TRUO <sub>2</sub> )	64	108	264	
U enrichment in UO <sub>2</sub> pin (wt%)	4.50	4.50	-	
U enrichment in MOX pin (wt%)	0.20	0.20	4.95	
Pellet density (g/cm <sup>3</sup> )	10.430 (96 %TD)	10.430 (96 %TD)	10.430 (96 %TD)	
Pellet radius (cm)	0.4095	0.4095	0.4095	
Cladding material	Zircaloy-4	Zircaloy-4	Zircaloy-4	
Cladding thickness (cm)	0.0570	0.0570	0.0570	
Gap thickness (cm)	0.0085	0.0085	0.0085	
Rod radius (cm)	0.4750	0.4750	0.4750	
Pin pitch (cm)	1.2234	1.2234	1.2234	
Assembly pitch (cm)	20.879	20.879	20.879	



Fig. 1. Configurations of the fuel assemblies For the CASE 1 and CASE 2, two different TRU recycling

options are considered: 1) recycling of TRUs only from MOX fuel pins (OPTION 1) and 2) recycling TRUs both from UO<sub>2</sub> and MOX fuel pins (OPTION 2). In all the cases, the depletion calculations are performed up to 45 MWD/kg which was set to the cycle length even if the infinite multiplication factors are below 1.0 for some depletion time interval because this discharge burnup is within the typical range of the PWR fuel assemblies. The initial weight percentage of TRUO<sub>2</sub> in MOX fuel pins are fixed to 9.5 wt% for all the cases but the TRUO<sub>2</sub> contents at BOC for the subsequent cycles are determined with the different ways depending on the recycling options. The cooling time between discharge and recharge is assumed to be 7 years. For the recycle OPTION 1, the initial uranium enrichment of UO2 pins is fixed to 4.5 % for every cycle and the fuel assembly is depleted up to 45 MWD/kg, which is followed by the 7 years cooling calculation using ORIGEN-2. The TRUs from the MOX pins are recycled and the reduced actinide inventory is supplemented by depleted uranium in MOX pins. On the other hand, the OPTION 2 is the same as the OPTION 1 except for the fact that all the TRUs from UO<sub>2</sub> and MOX pins are recycled. The CASE 3 fuel assembly is loaded only with MOX pins and all the TRUs remained in the MOX pins are recycled into the next cycle and the reduced amount of actinides are made up by the 4.95 % enriched uranium in MOX pins.



Fig. 2. Comparison of k-inf values for each cycle in CASE 1

The evolutions of  $k_{inf}$  up to 7<sup>th</sup> cycle for two recycling options of CASE 1 are compared in Fig. 2. It is shown in Fig. 2 that the  $k_{inf}$  values decrease as the cycle proceeds and they approach the equilibrium state. In particular, it is noted that the recycling OPTION 2 leads to smaller reductions of  $k_{inf}$  in the subsequent cycles after 1<sup>st</sup> cycle than in the recycling OPTION 1, which comes from the fact that the depleted uranium (DU) content is reduced (i.e., higher TRU content) in MOX pins for OPTION 2 than that for OPTION 1. Also, it should be noted that the TRU contents in MOX pins decreases as the recycling proceeds for OPTION 1 while the one increases for OPTION 2. Fig. 3 compares the evolutions of  $k_{inf}$  values for the recycling OPTION 1 and 2 for the CASE 2 fuel assembly. Fig. 3 shows that the reduction of  $k_{inf}$  values for the cycles after 1<sup>st</sup> cycle is larger than in the CASE 1 fuel assembly due to its larger number of MOX pins. So, these results show that the recycling of TRUs both from UO<sub>2</sub> and MOX pins are more effective in reducing the loss of cycle length than the recycling of TRUs only from MOX pins.



Fig. 4 compares the evolutions of  $k_{inf}$  values for the full MOX case (i.e., CASE 3). For this case, the reduced amount of actinides in MOX pins are supplemented with 4.95 % enriched uranium in MOX pins. For this full MOX pin case, it is expected that the TRU content in MOX pins decreases as the recycling proceeds. Fig. 4 shows that the subsequent cycles after 1<sup>st</sup> cycle have much more reduced  $k_{inf}$  values than in the above cases. The significant reduction in  $k_{inf}$  for the subsequent cycles after 1<sup>st</sup> cycle is due to the reduction of fissile plutonium nuclides in the recycled TRUs.

The changes of major TRU nuclides' inventories over the cycles for the CASE 2 with recycling OPTION 2 and the CASE 3 are compared in Fig. 5 and Fig. 6, respectively. Specifically, the inventories given in these figures are the ones at EOC of the cycles. For the CASE 2 with recycling of TRUs both from UO<sub>2</sub> and MOX pins, <sup>239</sup>Pu decreases initially but rapidly stabilized as the recycling proceeds. The other fissile nuclide <sup>241</sup>Pu very slowly decreases but its change is very small. On the other hand, the fertile Pu nuclide increases as recycling proceeds. In particular, <sup>242</sup>Pu initially increases and then stabilized from EOC of 6<sup>th</sup> cycle while <sup>240</sup>Pu slowly increases but its change is very small up to 7<sup>th</sup> cycle and <sup>238</sup>Pu also initially increases but it is rapidly stabilized from 3<sup>rd</sup> cycle. <sup>241</sup>Am shows only a small change while <sup>237</sup>Np slowly decrease and it is stabilized shortly. On the other hand, the CASE 3 assembly with TRU recycling shows different trends

of the changes in the nuclide-wise inventories from the above case. For this case, the both fissile Pu nuclides decrease as recycling. In particular, <sup>239</sup>Pu rapidly decreases. This different trend from that of the CASE 2 with recycling of TRU from UO<sub>2</sub> pins and MOX pins is due to the fact that the TRUs are not fed from the existing PWR spent fuels in the CASE 3. In particular, it is noted that <sup>240</sup>Pu decreases as recycling and it is not stabilized up to 7<sup>th</sup> cycle and <sup>238</sup>Pu initially increases up to 3<sup>rd</sup> cycle and then slowly decreases. <sup>241</sup>Am and <sup>237</sup>Np decreases more rapidly than the CASE 2 with recycling of TRU from UO<sub>2</sub> pins and MOX pins.



Fig. 4. Comparison of k-inf values for each cycle in CASE 3



Next, the reactivity coefficients (i.e., MTC and FTC) are analyzed and compared. Figs. 7 and 8 compare the MTC values of the CASE 2 assembly with recycling TRUs from MOX pins and UO<sub>2</sub> pins and the CASE 3 for all the recycling cycles up to 7<sup>th</sup> cycle. These figures show that MTC values become more negative as recycling proceeds. In particular, the

full MOX case (i.e., CASE 3) have overall less negative MTC values than the CASE 2 with recycling OPTION 2 for all the cycles up to  $7^{\text{th}}$  cycle.



Fig. 7. Comparison of MTC for recycling cycles in CASE 2 (OPTION 2)



Fig. 8. Comparison of MTC for recycling cycles in CASE 3

The FTC values for the above two cases are compared in Figs. 9 and 10, respectively. As shown in these figures, the FTC values also become more negative as recycling proceeds. In particular, it is noted that the full MOX case has relatively smaller changes as burnup within each recycle than in the CASE 2 assembly with recycling OPTION 2.



(OPTION 2)



Next, the detailed mass balance for PUs and MAs are analyzed over the recycling cycles. Table II summarizes the results of the TRU nuclides' inventory analysis for the CASE 2 assembly. Also, the inventory analysis for the reference assembly comprised of only  $UO_2$  pins are considered for comparison in Table II. From Table II, it is shown that the net destructions of Pu and MA nuclides are observed in the

recycling OPTION 1 but the amount of destructions decreases as recycling proceeds while Pu and MA nuclides in the recycling OPTION 2 increase but their increased amounts are quite smaller than the reference UO<sub>2</sub> case. But it should be noted in Table II that the discharge amount for the recycling OPTION 1 considers only the TRUs from the MOX pins (not the TRUs generated in the  $UO_2$  pins). That is to say, the net increase of TRUs for the recycling OPTION 1 means TRUs mass change only for the MOX pins. For example, the CASE 2 assembly generates net amount of 0.96 kg TRU through 7<sup>th</sup> cycle while the reference case having all UO<sub>2</sub> pins generates 6.63 kg TRU, which means that the recycling of TRUs significantly reduces the production of TRUs. Table III summarizes the mass balances for the CASE 3 assembly. Table III shows that the recycling of TRUs for the full MOX case leads to more substantial net decreases of TRU (both Pu and MA) and the amount of net decreases of TRU decreases as recycling proceeds. The net decreases of TRU through 1st and 7<sup>th</sup> cycles are estimated to be 4.28 kg and 1.19 kg which correspond to 9.24 % and 3.8 %, respectively.

Cycle		Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5		Cycle 6		Cycle 7		Pafaranaa	
	Option	01	02	01	02	01	02	01	O2	01	02	01	O2	01	O2	(All UO <sub>2</sub> )	
	Charge (kg)	15.94	15.94	11.89	16.00	9.48	16.29	7.89	16.63	6.83	16.95	6.11	17.25	5.62	17.54	0.00	
PU	Discharge (kg)	11.89	16.00	9.48	16.29	7.89	16.63	6.83	16.95	6.11	17.25	5.62	17.54	5.29	18.01	6.16	
	Net increase (kg)	-4.05	0.06	-2.41	0.29	-1.58	0.34	-1.07	0.32	-0.72	0.30	-0.49	0.29	-0.34	0.47	6.16	
	Charge (kg)	3.00	3.00	2.47	2.90	2.15	3.02	2.01	3.20	1.97	3.38	1.94	3.57	1.91	3.72	0.00	
MA	Discharge (kg)	2.47	2.90	2.15	3.02	2.01	3.20	1.97	3.38	1.94	3.55	1.91	3.72	1.86	3.78	0.47	
	Net increase (kg)	-0.53	-0.11	-0.32	0.12	-0.14	0.18	-0.05	0.18	-0.03	0.17	-0.03	0.15	-0.05	0.06	0.47	
TRU	Charge (kg)	18.94	18.94	14.37	18.90	11.61	19.31	9.92	19.83	8.79	20.33	8.05	20.82	7.53	20.82	0.00	
	Discharge (kg)	14.37	18.90	11.61	19.31	9.92	19.83	8.79	20.33	8.05	20.80	7.53	21.26	7.15	21.78	6.63	
	Net increase (kg)	-4.57	-0.05	-2.76	0.41	-1.69	0.52	-1.13	0.50	-0.75	0.47	-0.52	0.44	-0.38	0.96	6.63	

Table II: Comparison of mass balances (kg) in the CASE 2 fuel assembly

Table III: Comparison of mass balance in the CASE 3 and REFERENCE assemblies

	Cycle	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	Cycle 6	Cycle 7	Reference (All UO <sub>2</sub> )
PU	Charge (kg)	38.98	34.95	32.22	30.07	28.29	26.78	25.51	0.00
	Discharge (kg)	34.95	32.22	30.07	26.78	26.78	25.51	24.44	6.16
	Net increase (kg)	-4.05	-2.65	-2.15	-1.49	-1.26	-1.26	-1.07	6.16
MA	Charge (kg)	7.35	7.10	6.66	6.07	5.90	5.90	5.76	0.00
	Discharge (kg)	7.10	6.66	6.31	5.90	5.76	5.76	5.64	0.47
	Net increase (kg)	-0.23	-0.44	-0.34	-0.18	-0.14	-0.14	-0.11	0.47
TRU	Charge (kg)	46.33	42.05	38.86	34.35	32.67	32.67	31.27	0.00
	Discharge (kg)	42.05	38.86	36.38	32.67	31.27	31.27	30.08	6.63
	Net increase (kg)	-4.28	-3.19	-2.48	-1.68	-1.40	-1.40	-1.19	6.63

Finally, the characteristics of the 1kg TRU nuclides that are discharged and recycled are evaluated using ORIGEN-2 and inter-compared. These characteristics include the decay heat, radiotoxicity, and radioactivity. Figs. 11, 12, and 13 compare the radioactivities (curies), decay heats (W), and radiotoxicities (radioactive ingestion hazard,  $m^3$  water), respectively, at EOC of 7<sup>th</sup> cycle for all the cases including the reference one having only UO<sub>2</sub> pins. These figures show that the TRUs from all the cases considering TRU recycling have

much higher values of these quantities than the reference case, which means that the fabrications of the MOX fuels are much more difficult due to higher radioactivity and decay heat than the reference  $UO_2$  fuels. These high radioactivities, radiotoxicities, and decay heats for the recycled TRUs are resulted from the higher contents of MA nuclides and the even mass number plutonium nuclides.



Fig. 11. Comparison of radioactivity at EOC of 7<sup>th</sup> cycle versus cooling time (1 kg recycled TRU)



Fig. 12. Comparison of decay heat at EOC of 7<sup>th</sup> cycle versus cooling time (1 kg recycled TRU)



versus cooling time (1 kg recycled TRU)

### 3. Summary and Conclusions

In this work, the recycling of TRU nuclides in PWR fuel assemblies were analyzed using the fuel assemblylevel depletion using DeCART2D and ORIGEN2 decay calculations to understand the neutronic characteristics including the characteristics of recycled TRUs such as radioactivity, decay heat, and radiotoxicity. In particular, three different loading ways of MOX pins including one fuel MOX pin case are considered for the 17×17 fuel assembly. From the analysis, it was found that 1) the recycling of TRUs from only MOX pins or from MOX and UO<sub>2</sub> pins significantly reduces the reactivity and so the cycle length for the subsequent cycles after 1<sup>st</sup> cycle, and significantly reduces the net TRU production in comparison with the no recycling case, 2) the MTC and FTC values become more negative as the recycling proceeds, 3) the full MOX case showed the considerable net destruction of TRU up to 7<sup>th</sup> cycle (e.g., 1.19 kg TRU during 7<sup>th</sup> cycle), and 4) the radioactivity, radiotoxicity, and decay heat of the recycled TRUs are significantly higher than those of the reference no recycling case, which would make it difficult to fabricate the fuel with recycled TRUs.

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