Study on Characteristics of Spent PWR Cladding Hull for Categorizing into Non-TRU waste

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1. Introduction

AFCI and GEN-IV programs aim for decreasing the high level radioactive wastes to be disposed. They also try to get valuable materials to recycle as resources such as uranium and plutonium. On the other hand, cladding hull expected to be one-thirds in volume of spent fuel assembly has not studied so much in the point view of recycling to reuse. Since traditional process of reprocessing was wet process, cladding hull generating through the reprocessing process was unavoidably contaminated with TRU by acid solvent during the process[1]. Therefore, cladding hull has been classified into TRU wastes or high level wastes.

According to the strategy for TRU high level radioactive wastes of USA as well as Korea, it regulates in two respects. One is activity and the other is heat generation. In respect of activity, TRU waste contains more than 100 nCi/kg of alpha emits with longer half life than 20 years and higher than 92 in atomic number. Also, wastes are categorized into TRU waste when it generates higher than 2kW/m^3, in the respect of heat generation.

Our results as well as literatures, almost all of TRU nuclides in the cladding hull are responsible for remained uranium and plutonium owing to pellet-cladding interaction. In addition, recoiled fission products on the surface of the cladding hull serve as heat generator. Up to now, decontamination of the cladding hull generating from the reprocessing of wet process is regarded as valueless and un-economic works owing to the amount of second waste produced[2].

2. Characteristics of the cladding hull from conventional process

Restani et al.[3] investigated the characteristics of the cladding hull of PWR fuel whose burn-up of 30,000 MWd/MTU and cooling time was 5 years. The dose rate and radioactivity are shown in Table 1. The γ radio-activities of the hull were measured with a high purity Ge detector. The γ dose rate at a distance of 0.5m was about 10~50mR/hr(average 25±8mR/hr). The species of the fission products are identified from the analysis of the gamma ray spectrum obtained by the gamma spectrometry measurement system. β and γ radioactivity from the hulls are dominated by Sb-125 from the alloying element tin.

The contents of actinides are obtained by the isotope dilution analysis method. A specimen of the zircaloy-4 hull was dissolved in 3M HNO_3/2M HF. The results are shown in Table 2. Table 2 shows that the average contents of uranium and plutonium are 1,130mg U/kg-Zry and 13.6 mg Pu/kg Zry, respectively. In the mean time, the result of the α- autoradiography to evaluate the qualitative TRU distribution shows that the contents of uranium and plutonium are 380~2130mg U/kg Zry and 6.3~25.3 mg Pu/kg Zry, respectively.

The concentration depth profile of the radio-nuclides is analyzed with a shieled secondary ion mass spectrometer by Restani. The SIMS analysis shows that uranium adheres mainly to the zircaloy surface and that the concentration in the oxide layer drops sharply. The fission product penetrates at a considerable distance to about 10 μm as a form of recoil particles. The lightest element, Sr-88, penetrates the farthest distance of 12 μm.

3. Results and Discussions

In this study, the characteristics of cladding hull which are generated from the dry processes such as DUPIC and metal transformation of spent fuel was investigated[4]. The results were quite different comparing to the wet process of conventional treatment method. For this, three kinds of hulls of different burn-up spent PWR fuels - 27,000MWd/tU, 50,000MWd/tU and 60,000MWd/tU - were investigated. The results said that the cladding hull is layered with attached uranium and plutonium on the outer surface of the cladding hull, oxide layer in thickness of 2~10 ㎛, and recoiled fission products about 12 ㎛ depth in order.

4. Conclusion

Oxide layer of the cladding hull was not so strong comparing to the zircaloy metal that controlling the thickness or degree of oxidation of the oxide layer would like to provide a clue to peel off the contaminated layer of the cladding hull. Some approaches were carried out in cold tests, and valuable results were attained.

Calculation results by ORIGEN code for de-laminated cladding hull by dry process showed that cladding hull is possible to categorize into non-TRU wastes after treatment.

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Table 1. Activities of fission products, activation products and α-nuclides in Zircaloy-4 hulls from reprocessing process(cooling period: 5years, burn-up: 30,000MWd/tU)

Fission Products		Activation Products		α -nuclides	
nuclide	Radioactivity (mci/kg-Zry)	nuclide	Radioactivity (mci/kg-Zry)	nuclide	Radio-activity (mci/kg-Zry)
Cs-137	542	Sb-125	760	Pu(t)	4.51
Ru-106	458	Co-60	64	Cm-244	1.24
Cs-134	162	Mn-54	3.4	Am-241	0.90
Ce-144	70				
Eu-154	21				
total	1253		total	827.4	total

Table 2. Uranium, Plutonium concentrations and isotopic compositions in Zircaloy-4 hull

Uranium		1,132 mgU/kg-Zry	Plutonium		13.6mgPu/kg-Zry
Uranium Isotopes	U-234	0.025 wt%	Plutonium Isotopes	Pu-238	1.33 wt%
	U-235	1.19 wt%		Pu-239	61.01 wt%
	U-236	0.38 wt%		Pu-240	23.12 wt%
	U-238	98.41 wt%		Pu-241	10.08 wt%
	-	-		Pu-242	4.46 wt%
	total	100wt%		total	100 wt%

Table 3. Radioactivity of hull calculated by ORIGEN-II code (Ci/ton-Zry)

ASSY	DISCHARGE	5.0YR	10.0YR	15.0YR	20.0YR	25.0YR	30.0YR	35.0YR	40.0YR	45.0YR	50.0YR
U	1.49E+00	2.16E-07	1.79E-07	1.50E-07	1.28E-07	1.11E-07	9.73E-08	8.69E-08	7.89E-08	7.28E-08	6.81E-08
NP	1.48E+00	1.50E-06	1.50E-06	1.50E-06	1.50E-06	1.50E-06	1.50E-06	1.50E-06	1.50E-06	1.50E-06	1.50E-06
PU	3.98E-02	7.33E-03	5.78E-03	4.57E-03	3.62E-03	2.86E-03	2.27E-03	1.81E-03	1.44E-03	1.16E-03	9.28E-04
AM	1.58E-02	7.76E-05	1.28E-04	1.67E-04	1.97E-04	2.21E-04	2.38E-04	2.52E-04	2.62E-04	2.69E-04	2.74E-04
CM	3.49E-03	1.48E-04	1.21E-04	1.00E-04	8.29E-05	6.87E-05	5.69E-05	4.71E-05	3.91E-05	3.24E-05	2.69E-05
BK	2.54E-10	1.08E-12	2.07E-14	3.97E-16	7.61E-18	1.30E-19	2.13E-22	2.13E-22	2.13E-22	2.13E-22	2.13E-22
CF	1.68E-12	9.83E-13	6.85E-13	5.32E-13	4.35E-13	3.67E-13	3.15E-13	2.76E-13	2.45E-13	2.21E-13	2.03E-13
SUM(ci)	3.03E+00	7.55E-03	6.03E-03	4.84E-03	3.90E-03	3.15E-03	2.57E-03	2.11E-03	1.75E-03	1.46E-03	1.23E-03
Weight(g)	3.18E+05	3.18E+05	3.18E+05	3.18E+05	3.18E+05	3.18E+05	3.18E+05	3.18E+05	3.18E+05	3.18E+05	3.18E+05
Ci/ton	9.53E+00	2.37E-02	1.89E-02	1.52E-02	1.22E-02	9.91E-03	8.07E-03	6.63E-03	5.48E-03	4.58E-03	3.87E-03

Table 4. Heat generation of hull calculated by ORIGEN-II code (W/kg-Zry)

ASSY	DISCHAGE	5.0YR	10.0YR	15.0YR	20.0YR	25.0YR	30.0YR	35.0YR	40.0YR	45.0YR	50.0YR
AP+ACT+F	8.162688	0.019246	0.008439	0.003948	0.001924	0.000964	0.000491	0.000253	0.000132	6.98E-05	3.76E-05

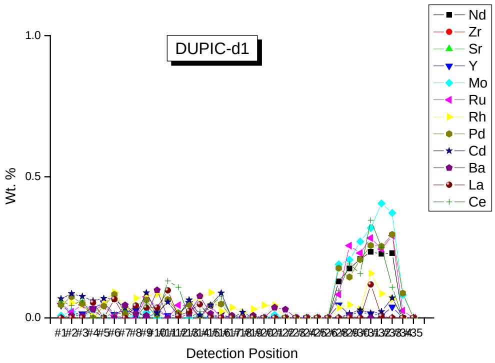

Fig. 1. Concentration profile of fission products of cladding hull with 27,000MWd/tU burn-up.

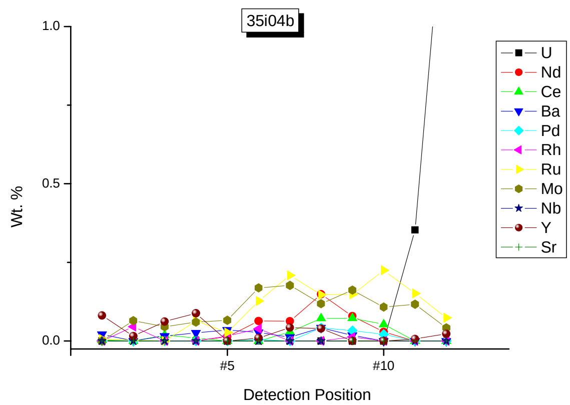

Fig. 2. Concentration profile of fission products of cladding hull with 60,000MWd/tU burn-up.