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Toxicity of Radioactive Wastes Generated from PEACER Spent Fuel

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Abstract – *Assessment on the back end fuel cycle, in PEACER (Proliferation-resistant Environmental-friendly Accident-tolerant Continuable and Economical Reactor) that was designated as a new transmutation concept, was performed. Recovery system of uranium and TRU for PEACER is based on pyroprocessing. In the assessment of long-lived fission products (LLFP) wastes, initially ^{90}Sr and ^{137}Cs are dominant contributor nuclides until 30 years and especially ^{90}Sr and ^{137}Cs have the highest activity and decay heat than other LLFP. In this study, recovery of ^{90}Sr and ^{137}Cs is recommended for reducing of wastes loading. The acceptable decontamination factor is investigated by the toxicity of PEACER spent fuel. The acceptable decontamination factor is about 1.02E+05 for the actinides from PEACER spent fuel after 10 years cooling, 4.26E+05 after 100 years cooling, 1.97E+04 after 300 years cooling, 9.52E+03 after 1000 years cooling.*

1. INTRODUCTION

The long-term hazard of radioactive wastes arising from nuclear energy production is a matter of continued discussion and public concern in many countries. By the use of partitioning and transmutation of the actinides and some of the long-lived fission products (LLFP), the radiotoxicity of the high-level waste and, possibly, the safety requirements for its geologic disposal can be reduced compared with the current once-through fuel.

P&T method of radioactive waste from spent fuel is more attractive because of highly concerning on the protection and the difficulty in radioactive waste disposal site selection in Korea. In the previous works SNU (Seoul National University) proposed a new transmutation concept designated as PEACER (Proliferation-resistant Environmental-friendly Accident-tolerant Continuable and Economical Reactor). PEACER includes the concept of pyroprocess-based partitioning system and lead-bismuth cooled transmutation reactor.

In order to make this concept more attractive, it is hoped to convert all the final waste into the class of low-level waste (LLW). The waste problem will become severer in the future than present time, since the long-life radioactive waste will accumulate as time passes even in this situation.

In this paper, it was studied on the back end fuel cycle in PEACER and analyzed toxicity of total wastes from pyroprocessing in PEACER. Furthermore the principal goal of this study is to investigate the feasibility of converting PEACER wastes into LLW based on pyroprocess technology and practical acceptable range of decontamination factor (DF).

2. TRANSMUTATION REACTOR

The earlier conceptual design of PEACER was developed by combining the Integral Fast Reactor(IFR) approach with the heavy liquid metal cooled reactor technology. As its basic core design, an LWR-type square-lattice is employed with metallic fuel elements having high pitch-to-diameter ratio in order to accommodate the viscosity nature of lead-bismuth coolant. Both uranium and TRU were used as fuel

materials and its thermal power output has 1,560 MWt.[1] PEACER reactor design parameters are shown in Table I. Also composition of charged PEACER fuel and discharged to pyroprocessing is shown in Table II.

Table I. Reactor design parameters

Thermal power output	1,560 MWt
HM(Kg) 1/4 Core	3.69E+03
Batch	3
Fuel cycle (day)	365

Table II. PEACER fuel composition(1/4 Core)

ISOTOPE	Charged(gram)	Ddischarged(gram)
TH232	8.16E-06	8.00E-06
PA233	1.36E-20	1.75E-14
U233	1.14E-04	1.14E-04
U234	3.14E+00	2.96E+00
U235	1.79E+00	1.33E+00
U236	3.72E+00	3.46E+00
U238	8.22E+02	7.68E+02
NP237	1.92E+01	1.33E+01
PU238	1.51E+01	1.35E+01
PU239	1.94E+02	1.52E+02
PU240	1.67E+02	1.50E+02
PU241	4.16E+01	3.36E+01
PU242	3.96E+01	3.68E+01
AM241	1.08E+00	7.00E-01
AM243	1.29E+01	1.22E+01
CM242	6.17E-03	4.98E-03
CM243	4.24E-03	3.83E-03
CM244	6.43E+00	6.54E+00
CM245	1.70E+00	1.68E+00
CM246	9.22E-01	9.20E-01
TOTAL	1.33E+03	1.20E+03
U	8.30E+02	7.76E+02
PU	4.57E+02	3.86E+02
AM	1.40E+01	1.29E+01
CM	9.06E+00	9.15E+00
NP	1.92E+01	1.33E+01
TRU	4.99E+02	4.21E+02
HM	1.33E+03	1.20E+03

Waste Stream

In order to evaluate the produced actinide wastes from PEACER, it is analyzed in equilibrium state by REBUS code. Table III shows annually total discharged actinide wastes from PEACER 1,560 MWt reactor.

TABLE III. Disharged actinide wastes annually from PEACER 1,560 MWt Reactor

U		3,820kg
TRU	PU	1,900kg
	AM	63.3kg
	CM	45.0kg
	NP	65.5kg
	Sub total	2,073.8kg
Total HM		5,893.8kg

On the PEACER conceptual design, it is aimed to convert all the final waste into the class of low-level waste and it is also required to transmute two important fission products Tc-99 and I-129. Figure 1 shows the simplified flow sheet of back end fuel cycle in PEACER.

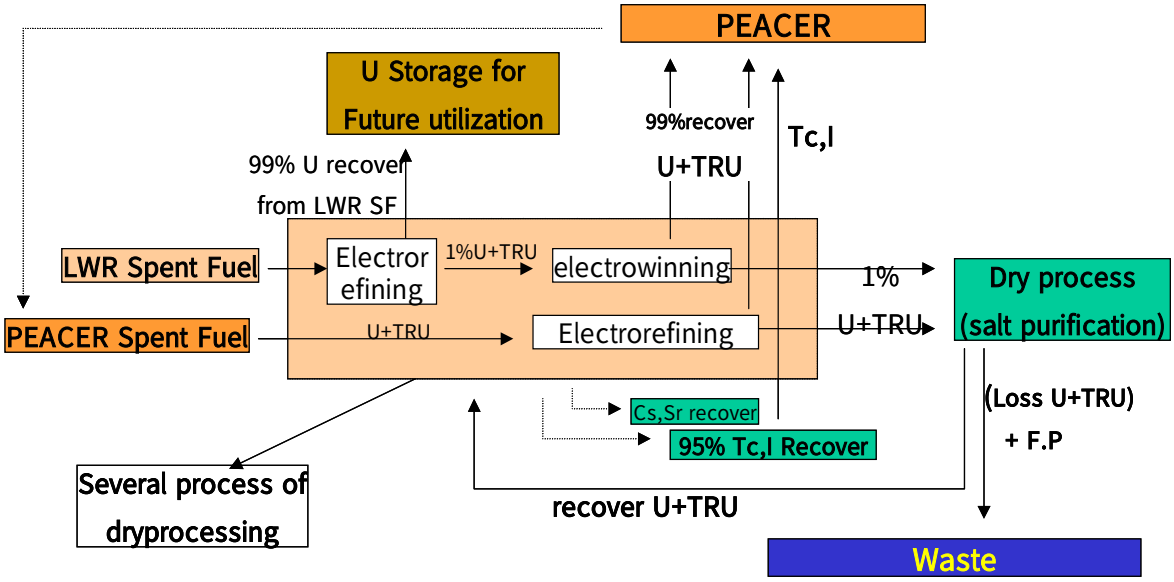

Fig. 1. Flow sheet of back end fuel cycle in PEACER

A reference pyrochemical process for PEACER is based on LiCl-KCl molten salt and liquid cadmium cathode. LiCl-KCl based pyroprocess has been developed at ANL and recently has been redesigned for ATW system. LiCl-KCl based process has been also developed at CRIEPI. Key processes of Pyrochemical partitioning process are electrolysis process for TRU recovery that is electrorefining or electrowinning and salt purification including reductive extraction process for waste treatment Decontamination of TRU in LLW is subject to the combination of electrolysis and reductive extraction process. Decontamination factor (DF) is introduced for indication of process performance. Overall DF in pyrochemical partitioning is defined as the ratio of mass of TRU loaded into the process to TRU lost into waste stream expressed as follows;

$$DF_t = \frac{\text{The loaded TRU into pyrochemical process}}{\text{The lost TRU into waste stream}} \quad (1)$$

DF_t is a function of TRU loss fractions in electrorefining process and reductive extraction process. Asymptotically, DF_t is expressed as a reciprocal of a product of TRU loss fractions in electrorefining process and in reductive extraction process. In early study, PEACER pyroprocessing system that has 10^5 of DF_t was conceptually designed.[2]

3. LONG-LIVED FISSION PRODUCTS

In order to evaluate the produced total wastes from pyroprocessing, we assumed that LWR has 1 GWe capacity, 40 years lifetime with spent fuels discharged at 33,000 MWD/MTU burnup and 30 years cooling time. And as it can see Figure 1, by the PEACER pyrochemical partitioning process, about 99% of uranium in the spent LWR fuel is recovered and stored in the metallic form for the future utilization. The nuclide inventory of LWR was obtained by ORIGEN2 code. Table IV shows that the total waste production of long-lived fission products (LLFP) that are generated from 20 LWR's with total electricity of 800 Gwe-yr.

It was also obtained by ORIGEN2 code. To improve repository performance, ^{99}Tc and ^{129}I need to be separated from waste stream and to be transmuted to stable nuclides. According to early conceptual PEACER design, these two fission products are assumed to be recovered with 95% efficiency.[3]

Table IV. Total waste production generated from LLFP

Nuclide	Mass [g]	Activity [Ci]	Heat load [watts]
SE 79	2.05E+05	1.43E+04	3.55E+00
SR 90	9.33E+06	1.24E+09	5.13E+06
ZR 93	2.51E+07	6.30E+04	7.33E+00
TC 99	1.35E+06	2.28E+04	1.14E+01
PD107	7.62E+06	3.92E+03	2.32E-01
SN126	9.56E+05	2.71E+04	3.38E+01
I129	3.12E+05	5.51E+01	2.55E-02
CS135	1.05E+07	1.21E+04	4.02E+00
CS137	1.79E+07	1.87E+09	1.11E+06
SM151	3.79E+05	9.97E+06	1.17E+03
TOTAL	7.36E+07	3.11E+09	1.34E+07

In the sense of low-level wastes (LLW) disposal site, heat load must be one of the most important factors as much as concentrations of nuclides. IAEA has classified LLW as wastes with less than 2 kW/m^3 heat load. Figure 2 shows decay heat that was generated from each long-lived fission products. As it can see Figure 5, initially ^{90}Sr and ^{137}Cs have the highest heat load. The most important thing that was required to meet the regulation for LLW disposal is concentration of nuclides, also known as activity. Figure 3 shows the trends of activity in time evolution. Initially ^{90}Sr, ^{137}Cs and ^{151}Sm are dominant contributor nuclides until 30 years and especially ^{90}Sr and ^{137}Cs have the highest activity than other LLFPs. It will be required very large dilution volume to meet the regulation for LLW disposal site due to high activity and heat of these two nuclides if it could be wanted to dispose initially all of LLFPs in LLW disposal site. If ^{90}Sr and ^{137}Cs are separated from waste for cooling, the dilution volume that was required to dispose in LLW disposal site will be reduced as a large amount. Therefore in this study, recovery of ^{90}Sr and ^{137}Cs as well as ^{99}Tc and ^{129}I is recommended for reducing of wastes loading.

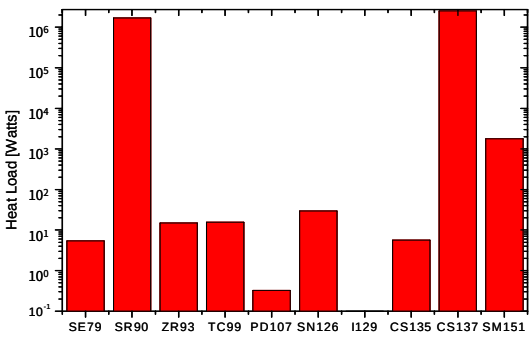

Fig. 2. Decay heat generated from LLFP

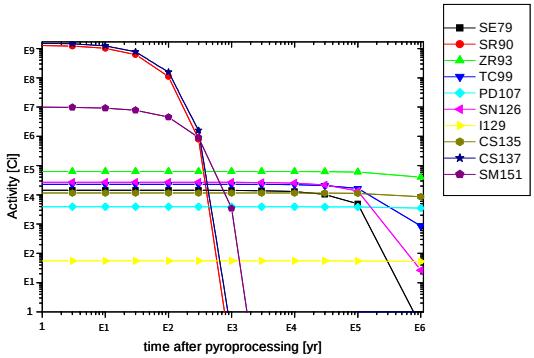

Fig. 3. Activity generated from LLFP

4. TOXICITY

The risk may be the best index for evaluation of this system, but it depends on the disposal method and environmental conditions, and furthermore the analysis for evaluation has usually large uncertainties and errors. On the other hand, toxicity per unit radioactivity is given for each of most nuclides, and it can be used like a nuclear data. These have several kinds of toxicity units and meet some ambiguity for choice. However the relative difference among these units is allowable.[5] Long-term tendency of radio-toxicity (defined as the volume of the water which would have to be used to dilute a given quantity of waste(key actinide nuclides) so that the water could be used as drinking water like following equation.

$$Toxicity = \sum_i \frac{\lambda_i N_i}{C_i} \qquad (2)$$

In this paper the annual limit on intake (ALI) is employed as the radioactive ingestion hazard.

All of the actinides are recycled into the reactor and confined in the system. However it is impossible to confine them perfectly in the system, but a small part may leak from the system finally into the biosphere. The highest possible mechanism for leakage may be contamination to be discharged to the environment. In this paper, the upper limit on leakage at the pyroprocessing process acceptable from the point of toxicity balance between production from PEACER and pure natural uranium is discussed. In other words the leakage rate means a reciprocal of decontamination factor (DF).

Toxicities expressed in ALI Ingestion Hazard Index of actinides discharged annually from PEACER 1,560 MWt Reactor are shown in Table V. The toxicity of actinides changes along the time after leaving the reactor as shown in Figure 4 for typical system. The maximum acceptable leakage rate depends on the time after leakage as shown in Table VI. And also Table VI shows the acceptable decontamination factor(DF) that total toxicity of actinides is the same corresponding equilibrium pure natural uranium toxicity. From the results, the acceptable DF is about 1.02E+05 for the actinides from PEACER spent fuel after 10 years cooling, 4.26E+05 after 100 years cooling, 1.97E+04 after 300 years cooling, 9.52E+03 after 1000 years cooling.

TABLE V. Toxicities in ALI Ingestion Hazard Index of actinides annually from PEACER 1,560 MWt Reactor

year	1	100	300	1000
RA	4.18E+06	1.35E+07	2.95E+06	1.45E+07
TH	1.24E+06	3.96E+06	8.99E+05	1.33E+06
U	5.10E+06	7.35E+06	9.81E+06	1.06E+07
NP	4.66E+08	1.37E+08	1.41E+08	1.45E+08
PU	2.32E+11	1.01E+11	4.70E+10	3.09E+10
AM	1.64E+10	9.21E+10	6.80E+10	2.43E+10
CM	5.84E+11	1.24E+10	9.03E+08	7.82E+08

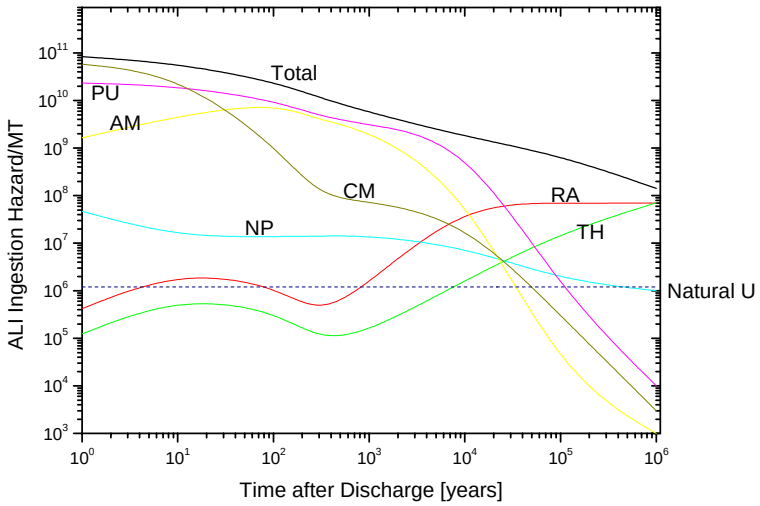

Fig. 4. Toxicities expressed in ALI of actinides per year from PEACER

TABLE VI. Acceptable Decontamination Factor(DF) of actinides at pyroprocessing

Decay time (years)	Leakage rate	DF
10	9.80E-06	1.02E+05
100	2.35E-06	4.26E+05
300	5.08E-05	1.97E+04
1,000	1.05E-04	9.52E+03

5. CONCLUSIONS

Assessment on the back end fuel cycle, in PEACER (Proliferation-resistant Environmental-friendly Accident-tolerant Continuable and Economical Reactor) that was designated as a new transmutation concept, was performed. While the high-level radioactive waste problem may be one of the most important problems for the future, PEACER can propose some systems where the toxicity in the environment is reducing. To reduce the toxicity level of leaked actinide, the leakage rate should be very small. After 10 years cooling, the acceptable decontamination factor is 1.02E+05. And the acceptable DF is about 4.26E+05 after 100 years cooling, 1.97E+04 after 300 years cooling, 9.52E+03 after 1000 years cooling. Though this period seems to be too long for human control, it may be a confirmed period for the waste confinement by underground artificial barrier judging from archaeological studies.

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