INVESTIGATION OF RADIOACTIVE INVENTORY IN THE TRU SOLID WASTES GENERATED FROM THE TOKAI REPROCESSING PLANT

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ABSTRACT

Japan Nuclear Cycle Development Institute (JNC) has started a project for investigation of radioactive inventory in the TRU solid wastes generated from the Tokai Reprocessing Plant (TRP) since 2003, as a preparation step for disposal of TRU wastes.

The investigation procedure was discussed considering the property of nuclides deposited on the TRU wastes. The sampling optimization was discussed considering the feature of wastes for the efficient evaluation. Nuclide composition analysis was conducted for 6 samples of TRU solid wastes, and the existing ratios to some non-destructively analyzable nuclides (key nuclides) were discussed.

The correlation on the concentration between Cs-137 and other fission products was observed except for H-3.The correlation between Am-241 and other TRU nuclides was also observed.

INTRODUCTION

In Japan, it is planned to dispose of TRU wastes in underground. In order to dispose of the TRU wastes in underground, it is necessary to verify radioactive inventory in the TRU wastes.

To develop the method of verifying the quantities of nuclides in the TRU wastes, it is necessary to investigate the correlation on the concentration between a key nuclide and a scaled nuclide which is difficult to measure by non-destructive measurement method of the TRU solid wastes generated from TRP.

In Japan, the scaling factor method has been applied to verify the radioactivity of law level waste packages generated from nuclear power plants.

On the other hand, the reprocessing plant is a chemical plant to recover plutonium and uranium from spent fuels.

Therefore it seems to be difficult to apply the scaling factor method for verification of the TRU wastes generated from the reprocessing plant, because of the complex nuclide composition of TRU solid waste.

We investigated the radioactive inventory in the TRU solid wastes generated from TRP to apply the scaling factor method for the verification of radioactive inventory in the TRU solid wastes generated from TRP.

RADIOACTIVITY INVESTIGTION

To apply the scaling factor method for the verification of the radioactive inventory in the TRU solid wastes, it is necessary that a key nuclide and a scaled nuclide have both similar generation mechanism in reactor and similar behavior in the reprocessing plant. In addition, the correlation on the concentration between the key nuclide and the scaled nuclide should be observed by radiochemical analysis of the wastes.

Therefore, for evaluation of scaling factors of TRU wastes, it is necessary to characterize nuclides in the wastes by both theoretical method and radiochemical method.

Theoretical characterization of nuclides in TRU wastes was carried out from the viewpoint of their generation in reactors and their behaviors in the reprocessing plant. The nuclides having similar property are categorized into the same group to estimate the correlation between scaled nuclides and key nuclides.

The radiochemical analysis of the TRU wastes was carried out for the waste samples collected from the typical sampling points in the reprocessing plant.

Key Nuclide

The key nuclide must be measurable by non-destructive method and be correlated with the scaled nuclide.

There are three non-destructive measurement methods of the key nuclides in a waste package, passive gamma measurement method, passive neutron measurement method, and active neutron measurement method.

The nuclides analyzable by passive gamma measurement method are Co-60, Cs-137, and Am-241. The nuclides analyzable by passive neutron measurement method are even-mass plutonium isotopes (Pu-238, Pu-240, and Pu-242). The nuclides analyzable by active neutron measurement method are fissile nuclides (U-235, Pu-239, and Pu-241). The neutron measurement methods cannot analyze each nuclide individually, but sum of nuclides.

Those nuclides are able to be key nuclides.

Mechanism of generation of nuclide in reactor

Fission products and TRU nuclides are generated by irradiation of uranium in reactor. Therefore, those nuclides have similar generation mechanism in reactor.

Activation products such as Co-60, Ni-59/63, and Nb-94 are generated by activation of corrosion products deposited on fuel assembly.

Carbon-14 is produced by the (n,p) reaction on N-14 in fuel and clad.

Therefore, from the viewpoint of the generation mechanism in reactors, the nuclides in the TRU wastes are categorized into 3 groups, (1) fission products and TRU nuclides, (2) activation products, and (3) C-14.

Behavior of Nuclide in the Reprocessing Plant

TRP uses a purex process. The reprocessing plant recovers plutonium and uranium and removes fission products and other TRU nuclides from spent fuels.

From the viewpoint of the behavior in the reprocessing plant, the nuclides in spent fuel are categorized generally into some groups.

Plutonium isotopes flow into the stream of purified plutonium. Uranium isotopes flow into the stream of recovered uranium.

Other TRU nuclides, fission products, and activation products, flow into the aqueous waste stream. Furthermore, nuclides which flow into the aqueous waste are categorized to volatile nuclides such as H-3, C-14, and I-129 which flow into the off-gas treatment step and to non-volatile nuclides which flow into the high level aqueous waste (HLW) stream. For example, Co-60 and Cs-137 flow mainly into HLW stream.

Tritium is carried in reprocessing plant as tritiated water, and the behavior of H-3 is different from other nuclides.

In TRP, dissolver off gas is treated with alkaline trap. Carbon-14 and I-129 which contained in dissolver off gas, is carried through the off gas treatment step to the low-level aqueous waste (LLW) treatment step.

The behavior of Am-241 in the reprocessing plant appears to be similar to plutonium rather than other americium isotopes, because Am-241 is generated as a daughter of Pu-241 in the reprocessing plant.

Neputunium-237 flows into both purified plutonium stream and HLW stream, because neptunium has many oxidation states and complex behavior in the purex process. Therefore, Np-237 is seems to be correlated with plutonium or Cs-137.

Therefore, from the viewpoint of the behaviors in the reprocessing plant, the nuclide in the TRU wastes are categorized into 7 groups, (1) plutonium isotopes, (2) uranium isotopes, (3) H-3, (4) C-14 and I-129, (5) Am-241, (6) Np-237, (7) other fission products, TRU nuclides, and activation products. The property of key nuclides and scaled nuclides are shown in Table I.

Table I. The Property of Key Nuclides and Scaled Nuclides

Scaled nuclide	Key nuclide	Generation mechanism	Main pass way in TRP
H-3	none	Ternary fission	Acid recovery or release to ocean
C-14	none	N-14 (n,p) reaction	→ LLW treatment
I-129	none	Fission	→ LLW treatment
Ni-59/63 Nb-94	Co-60	Activation	→ HLW treatment
Sr-90, Tc-99, Sn-126, Zr-93,	Cs-137	Fission	→ HLW treatment
Am-242m/243, Cm-242/244	Cs-137	Neutron capture	→ HLW treatment
Am-241	Pu	Neutron capture	→ Pu purification
Np-237	Cs-137 Fissile	Neutron capture	→ HLW treatment or Pu purification
U-isotopes	Fissile	Neutron capture	→ U purification
Pu-isotopes	Fissile Pu	Neutron capture	→ Pu purification

Fissile : U-235+Pu-239+Pu-241
Pu : Pu-238+Pu-240+Pu-242

Calculation of Scaling Factor

The scaling factors were calculated considering the spent fuel inventory and the radioactivity balance in the process of TRP. The spent fuel inventory was calculated with ORIGEN2.1 computer code. The radioactivity balance in the process of TRP is based on the recent review of the design data of TRP.

As the ratio of a scaled nuclide to a key nuclide is changed by the kind of the step, the scaling factor appears to be variable in TRP. The calculated maximum and minimum value of the scaling factors are shown in Table II.

Table II. Calculated Scaling Factors

Scaling factors	Maximum	Minimum
Sr-90/Cs-137 (Bq/Bq)	1.83E+01	6.84E-02
Tc-99/Cs-137 (Bq/Bq)	5.96E+00	3.37E-05
Sn-126/Cs-137 (Bq/Bq)	2.76E-03	7.18E-06
Zr-93/Cs-137 (Bq/Bq)	3.48E-04	9.63E-06
Am-243/Cs-137 (Bq/Bq)	7.57E-05	1.55E-05
Cm-244/Cs-137 (Bq/Bq)	4.73E-03	9.71E-04
Np-237/Cs-137 (Bq/Bq)	2.60E+01	3.46E-06
U-238/fissile (Bq/g)	7.90E+05	1.06E+03
Pu-238/fissile (Bq/g)	9.24E+09	3.44E+02
Np-237/fissile (Bq/g)	1.13E+09	6.45E+04
Np-237/Pu (Bq/g)	5.30E+12	1.88E+06

fissile : U-235+Pu-239+Pu-241

Pu : Pu-238+Pu-240+Pu-242

SELECTION OF SAMPLING POINTS

To optimize the sampling for the wastes, it is important to select the sampling point carefully. The sampling points are selected from the viewpoint of the nuclide composition of the wastes and the amount of waste generation.

Nuclide Composition of Waste

The nuclide composition of the TRU wastes generated from TRP depends on radioactive inventory of the spent fuel reprocessed in TRP and the nuclide composition of process fluids of each step from which the waste were generated.

The spent fuels reprocessed in TRP were irradiated in light water reactors and the advanced thermal reactor "FUGEN". Burn up of almost these fuels is about from 6,000 to 35,000MWd/t, and cooling time is up to about 20years. In this condition, the change of the nuclide composition of spent fuel is less than 1 order of magnitude.

On the other hand, the decontamination factor of uranium from fission products is 10^7 in the purex process.

In other wards, the nuclide composition of TRU wastes is determined by the place of the generation of the wastes.

There are about 30 steps in TRP from the viewpoint of the radioactivity balance in the processes and the equipment arrangement.

The steps in TRP were categorized into 9 groups in consideration of the change of nuclide composition of process fluids, and the amount of the waste generated.

To estimate the scaling factor, it is important to collect the waste samples from both the steps which have higher and lower value of calculated scaling factor, respectively.

In case Cs-137 is key nuclide, the calculated scaling factors, Tc-99/Cs-137 and Np-237/Cs-137 change widely, because technetium and neptunium are less decontaminated from plutonium and uranium than cesium in the purex process. Other calculated scaling factors do not change so widely, because other nuclides are decontaminated as well as cesium.

In case even-mass plutonium isotopes (Pu-238, Pu-240, and Pu-242), or fissile nuclides (U-235, Pu-239, and Pu-241), are key nuclides, the calculated scaling factors of Np-237 to key nuclides change widely.

The sampling points were selected considering the change of the calculated scaling factors about Tc-99 and Np-237.

In addition, the sampling points were also selected considering the main pass way of the nuclides, H-3, C-14, and I-129 which have unique behavior in TRP.

Amount of Waste Generation

To investigate radioactive inventory in the TRU solid wastes, it is necessary to collect waste samples from the steps where the large amount of solid wastes are generated in TRP.

In TRP, the facility and the room number where the solid wastes were generated are recorded. As a result of research on the records of the solid wastes, the largest amount of waste is generated from Main Plant (MP) which has principal steps of TRP. The next largest amount of wastes is generated from Auxiliary Active Facilities (AAF) which treats LLW.

In MP, the receiving and storage of fuel elements step and the maintenance area of the sharing machine generate large amount of solid wastes by operating of TRP. The fuel dissolution step, the clarification step, the acid recovery step, and off-gas treatment step generate large amount of solid wastes by repairing and replacing of the equipments of TRP

The selected sampling points are listed in Table III.

Table III. Selected Sampling Points

Steps in TRP	Nuclide composition	Amount of wastes	Selected sampling point
Receiving and storage of fuel	○	○	○
Sharing machine maintenance		○	○
Fuel dissolution and clarification	○	○	○
Off-gas treatment	○	○	○
Codecontamination			
Uranium and plutonium partition			
Uranium purification			
Plutonium purification			
Acid recovery	○	○	○
Intermediate storage of LLW			
Rework			
Uranium denitration	○		○
Plutonium concentration	○		○
HLW treatment and vitrification	○		○
Treatment of solvent waste			
Mixed conversion of plutonium and uranium	○		○
LLW treatment	○	○	○

Selection of Nuclides for Analysis

The nuclides for analysis were selected considering the importance for the safety assessment of disposal. Those nuclides are as follows:

H-3, C-14, Co-60, Ni-59/63, Sr-90, Zr-93, Nb-94, Tc-99, Sn-126, I-129, Cs-137, U-234/235/236/238, Np-237, Pu-238/239/240/241/242, Am-241/242m/243, Cm-242/244

RESULTS

In 2003, 6 samples were collected and analyzed. The samples were the following:

No.1□A piece of paper wiper used for the decontamination of a spent fuel-shipping cask. It was contaminated with crud.

No.2□A piece of paper wiper contacted with the pulse filter casing. It was contaminated with purified dissolver solution.

No.3□A piece of paper wiper used for maintenance of a manipulator in the vitrification facility. It was contaminated with HLW.

No.4□A parts of a valve on a HLW storage vessel. It was contaminated with HLW.

No.5□A piece of paper wiper used for maintenance of a LLW evaporator. It was contaminated with LLW.

No.6□A piece of paper wiper contacted with sampling bench of the receiving step in solvent waste treatment facility. It was contaminated with the solvent waste.

The observed correlations between key nuclides and scaled nuclides are as follows.

As shown in Figure 1, tritium concentration in the solid wastes is independent of Cs-137, and appeared to be in the constant range.

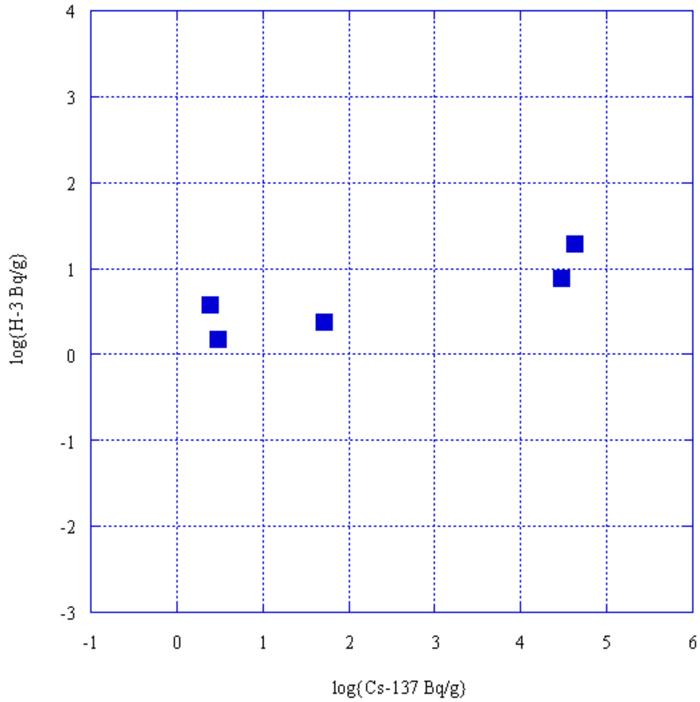

Fig. 1. The Concentration Ratio of H-3 to Cs-137

As shown in Figure 2, the concentration of Sr-90 and Tc-99 in the solid wastes is suggested to be correlated with Cs-137.

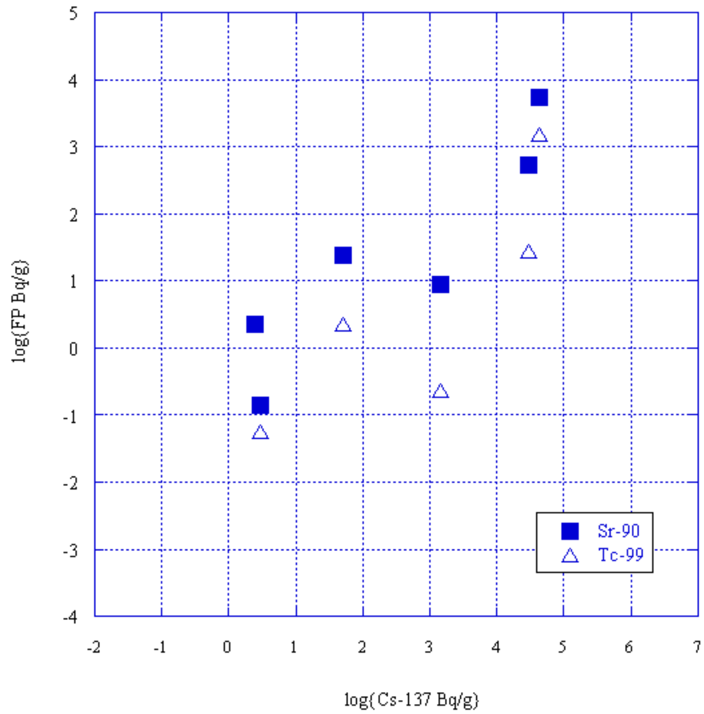

Fig. 2. The Correlation between Fission Products and Cs-137

As shown in Figure 3, the correlation between Am-243 and Cs-137 is not cleared.

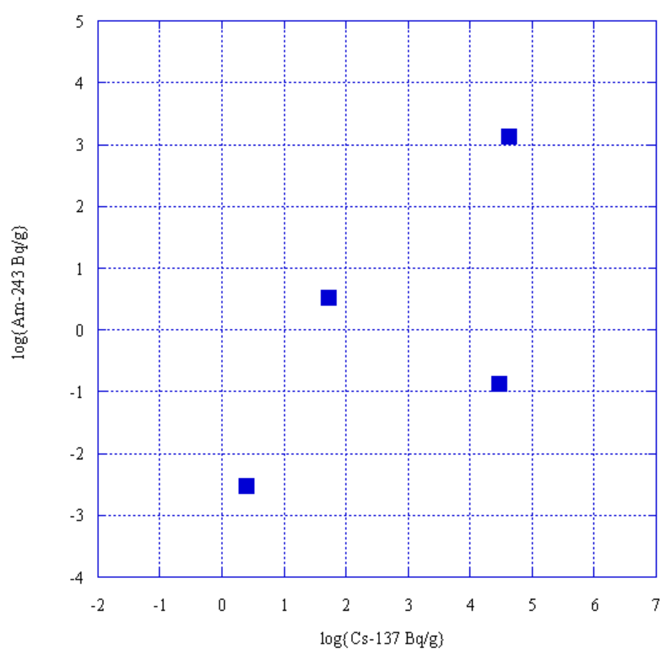

Fig. 3. The Concentration Ratio of Am-243 to Cs-137

As shown in Figure 4, the correlation between Am-241 and other TRU nuclides is suggested.

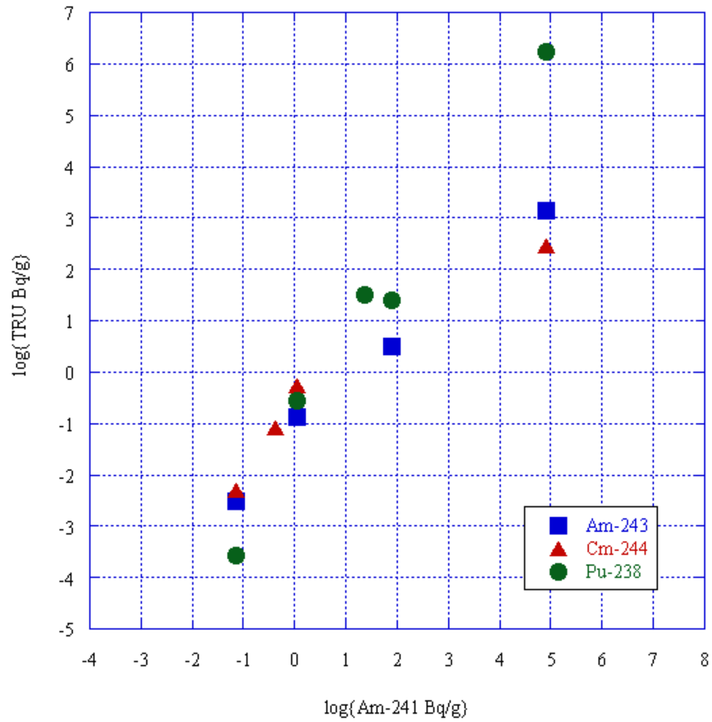

Fig. 4. The correlation between TRU and Am-241

CONCLUSION AND FUTURE PLAN

The preliminary conclusions are follows.

The scaling factors of the TRU solid wastes are calculated by considering of generation mechanism and behavior in TRP of each nuclide.

The following results were obtained from analyzing 6 waste samples:

1. Tritium concentration in the solid wastes is constant.
2. The correlation between Sr-90, Tc-99 and Cs-137 is suggested.
3. The correlation between Am-243 and Cs-137 is not cleared.
4. The correlation between Am-241 and other TRU nuclides is suggested.

In order to develop the method for the verification of the nuclide inventory in the TRU solid wastes, it is necessary to analysis more waste samples.

In JNC, as the first step of development of the scaling factor method, radioactive inventory in the

TRU solid wastes generated from TRP will be investigated by analyzing about 50 waste samples from 2003 to 2005.

REFERENCES

1. Japan Nuclear Cycle Development Institute (JNC). 1999. Review of Design Data for Safety Assessment of Tokai Reprocessing Plant. JNC TN8410 99-002. (In Japanese).