

PARTITIONING-TRANSMUTATION TECHNOLOGY: A POTENTIAL FUTURE NUCLEAR WASTE MANAGEMENT OPTION

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Abstract

Partitioning-transmutation technology (PT) will produce radioactive wastes of different physical and chemical properties and in different amounts from those generated in the current nuclear fuel cycle. To assess quantitatively the effects of PT on waste disposal, we first analyzed the amounts of the PT wastes, assumed conditioning for each type of the waste, and then made an attempt to estimate the repository area for disposal of the PT wastes. The properties of the “hot” Sr-Cs waste form are controlling factors in determining the size of the geologic repository. The disposal area could be reduced if the “Sr-Cs” fraction is disposed in a different subsurface repository or by long-term storage of the waste under institutional control. Disposal in a subsurface repository was found to comply with the Japanese law in terms of radioactivity constraint, through a performance assessment for disposal of the “Sr-Cs” fraction.

Introduction

Partitioning of high-level radioactive liquid waste (HLLW) produces different, chemically separated fractions--a highly radioactive Sr-Cs fraction, a recyclable fraction and a waste fraction of other metals & rare earths. If long-lived elements are transmuted into short-lived wastes, the lifetime of high-level radioactive waste (HLW) may be reduced. On the other hand, the processes used during the application of partitioning-transmutation technology (PT) generate other radioactive processing wastes and secondary wastes. The chemical compositions and amount of these wastes affect the treatment, total volume, and cost for waste storage and final waste disposal.

The PT, therefore, should not be intended for application only to HLW management, but must be a tool that provides integrated waste management options for the future nuclear energy system. We can choose suitable treatment and disposal options for the resulting wastes according to their chemical and physical properties.

To assess how wastes should be managed after introduction of PT, the effects or contribution need to be quantified based on wastes generated in a partitioning-transmutation system. We first identify the chemical and physical properties and quantify the amounts of radioactively contaminated wastes that may be generated in the partitioning-transmutation cycle that Japan Atomic Energy Research Institute (JAERI) has proposed [1]. The volume of generated wastes is one index for quantifying the effects of PT on waste management.

The design of a waste storage/disposal facility depends on waste package properties such as the quantity, volume, heat generation and radioactivity. The deep underground repository, originally designed for high-level radioactive waste, does not have to accept all types of waste packages. If some types of the waste packages can be disposed of in a different repository than the deep underground one, then capacity for high-level radioactive wastes in the deep underground repository would be substantially increased. The saving in capacity is another index for quantifying the effects of PT on waste management.

Radioactive Wastes generated by PT cycle

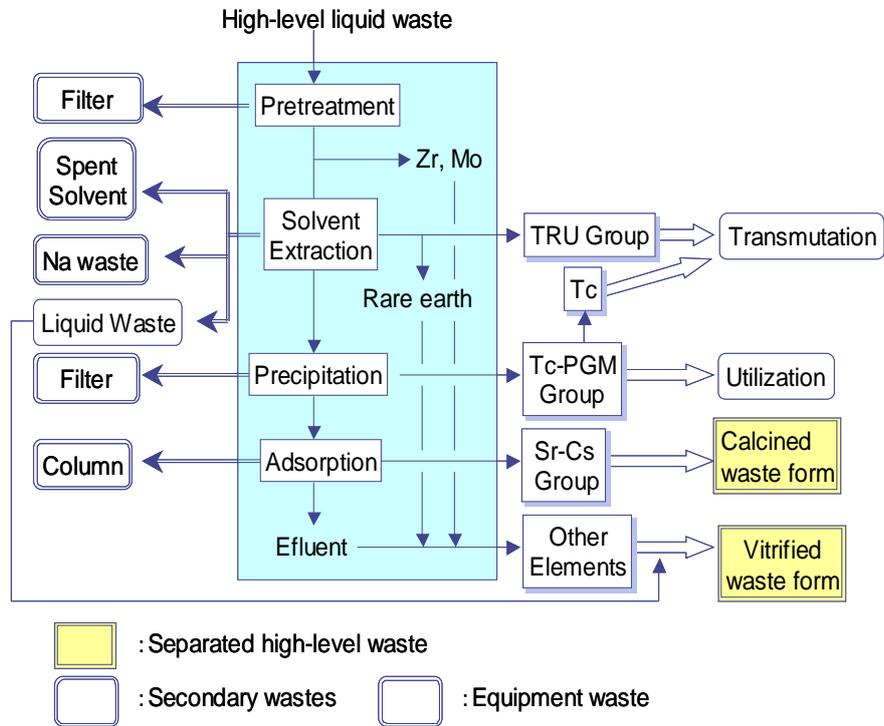
Partitioning

Japan Atomic Energy Research Institute has developed four-group partitioning. Radionuclides contained in HLLW are separated into four different groups: transuranic elements (TRU); technetium (Tc) and platinum group metals (PGM); Sr-Cs; and the rest, "other elements", which includes zirconium, molybdenum, iron and rare earth elements.

The source and waste form of wastes generated by the partitioning process are shown in Figure 1. Of these four groups, TRU elements from the "TRU Group" and Tc from "Tc & PGM¹ Group" become fuel for transmutation. The other two, "Sr-Cs" and "other elements", are wastes of interest as partitioned high level wastes. The highly radioactive Sr and Cs are adsorbed on zeolite or titanate acid, and stabilized as a calcified waste form. The "other elements" group radionuclides are vitrified into glass waste form.

¹ Platinum group metal

Figure 1. Wastes from JAERI's four-group partitioning process: Source and waste form [2]



Numerical data on the four-group partitioning are shown in Table 1[2]. This table shows, for example, that if 1 ton of 45 GWd/t LWR spent fuel is partitioned, 1.69 kg of minor actinide oxide, with a volume of 0.169 litres, is produced. The radioactivities and the heat density can be calculated.

Table 1. Products and wastes from 1 ton of 45 GWD/t LWR spent fuel by the four-group partitioning (updated from [3])

	Np, Pu, Am, Cm	U	Tc	PGM	Waste		Non- partitioned vitrified waste
					Sr-Cs	Other elements	
Chemical form or waste form	oxide	oxide	Metal	metal + oxide	calcified	vitrified (30 wt%)	vitrified (12 wt%)
Weight (kg/MTU)	1.69	5.34	1.33	4.97	71.9	103	453
Density (kg/L)	10	10	11.5	8	4.2	2.67	2.67
Volume (L/MTU)	0.169	0.513	1.16	0.621	22.1	38.7	170
Radioactivity (Bq/ton)							
α	1.3×10^{17}	8.5×10^{10}	9.1×10^7	2.3×10^{11}	1.4×10^{11}	5.5×10^{11}	4.9×10^{14}
βγ	5.4×10^{16}	2.3×10^{13}	4.8×10^{16}	2.1×10^{17}	2.4×10^{17}	4.0×10^{16}	5.0×10^{16}
Total	1.9×10^{17}	2.3×10^{13}	4.8×10^{16}	2.1×10^{17}	2.4×10^{17}	4.0×10^{16}	5.1×10^{16}
Heat density (W/L)	1,240	0.024	50.4	209	76.3	7.53	13.7

Wastes are listed in the three right-hand columns; 71.9 kg or 22.1 litres of calcined Sr-Cs fraction are generated. This fraction carries 76% of the radioactivity in HLW. The “other elements” fraction is assumed to be vitrified into 38.7 litres of waste form with the waste loading of 30 wt%. This high loading, 30 wt%, is possible in vitrified waste mainly because of removal of the “hot” constituents from the HLW, the Sr and Cs. The total volume of the high-level waste form after partitioning is $22.1 + 38.7 =$ about 60 litres. This volume is about a third that in the current Japanese-type glass waste form of HLLW, with a 12 wt% loading (170 litres).

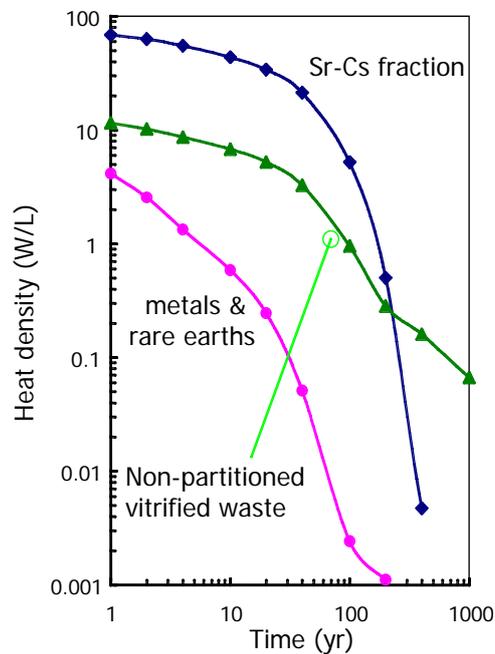
The Sr-Cs fraction is very hot; because the waste form volume is small, the heat density is high. The heat density is about 6 times higher than that of the non-partitioned vitrified waste form, and decreases rapidly with radioactive decay of ^{90}Sr and ^{137}Cs , as shown in Figure 2. Of significance is that the heat density of the low-level “other elements” fraction, referred to as “metals & rare earths” in Figure 2. The heat density of this fraction decreases to 4.2 W/L after the first year, which is almost the same as the heat density of non-partitioned vitrified waste after 30 years, 4.1 W/L. The small heat density of the low-level “other elements” fraction could affect the time period and capacity of interim storage before disposal, as discussed later.

The four-group partitioning process also generates secondary wastes. Their amounts and properties are summarised in Table 2. The secondary wastes are typically two types. One is used or spent solvent, DIDPA-TBP-dodecane, and the other type is sodium salt solutions, sodium carbonate and sodium nitrate. These wastes are stabilized into calcium phosphate ($\text{Ca}_2\text{P}_2\text{O}_7$) and sodium nitrate (NaNO_3) pellets, respectively, as their waste forms. These waste forms are the same as those planned for the Rokkasho reprocessing plant, and therefore it is assumed these will be solidified into a cemented waste form.

Table 2. Secondary wastes generated by four-group partitioning after reprocessing of 1 ton of 45GWD/t LWR spent fuel at Rokkasho (updated from [2])

	Used Solvent (DIDPA-TBP- dodecane)	Washing liquid (sodium salt solutions)
Solid waste weight (kg/MTU)	7.90 (Ca ₂ P ₂ O ₇)	288 (NaNO ₃)
Solidification	cement	cement
Cemented waste form		
Weight (kg/MTU)	79.0	577
Density (kg/L)	1.7	2.0
Volume (L/MTU)	46.5	288
Radioactivity (Bq/t)		
A	6.89×10^6	7.73×10^6
By	5.32×10^{11}	2.06×10^{13}
Total	5.32×10^{11}	2.06×10^{13}
Heat density (W/L)	8.68×10^{-5}	4.19×10^{-3}

Figure 2. Heat density of partitioned radioactive wastes and non-partitioned vitrified waste



As listed in Table 2, the amounts of these secondary wastes are not great. For instance, the volume of the used solvent waste, about 200 drums per year, is about 40% of that generated from the Rokkasho reprocessing plant. This means that partitioning would increase the total volume of used (spent) solvent waste to about 1.4 times by combination of the Rokkasho plant and the partitioning plant. The radioactivity of the secondary wastes is fairly low. Based on radioactivity, these wastes can be disposed of at the now operational Rokkasho low-level waste site.

Transmutation

The long-lived waste fraction produced by the partitioning process contains minor actinides and Tc. These elements are transmuted into short-lived nuclides in the transmutation cycle. The transmutation cycle JAERI proposes consists of nitride fuel fabrication, accelerator-driven transmutation of the nitride fuel by an accelerator-driven, sub-critical system (ADS), and pyrochemical fuel reprocessing of the nitride fuel[1]. Radioactive waste arisings from the transmutation cycle are shown in Figure 3, and their waste form and quantities are listed in Table 3. Process wastes typical of the transmutation cycle are spent fuel pins, used KCl-LiCl salt and “rare metal FP”. The “rare metal FP” is the insoluble fraction in the salt phase and separated as a metal phase during the electro-refining process. The “FP in salt” in Table 3 is a soluble fraction in the salt phase and contained in the “Used salt”. It is assumed for this FP fraction that FPs are separated from the used salt with no further secondary wastes, and that the separated used salt is recycled in the fuel reprocessing.

Figure 3. Radioactive wastes generating from JAERI’s transmutation cycle. Yellow hatched wastes are high-level waste. An: actinides, FP: fission products, SP: spallation products, AP: activation products [2]

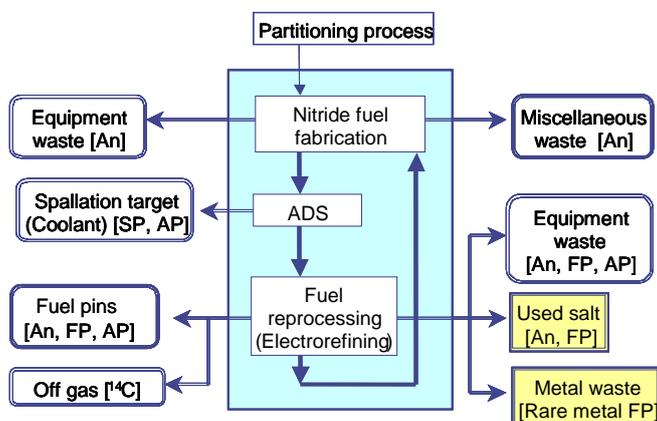


Table 3. Wastes generated from JAERI's transmutation cycle for 1 ton of 45 GWD/t LWR spent fuel reprocessing at Rokkasho

Waste	Rare Metal FP	FP in Salt	Fuel Pins
Raw waste (kg/MTU)	0.47	0.58	11
Solidification	zirconium alloyed/cementation	sodalitization/cementation	cementation
Volume of waste form (L/MTU) (waste wt%)	1.75 (20 wt%)	2.15 (10 wt%) 4.30 (5 wt%)	6.9 (50 wt%)

The "rare metal FP" is zirconium alloyed and cemented. The "FP in salt" is solidified into sodalite followed by cementation. Fuel pins are solidified in cement. High-level radioactive wastes are separated FP: zirconium-alloyed "rare metal FP" and cemented "FP (sodalite) in salt".

Because the design of the fuel fabrication processes has not progressed as far as the partitioning process, chemical engineering data are not yet available to estimate the quantity and characteristics of the wastes. Table 3 lists the amounts of radioactive wastes per ton of 45GWD/t LWR spent fuel reprocessing at Rokkasho estimated on a tentative, semi-quantitative basis. As a result of reprocessing of 800 tons of spent nuclear fuel at Rokkasho, 1.4 m³ of zirconium-alloyed rare metal FP waste, 3.4 m³ of cemented FP sodalite (5 wt% waste loading), and 5.55 m³ of cemented fuel pins would be generated a year. These amounts are small compared with the wastes from partitioning. The wastes generated from the partitioning process are the dominant wastes in the PT cycle.

Wastes from JAERI's PT cycle - Summary

Figure 4 summarizes the kind of wastes and the quantities that would be generated from JAERI's PT cycle. The high-level waste would be reduced in volume to about a third of the currently planned glass waste form². Although not explained previously, a different waste arising is from the maintenance/replacement and decommissioning of the plants. The types and amounts of these wastes have not yet been estimated because of the lack of data on ADS plant designs. However, radioactive contamination of some of the components of accelerator internals can be estimated on at least a semi-quantitative basis; about 7,000 tons of Pb-Bi coolants of the ADS are a source of spallation products. The coolant is contaminated with ²¹⁰Pb (22 yr) and long-lived ²⁰²Pb/²⁰²Tl and ^{210m}Pb/²⁰⁶Tl.

² Waste containing transuranic nuclides, "TRU waste", is not shown in Figure 4 and not included in wastes for geologic disposal.

Figure 4. Wastes generated from JAERI's partitioning-transmutation cycle for 800 tons of 45 GWD/t LWR spent fuel reprocessing at Rokkasho

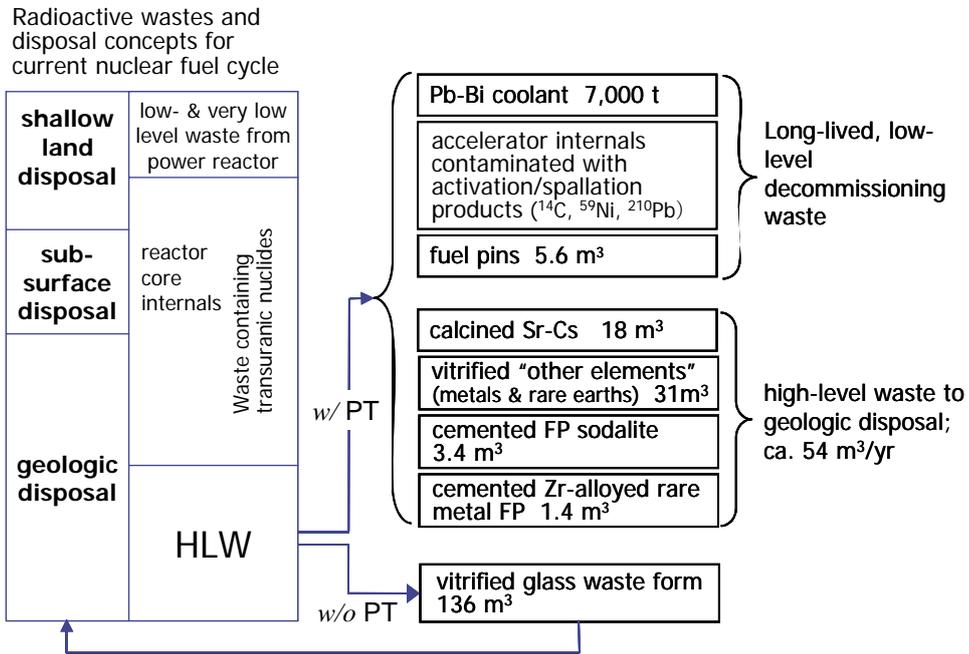
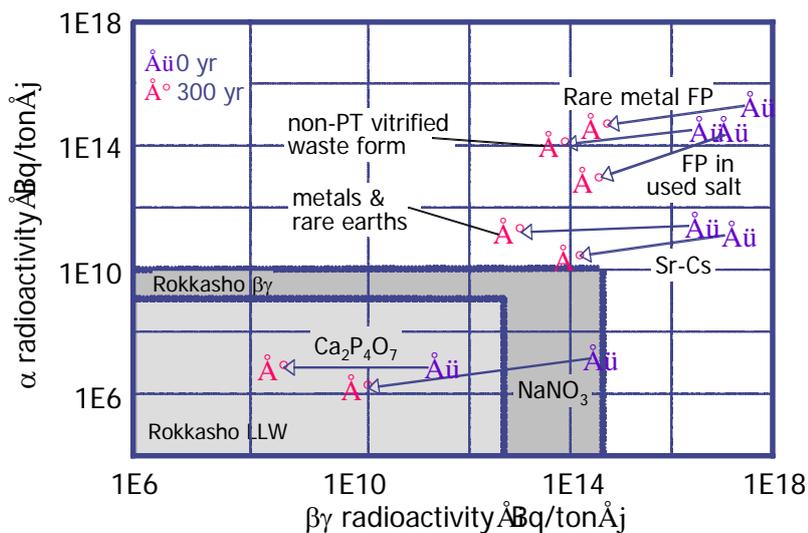


Figure 5 is a diagram of alpha and beta-gamma radioactivities of wastes from JAERI's PT cycle, and comparison with the Japanese criteria for the current Rokkasho waste disposal. The "non-PT vitrified waste form" is the current glass waste form. The beta-gamma radioactivity would decrease by about three orders of magnitude after the first 300 years. The alpha radioactivity would not decrease much.

Figure 5. Alpha and beta-gamma radioactivities of wastes from the JAERI's PT cycle



The beta-gamma radioactivity of the glass waste form is divided into the two fractions: one is “Sr-Cs” fraction, and the other is “other elements” fraction, referred to as “metals & rare earths” in Figure 5. The gap in the alpha radioactivity between the “non-PT glass waste form” and these two fractions is two to three orders of magnitude, and this part went to the nitride fuel of minor actinides. The beta-gamma radioactivity of both of these fractions (“Sr-Cs” and “metal & rare earth”) would decrease by the same quantity, about two to three orders of magnitude, after the first 300 years. The beta-gamma radioactivity of the “rare metal FP” and “FP in used salt” would be even higher than that of the current glass waste form.

Backend for PT wastes

Interim storage

Currently, vitrified waste forms are placed for under 50-year interim storage at the Rokkasho site and are awaiting disposal in a deep underground repository. For the storage of the waste generated from the 50-year operation of the Rokkasho plant, the interim storage facility needs a capacity of about (170L/MTU x 800 MTU/yr x 50 yr =) 68 hundred cubic meters or 45 thousand 150-liter glass waste packages. After partitioning, if the calcined “Sr-Cs” waste is under storage for 50 years like the vitrified glass waste form, the volume of the “Sr-Cs” waste is about (22.1 L/MTU x 800 MTU/yr x 50 yr =) 880 cubic meters. Vitrified “metals & rare earths” are short-lived and do not need to be stored long. If the waste needs only 5 years, the volume for storage of this type of waste is about (38.7 L/MTU x 800 MTU/yr x 5 yr =) 150 cubic meters. Partitioning can reduce the amount of high-level waste to about 880 + 150 =) 1,000 cubic meters, which is about a seventh of the current glass waste form.

The “Sr-Cs” fraction is hot waste, and the heat density is much higher than that of non-partitioned vitrified waste as shown in Figure 2. Although the volume of the “Sr-Cs” waste is smaller than that of non-partitioned vitrified waste, the “Sr-Cs” waste has larger space requirements per unit volume in terms of ventilation efficiency of the storage facility. A thermal analysis is needed to determine the necessary space requirements. The heat density of the “metals & rare earths” fraction decreases quickly, and the space requirements per unit volume of this type of waste are much smaller than that of the “Sr-Cs” fraction.

Disposal

As shown in Figure 4, the volume of arising high-level radioactive wastes from the PT cycle is about 40 % of that from the current Japanese nuclear fuel cycle; about 136 m³/yr of vitrified glass waste is reduced to about 54 m³/yr. Based on the information shown above, we made an attempt to show a first-order quantitative approximation for the HLW repository area, in comparison with the current Japanese design.

According to the current Japanese rule, radioactive waste containing more than 1GBq/ton of alpha emitters must be disposed of in a deep underground repository [4]. The calcined Sr-Cs waste form, the vitrified metal & rare earth waste form (from partitioning), cemented rare metal FP, and cemented FP sodalite (from transmutation) would be placed in this deep repository.

The necessary disposal areas for each of the waste forms would depend on the heat generation and dimensions of the waste packages. We performed thermal analysis to calculate the pitch (allowance) between two waste forms, and then estimate the necessary areas for placement of waste packages [5,6]. For comparison with the currently designed Japanese HLW repository for 40,000 glass waste forms by Japan Nuclear Cycle Development Institute[7], the design of engineered barriers for the PT wastes are based on the JNC designs in most cases; which means that, for example, all of the waste forms are encompassed by 75-cm thick bentonite-based buffer materials. Thermal properties of the host rock (granite), backfill materials, buffer materials and overpack, and mathematical configuration for the analysis followed those of the JNC report [7]. The pitch is determined by the temperature constraint; the upper limit temperature of buffer materials, being 100°C.

Because the pitch between the waste packages is sensitive to heat flux from the waste form, the dimension of the cylindrical “Sr-Cs” waste form affects the pitch and the necessary area, as shown in Table 4. If the engineered barrier configuration of the current Japanese repository design is employed, a large part of the repository area of this disposal option would be dominated by the Sr-Cs waste packages. The effect of decrease in heat density during the prolonged interim storage (100 years) is large. Strontium-90 and ¹³⁷Cs, both with half-lives of about 30 years, decay to about 1/1000 during the 100-hundred year storage, and occupy a much smaller area than that for 50-year interim storage. The results, shown in Table 4, indicate the possibility to optimize repository designs depending on waste package properties, such as dimension and heat density (radioactivity).

Table 4. Comparison of disposal area requirements for deep geologic disposal of PT waste and non-PT vitrified waste (km²)

PT	Calcined Sr-Cs (D: diameter, height: 45cm)	50-year Interim Storage			100-year Interim Storage			
		D = 10	D = 20	D = 30	D = 20	D = 30	D = 30	
		(70 cm thick buffer materials)					(no buffer)	
		2.75	0.81	1.01	0.73	0.34	0.24	
	Zirconium alloyed rare metal	0.008						
	Cemented used salt + vitrified metals & rare earths	0.13						
	Total	2.88	0.94	1.14	0.87	0.48	0.38	
Vitrified HLW (40,000)		1.46						

Most of the radioactivity of the “Sr-Cs” fraction is ⁹⁰Sr and ¹³⁷Cs, and they are short-lived. This fraction, however, contains a small amount of long-lived ¹³⁵Cs (2.3x10⁶ yr). Because of the presence of this long-lived nuclide, the “Sr-Cs” fraction must be disposed of in a deep underground repository. If this fraction does not need to go into a deep repository, the necessary area could be significantly reduced.

Japan's waste disposal concept consists of shallow land disposal, subsurface (or *intermediate-level*) disposal, and geologic disposal (Figure 4). The subsurface repository, which will be constructed between 50 to 100 meters below surface, is for disposal of waste with comparatively high radioactivity such as reactor core internals. The compliance requirement by Nuclear Safety Committee (NSC) of Japan for the subsurface repository is a dose to a maximally exposed individual of 10 μ Sv/yr. The acceptable nuclides and their concentrations in radioactivity are also limited. Radionuclides contained in the "Sr-Cs" fraction do not meet this requirement³.

However, to assess the possibility of disposal of the "Sr-Cs" fraction into the subsurface repository on the radioactivity constraint basis, we conducted a system performance assessment for the "Sr-Cs" fraction using the same scenarios, mathematical models and parameters as those employed in determining the NSC criteria [8]. Long-lived actinides and ¹³⁵Cs were of interest for assessment because short-lived ⁹⁰Sr and ¹³⁷Cs will decay out during the institutional control period of a few centuries.

Concentrations of nuclides, which give a maximally exposed individual dose of 10 μ Sv/yr, were calculated to be in the same order (¹³⁵Cs) or much higher (actinides) than those contained in the partitioned "Sr-Cs" fraction. The "Sr-Cs" fraction, therefore, can be disposed of in the subsurface repository on the radioactivity constraint basis. The mathematical models and parameters used in Ref. [8] are conservative, and introducing more reasonable data such as the lower leach rate for calcined "Sr-Cs" waste form may ensure the acceptance of this fraction in a subsurface repository. If this fraction is not disposed of in a deep geologic repository, the necessary area will be greatly reduced. Reduction in area leads to an extended service life of a repository. Such a "long-lived" repository will be needed for an anticipated, prolonged nuclear age.

Table 5. Concentrations of some selected long-lived nuclides contained in the "Sr-Cs" fraction for disposal into the subsurface repository

Nuclide	Concentration corresponding to the dose limit of 10 μ Sv/yr (Bq/t)		Concentration in the "Sr-Cs" fraction
	This study ¹⁾	NSC criterion[8]	
²³⁶ U (4N series)	5.4 x 10 ¹⁰	(not considered)	3.8 x 10 ⁴
²³⁷ Np (4N+1 series)	7.3 x 10 ⁹	1.3 x 10 ^{10 2)}	1.0 x 10 ⁸
²³⁸ U (4N+2 series)	9.1 x 10 ⁹	(not considered)	3.6 x 10 ⁴
²³⁵ U (4N+3 series)	2.1 x 10 ⁹	(not considered)	2.1 x 10 ³
¹³⁵ Cs	2.4 x 10 ¹¹	(not considered)	3.1 x 10 ¹¹
⁹⁹ Tc	---	8.2 x 10 ¹⁰	1.8 x 10 ⁷

1) Daughter nuclides are taken into account. 2) Daughter nuclides were assumed to be in equilibrium with the parent nuclides anytime

³ For the similar reason, the secondary wastes from the partitioning process appear to be acceptable into the Rokkasho site for disposal on the radioactivity constraint basis (Figure 5), but specific radionuclides contained in the secondary wastes are not accepted.

A different way to avoid the “Sr-Cs” fraction disposal in a geologic repository is long-term interim storage under institutional control in a surface facility. Century-long storage will result in decreased radioactivity of the fraction and the remaining long-lived waste containing ^{135}Cs and actinides can be disposed of in a deep repository.

Figure 4 also shows that application of PT produces radioactive wastes of different physical and chemical properties from those generated in the current nuclear fuel cycle. The wastes include structural materials of ADS contaminated with activation and spallation products such as Pb-Bi coolant, accelerator internals and fuel pins, and salt used in the nitride fuel reprocessing plant. They are generally low in radioactivity, but long-lived and chemically toxic (Pb-Bi). Disposal for these wastes also have to be taken into account from the application of P&T in the nuclear fuel cycle.

Partitioning-transmutation technology is a potential method to provide waste management options for the future nuclear energy system. The options should integrate all of the nuclear-related wastes. Based on our analysis, in the context of comparison with the currently planned management of HLW, dealing with the Sr-Cs fraction would be one factor in considering the effects of partitioning-transmutation technology on waste management strategy.

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