

# Chapter 2

## Generation and Characteristics of Radioactive Wastes

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### 2.1 Wastes from Uranium Mining and Melting Facilities

Mining refers to the act of collecting ore containing a target metal from mines. This process generates gangue—rock that is commercially valueless and therefore subject to disposal. The process of extracting metal from its ore is called melting. Uraninite, pitchblend and brannerite are uranium containing ores, and they contain uranium in the form of oxide. Their grade generally ranges from 0.1 to 0.3 % triuranium octoxide ( $U_3O_8$ ) equivalent. Although extracted ore of ordinary metals undergoes “dressing,” the process of separating valueless rocks based on physical or chemical characteristics by such methods as fire refining and aqueous refining, the dressing process is not effective for low-grade uranium ore. This kind of ore is crushed into pieces and dissolved in acid or alkali solution. Then uranium is refined and concentrated, followed by precipitation using strong alkali. An intermediate from this milling process is uranium concentrate, which is  $U_3O_8$  powder, called yellow cake for its color. Its uranium content ( $U_3O_8$  content) is about 70–80 %. This material is further refined (or purified) to increase purity and is converted to forms such as  $UF_6$ ,  $UO_2$  or metallic uranium, suitable for use as reactor fuel in the next process at fuel fabrication facilities. In this connection, refining means increasing the purity of metal resulting from melting by electrolysis or other processes; in the uranium melting process, purification corresponds to refining.

Wastes generated from mining and melting processes mainly includes, in addition to gangue, refining effluent, tailings of ore after uranium extraction (also called

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The nuclear regulatory system in Japan has been changed significantly after the Fukushima Daiichi Nuclear Power Station accident in March 2011. Descriptions in this chapter have been translated from the book originally published in Japanese before the accident, with minimal update where appropriate.

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mill tailings), and wastes from facility maintenance. These wastes are managed under the Mine Safety Law and not classified as radioactive wastes in Japan. Melting effluent is neutralized and diluted before being released.

When a light water reactor with a rated electrical output of 1,000 MW is operating at an 80 % loading factor, 40,000 MW/d burn-up and 33 % thermal conversion rate, it consumes about 27 tons of low-enriched uranium fuel (4 %) annually. How much waste is generated from the mining and melting processes when this standard reactor is operated for 1 year can be estimated like this. For the sake of simplicity, it is assumed that uranium recovered by reprocessing is not recycled. Provided that the  $^{235}\text{U}$  content in the depleted uranium generated by enrichment is 0.3 %, the amount of natural uranium needed for a 1 year period calculated from the mass balance in the enrichment process is approximately 244 tons (see Exercise 1 at the end of this chapter). Assuming that the uranium recovery rate during mining and melting is 93 % and the ore grade is 0.2 %, 130,000 tons of ore would have to be extracted and smelted.

Also included in the waste from mining and melting, besides residual uranium, is almost the entire amount of progeny nuclides derived from isotopes of uranium that decayed in the ore. These progeny nuclides are considered to be in the state of secular equilibrium with uranium isotopes while confined in ore. Once secular equilibrium is broken by melting, radiation thereafter attenuates in accordance with the half-life (about 80,000 years) of  $^{230}\text{Th}$ , a progeny nuclide of  $^{238}\text{U}$ . Among the progeny nuclides, volatile  $^{222}\text{Rn}$  has a short half-life of 3.7 days, but it continues to be produced because of the decay of the parent nuclide. Special attention should be paid to  $^{222}\text{Rn}$  in radiation control because it has high mobility in the environment. Waste generation from mining and melting is not a major issue in Japan, which has no large uranium mines. However, in Canada, one of the major uranium producing countries, tailings in storage have reached about 225 million tons, and their confinement and stabilization, as well as closure work at abandoned mines, are recognized as serious problems.

In veins in Kazakhstan that have attracted attention recently, where deposits are relatively low-grade and therefore mining and melting is not financially viable, “in-situ leaching” is used, which injects acidic solutions directly into the underground vein to dissolve uranium and draw up uranium solution out of downstream wells. Although this method does not generate waste as described above, the injected acid cannot be collected entirely. The operator must expect a certain level of groundwater contamination.

The collection of seawater uranium, a prospective method, is expected to allow the avoidance of uncollectable waste generation and environmental pollution problems.

## 2.2 Wastes from Fuel Fabrication Facilities

Fuel fabrication facilities are where nuclear fuel materials, such as uranium and plutonium, are physically or chemically treated to fabricate them into a form or a composition ready for use as reactor fuel. These are divided into two types: uranium processing facilities and mixed oxide (MOX) fuel fabrication facilities.

**Table 2.1** Characteristics of wastes from fuel fabrication facilities [1]

Source facility	Conversion and enrichment facilities	Enrichment, reconversion and fabrication facilities	Enrichment facilities	MOX fuel fabrication facilities
Waste type	Uranium waste contaminated with natural uranium	Uranium waste contaminated with enriched uranium	Uranium waste contaminated with depleted uranium	TRU waste
Composition of isotopes of uranium	$^{238}\text{U}$ : 99.2739 %	$^{238}\text{U}$ : 95 % or more	$^{238}\text{U}$ : 99.7 % or more	—
	$^{235}\text{U}$ : 0.7204 %	$^{235}\text{U}$ : 5 % or less	$^{235}\text{U}$ : 0.3 % or less	
	$^{234}\text{U}$ : 0.0057 %	$^{234}\text{U}$ : around 0.1 %	$^{234}\text{U}$ : around 0.005 %	
Coexisting nuclides	Uranium series nuclides, actinium series nuclides	Progeny nuclides ( $^{231}\text{Th}$ , $^{234}\text{Th}$ , $^{234}\text{Pa}$ )	Progeny nuclides ( $^{231}\text{Th}$ , $^{234}\text{Th}$ , $^{234}\text{Pa}$ )	Progeny nuclides ( $^{231}\text{Th}$ , $^{234}\text{Th}$ , $^{234}\text{Pa}$ ), FPs

MOX mixed oxide, TRU transuranium, FP fission product

Uranium processing facilities consist of conversion facilities, which convert uranium oxide ( $\text{U}_3\text{O}_8$ ) into uranium hexafluoride ( $\text{UF}_6$ ); enrichment facilities, which enrich  $\text{UF}_6$ ; reconversion facilities, which reconvert enriched  $\text{UF}_6$  into enriched uranium oxide ( $\text{UO}_2$ ); and fabrication facilities, which fabricate enriched uranium oxide into fuel assemblies. MOX fuel fabrication facilities are where uranium and plutonium which are recovered from reprocessing of spent fuels are converted and fabricated.

The operation of these facilities generates radioactive waste containing uranium or plutonium, or both. The generators of such wastes are limited to three: fuel fabricators, Japan Nuclear Fuel Ltd. (JNFL), and the Japan Atomic Energy Agency (JAEA), which operates enrichment facilities. Their wastes can be classified by the characteristics of contained radioactive material as shown in Table 2.1 [1]. The components contained in uranium wastes are essentially limited to uranium isotopes and their progeny nuclides, as far as radionuclides are concerned, although they can be natural uranium, enriched uranium or depleted uranium depending on the type of facility. And they contain no radionuclides produced by activation or nuclear fission. It should be noted that the uranium wastes from enrichment facilities in this table include those contaminated with depleted uranium, but depleted uranium itself is not considered as waste because it is a resource for future use as fast reactor fuel.

Wastes can be mainly classified into the following types according to their properties: used off-gas filters from gaseous waste treatment, sludge as a result of liquid waste treatment, combustible solid waste (e.g., used work clothes and gloves and waste wood), and used centrifugal separators. The amount of generation of these wastes is shown in Table 2.2 [1]. It is safe to say that the amount of uranium wastes generated is almost in proportion to the amount of fuel fabricated. In Japan,

**Table 2.2** Uranium wastes generated per unit amount of uranium fuel fabrication (in the number of 200-liter drums per ton of uranium) [1]

Combustible waste	Incineration ash	Sludge	Incombustible miscellaneous solid wastes	Filters	Total (excluding combustible waste)
(1.36)	0.05	0.24	1.00	0.49	1.78

by the end of FY 2050, the cumulative amount of these wastes is estimated to reach approximately 115,000 tons (about 500,000 200-liter drums), including an estimated amount of clearance wastes of approximately 90,000 tons [2]. Table 2.3 indicates the average uranium content in different types of uranium wastes. The amount of incineration ash generated is small as shown in Table 2.2, whereas its uranium content is high as shown in Table 2.3.

Uranium-containing radioactive wastes from fabrication facilities are characterized as follows in comparison with other types of radioactive wastes [3].

1. The radionuclide content is relatively low (ranging from  $10^4$  Bq/g to 1 Bq/g, which is close to the natural content), and the contents of beta- and gamma-emitting nuclides are especially low.
2. Since uranium isotopes, which are primary radionuclides, have very long half-lives, the radionuclide content in wastes does not essentially decline. The radioactivity concentration will gradually increase with the generation of progeny nuclides and reach its peak in a few hundreds of thousands of years.
3. The gaseous progeny nuclide,  $^{222}\text{Rn}$ , is generated, incurring the risk of a significant radiation dose depending on the condition.
4. The possibility that there is an effect from non-radiological factors (e.g., properties of uranium as a heavy metal) must be taken into consideration.
5. Consideration should be given to criticality control.

There is no special treatment method for uranium wastes that focuses on its radiological characteristics; uranium wastes are treated in the same methods as those used at nuclear power plants, according to the physicochemical characteristics of each radioactive waste. Meanwhile, to effectively use uranium resources and reduce the environmental impact of burial disposal, there are various decontamination methods to remove and recover uranium from generated radioactive wastes. For example, the ultrasonic washing method and the wet blast method are used for flame-resistant wastes, and the electropolishing method is additionally applied to carbon and stainless steels, allowing decontamination even to the background level in some cases [2].

The clearance levels for the metal wastes from uranium handling facilities were calculated by the former Nuclear Safety Commission of Japan in 2009 [4] (see Sect. 4.6.1, Clearance for uranium-handling facilities). Building on the calculation results, legislation is now being considered for uranium clearance levels. Metals below the statutory clearance levels will not be considered as radioactive wastes. By contrast, metals contaminated beyond the clearance levels will be treated as radioactive wastes and be disposed of by the appropriate method chosen from the existing disposal concepts according to their uranium content.

Table 2.3 Uranium content and amount in wastes from fuel fabrication [1]

	Combustible wastes	Incineration ash	Sludge	Incombustible miscellaneous solid wastes			Filters	Aggregate average
				Flame-resistant	Incombustible	Total		
Uranium content (g-U/kg)	4.2	41	8.0	2.1	0.6	1.0	16	5.5
Uranium amount (g-U/200-liter drum)	290	3836	806	138	43	86	481	460

## 2.3 Wastes from Nuclear Power Plants [5]

Nuclear power plants generate all forms of wastes—gaseous, liquid, and solid wastes. Their characteristics vary depending on the reactor type. The following sections describe the waste generated by boiling water reactors (BWRs) and pressurized water reactors (PWRs).

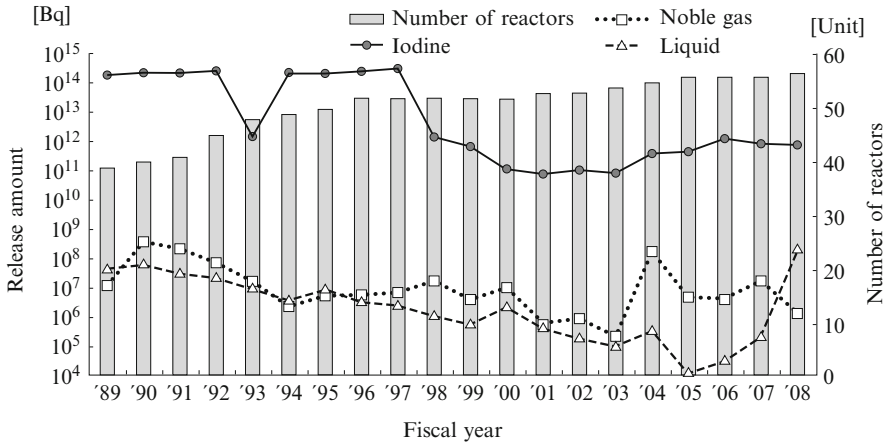
### 2.3.1 Gaseous Wastes

Gaseous wastes that should be considered at nuclear power plants are gaseous or volatile fission products. The radionuclides include  $^{85\text{m}}\text{Kr}$  (half-life: 4.4 h),  $^{87}\text{Kr}$  (78 min),  $^{88}\text{Kr}$  (2.77 h),  $^{133}\text{Xe}$  (5.27 d),  $^{133\text{m}}\text{Xe}$  (2.3 d),  $^{135}\text{Xe}$  (9.13 h) and  $^{138}\text{Xe}$  (17 min), which are all radioactive noble gases, and  $^{131}\text{I}$  (8.02 d), which is a halogen. Radioactive noble gases often have a relatively short half-life.

The gaseous waste treatment system of a nuclear power plant mainly consists of three subsystems: the reactor off-gas system, the standby gas treatment system, and the building air conditioning and ventilation system.

The filters used in the standby gas treatment system are designed to provide sufficient efficiency to remove 99.9 % or more of the iodine and particulate matter from the air released from the reactor building to the stack in the event of an accident. In a BWR, off-gas from the reactor is transferred to the turbine together with steam through the main steam pipe. The steam is condensed to water in the main condenser, while the off-gas is drawn with the air ejector, transferred to the gaseous waste system, goes through the decay pipe and the activated carbon type noble gas hold-up system for sufficient radioactive decay, before being released from the stack. In the event of a leak from a pump, valve or other components, the ventilation air in the reactor building or the turbine building may contain a trace amount of radioactive material. This air is released from the stack through the filter in the air conditioning and ventilation system.

A PWR has a closed circuit reactor cooling system, and it does not continuously release off-gas like the BWR. The amount of off-gas from the PWR is not as large as that from the BWR, and it is stored in the gas decay tank for a few tens of days for the decay of nuclides with short half-lives. In the PWR, hydrogen is added to the reactor coolant as a corrosion inhibitor. To keep its content at the proper level, hydrogen gas is injected in the volume control tank to maintain the specified pressure. At the startup and shutdown of the plant, hydrogen gas is replaced by nitrogen gas in the volume control tank. During this gas replacement process, noble gas in the primary coolant is discharged out of the system, along with hydrogen gas and nitrogen gas. To reduce the radioactivity of this noble gas, hydrogen gas in the off-gas is separated by the hydrogen recombiner first, and the remaining off-gas containing noble gas is stored in the gas decay tank for one to one and a half months for radioactive decay before being released to the environment. The latest PWR



**Fig. 2.1** Amount of radioactive gaseous and liquid wastes released from commercial power reactors and the number of reactors in Japan [6]

model uses the activated carbon type noble gas hold-up system, which requires simpler equipment than the volume control tank.

Figure 2.1 shows the trends in the total amount of gaseous wastes released from reactors in Japan. Since the beginning of this century, the released amount has been smaller by three to four orders of magnitude than in the 1990s. This is attributable to the shutdown of the Tokai Power Station of the Japan Atomic Power Company (JAPC). The reactor at the Tokai Power Station was a gas-cooled reactor, which, unlike light water reactors, directly released air used for cooling around the core to the environment and therefore released air containing  $^{41}\text{Ar}$  (110 min).

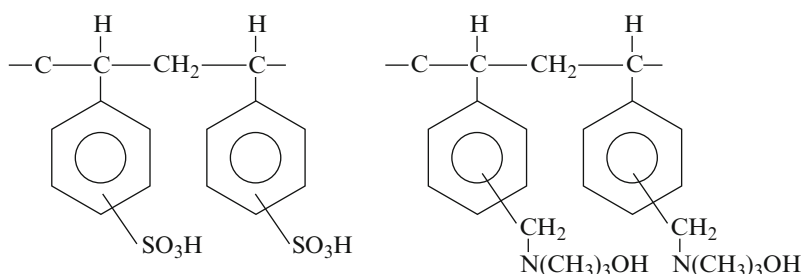
### 2.3.2 Liquid Wastes [7, 8]

The liquid waste treatment system separates both radioactive effluent and effluent potentially contaminated with radioactive material generated in the power plant according to characteristics, then collects and treats them. The treated water is reused in the power plant or discharged to the environment after assessment of the radioactivity concentration. The total amount of liquid waste released from commercial reactors in Japan to the environment is shown in Fig. 2.1. The amount of liquid waste has increased since 2007 because of the contribution of JAPC's Tokai Daini Power Station. In the volume reduction process for miscellaneous solid wastes employed by this facility, radionuclides migrate to the off-gas where they become trapped in the washing water of the off-gas scrubber before being released as effluent.

**Table 2.4** Example design specifications for the BWR condensate demineralizer using ion exchange resin

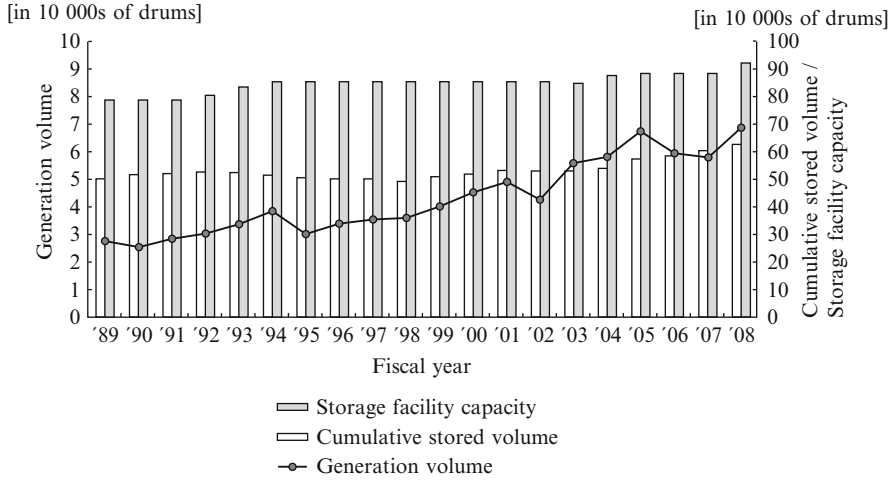
Condensate flow rate [ $\text{m}^3/\text{h}$ ]	6,345
Number of demineralizer tanks	10 (nine demineralizers in operation, one on standby)
Volume of cation exchange resin [ $\text{m}^3/\text{tank}$ ]	4.25
Volume of anion exchange resin [ $\text{m}^3/\text{tank}$ ]	2.65
Total resin volume <sup>a</sup> [ $\text{m}^3$ ]	70.5

<sup>a</sup>Including the volume of intermediate resin for chemical regeneration

**Fig. 2.2** Chemical structure of two ion exchange resins: *left*, cation exchange resin; *right*, anion exchange resin

In nuclear power plants, granular and powdery ion exchange resins are widely used in equipment such as the condensate demineralizer, condensate filter demineralizer system, reactor water purification system and effluent treatment system. Table 2.4 shows example design specifications for the BWR condensate demineralizer using ion exchange resin. The volume of ion exchange resin used for liquid waste treatment is as large as about  $70 \text{ m}^3$ . The chemical structure of an example ion exchange resin is shown in Fig. 2.2. It is based on a copolymer of styrene and divinylbenzene to which a sulfone group or a quaternary ammonium group has been introduced. Each group is capable of exchanging positive and negative ions. Radioactive material in liquid is fixed onto the ion exchange resin, which will then be treated as solid waste.

Liquid wastes are subject to filtration through which the solid component of effluent is separated and removed. The filter primarily employed for filtration in PWRs is the “hollow fiber” filter (HFF), which uses a hollow, filamentous membrane with microscopic pores on the surface. The HFF can be downsized by bundling filamentous filters while being able to minimize pressure losses arising from the passage of water. It also delivers such benefits as not easily clogging due to the small pore size, high performance and long life, thus reducing secondary waste generation. These treatment technologies are elaborated on in Chap. 5, “Radioactive Waste Treatment Technologies.”



**Fig. 2.3** Generation volume and cumulative stored volume of radioactive solid wastes from commercial power reactors in Japan [6]

### 2.3.3 Solid Wastes

Solid wastes from nuclear power plants are classified into the following types and managed according to their properties.

1. Sludge: This is stored in the sedimentation tank in the water purification system or solidified with cement or other material after storage for radioactive decay.
2. Spent resin: This is stored in the spent resin tank for radioactive decay, before being incinerated at miscellaneous solid waste treatment facilities or solidified with cement or other material. The annual generation volume of spent exchange resin per reactor is 20 m<sup>3</sup> for a BWR and 4–7 m<sup>3</sup> for a PWR.
3. Combustible miscellaneous solid wastes: These are incinerated in incineration facilities for volume reduction. Incineration ash is stored in metal drums.
4. Incombustible miscellaneous solid wastes: These are compacted for volume reduction if possible. In some power plants they may be subjected to melting.
5. Relatively high level radioactive solid wastes: Activated components, such as spent control rods, have a relatively high level of radioactivity and are therefore stored in the fuel pool for radioactive decay. For effective storage of fuel in the fuel pool, some power plants have a side bunker facility as a storage pool dedicated to receiving wastes.
6. Solid wastes are usually stored in 200-liter drums or metal containers of around 1 m<sup>3</sup>, which are called square containers.

Figure 2.3 shows example data regarding the generation of solid wastes from the Japanese plants. In FY 2008, 734 m<sup>3</sup> of waste was generated in total from three power plants due to the replacement of steam generators, reactor vessel upper heads

and reactor core internals. The graph indicates a gradual increase in the annual waste generation volume because of the increase in the number of operating plants and the extension of periodic inspection periods as plants age.

In FY 2008, commercial power reactors in Japan generated approximately 67,600 200-liter drums of low-level radioactive wastes. However, the cumulative stored volume increased only about 21,700 drums. This is attributable to activities other than waste volume reduction, such as shipment to the Low-Level Radioactive Waste Disposal Center of JNFL, located in Rokkasho Village, Aomori Prefecture, and clearance by JAPC, along with volume reduction through incineration and compaction. The cumulative stored volume in FY 2008 was about 624,300 drums, which accounts for 68.3 % of the capacity of solid waste storage facilities, 914,600 drums.

The data in Fig. 2.3 shows statistics on commercial power reactors only, excluding waste from Fugen and Monju, which are R&D phase reactors at JAEA. Fugen, which is now being decommissioned, generated about 160 200-liter drums of low-level radioactive waste in FY 2009; its cumulative stored volume has reached approximately 19,100 drums, representing 88 % of the storage capacity [9]. Ion exchange resin and filter sludge are stored in the tanks, and spent control rods and neutron detectors in the spent fuel pool. Monju, which resumed operation in May 2010 after an interval of 14 years, generated 584 drums of waste in FY 2009 even while out of service; its cumulative stored volume is 4,432 drums [9].

Solid wastes generated as a result of reactor operation and carried out of power plants are subject to landfill disposal, concrete vault disposal or subsurface disposal according to the radioactivity level (Chaps. 1 and 6).

## 2.4 Wastes from Reprocessing

Radioactive wastes from reprocessing plants are characterized by a greater content of TRU nuclides than other types of radioactive wastes. This section describes radioactive wastes from reprocessing plants using the tried-and-tested PUREX (Plutonium and Uranium Recovery by EXtraction) method. Spent fuel (including cladding and other components of fuel assemblies), which is the subject of reprocessing, is highly radioactive, containing approximately 31.6 PBq ( $=31.6 \times 10^{15}$  Bq) of radioactivity per ton of waste, as exemplified in Table 2.5 [10].

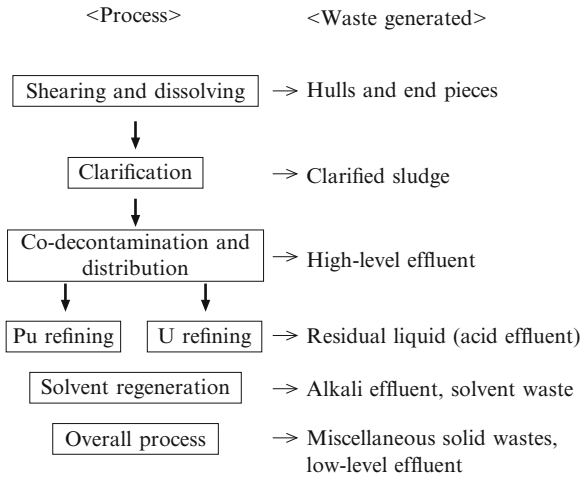
The PUREX method separates uranium and plutonium, as nuclear material, from fission products by dissolving spent fuel with nitric acid into a solution, which then repeatedly undergoes the same extraction process. Since it is an aqueous method, large amounts of liquid wastes are produced; the liquid wastes contain most of the fission products that were in the spent fuel, unrecovered uranium and TRU nuclides.

Figure 2.4 shows radioactive wastes generated in different stages of reprocessing based on the PUREX method. Radioactive liquid wastes mainly consist of high-level effluent and acid effluent from solvent extraction process. The high-level effluent is vitrified as high level solid radioactive waste. Various kinds of chemicals are used in reprocessing, such as nitric acid to dissolve the fuel, tributyl

**Table 2.5** Data on spent fuel [10]

Fuel	Weight	Uranium	957 kg
		Plutonium	9 kg
		Fission products	34 kg
	Radioactivity	Alpha- emitting nuclides	4.0 PBq
		Beta- and gamma-emitting nuclides	27.4 PBq
	Calorific power		2.96 kW
Fuel assembly components	Weight	Zircaloy	292 kg
		Stainless steel	106 kg
		Inconel	24 kg
		Alumina	2 kg
	Radioactivity	Activated nuclides	0.2 PBq*

Data are per ton of new fuel uranium, for PWRs, at a burn-up of 33,000 MWd, and for fuel having been cooled for 3 years after being unloaded from the reactor (except were marked with an asterisk [\*], which is for fuel cooled for 10 years after being unloaded from the reactor)



**Fig. 2.4** Major radioactive wastes from reprocessing based on the PUREX method [11]

phosphate (TBP) to extract uranium and plutonium, dodecane as an organic solvent to dilute the TBP, and sodium carbonate as a cleaner for the organic solvent. Although some of these are regenerated and reused, all will ultimately become radioactive wastes in the form of spent solvents.

Solid wastes can be divided into several types: end pieces of fuel structural components generated during the shearing and dissolution processes, and hulls used as cladding, which are both wastes with a relatively high level of radioactivity; and combustible or incombustible low-level radioactive miscellaneous solids, which consist of waste components and materials from plants. In addition, repaired or replaced items from different stages of reprocessing, such as primary equipment, auxiliary equipment and purification equipment, are contaminated with

radioactive materials and treated as radioactive wastes. Reprocessing plants are decommissioned after their service life, leading to the generation of dismantling waste.

The reprocessing plant of JNFL has the capacity to reprocess 800 tons of spent fuel annually and is expected to generate  $5,600 \text{ m}^3$  of waste every year.

### 2.4.1 Gaseous Wastes

When spent fuel is sheared and dissolved, radioactive gas trapped in the fuel pin plenums or fuel pellets is released. This gas constitutes a large part of the radioactive gaseous wastes from reprocessing plants and its major nuclides are Kr and I.

More than 80 % (in molar ratio) of the gas generated as fission products in nuclear fuel is Xe. However, due to its short half-life (9.1 h for  $^{131}\text{Xe}$ , 5.27 d for  $^{133}\text{Xe}$ ), most of it decays during the cooling period before reprocessing starts. Among the major nuclides of Kr and I in gaseous wastes, the dominant one is  $^{85}\text{Kr}$  (10.3 y). Although its collection was considered for the purpose of reducing the environmental release of radioactivity, such a measure has not been adopted by JNFL's reprocessing plant. To collect and solidify Kr, highly enriched krypton gas would need to be accumulated in the plant. The above decision not to recover Kr was made because dilution and diffusion through release from the main stack is more effective in reducing radiation doses to residents around the plant. Likewise, Kr is not collected in reprocessing plants in France and the U.K. either, where commercial reprocessing takes place [12].

JNFL's reprocessing plant is estimated to generate  $1.23 \times 10^{12} \text{ Bq}$  (approx. 189 kg) of  $^{129}\text{I}$  (15.7 million years) annually. On the other hand,  $^{131}\text{I}$  (8.02 d) and other radioactive iodine nuclides with short half-lives disappear for the most part during the cooling period before reprocessing, at least temporarily. However, a very small amount, or  $1.86 \times 10^{11} \text{ Bq}$  (approx. 40  $\mu\text{g}$ ) annually, of  $^{131}\text{I}$  is estimated to be generated due to nuclear fission in high level liquid waste tanks and other parts of the plant [13].

Iodine can be removed either by the washing method or the solid adsorption method. A common washing method is the alkali washing method, which uses sodium hydroxide. Solid adsorbents being considered or actually used are silver adsorbents such as silver zeolite, silver silica gel, and silver alumina. The alkali washing method usually brings off-gas, containing iodine, in contact with 1–2 mol/L of sodium hydroxide (NaOH) to remove iodine as sodium iodide (NaI) or sodium iodate ( $\text{NaIO}_3$ ).



Washing with alkali solution is effective for removing inorganic iodine but not for organic iodine, which is difficult to capture.

Impregnated charcoal, which is made by adding a small amount of potassium iodide to activated carbon, is used as the iodine filter at nuclear power plants, but not at reprocessing facilities because iodine may react with  $\text{NO}_x$  in the off-gas and create an explosive compound. Unlike silver adsorbents, activated carbon only physically adsorbs iodine to temporarily hold it. Although adsorption efficiency increases as temperature drops, a reduction in the adsorption amount or desorption of the adsorbed iodine occurs if temperature rises or if other materials, such as water, are adsorbed. Therefore, if humidity is high and an adsorptive gas (e.g., acid gas and organic gas) coexists, the iodine capture efficiency significantly drops [10].

### 2.4.2 Liquid Wastes

Acidic PUREX raffinate from co-decontamination and partition processes during reprocessing contains most of the nonvolatile fission products included in spent fuel, actinides, and trace amounts of uranium and plutonium (about 1 % of the amount in spent fuel), as well as other corrosion products, and is highly radioactive, at about  $10^7$  GBq/m<sup>3</sup>. Reprocessing using the PUREX method produces about 5 m<sup>3</sup> of the raffinate from every ton of spent fuel. Its volume is reduced to about 1/10 through evaporation. The volume reduction rate is closely related to the burn-up and the cooling time of the fuel to be treated. If the cooling period is long enough, the limiting factor of the extent of volume reduction is the heat generation in the concentrated solution and the amount of solids in the liquid waste, which contain fission products and chemicals used for reprocessing. The appropriate volume reduction assumed for the design of the storage tank capacity at a reprocessing plant is a reduction to around 0.6 m<sup>3</sup> per ton of spent fuel. To prevent metallic corrosion of the evaporator, the raffinate undergoes treatment to lower the nitric acid concentration by reducing the nitric acid in the solution to  $\text{NO}_x$  by adding formaldehyde and sucrose. Then vitrification, described in the next section, takes place.

Liquid wastes from other reprocessing processes, such as uranium refining and plutonium refining, also undergo evaporation. In overseas reprocessing plants, where liquid wastes have high  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  contents, a coagulation-sedimentation treatment is used to coprecipitate  $^{90}\text{Sr}$  with sulfate and  $^{137}\text{Cs}$  with nickel ferrocyanide for removal. Concentrated liquid waste and sludge generated in the course of concentration of these liquid wastes are usually subjected to bituminization. Due to differences in radiation levels in liquid waste, bituminization uses straight asphalt at nuclear power plants and blown asphalt at reprocessing plants in general. Blown asphalt is characterized by smaller cubical expansion caused by gas generated by radiolysis than straight asphalt. At JNFL's reprocessing plant, concentrated liquid waste from the evaporation of low-level radioactive liquid waste is dried and compaction-molded before being stored in metal drums.

The solvent used in reprocessing contains impurities which have been generated by chemolysis or radiolysis such as degradation products of TBP and dodecane.

The degradation products of TBP and dodecane include dibutyl phosphate, butyl nitrate and 1-butanol, and lauric acid, respectively. Solvent waste as a result of purifying spent solvent was once treated by incineration. However, since it causes significant corrosion to the incinerator, other methods, including vacuum distillation method and the TBP extraction method using phosphoric acid, are used today. Solvent waste mainly consisting of TBP separated by any of these methods is thermally decomposed, and the remaining phosphoric acid is neutralized with calcium hydroxide to get an inorganic salt. At JNFL's reprocessing plant, solvent waste from the solvent treatment system in the solvent collection facility is thermally decomposed, compaction-molded, and stored in metal drums or other containers.

### 2.4.3 Solid Wastes

Solid wastes, such as cladding (hulls) and fuel assembly components which are insoluble in nitric acid, are contaminated with not only undissolved fuel and fission products but also activated products that are radioactive as a result of irradiation with neutrons. The zircaloy hulls for light water reactors occlude tritium. Although discussions have been held on volume reduction methods for these solid wastes, they are stored without being treated at present. Other solid wastes generated during the reprocessing process undergo volume reduction by incineration, compaction or other methods.

The primary wastes from reprocessing plants are so called "TRU waste" and high-level radioactive wastes, as mentioned above. The former includes spent silver adsorbent, which has solidified  $^{129}\text{I}$ , TRU waste from MOX fuel fabrication facilities (Sect. 2.2), and repatriated low-level radioactive wastes that have been generated in reprocessing projects contracted to overseas facilities. Repatriated wastes are elaborated on in Sect. 2.6.

As explained in Chap. 1, TRU wastes from the reprocessing process are characterized as being contaminated with both nuclear fission products and TRU nuclides and as containing diverse waste forms; therefore they range widely in radioactivity concentration. In these aspects, TRU wastes are unlike wastes from fuel fabrication facilities and from nuclear power plants, which generate wastes of almost uniform composition. The radioactivity concentrations in TRU wastes (Fig. 1.1) from alpha-emitting nuclides are from 10 kBq/ton to 1 TBq/ton and from beta- and gamma-emitting nuclides, 1 MBq/ton to 1 PBq/ton. The appropriate disposal method is selected for each type of TRU waste in consideration of such radioactivity concentrations and confinement performance for the given waste form.

Another waste specific to reprocessing is high-level radioactive liquid waste. This waste is mixed with glass material and solidified in stainless steel canisters after which it is converted into vitrified waste. In Japan, the term high-level radioactive wastes always refers to vitrified wastes. Table 2.6 shows major radio-nuclides contained in vitrified wastes and their radioactivity. They can be broadly

**Table 2.6** Major radionuclides contained in vitrified wastes and their radioactivity<sup>a</sup> [14]

Half – life (year)	Fission products			Transuranium		
	Nuclide	Half – life (year)	Radioactivity (Bq)	Nuclide	Half – life (year)	Radioactivity (Bq)
2–100	<sup>90</sup> Sr - <sup>90</sup> Y	29.1	$6.41 \times 10^{15}$	<sup>238</sup> Pu	87.8	$3.39 \times 10^{11}$
	<sup>125</sup> Sb - <sup>125m</sup> Te	2.8	$1.32 \times 10^{14}$	<sup>241</sup> Pu	14.4	$1.01 \times 10^{13}$
	<sup>134</sup> Cs	2.1	$1.74 \times 10^{15}$	<sup>243</sup> Cm	28.5	$8.95 \times 10^{11}$
	<sup>137</sup> Cs - <sup>137m</sup> Ba	30.2	$9.32 \times 10^{15}$	<sup>244</sup> Cm	18.1	$1.67 \times 10^{14}$
	<sup>147</sup> Pm	2.6	$1.97 \times 10^{15}$			
	<sup>151</sup> Sm	90.0	$1.38 \times 10^{13}$			
	<sup>152</sup> Eu	13.6	$6.85 \times 10^{10}$			
100–1,000	<sup>154</sup> Eu	8.6	$1.94 \times 10^{14}$			
	<sup>155</sup> Eu	5.0	$8.99 \times 10^{13}$	<sup>241</sup> Am	432	$5.52 \times 10^{13}$
				<sup>242m</sup> Am	152	$2.86 \times 10^{11}$
1,000–10,000				<sup>240</sup> Pu	6,540	$4.51 \times 10^{10}$
				<sup>243</sup> Am	7,380	$1.26 \times 10^{12}$
				<sup>245</sup> Cm	8,500	$3.13 \times 10^{10}$
				<sup>246</sup> Cm	4,730	$5.81 \times 10^9$
	<sup>79</sup> Se	$6.50 \times 10^4$	$1.59 \times 10^{10}$	<sup>239</sup> Pu	$2.41 \times 10^4$	$2.78 \times 10^{10}$
	<sup>99</sup> Tc	$2.13 \times 10^5$	$6.59 \times 10^{11}$	<sup>242</sup> Pu	$3.87 \times 10^5$	$2.18 \times 10^8$
10,000–1 million	<sup>126</sup> Sn	$1.00 \times 10^5$	$3.22 \times 10^{10}$			
	<sup>93</sup> Zr	$1.53 \times 10^6$	$8.95 \times 10^{10}$	<sup>237</sup> Np	$2.14 \times 10^6$	$1.32 \times 10^{10}$
	<sup>107</sup> Pd	$6.50 \times 10^6$	$5.96 \times 10^9$			
	<sup>135</sup> Cs	$2.30 \times 10^6$	$2.22 \times 10^{10}$			
Total radioactivity		$2.28 \times 10^{16}$ Bq				
Element weight		38.4 kg		Np 506 g, Pu 22.1 g, Am 583 g, Cm 61.2 g		

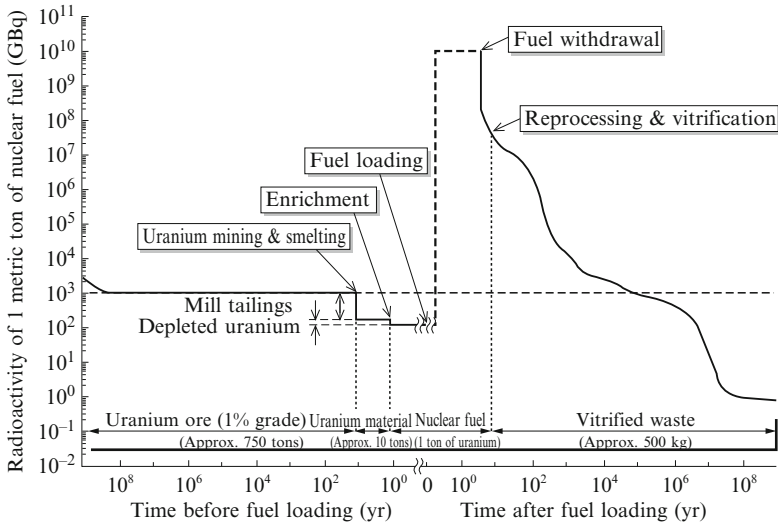
<sup>a</sup>The figures are per ton of spent fuel at a 45,000 MWd/ton burn-up in a PWR after a 5-year cooling period. The ratios transferred to wastes through reprocessing are assumed to be 0.2 % for U, 80 % for Np, and 0.2 % for Pu. Although <sup>129</sup>I ( $1.57 \times 10^7$  years,  $3.16 \times 10^7$  Bq) contained in spent fuel is an important nuclide, it is not contained in vitrified wastes

divided into three types: fission products of uranium and its fissile products such as plutonium, TRU which are generated when uranium absorbs neutrons, and activation products which are produced from small amounts of impurities in fuel. Radioactivity levels of vitrified waste at the time of fabrication is dominated by nuclides with relatively short half-lives, such as  $^{90}\text{Sr}$  (29.1 y),  $^{137}\text{Cs}$  (30.2 y) and  $^{244}\text{Cm}$  (18.1 y), with a total radioactivity of about  $1 \times 10^{16}$  Bq/ton. After these nuclides decay in a few hundreds of years, radioactivity remains from nuclides with long half-lives, such as  $^{99}\text{Tc}$  (213,000 y),  $^{93}\text{Zr}$  (1.53 million years) and  $^{237}\text{Np}$  (2.14 million years), although they are present in small amounts.

When spent fuel is reprocessed within a few years after its unload from the reactor core, the decay of  $^{241}\text{Pu}$  (14.4 y) is negligible. However, if reprocessing does not take place for 10 years or more after unload from the reactor core, generation of  $^{241}\text{Am}$  from the beta decay of  $^{241}\text{Pu}$  is not negligible. In its half-life of 432 years,  $^{241}\text{Am}$  undergoes alpha decay to produce  $^{237}\text{Np}$ . Reprocessing of spent fuels at an early point after unload and early Pu recycling after reprocessing are preferable because, from the viewpoint of Pu resource utilization, useful  $^{241}\text{Pu}$  which has the large cross section of fission decreases by its decay and, from the viewpoint of the safety of geological disposal, the heat generated from  $^{241}\text{Am}$  remains dominant for more than 1,000 years, and long-lived, toxic  $^{237}\text{Np}$  is generated.

Vitrified wastes produced after reprocessing have very high levels of radioactivity and dose equivalent rates at the time of production are estimated as 14,000 Sv/h on the waste surface and 420 Sv/h at 1 m apart from the surface. Therefore, verification of canister integrity is performed remotely as a matter of fact. Because of high calorific values, vitrified wastes are cooled for 30–50 years after vitrification, followed by transportation to a geological disposal site. The cumulative generation amount of vitrified wastes is estimated to reach about 40,000 canisters, or about  $7,000 \text{ m}^3$ , by 2020 in Japan.

Vitrified wastes have a very long “life” because of the long-lived nuclides contained in the high-level radioactive wastes. This is the fundamental reason why geological disposal, which requires no further direct control after closure, is chosen. Figure 2.5 illustrates changes in radioactivity level in nuclear fuel throughout its life cycle, from uranium ore to power generation and disposal. To supply 1 ton of uranium for fuel, 750 tons of 1 % grade uranium ore is needed, which has a radioactivity of 1,000 GBq (1 % is a fairly high grade, and Canada has ore deposits with a uranium grade of this level). When fabricated into fuel and loaded in the reactor, radioactivity per ton of fuel drops to 100 GBq, but once fuel is used for power generation and is irradiated in the reactor, its radioactivity steeply rises by eight orders of magnitude, or 100 million-fold. During the 4-year period between unload from the reactor and reprocessing and vitrification, radionuclides with very short half-lives decay and disappear, reducing radioactivity by about two orders of magnitude. This is considered the radioactivity level for vitrified waste. Its radioactivity slowly declines by about three orders of magnitude for the first few hundred years because of the decay of radionuclides that played a dominant role in the initial radioactivity, such as  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{244}\text{Cm}$ . Although the radioactivity level



**Fig. 2.5** Changes in radioactivity per ton of uranium throughout the nuclear fuel cycle (for 1 metric ton of 4.5 % enriched nuclear fuel) [15]

continues to drop thereafter as well, it takes about 100,000 years after terminating power generation until the level equivalent to the original uranium ore is restored.

Figure 2.5 also indicates that specific radioactivity 100 million or more years ago was higher than it is now. This is because the isotopic ratio of  $^{235}\text{U}$ , with a 700 million year half-life, was greater at the birth of the Earth. It is easily calculated that the isotopic ratio of  $^{235}\text{U}$  about two billion years ago was a few percent, far higher than it is now, 0.72 %. At that time, unevenly distributed uranium in underground veins and surrounded with groundwater and rocks (mainly silica) that were effective neutron moderators, created a “natural reactor.” It has been confirmed that in the Oklo mine in the Gabonese Republic, Africa, a chain reaction of nuclear fission had occurred intermittently for a few hundreds of thousands of years [16].

## 2.5 Wastes from R&D and Radioisotope Uses

Radioisotopes (RIs), radiation generating devices, and nuclear fuel material are used not just in nuclear power generation and other energy fields but also for many other purposes, including R&D, agriculture, industry and health care.

RIs and radiation generating devices are used in industrial fields such as paper-making, steel sheet manufacturing (to make thickness measurements) and semiconductor fabricating, and in medical fields such as nuclear medical diagnosis (e.g., positron emission tomography-computed tomography, PET-CT) and the

sterilization of injection needles and other medical equipment. Universities and research institutes conduct studies by using RIs as tracers and employ accelerators as radiation generating devices. The operation of these RI-using facilities and radiation generating devices generates RI-containing wastes such as test tubes, injectors, paper towels and off-gas filters. The dismantling of these facilities also generates such wastes as metals and concrete contaminated with RIs and activated products of these materials. These are collectively called RI wastes.

Typical radionuclides contained in RI wastes are  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{32}\text{P}$  and  $^{35}\text{S}$  in the R&D and industrial fields and  $^{99\text{m}}\text{Tc}$ ,  $^{125}\text{I}$  and  $^{201}\text{Tl}$  in the medical field. Most RI wastes generated in Japan are collected and stored by the Japan Radioisotope Association (JRIA).

R&D activities at universities and research institutes produce a wide variety of wastes contaminated with nuclear fuel materials. Likewise, the dismantling of facilities handling nuclear fuel materials generates contaminated metals and concrete as well as activated reactor core internals. These are classified as radioactive wastes generated from research, industrial and medical facilities. Wastes from JAEA's prototype advanced thermal reactor Fugen and the prototype fast breeder reactor Monju are included in the wastes because they are in the R&D phase and not commercial reactors (Sect. 2.3.3). Typical radionuclides in the radioactive wastes are  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{238}\text{U}$ . Most of these wastes are stored at the business site of the source facilities.

RI wastes and radioactive wastes from laboratories are together called radioactive wastes from research facilities. These two types of wastes are often discussed separately because each is governed by a different set of regulations in Japan. The use of RIs is regulated by the Law Concerning Prevention from Radiation Hazards due to Radioisotopes, etc. (Radiation Hazards Prevention Law), the Medical Service Act, the Pharmaceutical Affairs Act, the Veterinary Practice Act, and the Clinical Laboratory Technician Act. The Radiation Hazards Prevention Law applies to about 4,600 business sites, of which about 1,000 are generators of RI wastes. The Medical Service Act, the Pharmaceutical Affairs Act, and the Clinical Laboratory Technician Act apply to about 1,300 facilities handling radioactive drugs. The use of nuclear fuel materials is governed by the Act on the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors. Radioactive wastes from laboratories are generated from about 170 organizations, including JAEA, universities, and private enterprises.

It was decided in 2008 that radioactive wastes from research facilities should be disposed of by JAEA, and the Japan Atomic Energy Agency Act was revised accordingly. Survey results on the generated amounts of such wastes expected to be disposed of by FY 2048 are shown in Table 2.7 [17]. One notable characteristic of radioactive wastes from research facilities is the variation in radioactivity concentration in the waste forms, which ranges from somewhere around clearance levels to above the upper concentration limits for concrete vault disposal, because of the wide variety of sources. The disposal method appropriate for the radioactivity concentration of each waste must be selected accordingly.

**Table 2.7** Estimated disposal amount of radioactive wastes research facilities [17]

Institution	Landfill disposal	Concrete vault disposal	Subsurface disposal
JAEA	244,400	188,700	56,200
JRIA	33,300	9,100	—
Universities, businesses, etc.	42,800	2,300	4,400
Total	320,500	200,100	60,600

Data are expressed in the number of 200-liter drums and indicate cumulative totals expected by FY 2048. The figures are not generation amounts at facilities but the number of disposal waste forms that will have undergone incineration, compaction or other treatment after generation

## 2.6 Repatriated Wastes

As a transitional measure until the inauguration of JNFL's reprocessing plant in Rokkasho Village, Aomori, Japanese electric utilities have outsourced the reprocessing of their spent fuel to AREVA NC (formerly COGEMA), based in France, and British Nuclear Group Sellafield Ltd. (BNGS, formerly BNFL). Approximately 7,100 tons of spent fuel has been sent overseas reprocessing by 2007. The recovered uranium and plutonium are repatriated from the reprocessing plants as fuel. Wastes generated during reprocessing are also returned to Japanese utilities because both European countries follow the policy of repatriating radioactive wastes from reprocessing to the country of origin, in principle. Such wastes are called repatriated waste.

Vitrified high-level radioactive wastes have been shipped to Japan 13 times by March 2010, with 1,338 canisters accepted in total since the first shipment of 28 canisters in April 1995 from the Port of Cherbourg, France, to Mutsu-Ogawara Port in Aomori. The ship used is owned by PNTL, an affiliated company of BNGS. After arrival at the port, waste forms are taken to JNFL's Vitrified Waste Storage Center in Rokkasho Village by a special land carrier. Approximately 850 canisters of vitrified high-level radioactive wastes are expected to be repatriated from the U.K. The Vitrified Waste Storage Center's storage facility employs a multi-stack vault storage structure with an air cooling system and has a storage capacity of 1,440 canisters, which is expected to increase to 2,880 in the future.

Low-level radioactive wastes are expected to be duly repatriated as well. France and the U.K. proposed changes in the specifications of repatriated wastes regarding this. The former proposed that the form of the TRU wastes (low-level radioactive wastes subject to geological disposal) be changed from the originally planned bituminized form (approx. 1,100 drums) to 28 canisters of vitrified waste. The U.K. proposed that a certain numbers of low-level radioactive waste forms—namely, about 4,500 drums of cemented TRU wastes subject to geological disposal and about 6,000 drums of heterogeneous solid wastes subject to shallow underground disposal—be exchanged with about 150 canisters of vitrified high-level radioactive wastes for repatriation. These proposals are expected to benefit Japan in

that they will significantly cut back transportation costs because of the reduced volume of the waste to be repatriated and thus a smaller number of shipments will be required; as well they will create an extra allowance in Japan's storage capacity.

However, before these proposals could be accepted, the Japanese government had to revise laws concerning geological disposal. The British proposal on TRU wastes called for "exchanging with small amounts of high-level radioactive wastes (originating in overseas reactors) with equivalent radiation" (equivalent exchange). The then-applicable Designated Radioactive Waste Final Disposal Act [18], enforced in 2000, assumed accepting only high-level radioactive wastes generated at domestic power reactors and not those from overseas reactors. In 2005, the Act was amended to add TRU wastes to the list of the specified radioactive wastes and to oblige JNFL and JAEA, as the generators of TRU wastes, as well as electric utilities as the importers of high-level radioactive wastes to be received in place of TRU wastes, to contribute toward disposal expenses. This has legally enabled the acceptance of repatriated wastes.

## 2.7 Wastes Below the Clearance Limit

Clearance is discussed in further detail in Chap. 4. The clearance level is a radioactivity concentration for which radiation effects on the human body are small enough to be negligible, and materials with a radioactivity concentration below this level do not have to be classified as radioactive materials. Wastes with radioactivity below the clearance limit are outside the scope of regulation and are no longer considered as radioactive wastes.

Some of the wastes generated during nuclear facility dismantling are clearance items. When a 1,100 MW light water reactor is dismantled, approximately 500,000 tons of waste is generated, of which cleared items account for 2–5 %.

Meanwhile, there is the concept of "non-radioactive wastes." These are a kind of waste that is generated within a controlled area but they are distinguished from radioactive wastes because of usage records indicating no possibility of contamination with radioactive materials or no need for consideration of the impact of neutron-induced activation. Non-radioactive wastes account for 92–96 % of the wastes from the dismantling of a 1,100 MW light water reactor. This means that as far as the reactor is concerned, wastes below the clearance limit almost always refer to non-radioactive wastes.

## 2.8 Wastes from Partitioning and Transmutation

Partitioning and transmutation (P&T) technology separates long-lived radionuclides contained in high-level radioactive wastes and converts them into shorter-lived or stable nuclides through transmutation. Its primary goal is to reduce the

long-term burden of geological disposal by attenuating the radioactivity of the high-level radioactive wastes in shorter time periods. The partitioning of elements contained in wastes is sometimes conducted, even without transmutation, for the purpose of optimizing treatment and disposal by developing the optimal waste form for each element, as in the case of the Hanford Site in the U.S., where high level effluent from nuclear weapons production is treated. However, the mainstream trend in the world is R&D focusing on systems combining both P&T, since transmutation has the potential to change long-lived nuclides into shorter-lived nuclides and partitioning is indispensable to transmutation.

How the introduction of this technology could change the properties of high-level radioactive waste, that is to say, how it could change radiation attenuation, has been well reported for many years [19]. However, introducing P&T technology means changing the amounts and flows of materials in the nuclear fuel cycle, which would result in the generation of radioactive wastes that are different in composition, radioactivity and chemical characteristics than wastes generated in the current nuclear fuel cycle. Just as all types of current radioactive wastes are ensured safe disposal in accordance with their characteristics on the basis of comprehensive discussions, all the radioactive wastes from the nuclear fuel cycle involving P&T call for consistent waste management policies. Furthermore, it is necessary to present benefits large enough to offset the economic and social cost of introducing the additional treatment of P&T.

This section first describes P&T technology and then analyzes and discusses radioactive wastes that would be generated if this technology were to be introduced. Medical and industrial applications of radionuclides separated from high-level radioactive liquid waste, or so-called effective utilization of waste, have been studied for a long time. Reference [20] is a starting point to provide further information.

### 2.8.1 Partitioning

This is a process to separate components subject to transmutation from spent fuel and high-level radioactive liquid waste generated from spent fuel reprocessing. The nuclides it mainly deals with are such actinide elements as  $^{237}\text{Np}$  (2.14 million years),  $^{243}\text{Am}$  (7,380 y) and  $^{245}\text{Cm}$  (8,500 y) as well as such fission products as  $^{99}\text{Tc}$  (213,000 y),  $^{129}\text{I}$  (15.7 million years) and  $^{135}\text{Cs}$  (2.3 million years). These are all long-lived radionuclides. Np, Am and Cm are called minor actinides because their weight content in spent fuel from light water reactors is smaller than that of Pu by more than one order of magnitude. Although such nuclides as  $^{79}\text{Se}$  (65,000 y),  $^{93}\text{Zr}$  (1.53 million years),  $^{107}\text{Pd}$  (6.5 million years) and  $^{126}\text{Sn}$  (approx. 100,000 y) merit attention from the viewpoint of the nuclide confinement capability of the geological

disposal system, the above-mentioned  $^{99}\text{Tc}$ ,  $^{129}\text{I}$  and  $^{135}\text{Cs}$  are considered for partitioning because of their volume of production. Other important nuclides for partitioning are  $^{90}\text{Sr}$  (29.1 y) and  $^{137}\text{Cs}$  (30.2 y), which have relatively short half-lives and are therefore not subject to transmutation, but are limiting factors in disposal site design because of their calorific value.

Thus a wide variety of elements is considered for partitioning, and different partitioning technologies would be used depending on the type of waste. Aqueous partitioning technology would be employed for separation from high-level radioactive liquid waste, while an improved reprocessing technology based on the PUREX method would primarily be used for direct separation from spent fuel. Since transmutation efficiency is generally low, the transmutation process must be repeated. This means spent fuel that has undergone transmutation would be reprocessed. Since it would contain a drastically increased content of minor actinides, consideration on material degradation due to irradiation (e.g., damage to solvent) and criticality control measures would be needed. Because of solvent degradation, dry partitioning technology not using organic solvent might be a better candidate.

## 2.8.2 *Transmutation*

The transmutation method is essentially limited to one that uses neutrons. In other words, the primary nuclear reaction is nuclear fission in minor actinide transmutation, and neutron absorption in fission product transmutation. Since multiple nuclear reactions take place simultaneously in the reactor, the number of neutrons involved in nuclear fission reactions is important to increase the efficiency of the target transmutation. Fast neutrons are especially effective because minor actinides are more likely to cause nuclear fission than absorption in the 700 keV or higher neutron energy range. Therefore, transmutation requires a high-intensity, high-energy neutron source. The two most likely sources are fast neutrons in fast reactors and fusion reactors, and neutrons generated by nuclear spallation reactions in accelerators.

The method using fast reactors has the advantage of requiring fewer items to be developed because it is an extension of current reactor technology and nuclear fuel cycle technology. By contrast, the idea of using the accelerator involves many new R&D challenges, such as developing targets inducing the nuclear spallation reactions and the steady operation of the accelerator. However, the latter does not impose as difficult safety requirements on criticality as the former because the accelerator would maintain steady output in a subcritical state. A system that induces transmutation externally by an accelerator is called an accelerator-driven system (ADS).

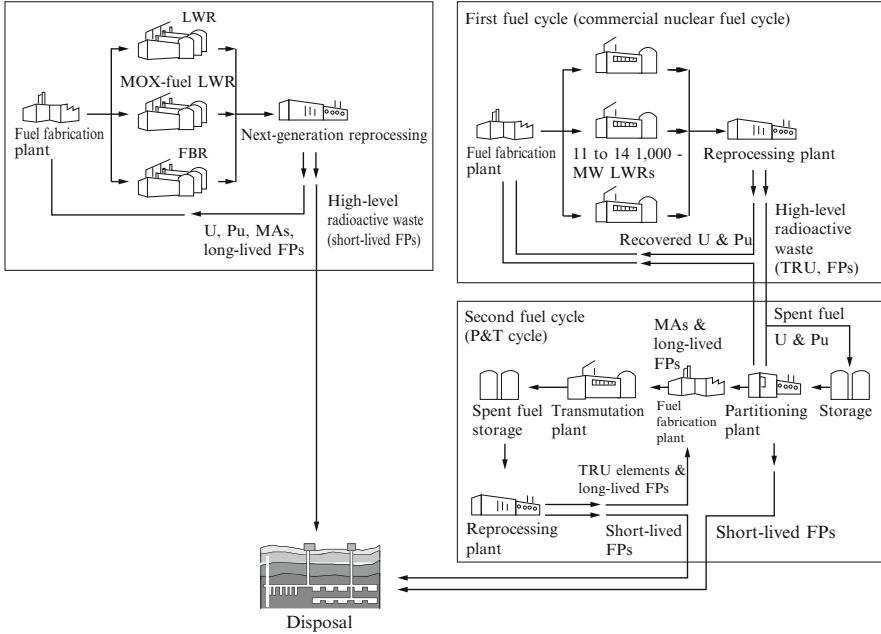
### 2.8.3 *Fuel for Transmutation*

The chemical composition requirements for fuel used for transmutation are essentially the same as those for candidate materials for the current fast reactor fuel, such as oxides, metals and nitrides, although depending on the transmutation method to be used. The only difference is the content of minor actinides. To achieve high efficiency in transmutation, fuel must have greater minor actinide content and a higher burn-up. Since such fuel would accumulate more He due to alpha damage and alpha decay, consideration must be given to effects on fuel integrity. In addition, the fabrication and reprocessing of such fuel would require higher safety, including criticality control and shielding against neutrons and other radiation sources.

Transmutation systems can be mainly divided into two types: those using power reactors and those using burner reactors specially designed for transmutation. The fuel design concept widely differs between them. The fuel design for the first type is restricted by efficiency and economy as a commercial reactor and therefore fuel having uniform composition yet low minor actinide content would be used. Given the primary requirement of maintaining reactor output at a specified level, it would be difficult to set a high transmutation rate for minor actinides. By contrast, fuel for the second type is designed to fit the reactor for the purpose of maximizing the transmutation rate of the target nuclide. For this reason, fuel would generally be able to have higher content of minor actinides. Such fuel would be placed in a limited location in the core of the burner reactor. However, requirements for fuel are stricter because of a greater change in fuel composition expected during burning.

### 2.8.4 *P&T System*

The incorporation of P&T technology into a nuclear fuel cycle would require not only neutron sources, such as the reactor and the accelerator, but also processes for partitioning, fuel fabrication and spent fuel reprocessing. Since transmutation efficiency would not be high enough to complete conversion in a single burning process, a repetition of spent fuel reprocessing and transmutation—or the multiple recycling of minor actinides—would be necessary. This suggests that it would take a significantly long time, like 100 years, to reduce the total amount of minor actinides produced by power generation by means of transmutation. This means that P&T technology must be addressed in a long-term development program for nuclear power generation, such as a reactor design strategy. International studies have been conducted on introduction scenarios for P&T technology [21]. Two major scenarios are shown in Fig. 2.6 [16], which have been studied in Japan: the advanced nuclear fuel cycle using power reactors and the tiered nuclear fuel cycle using burner reactors dedicated to transmutation, as already described.



**Fig. 2.6** Major partitioning and transmutation systems (*left*: advanced nuclear fuel cycle, *right*: tiered nuclear fuel cycle) [19] – LWR light water reactor, MA minor actinide, MOX mixed oxide fuel, TRU transuranium, FP fission product

In the first scenario, the light water reactor or the fast reactor for commercial power generation is used for minor actinide transmutation, followed by the next-generation reprocessing with an advanced PUREX method for the recovery of minor actinides along with uranium and plutonium. A commercial nuclear fuel cycle serves as a P&T cycle as is, which forms a large-scale transmutation system whereby minor actinides are handled within the nuclear fuel cycle. This is a concept that intends to implement a fuel cycle in a relatively short period of time by advancing existing technologies. As opposed to this, the second concept, the tiered nuclear fuel cycle, seeks to achieve high efficiency transmutation based on newly conceived technologies. In this scenario, minor actinides and other materials used for transmutation are extracted during the partitioning process that follows the reprocessing process in the commercial nuclear fuel cycle, and transmutation is induced in a special plant. Because it combines a separately formed P&T cycle with a commercial nuclear fuel cycle, its whole structure is called the tiered cycle. A benefit of this cycle is the ease of ensuring nuclear safety, such as preventing nuclear proliferation due to the confinement of minor actinides within a small P&T cycle, preventing minor actinides from spreading throughout the commercial nuclear fuel cycle. Most of today's research focuses on the ADS as a special transmutation plant in this cycle.

### 2.8.5 Wastes from P&T

What kind of wastes would replace high-level radioactive wastes if the P&T system were introduced to the nuclear fuel cycle? Although the amount of long-lived minor actinides contained in high-level radioactive liquid waste from the commercial nuclear fuel cycle would decrease as a result of the introduction of the P&T system, unconventional wastes would be generated from partitioning, transmutation and reprocessing. The adoption of P&T technology would change the flow of materials in the nuclear fuel cycle as well as the kind of wastes generated.

The partitioning and reprocessing processes would generate high-level radioactive wastes that would contain most of the fission products, and secondary wastes including small amounts of actinides and fission products. The loss of actinides would be inevitable in the fuel fabrication process, and activation products and nuclear spallation products would be generated in the transmutation process as radioactive wastes. The wastes expected from the tiered nuclear fuel cycle (Fig. 2.6) are analyzed below for each process.

The 4-group partitioning [22] is assumed as the partitioning process. This separation process divides the elements in high-level radioactive liquid waste into four groups: the transuranic group to be used as fuel for transmutation; the Sr and Cs group including the heat-generating nuclides of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ; the Tc and Pt group for effective utilization; and the group of other elements including iron and rare earth elements. Among these four, the Sr and Cs group and the group of other elements are considered as wastes. Sr and Cs would be adsorbed by titanium oxide and zeolite, respectively, for separation from high-level radioactive liquid waste. These adsorbents, along with adsorbed Sr and Cs, would be burned into a solid, which would then be cemented into metal drums or other containers for conversion into waste form. Vitrification is assumed for the group of other elements. Secondary wastes from this process would be solvent waste and sodium waste; the latter consists of sodium used for solvent washing. Solvent waste would be converted into calcium phosphate by pyrolysis; sodium waste would be converted into sodium nitrate by drying, before finally being solidified.

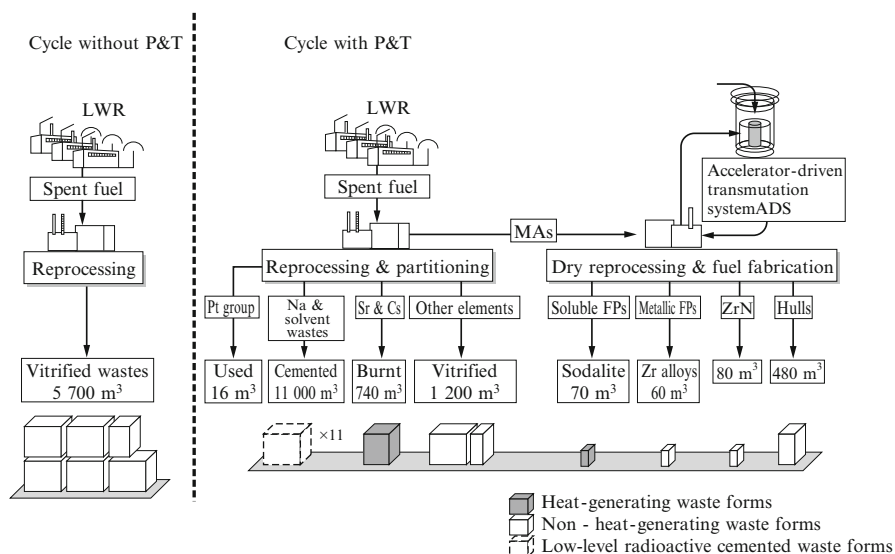
Transmutation would take place in a special ADS. The accelerator-driven reactor is assumed to have a thermal output of 800 MWt, and lead-bismuth (Pb-Bi) molten salt would serve as both the nuclear spallation target and coolant. The transmutation cycle would be operable only by the electrical output from the ADS. Since fuel would be minor actinide nitride, zirconium nitride would be used as a diluent. The initial load of minor actinides would be 2.5 tons. With the annual transmutation rate of 10 wt%, the system would be capable of transmuted 0.25 tons of minor actinides annually. This amount of minor actinides is equivalent to the amount generated from 195.7 tons of light water reactor spent fuel at 45,000 MWd/t after 4 years of cooling. Considering that JNFL's reprocessing plant can reprocess 800 tons of spent fuel annually, the operation of four ADSs is assumed here. The total reprocessing amount is assumed to be 32,000 tHM, which corresponds to 40 years of operation.

During this transmutation process, nuclear spallation products and activation products would be generated in the nuclear spallation target, and activation products in the coolant. Since the target would double as coolant, these products would be completely mixed and then extracted as dismantling waste when the service life of the transmutation reactor would expire. The amount is estimated to be about 6,500 tons.

The fuel withdrawn from the transmutation reactor would undergo fuel reprocessing, during which the residual minor actinides would be separated and recovered from fission products. It is assumed that this reprocessing would employ the fused-salt electrolysis process, which uses lithium chloride-potassium chloride (LiCl-KCl) salt and liquid cadmium. Major wastes generated here would be salt waste containing the majority of fission products and trace amounts of unrecovered minor actinides; metal waste containing rare fission products, such as platinum group elements; hulls containing activation products; and exhaust including  $^{14}\text{C}$  generated from  $^{14}\text{N}$ .

Since the fuel cladding, which would be turned into waste hulls, is made of stainless steel, it would be solidified with cement. Rare metal fission products (e.g., Ru, Rh, Pd and Tc) that would not be transformed into chlorides during spent fuel melting would become metal waste and be solidified as a zirconium alloy containing up to 4 % of them. As a means to solidify LiCl-KCl salt waste containing soluble fission products other than rare elements and noble gases, the sodalite solidification method would be used [23].

Figure 2.7 shows the results from analysis of types and generated amounts of radioactive wastes from the above P&T system, which may be called P&T system



**Fig. 2.7** Radioactive wastes from the tiered nuclear fuel cycle [24] –LWR light water reactor, MA minor actinide, FP fission product, ZrN zirconium nitride

wastes and would be generated instead of high-level radioactive vitrified wastes. The analysis is based on the calculation of material balance and radioactivity balance in the P&T system. Among the wastes identified, those with radioactivity levels equivalent to the radioactivity levels of today's high level radioactive vitrified wastes are cemented waste forms produced by burning Sr and Cs generated during the partitioning process; and sodalite waste produced from soluble fission products generated during the reprocessing of transmutation spent fuel. It is suggested that their total volume would be reduced to about 1/7 of that of vitrified wastes generated from the current cycle without P&T. Other wastes would be considered as TRU or low-level radioactive wastes. The type of waste that would particularly be large in generated amount would be the cemented form of the secondary waste (sodium waste and solvent waste) from the partitioning process, which would have radioactivity levels high enough to require concrete vault disposal or subsurface disposal today.

Wastes not included in the chart are operational wastes (e.g., devices replaced during facility operation or maintenance) and facility dismantling wastes. Items contaminated with minor actinides lost during fuel fabrication are not included either. Although activated Pb-Bi coolant (6,500 tons) in the transmutation reactor would be one of the dismantling wastes as mentioned above, studies have not reached the point where quantitative estimations concerning the dismantling of P&T and other related facilities are possible.

All radioactive wastes must be disposed of safely and rationally. P&T technology has the potential to allow high-level radioactive liquid wastes, which would otherwise be vitrified entirely, to be separated according to nuclide properties and thereby enables the design of more rational disposal sites than today's site designs. However, some wastes, like activated Pb-Bi coolant, could raise the issue of chemical toxicity of metals, in addition to being radioactive.

The radioactive wastes addressed in studies on P&T technology is essentially limited to current high-level radioactive wastes. The current nuclear power generation system produces several types of radioactive wastes other than high-level radioactive wastes. Efforts have been made to safely dispose of them by the method suitable for the characteristics of each waste type, such as the types and contents of radionuclides included (Chap. 6). A challenge in the future development of P&T technology is to establish holistic measures for radioactive waste management for the wastes expected from the nuclear fuel cycle after the introduction of P&T.

### ***2.8.6 P&T as a Waste Management Policy***

Many countries and international organizations have been eagerly conducting R&D on P&T technology. Japan was one of the first countries to start it, with basic research on partitioning and fast reactor transmutation launched around 1973. In 2009, the Japan Atomic Energy Commission decided that R&D in this area should be continued steadily [25].

With many countries aggressively promoting R&D in this sector, France is the only country that has adopted P&T as a national radioactive waste management policy, putting it on the table for national discussion. To discuss future radioactive waste management policies, France enacted in 1991 the Law of 30 December 1991 Concerning Research Activities on the Management of Radioactive Waste (1991 Law) [26] to stipulate that research should be conducted in three fields: P&T of long-lived radionuclides, and conditioning of radioactive wastes, long-term storage on the ground surface of spent fuel and radioactive wastes, and geological disposal.

In 2005, 15 years later, the results of the research were presented to the French government, which then established the Planning Act on the Sustainable Management of Radioactive Materials and Waste based on them in June 2006 [27]. The greatest advancement from the 1991 Law was that the new Planning Act stipulates that R&D on long-lived, medium- to high-level radioactive waste should principally focus on geological disposal and that P&T and long-term storage should be studied as complementary measures. It is also noteworthy that the French government formulated a clear policy for such R&D with an eye toward commercialization and industrialization. More specifically, with respect to geological disposal, the goal set in the policy is applying for a license to establish a disposal site by 2015 and starting operation by 2025. Regarding P&T, the policy seeks to promote surveys and research on the fourth-generation reactor and the ADS in coordination to complete evaluation of the outlook for industrialization by 2012 and start of operation of a prototype reactor by 2020. Additionally, the policy requires the promotion of long-term storage with the goal of either constructing new storage facilities or modifying existing facilities by 2015 at the latest so that storage capacity and period needs can be met.

In this way, France has been discussing P&T with attention to its potential to contribute to radioactive waste management. The results suggest that the policy on future P&T cannot be considered without addressing the issue of industrialization. Arguments in previous sections show that the introduction of P&T technology has an impact on the technology base for the entire nuclear fuel cycle. The issue is what kind of role this technology should play in the development of the nuclear power generation system over the next century.

## Exercises

1. The uranium enrichment process uses, as raw material, natural uranium or uranium recovered through reprocessing, and produces two products: enriched uranium with a  $^{235}\text{U}$  content higher than that in natural uranium (0.72 %); and depleted uranium with a  $^{235}\text{U}$  content lower than that in natural uranium, which is considered as waste. Answer the following questions about the material balance in the enrichment process.
  - (a) Assume that natural uranium is the only raw material input into the enrichment process and that the  $^{235}\text{U}$  content in waste is 0.3 %. How many tons of natural uranium are needed to produce 27 tons of 4 % enriched uranium? In this case, how many tons of depleted uranium will be generated?

- (b) Assume that, in addition to natural uranium, uranium recovered through reprocessing is used as a raw material input into the enrichment process. Provided that the  $^{235}\text{U}$  content in the recovered uranium is 0.8 % and that 25 tons of this uranium is supplied, how many tons of natural uranium are needed to produce 27 tons of 4 % enriched uranium? In this case, how many tons of depleted uranium will be generated?
  - (c) Compare the above two cases and explain the natural uranium saving effect of recycling uranium recovered through reprocessing and its impact on the amount of wastes generated during mining and melting processes.
2. Since the transmutation efficiency of minor actinide transmutation reactors, such as the accelerator-driven reactor, is not high enough to complete transmutation in a single burning cycle, a repetition of spent fuel reprocessing and transmutation—or the multiple recycling of minor actinides—is essential. Answer the following questions, requiring calculation of the amount of minor actinides transferring to wastes during multiple recycling.
- (a) Assume the transmutation efficiency in one burning cycle is  $\alpha$ . This means that  $\alpha$ , in tons, of minor actinides is transmuted from 1 ton of minor actinides loaded in the transmutation reactor. Then assume  $\beta$  is the ratio of transferring of spent minor actinides discharged from the transmutation reactor to waste through dry reprocessing. Calculate the total amount of minor actinides transferring to waste when 1 ton of minor actinides is transmuted.
  - (b) The value of  $\alpha$  in a typical accelerator-driven reactor is 10 %. Calculate the migration rate  $\beta$  needed to limit the amount of minor actinides transferring to waste to 1 % or less of the amount before transmutation.

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