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World-wide trend of long-lived radionuclides transmutation studies

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Abstract

The objective of this study is to review the concepts of partitioning and transmutation studies which are being conducted in several countries. This review was focused on the analysis of such areas as radiotoxicities of radwastes containing long-lived radionuclides, transmututon by reactors or accelerators, and separation of minor actinides. The world-wide trend of partitioning and transmutation studies was also investigated on the basis of each country's R & D activities in this area.

Introduction

Radioactive wastes containing long-lived radionuclides should be monitored even after being disposed of in underground repositories because their radioactivity is maintained for several hundred thousand years. According to the present scheme of high-level wastes management in a few countries, they will be disposed of in deep underground as a vitrified form after their radioactivity is considerably reduced by storing them for several decades. However, they would still have some possibility of being released into the biosphere due to the migration of radionuclides through underground media. Although there are some ways to install artificial barriers in order to block this migration, it would still raise concerns whether the radioactive materials could be safely encapsulated or not at the time of potential earthquakes or deterioration of the structural materials. In this regard, it may be a

long-term solution to separate long-lived radionuclides from wastes and then to transmute them into short-lived or stable nuclides by means of nuclear reactors and/or accelerators.

To achieve this goal of transmutation, intensive studies for the development of partitioning and transmutation technologies are being performed in a number of research institutes in the world. This study was aimed at reviewing the concepts of partitioning and transmutation studies as well as investigating the world-wide trend of R & D activities for the technology development.

Potential hazard of high-level waste

The change in the hazard index (HI_{ALI}) of high-level waste with time is shown in Fig.1. This hazard index was calculated on the basis of annual limit of intake(ALI) proposed by the ICRP with the high-level waste per ton of spent fuel (3.3% EU, thermal power : 30MW/MT, burnup : 33000MWD/MT). The HI_{ALI} is defined by

$$HI_{ALI} = \frac{\textit{Radioactivity of nuclide } i \textit{ (Bq)}}{\textit{ALI of nuclide } i \textit{ (Bq)}} \qquad (1)$$

As illustrated in Fig.1, strontium and cesium dominate the hazard for the first sereral hundred years, and curium is the next contributing element. Between the time of one thousand and one hundred thousand years, americium and plutonium are found to be the most hazardous elements. After 10^5 years, neptunium is then the major element exibiting a high HI_{ALI} value, while ^{229}Th and ^{225}Ra, which are radionuclides newly generated by radioactive decay, also show appreciable HI_{ALI} values. ^{99}Tc and ^{129}I show nearly constant values of HI_{ALI} until 10^5 years and 10^7 years, respectively. These are the major radionuclides among fission products in the evaluation of environmental impact on the biosphere because their radiotoxicity is maintained for a significantly long time and, moreover, their underground mobility is relatively larger than that of other elements.

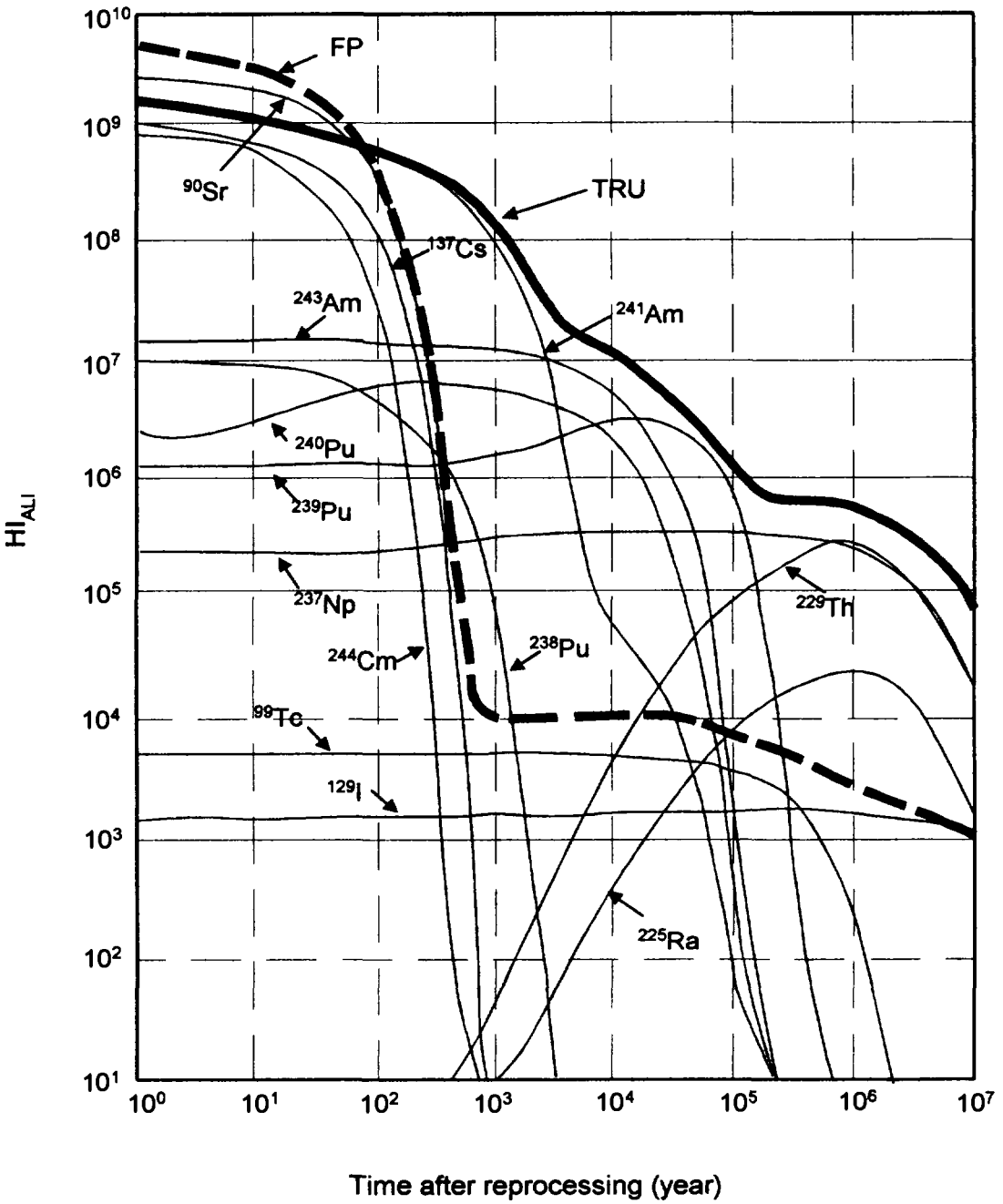

Fig. 1. Variation of HI_{ALI} with Time for Various Radionuclides in the High-level Waste

(PWR Spent Fuel ; 33,000 MWD/MT , 5yr Cooling)

Transmutation by nuclear reactors

1. An overview of minor actinides recycle into nuclear reactors

Minor actinides are transmuted in nuclear reactors mainly by nuclear fission or neutron absorption. Higher order actinides are generated in neutron absorption, generally increasing the radiotoxicity, which means that the reactor for transmutation should be optimized to have lower values of capture/fission. Table 1 gives the values of the capture cross section (σ_c), fission cross section (σ_f) and ratio, σ_c / σ_f , in a typical pressurized water reactor(PWR) and a fast neutron reactor(FNR). The ratios of σ_c / σ_f for various minor actinides, are found to be larger in the PWR than those in the FNR, indicating that net transmutation rates for the minor actinides will be higher in the FNR.

Table 1. Average Cross-Sections in a Typical PWR and FNR

(unit : barn)

Nuclide	PWR			FNR		
	σ_f	σ_c	σ_c / σ_f	σ_f	σ_c	σ_c / σ_f
^{237}Np	0.52	33	63	0.32	1.7	5.3
^{238}Np	134	13.6	0.1	3.6	0.2	0.05
^{238}Pu	2.4	27.7	12	1.1	0.58	0.53
^{241}Am	1.1	110	100	0.27	2.0	7.4
^{242}Am	159	301	1.9	3.2	0.6	0.19
^{242m}Am	595	137	0.23	3.3	0.6	0.18
^{243}Am	0.44	49	111	0.21	1.8	0.57
^{242}Cm	1.14	4.5	3.9	0.58	1.0	1.7
^{243}Cm	88	14	0.16	7.2	1.0	0.14
^{244}Cm	1.0	16	16	0.42	0.6	1.4
^{245}Cm	116	17	0.15	5.1	0.9	0.18
^{235}U	38.8	8.7	0.22	1.98	0.57	0.29
^{239}Pu	102	58.7	0.58	1.86	0.56	0.3

Table 2 shows the comparison of the effects of minor actinides recycled in the typical PWR and FNR fuel cycles. When 1% neptunium is recycled to the PWR fuel, a considerable increase in radioactivity is estimated at the beginning of the irradiation cycle. In the case of 1% americium, the effect is much greater, both at the beginning and end of the cycle. This means that homogeneous recycling of minor actinides to the PWR fuel is very unfavorable at the time of fuel manufacture, as well as fuel handling.

Recycling of 2.5% neptunium in the FNR fuel seems to have no significant effect on the fuel cycle, except to increase the dose rate at fabrication time due to the presence of ^{233}Pa ($T_{1/2}$ =27d ; β^-), which is in equilibrium with ^{237}Np. After irradiation in the FNR and 5 years of cooling, a limited increase in γ-activity and decay heat can be observed, meaning that homogeneous recycling of neptunium with a content of 2.5% does not cause any problem in the fuel cycle.

Recycling of Am, however, has a more remarkabe impact on the fuel cycle. At the beginning of the irradiation cycle, the intensity of γ-activity is 4.5 times greater compared with that of a normal fuel. After irradiation and 5 years of cooling, the intensities of the other sources are about 3 times greater, indicating that the homogeneous recycling of americium is very unfavorable. Thus, in the case of americium, a heterogeneous mode is recommended over a homogeneous mode.

In the case of curium, however, homogeneous recycling into the FNR fuel would be impossible in view of its effect on the radioactivity of fuel both at the beginning and end of the irradiation cycle. Therefore, for curium, long-term storage is a feasible management strategy because the half-life of ^{242}Cm is not so long and, moreover, ^{242}Cm decays into ^{238}Pu which can be recycled later.

As a result, it is recommended that neptunium be recycled into an FNR in a homogeneous mode whereas americium should be recycled in a heterogeneous mode. In order to follow this recommendation, neptunium, americium, and curium should be individually separated from one another, because each element must be directed to a different place. This will require an additional separation cost.

Table 2. Effects of Minor Actinides Recycle on the Fuel Cycle

		PWR		FNR		
Fuel material		UO_2		$(U\ Pu)O_2$		
MA addition		1% Np	1% Am	2.5% Np	2.5% Am	2.5% Cm
Beginning of cycle	α	×18	×58000	×1	+71%	×23
	β	×11	×3000	×1	×1	×1
	γ	×68	×80000	×1	×4.5	×15
	n	+16%	×1600	×1	+40%	×1600
	h	×19	×65000	×1	+70%	×24
End of cycle + 5 years	α	×5	×14		×3.0	×4.9
	β	+10%	+9%		−1%	+3.7%
	γ			+49%	×2.8	×3.2
	n	−30%	×28	−6.4%	×3.5	×82
	h	×5.3	×15	+69%	×3.0	×5.0

h : decay heat

2. Homogeneous recycle of neptunium

In the homogeneous mode, fuel material and long-lived nuclides are homogeneously mixed in the fuel fabrication process. This mode, however, has a few drawbacks in the fuel cycle. Firstly, it is impossible for the two components, namely, the fuel material and long-lived radionuclides, to have different irradiation periods in the reactors and thus both components must be recycled by reprocessing at each end of the irradiation cycle, whereas the target material, containing long-lived radionuclides, can be irradiated for more than one life cycle without reprocessing in the heterogeneous mode. Secondly, the fuel material always accompanies long-lived radionuclides in reprocessing, making itself more radioactive due to the radioactivities of long-lived radionuclides.

In the EFR (European Fast Reactor) type, various physical parameters of the core are estimated to change with the initial ^{237}Np content. The

following changes are estimated to occur in the core when ^{237}Np is introduced to the fuel instead of ^{238}U :

- Decrease in the initial core reactivity by the high neutron capture rate for ^{237}Np
- Decrease in the reactivity loss over the cycle by the transformation of ^{237}Np into the more fissile isotope, ^{238}Pu
- Decrease in the effective beta value due to the lower delayed neutron yield for ^{237}Np compared to ^{238}U
- Significant drop in the Doppler effect due to the hardening of the neutron spectrum
- Significant increase in the reactivity effect by sodium voiding because the neutron captures by ^{237}Np tends to amplify the variation of absorption during the voiding
- Increase in the ^{238}Pu generation with increased initial Np content

As a result, these effects limit the initial content of Np in the fuel. However, an introduction of a moderator to the core can increase the Doppler effect while reducing the voiding effect. In addition, the content of ^{238}Pu should be limited by less than 5% to reduce the impact of its radioactivity in the reprocessing, e.g., solvent radiolysis. This is also one of the factors that limit the initial content of neptunium. Consequently, the neptunium content is recommended to be limited by less than 2.5% in the EFR type core.

3. Heterogeneous recycle of americium

Long-lived radionuclides are made into separate targets other than the nuclear fuel assemblies in this mode. This would enhance the transmutation rate of the target material by irradiating them longer than the fuel material and also make it possible to have separate chains of reprocessing and fabrication for targets and fuel assemblies. However, it might require more partitioning cost in order to separate a certain pure component for target material. Therefore the selection of one of the two modes depends on the behavior of the nuclides in the core, as well as their impact on the fuel cycle.

Loading 100% Am in the blanket causes a significant void effect and also raises the generation rates of ^{238}Pu and ^{244}Cm. However, heterogeneous loading in the first radial blanket ring of the EFR is estimated to have little effect on the core at concentrations up to 50%.

4. A model for transmutation of long-lived radionuclide in the EFR

A model was suggested by scientists in the European commission for the transmutation of long-lived minor actinides and fission products in the EFR. The transmutation model, as well as the anticipated results, are summarised below.

Reactor capacity : 1500 MWe
Irradiation cycle : 1500 ENPD
Fuel : MOX (45% Pu)
Np loading : Homogeneous
5% Np in core
moderated by B_4C
Am loading : Heterogeneous
AmO_2 + MgO target
40% Am in radial blanket
Tc loading : Tc metal mixed with CaH_2
10% Tc
Lower axial blanket
I loading : CeI_3 mixed with CaH_2
20% I
Upper axial blanket
Np transmutation : 67% (1500 ENPD)
Am transmutation : 64% (3000 ENPD)
Tc transmutation : 19% (1500 ENPD)
I transmutation : 16% (1500 ENPD)

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* ENPD : Equivalent Nominal Power Day

45% of the plutonium content in the fuel was considered because it can be managed in the fuel cycle without any further advancement in the present technology using the maximum amount of plutonium possible. The use of B_4C as a moderating material is taken into account to overcome the impacts of the Doppler effect as well as the voiding reactivity. In the case of ^{237}Np, 67% of initial neptunium was calculated to be transmuted in the presence of the moderator, whereas 48% was obtained in the absence of the moderator.

The reason why the fission products are mixed with calcium hydride(CaH_2),

is so they can work as a moderator, shifting the neutron spectrum towards lower energies and provide more favorable conditions for the incineration of tecnetium and iodine. As for ^{99}Tc and ^{129}I, when designed to be heterogeneously recycled to the axial blankets, the net quantities transmuted were calculated to be 2.9 kg and 1.6 kg per Twh, respectively.

Accelerator-driven transmutation systems

Since transmutation rates tend to be relatively low or limited in nuclear reactors, multiple recycling is needed to raise the transmutation rates of a certain radionuclide. This would require a high reprocessing cost as well as a very long irradiation period to get a complete incineration. A hybrid system of a proton accelerator-subcritical reactor is therefore being considered as an alternative transmutation system. Several concepts of transmutation using proton accelerators are being studied in a few countries. The accelerator-driven transmutation concepts, dependant on the neutron energy spectrum, fuel form (solid or liquid), fuel cycle, coolant/moderator type, and accelerator type, are listed in Table 3.

Some scientists in LANL claim that the ADTT system will be able to eliminate the world inventory of reactor and excess-weapon plutonium in 30 years, as shown in Fig.2. This figure shows that the amount of reactor grade plutonium will reach about 2000 tons by the year 2010 in the world if no more new reactor constructions are made. The ratio of the number of ADTT units to that of existing LWRs should be about 1 : 6 in order to transmute all the plutonium during the time span of 100 years. Minor actinides and long-lived fission products can be also destructed together with the plutonium in the ADTT system.

If thorium is used in the ADTT system as a fertile fuel, nuclear energy can be generated without producing higher actinides waste. Therefore it would have a great advantage in terms of waste management compared with conventional reactors because it would reduce the requirements for long-lived waste treatment. A subcritical reactor system, for instance, would start with ^{232}Th and then convert it into the fissile fuel ^{233}U by neutron captures. This system would be composed of a proton accelerator, subcritical reactor, and fuel recycle system, etc., as shown in Fig.3. Unlike conventional power reactors, there would be no self-sustained chain reaction in this system because neutron loss is so great in the core due to transmutation. Part of

Table3. Various Concepts of Accelerator-driven Transmutation Systems

Organization	Program	Concept
LANL	ADTT	① Energy production - Th/U fuel - Molten salt - Thermal neutron - Linac driven ② Plutonium burning - U/Pu fuel - Molten salt - Thermal neutron - Linac driven
BNL	Phoenix	① MA Incineration - U/Pu fuel (solid) - Na or Pb/He cooled - Fast neutron - Linac driven
	APT	② Pu/MA/FP incineration - U/Pu C pebbles - He/D_2O cooled - Thermal neutron - Linac driven
JAERI	OMEGA	① MA incineration -U/pu fuel -Molten chlorides -Fast neutron -Linac driven ② MA incineration -U/Pu fuel -Na or Pb/He cooled -Fast neutron -Linac driven
ITEP	Weapon Pu burning	① Pu burning -Pu fuel -D_2O cooled -Thermal neutron -Linac driven ② MA/FP incineration -U/Pu D_2O solution -Thermal neutron -Linac driven

the neutrons would make up for the loss from an external neutron source, that is, a spallation target which produces neutrons by the proton beam coming from the accelerator.

The General Atomics Corporation suggested another type of plutonium incineration system : a helium-cooled, graphite-moderated reactor with ^{239}Pu particles suspended in graphite, operated to destruct ^{239}Pu during the first stage and then transfer the fuel to a subcritical reactor where the plutonium destruction continues by spallation neutrons.

Other nuclear fuels in the ADTT program considered are a molten salt type liquid fuel as well as on-line recycling of long-lived radionuclides. Such concepts might contribute to achieving an economical transmutation system .

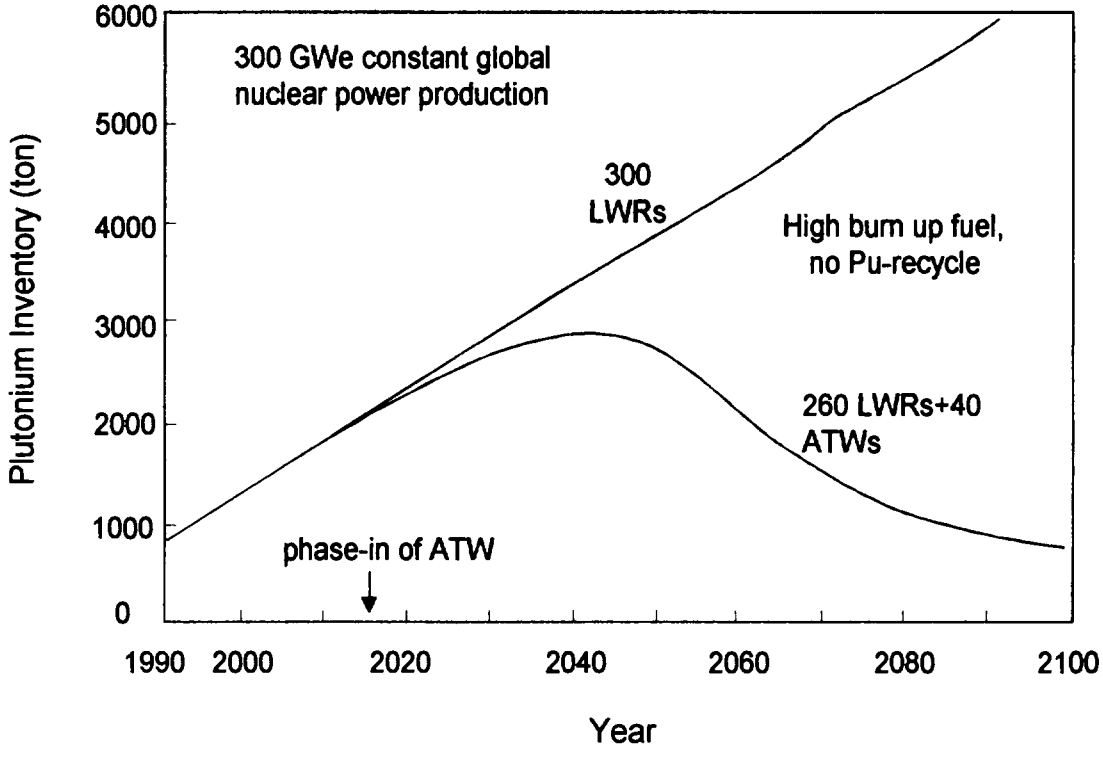

Fig. 2. Reduction of World-wide Plutonium Inventory When ATW is Implemented

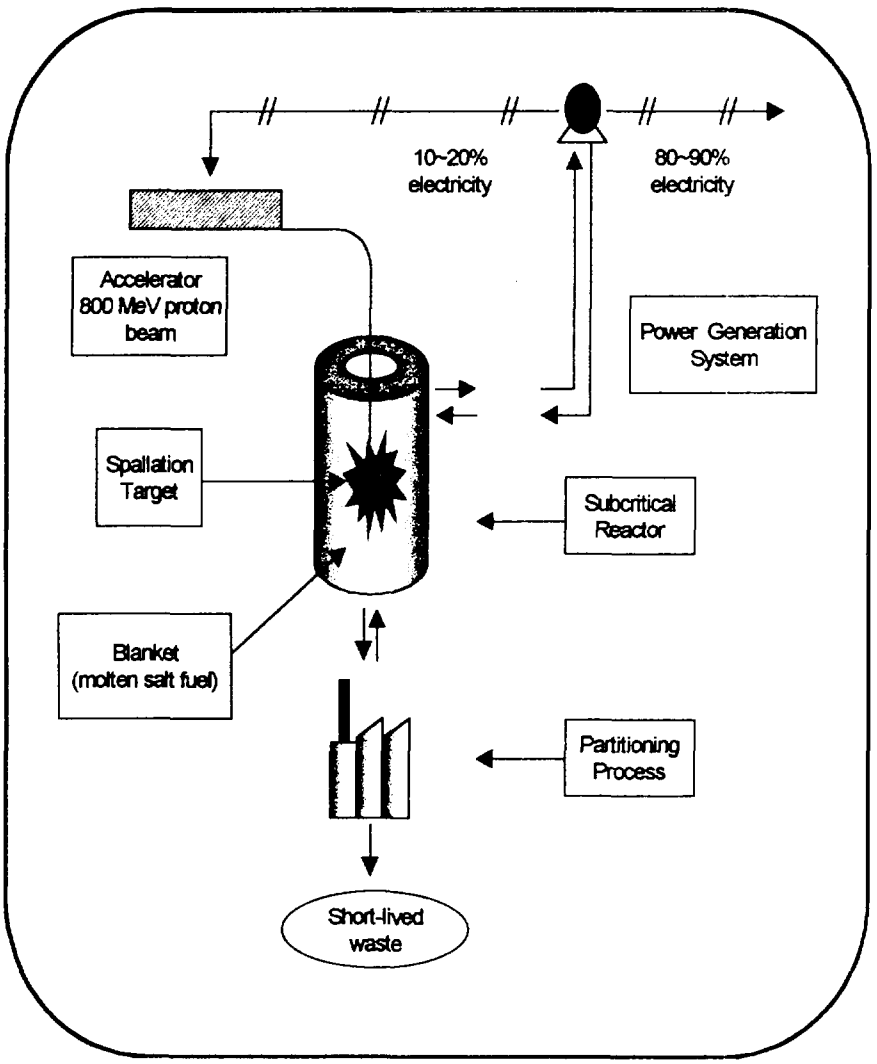

Fig. 3. Accelerator-driven Transmutation of Waste

Development of partitioning technology

Partitioning is the first step to be carried out for the transmutation of long-lived radionuclides. Development of partitioning technology depends on the grouping of various radionuclides contained in wastes. The radionuclides in a typical high-level waste are usually classified into such groups as minor actinides, Cs / Sr, platinum metals, and other residual elements on the basis of their chemical and radioactive properties. The groups are then partitioned from one another.

Up to now, several partitioning processes have been tested or are under development in a number of countries. The partitioning processes which have been or are being studied are listed with their characteristics in Table 4. Among these processes, none has been in commercial use yet. Selection of a suitable process, in each country, should be dependent on its transmutation strategy as well as fuel cycle strategy. In some countries, the modification of existing reprocessing processes is being considered to recover long-lived radionuclides such as ^{237}Np, ^{99}Tc, and ^{129}I. Once the process is renovated, these radionuclides would be recovered in the main stream of the reprocessing process in advance, without treatment of the high-level wastes. However, an additional cost would be needed for the separation of the radionuclides.

If the long-lived radionuclides are not recovered in the reprocessing, then they should be recovered by treatment of the high-level waste in a separate partitioning process. Among the minor actinides, both americium and curium are always recovered together in a mixture state during all the existing processes. However, they should be separated from each other to recycle americium only as a transmutation target and to store the curium without recycling to the reactor, as described in a previous part of this paper. To develop a separation method for this, a new technology is being studied in CEA; that is, americium is oxidized electrochemically from tri-valent to hexa-valent state, and then separated from curium by selective extraction of Am(VI) with an alkyl phosphoric acid. The americium is finally stripped from the solvent by use of a reducing agent. Dilute hydrogen peroxide, for instance, can be employed as a kind of reducing agent.

Recently, a pyrochemical process is being developed in a few countries as an alternative reprocessing method because it is known as a kind of nuclear proliferation-resistant technology and also as a more economical process than the existing wet processes. This method is, however, handled at high temperatures, thus having a more severe effect on the corrosion of equipment

materials. The treatment of secondary wastes is also a problem with this process.

Table 4. Various Partitioning Processes

Process	Research Institute	Present Status	Process Concept
TRUEX	ANL (USA)	Used for military wastes treatment	Coextraction of Am,Cm and RE with CMPO
CTH	Charmers Univ. (Sweden)	Studied in 1980s	Coextraction of Am,Cm and RE with HDEHP Selective stripping of Am and Cm with DTPA
DIAMEX	CEA (France)	Under development (SPIN program)	Coextraction of Am, Cm and RE with Diamide Selective stripping of RE with TPTZ
DIDPA	JAERI (Japan)	Under development (OMEGA program)	Coextraction of MA and RE with DIDPA Selective stripping of Am and Cm with DTPA
TRPO	Chungwha Univ. (China)	Under development	Coexraction of MA and RE with TRPO Selective extraction of MA with Cyanex 301
Pyroprocess	CRIEPI (Japan) ANL (USA)	Under development R & D stopped	Electrorefining in chloride medium Recovery of U, Pu and MA together
Pyroprocess	LANL (USA) Kurchatov(Russia) NRI (Czech)	Under development '' ''	Electrowinning in fluoride medium Recovery of transuranics and lanthanides together Recovery of uranium and zirconium together

Conclusion

A number of transmutation concepts, that is, transmutation by reactors, via accelerators, the applicability of various kinds of fuel types, and loading in the homogeneous or heterogeneous mode, etc., are currently being studied around the world. Although at present it is hard to determine the best way to incinerate long-lived radionuclides, in the future it will be easier to set up an effective strategy for partitioning and transmutation because relevant technologies are currently being developed in many countries. In each country, the appropriate strategy for partitioning and transmutation should be drawn out according to the country's individual nuclear industry situation, with regards to the power reactors it has, the presence or absence of nuclear fuel recycle facilities, as well as other factors.

List of abbreviations

ADTT : Accelerator-Driven Transmutation Technology
ALI : Annual Limits of Intake
ANL : Argon National Laboratory (USA)
APT : Accelerrator Production of Tritium
BNL : Brookhaven National Laboratory (USA)
CEA : Commissariat a l'Energie Atomique (France)
CMPO : Carbamoyl Phosphin Oxide
CRIEPI : Central Research Institute of Electric Power Industry (Japan)
DIAMEX : Diamide Extraction
DIDPA : Di-iso Decyl Phosphoric Acid
DTPA : Di-ethylene Tri-amine Penta Acetic acid
EFR : European Fast Reactor
ENPD : Equivalent Nominal Power Day
FNR : Fast Neutron Reactor
HDEHP : Di (2- Ethyl) Hexyl Phosphoric Acid
HI_{ALI} : Hazard Index based on ALI
ICRP : International Commission on Radiological Protection
ITEP : Institute of Teoretical and Experimental Physics (Russia)
JAERI : Japan Atomic Energy Research Institute (Japan)
LANL : Los Alamos National Laboratory (USA)
MA : Minor Actinide
MT : Metric Ton
MWD : Mega Watt Day
MOX : Mixed Oxide
NRI : National Research Institute (Czech)
OMEGA : Options Making Extra Gain from Actinides

PWR : Pressurised-Water Reactor
RE : Rare Earth
SPIN : Separation-Incineration
TPTZ : Tri-Pyridyl Tri-azine
TRPO : Tri-alkyl Phosphin Oxide
TRUEX : Transuranium Extraction
Twh : Tera Watt Hour

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