Proc. Conf. Advances in Radioactive Isotope Science (ARIS2014)
JPS Conf. Proc. **6**, 010004 (2015)
http://dx.doi.org/10.7566/JPSCP.6.010004

Transmutation of Long-lived Nuclear Wastes

Hiroyuki OIGAWA

Office of Strategic Planning, Japan Atomic Energy Agency,
2-2-2 Uchisaiwai-cho, Chiyoda-ku, Tokyo 100-8577, Japan

E-mail: oigawa.hiroyuki@jaea.go.jp

(Received October 23, 2014)

JAEA is conducting research and development on an Accelerator Driven System (ADS), aiming at reduction of burden for high-level radioactive wastes. To tackle technical challenges on ADS, JAEA is planning to build the Transmutation Experimental Facility as the Phase-2 program of J-PARC. Moreover, JAEA is considering the collaboration with the MYRRHA project proposed by Belgian Nuclear Research Center.

KEYWORDS: transmutation, nuclear waste, long-lived nuclide, minor actinide, fission product, accelerator driven system, ADS, J-PARC, spallation target, lead bismuth eutectic, subcritical core, proton beam

1. Introduction

Management of radioactive waste is one of the most critical issues to use nuclear energy. In Japan, spent nuclear fuel is to be reprocessed, and uranium and plutonium are to be recovered as resource. Remaining elements such as fission products and minor actinides in spent nuclear fuel are to be disposed of as high level waste (HLW). It has been, however, unsuccessful to find candidate sites for HLW disposal so far in Japan. When we turn our eyes to the world, only two counties, Finland and Sweden, seem successful to find the disposal site, where these two countries selected the direct disposal of spent fuel without reprocessing.

The difficulty in the HLW disposal is mainly caused by its long-lasting effect to human body. The HLW needs to be isolated from human activity area for a long period of time such as exceeding ten thousand years. This fact largely affects the public acceptance of HLW to their local area. If the long-lasting effect of HLW can be largely reduced, the burden to dispose of it would be also reduced. This is a motivation of the transmutation technology.

The Japan Atomic Energy Agency (JAEA) has been conducting the research and development of the transmutation technology. There are two major concepts; one is to use a commercial fast breeder reactor (FBR) and the other is to use an accelerator-driven system (ADS) as a dedicated transmutation system. Based on recent situation around the nuclear power in Japan, the transmutation technology using ADS has gotten a lot of attention.

In this paper, the impact and the present status of the transmutation technology using ADS are described.

2. Basic Idea for Transmutation

2.1 What to be Transmuted

The composition of the spent nuclear fuel is roughly 94% of uranium, 1% of plutonium, 0.1% of minor actinides (MAs) such as neptunium, americium and curium, and about 5% of fission products (FPs). Among FPs, the long-lived ones whose half-lives exceed 100 years are less than 10%. As shown in Table I, each nuclide has different dose coefficient which represents the magnitude of radiation effect to the human body. In general, values of the dose coefficients are large for plutonium and MAs in comparison with those for FPs because α-emitting nuclides have much larger effect to human organ in the case of intake. After the reprocessing, MAs and FPs are the main components of HLW, and its long-lasting potential influence to human body, which is called "radiological toxicity", is dominated by MAs. The first priority to transmute is, therefore, MA. Some of long-lived FPs (LLFPs) are also important because they are much more soluble to the ground water than MA. These LLFPs show the major contribution to the safety analysis of the repository site based on the ground water scenario.

Table I. Major long-lived nuclides in spent nuclear fuel.

Nuclide		Half-life (year)	Dose coefficient (μSv/kBq)	Mass (per 1tHM)
Uranium	^{235}U	7×10^{8}	47	10 kg
	^{238}U	4.5×10^{9}	45	930 kg
Plutonium	^{238}Pu	87.7	230	0.3 kg
	^{239}Pu	24,000	250	6 kg
	^{240}Pu	6,600	250	3 kg
	^{241}Pu	14.3	4.8	1 kg
Minor actinide (MA)	^{237}Np	2.1×10^{6}	110	0.6 kg
	^{241}Am	432	200	0.4 kg
	^{243}Am	7,400	200	0.2 kg
	^{244}Cm	18.1	120	60 g
Fission product (FP)	^{79}Se	3×10^{5}	2.9	6 g
	^{90}Sr	28.8	28	0.6 kg
	^{93}Zr	1.5×10^{6}	1.1	1 kg
	^{99}Tc	2.1×10^{5}	0.64	1 kg
	^{107}Pd	6.5×10^{6}	0.037	0.3 kg
	^{126}Sn	2.3×10^{5}	4.7	30 g
	^{129}I	1.6×10^{7}	110	0.2 kg
	^{135}Cs	2.3×10^{6}	2.0	0.5 kg
	^{137}Cs	30.1	13	1.5 kg

2.2 Combination of Partitioning and Transmutation

To achieve transmutation of MAs and FPs, they should be separated from HLW. Moreover, if we can separate elements in HLW into several groups, it may be possible to make use of valuable elements and to reduce the volume of HLW. This scheme is called "partitioning" as shown in Fig. 1, where HLW is separated into four groups; (1) MAs, (2) platinum group metals (PGMs) such as ruthenium, rhodium and palladium, (3) heat generators such as strontium and cesium, and (4) remaining elements. The PGMs can be utilized as catalyst. The heat generators can be used as radiation source and heat source, and can be disposed of in a compact manner after long-term cooling such as 300 years. Since the remaining elements do not contain large amount of heat generating nuclides nor long-lived α-emitting nuclides, they can be disposed of in a reasonable

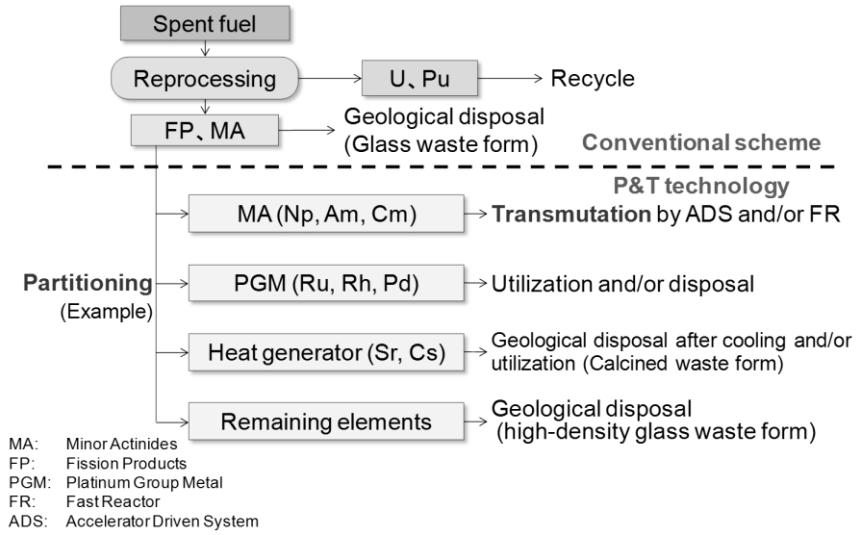


Fig. 1. Scheme of partitioning and transmutation technology.

manner; for example, the area of the disposal site can be reduced by a factor of 100.[1] It should be, however, noted that the remaining elements still contain LLFPs such as ^{93}Zr and ^{126}Sn, and hence the deep geological disposal needs to isolate these LLFPs from biosphere. To avoid the deep geological disposal, further partitioning of LLFPs and their transmutation into short-lived or stable ones are required, which seems very tough challenge from viewpoints of energy consumption and cost.

2.3 Impact of MA Transmutation

As described already, MAs are major contributors to the radiological toxicity of HLW. Figure 2 compares the time dependency of the radiological toxicity of spent nuclear fuel itself, HLW and the case of MA transmutation. The level of the radiological toxicity of natural uranium (9 ton) used as the raw material of the low-enriched uranium fuel (1 ton) is also shown in the figure. The radiological toxicity of the spent fuel decreases very slowly and becomes smaller than the level of the natural uranium after about 100,000 years. In the case of HLW, this time period is several thousand years, and the transmutation of MA further shortens the time period down to about 300 years.

2.4 Methods of Transmutation

To transmute nuclide, it is effective to use neutrons because neutron can easily convey energy to the nuclides for transmutation. The transmutation rate can be described as

$$R_A = N\sigma_A\varphi \ , \tag{1}$$

where R_A: reaction rate of reaction A ($cm^{-3}s^{-1}$),

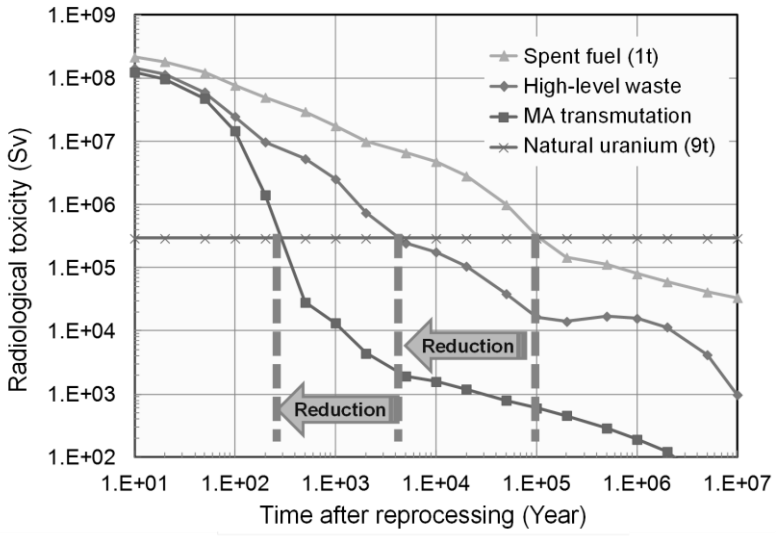


Fig. 2. Reduction of radiological toxicity by transmutation of minor actinide.

N: atomic number density (cm^{-3}),
σ_A: cross section of reaction A (cm^2 or barn=$10^{-24}cm^2$), and
φ: neutron flux ($cm^{-2}s^{-1}$).

If the other reactions and natural decay of nuclide can be neglected, the time-dependent atomic number density is simply described as

$$N(t) = N_0 e^{-\sigma_A \varphi t} \ , \qquad (2)$$

where N_0: initial atomic number density (cm^{-3}).

Then the reduced half-life of this nuclide by the reaction A can be described as:

$$\hat{T}_{A\,1/2} = {}^{\ln 2}/_{\sigma_A \varphi} \ , \qquad (3)$$

where $\hat{T}_{A\,1/2}$: reduced half life of the nuclide transmuted by reaction A.

Table II shows the reduced half lives of some long-lived nuclides for some reactions. The nuclear data file JENDL-4.0 [2] was used for the calculation.

^{241}Am can be transmuted by threshold fission reaction induced by fast neurons (>1MeV). The cross section of this reaction is in the order of 1 barn, and hence if we can provide fast neutron irradiation field of 10^{15} n/cm^2/s, the half life of this nuclide can be reduced to about 16 years. This level of the neutron flux is considered achievable by a fast spectrum fission reactor (FR) and an accelerator driven system (ADS). In the case of ^{99}Tc and ^{129}I, the transmutation by a capture reaction of thermal neutrons seems applicable because usual light water reactors have the neutron flux of about 10^{13} n/cm^2/s.

Table II. Examples of "Reduced Half-life".

Nuclide	Original half-life (year)	Reaction	Neutron energy	Cross section (JENDL-4.0) (barn)	Neutron flux (n/cm^2/s) 10^{13} Reduced half-life (year)	10^{14}	10^{15}
^{241}Am	432	Fission	Fission spectrum	1.378	1,600	160	16
^{99}Tc	2.1x10^5	Capture	Thermal	23.68	93	9.3	0.93
		n,2n	14 MeV	1.233	1,800	180	18
^{126}Sn	2.3x10^5	Capture	Thermal	0.09	24,000	2,400	240
		n,2n	14 MeV	1.686	1300	130	13
^{129}I	1.6x10^7	Capture	Thermal	30.33	72	7.2	0.72
		n,2n	14 MeV	1.464	1,500	150	15
^{135}Cs	2.3x10^6	Capture	Thermal	8.304	260	26	2.6
		n,2n	14 MeV	1.61	1,400	140	14
^{137}Cs	30.1	Capture	Thermal	0.27	8,100	810	81
		n,2n	14 MeV	1.549	1,400	140	14

The (n, 2n) reaction is another candidate to transmute nuclides. The value of cross section is almost the same among the nuclides, 1-2 barn by 14MeV neutrons, and it can be seen that the neutron source that can provide neutron flux of 10^{15}n/cm^2/s is necessary to achieve meaningful transmutation. The technical feasibility of such a strong neutron source, however, has not been verified yet.

Hereinafter, this paper discuss the present status of technical development of MA transmutation by ADS.

3. Accelerator Driven System for MA Transmutation

3.1 Principle of Accelerator Driven System (ADS)

An accelerator driven system (ADS) is a subcritical nuclear reactor whose fission chain reactions are kept by an intense external neutron source driven by an accelerator. Figure 3 shows an example of ADS for MA transmutation. A super-conducting linear proton accelerator (LINAC) supplies a proton beam of maximum 30 MW. The proton energy will be selected by considering the efficiency of neutron production, technical feasibility and economy. Our tentative reference design selected 1.5 GeV (20 mA for 30 MW), which is rather high value in comparison with other designs because higher energy with lower current reduces the burden to the beam window. The proton beam is

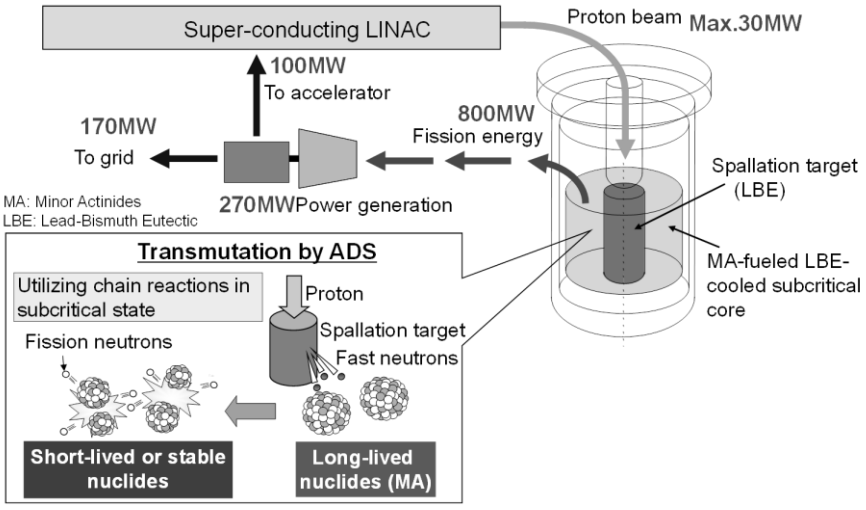


Fig. 3. Accelerator Driven System (ADS) for MA Transmutation.

bombarded into a spallation target made of liquid heavy metal and produces a number of fast neutrons. The first candidate of the spallatioin target is lead-bismuth eutectic (Pb-Bi, or LBE). MA-bearing fuel is placed around the spallation target, and the fast neutron induces fission reactions of MAs. A fission reaction of MA also produces a few neutrons, which are also used for MA transmutation. This is a kind of chain reactions, but it can be stopped by stopping the accelerator because the system is kept subcritical.

A fission reaction also produces energy of about 200 MeV. This ADS system driven by a maximum 30 MW proton beam can produce the thermal power of 800 MW. This energy is removed from the MA-bearing fuel to the coolant, and then converted to electricity of 270 MW, 100 MW of which is used for the operation of the accelerator, and the other 170 MW can be sold. Our first candidate of the coolant is also LBE. Water cannot be used as the coolant because it moderates the neutron energy which should be kept enough high to achieve efficient transmutation of MA.

A "critical" fast reactor (FR) also can supply a number of fast neutrons, and is considered as another candidate of MA transmutation. Large difference between FR and ADS is in the fuel composition that is acceptable to the system. From a safety point of view, FR can accept maximum 5% MA in the fuel, while MA fraction of ADS fuel is about 60 %. The fuel composition of FR is roughly 20 % of plutonium and 80 % of uranium. These major actinides produce another MAs, and hence the transmutation rate per unit power becomes much less than ADS; the transmutation rate of MA by FR is about 60 kg/GWth/y, while that by ADS is about 300 kg/GWth/y.[3] This means 5 times larger power is necessary for FR than ADS to transmute same amout of MA. On the contrary, the technology of FR is almost proven, though that of ADS is to be demonstrated in the coming decade or two.

3.2 Research and Development of ADS

To achieve ADS for MA transmutation, lots of technical challenges should be solved.

A high power proton accelerator is one of the key technologies for ADS. It should be reliable enough to drive a nuclear system. The trip rate should be reduced as low as possible, for example once a week.

The spallation target technology has been demonstrated by the international collaborative project MEGAPIE; an LBE target was installed into SINQ target of the Paul Schelere Institute of Switzerland and operated successfully at about 700 kW for 4 months in 2006.[4] Still more than one order gap is, however, existing between the current technology and the future 30 MW target. Especially the material for a beam window is a key technology because it is exposed to heavy irradiation of protons and neutrons, thermal stress, erosion and corrosion by LBE flow, and high pressure (about 1 MPa) of LBE. The beam window is, hence, exchanged periodically, but its expected lifetime is not clearly demonstrated.

It is also important to control the subcritical state of ADS. We do not have experience to operate such a high-power subcritical system nor such a nuclear system with large amount of MA. The nuclear data of MA do not have enough accuracy to predict evolution of fuel with good precision. The monitoring method of subcritical state is also to be developed and demonstrated.

The MA-bearing fuel is another technical challenge for ADS. Our first candidate is nitride; i.e., (MA, Pu)N diluted by ZrN. The nitrogen should be enriched by ^{15}N at about

99% to avoid production of ^{14}C which becomes long-lived waste. Since MA has high radiation and heat generation, such MA-bearing fuel should be fabricated by remote handling with cooling and shielding. After the irradiation of MA-bearing fuel, it should be reprocessed and re-fabricated to fresh fuel, i. e. fuel cycle, because only 20 % of MA will be transmuted by one irradiation for 2 years. We will use dry process for reprocessing of ADS spent fuel.

Finally, the separation process to recover MA from HLW is also to be developed and demonstrated.

3.3 Transmutation Experimental Facility of J-PARC

To overcome technical challenges for ADS, we have a plan to build the Transmutation Experimental Facility (TEF) as the Phase-2 of the J-PARC project.[5] As shown in Fig. 4, TEF consists of two facilities: one is the ADS Target Test Facility (TEF-T) and the other is the Transmutation Physics Experimental Facility (TEF-P). TEF-T is an irradiation facility with a 400 MeV – 250 kW proton beam. An LBE spallation target will be installed and material test for the beam window material will be mainly conducted. TEF-P is a critical / subcritical assembly to conduct reactor physics experiments for transmutation systems. A 400 MeV – 10 W proton beam can be introduced into the center of the assembly to simulate ADS driven by a spallation neutron source. Large amount of MA (order of 10 kg) will be available to simulate the MA transmutation system such as ADS and FR.

In parallel to TEF, we would like to collaborate with an ADS project in Belgium called MYRRHA.[6] MYRRHA is an LBE cooled subcritical / critical reactor with the thermal power of about 100 MW fuelled with U-Pu mixed oxide. The purposes of MYRRHA are to demonstrate engineering feasibility of an ADS and a heavy metal cooled FR, to provide a fast neutron irradiation field for material and fuel tests, to produce isotopes for medical use, and so on. As shown in Fig. 5, by combining elemental research and development at TEF and engineering demonstration at MYRRHA, we would like to realize ADS for MA transmutation around 2050.

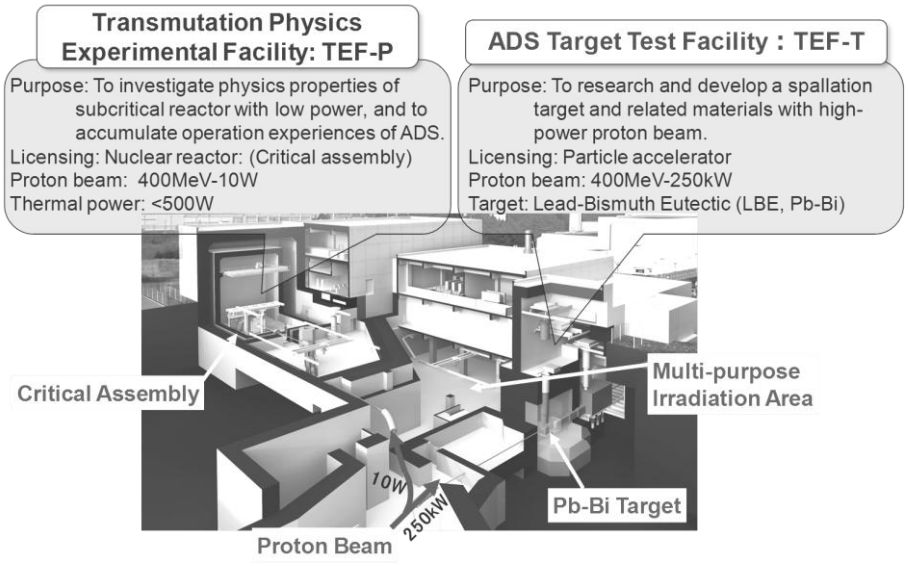
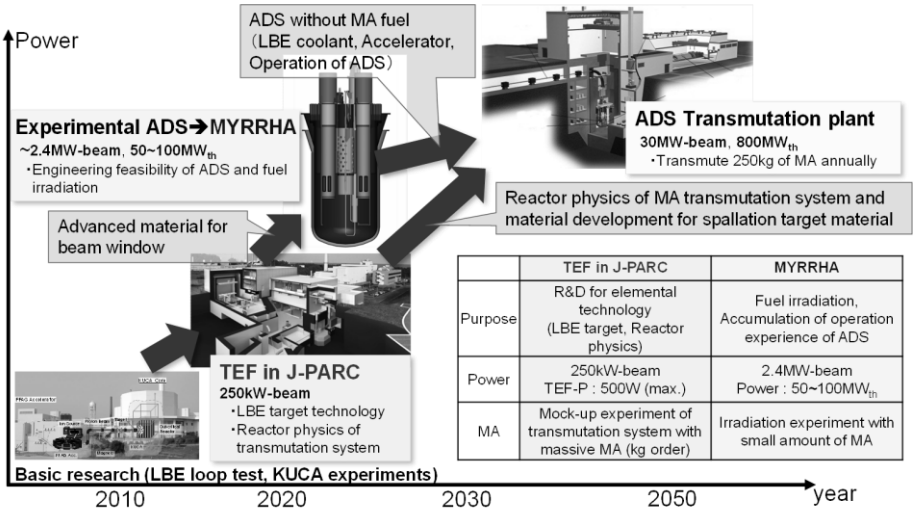


Fig. 4. Conceptual View of Transmutation Experimental Facility (TEF)



	TEF in J-PARC	MYRRHA
Purpose	R&D for elemental technology (LBE target, Reactor physics)	Fuel irradiation, Accumulation of operation experience of ADS
Power	250kW-beam TEF-P : 500W (max.)	2.4MW-beam Power : 50~100MWth
MA	Mock-up experiment of transmutation system with massive MA (kg order)	Irradiation experiment with small amount of MA

Fig. 5. International Collaboration with MYRRHA.

4. Conclusion

Although the transmutation technology is still in the fundamental stage, it has great potential to reduce the burden of radioactive waste management that is a globally common issue to utilize nuclear energy. It is, therefore, very important to tackle the technical challenges by international and interdisciplinary collaborations.

References

[1] K. Nishihara, S. Nakayama, Y. Morita, et al., J. Nucl. Sci. Technol., **45(1)** (2008) 84-97.
[2] K. Shibata, O. Iwamoto, T. Nakagawa, et al., J. Nucl. Sci. Technol., **48(1)** (2011) 1-30.
[3] OECD/NEA, "Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles: A Comparative Study" (2002).
[4] Ch. Latge, F. Groeschel, P. Agostini, et al., Proc. Ninth Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Nîmes, France, 25-29 September 2006, (2007) 407-419.
[5] http://j-parc.jp/index-e.html/
[6] http://myrrha.sckcen.be/