A Strategic Study of the Partitioning and Transmutation System being Developed at JAERI

H.YOSHIDA, M.KUBOTA, H.KATSUTA,
T.MUKAIYAMA and T.TAKIZUKA

Japan Atomic Energy Research Institute
Tokai-mura,Naka-gun, Ibaraki-ken, Japan

1. Introduction

The present HLW management is based on disposal HLW of in a deep geological formation after its solidification and cooling. The partitioning and transmutation (P-T) technology plays roles to mitigate the issues in the present HLW management,according to advance of the technology,such as a supporting technology by reduction of HLW volume and heat generation,a complemental technology by mitigation of natural barrier uncertainty,and a new technology different from the geological disposal.

Under the framework of OMEGA programme in Japan,The Japan Atomic Energy Research Institute(JAERI) has studied partitioning and transmutation (P-T) technologies which has a potential to provide a different HLW management from geological disposal. The technologies include a partitioning process to separate HLW into 4 elements groups together with minor-actinides group[1],and two different kinds of transmutation systems; minor-actinide burning fast reactor[2] and proton accelerator-based transmutation system [3,4],both of which have equivalent transmutation capability.

A preliminary strategic study has been carried out to investigate effectiveness of the above-mentioned P-T technologies to the HLW management. The study includes followings;

1) effect of long-lived nuclides separation to radioactive toxicity of HLW and,
2) effect of long-lived nuclides transmutation to their accumulation.

2. Effectiveness of Our Proposed Partitioning Technology

Target separation efficiency was preliminarily defined for long-lived minor actinides and fission products in HLW,comparing their radioactive toxicities with that of uranium ore. Based on these preliminarily defined separation efficiencies,separation capability of our proposed partitioning technology is discussed.

2.1 Priority of Long-lived Nuclides to be separated from HLW

A classification of TRU and fission-products on their half lives is given in Table 1. The composition of fission products in the table was calculated for an U-loaded 1000MWe PWR (unloading burnup:33000MWD/MT,cooling time before reprocessing:150 days) by the point-burnup calculation code SRAC-FPGS[5] with the nuclear

data library JENDL-2[6]. The table indicates that about 85% of fission products is stable or quasi-stable,and that there is no fission products with half-life from so years to 10000 years. This means that if we can remove TRU,long-lived fission-products with half-lives between 10^4 and 10^8 years,and Sr-90 and Cs-137, from HLW,the residual waste is free of both long-lived nuclides and high-decay-heat nuclides. Fig.1 shows variations of potential radioactive toxicities of long-lived nuclides with time. Here, potential radioactive toxicity is defined as the radioactivity (in Bq unit) of a nuclide to ALI value of the nuclide. Based on Table 1 and Fig.1,priority of nuclides to be separated for transmutation was decided as follow:

priority 1 ; **TRU** nuclides **including residual Pu**
2 ; **Tc-99 and 1-129**
3 ; Sr-90 **and** Cs-137
4 ; Zr-93,Cs-135 **and** Sm-151
5 : **long-lived activation products** (C-14,Ni-59 **etc.)**

If neutron reaction is applied to transmute long-lived nuclide to shorter-lived or stable nuclides,TRU can be effectively transmuted to fission products by fission reaction,and Tc-99 and 1-129 can be transmuted to stable nuclides by neutron capture reaction. However, it is difficult to transmute the other nuclides with lower priority by applying neutron reaction.

2.2 Target Separation Efficiency for Long-lived Nuclides in HLW

The allowable potential radioactive toxicity (DO) of residual waste including long-lived nuclide was determined so that potential radioactive toxicity should be balanced before and after reactor operation. According to Fig.2 which shows the potential radioactive toxicities before and after reactor operation,DO can be determined as;

$$DO = k1A + k2B - (1 - k3)C \qquad (1)$$

where, A,B and C are potential radioactive toxicities of depleted Unenriched U and Pu,and k1,k2 and k3 ($0<ki<1$) are transmutation rates of depleted Unenriched U and Pu,respectively. In the case of kl=k2=k3=l,the allowable toxicity DO is equivalent to the toxicity of natural U needed to produce enriched U. And in the case of kl=O and k2=k3=1,DO is equivalent to the toxicity of enriched U. These DOS are often used to normalize the toxicity of long-lived nuclide. Here, assuming 3 recycles of enriched U so that k1=0,k2=0.1 and k3=1, and also using the toxicity shown in Fig.1,the target separation efficiencies of important elements from HLW were determined preliminarily as follows;

Pu : 99.9% Np ; 99.5% Am ; 99.99%
Cm ; 99.9% Tc & I ; 99% Sr & Cs ; 99.9%

2.3 Separation Capability of Our Proposed Partitioning Process

A partitioning process to separate elements in the HLW into

four groups has been developed. In the mixer-settler experiments with the actual HLW, it was demonstrated that more than 99.99% of Am and Cm were extracted with the solvent DIDPA. It was also verified in a series of the tests with the synthesized HLW that more than 99.95% of Np was extracted when hydrogen peroxide was added. The separation of Tc has been demonstrated by adsorption with an activated carbon column. Over 99% of Tc was adsorbed from the synthesized HLW adjusted to 0.5 M nitric acid concentration and adsorbed Tc was eluted from the column very efficiently by using alkaline thiocyanate solution (2M KSCN-4M NaOH) as eluant. The Sr and Cs were also efficiently separated by adsorption with the inorganic ion exchangers of titanic and zeorite, respectively, and radioiodine will be recovered from the PUREX process offgas by adequate sorption methods.

These laboratory-scaled experiments shows that our proposed partitioning process has high possibility to realize the target separation efficiencies defined for the important elements in the HLW in the section 2.2, without big difficulty.

3. Effectiveness of Our Proposed Transmutation Technology

The TRU nuclides, and Tc-99 and 1-129 can be incinerated by fission reaction and neutron capture reaction, respectively, though incineration of Tc and I needs very high low energy neutron flux. However, effective mechanism has not been found yet to transmute the other long-lived nuclides mentioned in Section 2.1. In this section, the discussion will be limited to transmutation of the TRU nuclides.

3.1 A Forecast of Nuclear Electric Power Production in Japan

Basing on the tentative forecast of nuclear energy demand in Japan until the year 2010 submitted by the board of investigation on energy issues (June, 1989), were provisionally selected growth scenarios of nuclear power SUpply until the year 2100. These scenarios are shown in Fig.3, together with the total power sup ply. 3 scenarios of nuclear power growth after the year 2010 are used in the study; the high growth case with growth rate of 2%/y (case-1), the medium growth case with growth rate of 1%/Y (case-2) and the low growth case with growth rate of 0.5%/y (case-3). The strategic study on our proposed transmutation technologies was made for 3 cases, but is discussed only about the low growth case in this paper.

3.2 Accumulation of Minor-Actinides without Transmutation

Table 2 shows amount of minor-actinides annually generated from 1000 MWe nuclear power reactors; uranium fueled LWR (U-LWR), MOX fueled LWR (MOX-LWR) and MOX fueled LMFBR (LMFBR). Fig.4 shows accumulation of minor-actinides until the year 2100, where the nuclear power needed for the low growth case is supplied only by U-LWR and any transmutation is not made. The results show that the accumulation of total minor-actinides parabolically increases with time, and reaches more than 30 tons in the year 2010, more

than 150 tons in 2050 and more than 300 tons in 2100. As far as the total minor actinides production is concerned, there seems to be no significant difference between U-LWR and LMFBR as seen in Table 2. This means that the accumulation trend of total minor actinides will be similar, even if LMFBR is used to supply the nuclear power instead of U-LWR.

3.3 Capability of Our Proposed Transmutation Systems

The annual introduction rate of transmutation system was investigated as a function of its initial minor-actinides loading, annual transmutation rate and initial system introduction year (the year 2010 and 2020). The investigation was performed under the following conditions;

1) Amount of minor-actinides annually Produced from U-LWRs should be equivalent to their annually incinerated amount, in the year 2050, and
2) Out of pile storage capacity of minor-actinides should be zero in the year 2100.

The calculated results shows that the above mentioned conditions can be satisfied by introducing realistic transmutation systems together with their introduction rate as seen in Fig.5

Thermal and fast power reactors (LWR and LMFBR) are generally considered as realistic devices to transmute minor-actinides. In LWR, the minor-actinides are transmuted by fission event mostly after one or two neutron capture and then the build-up of higher actinides is also large. Even in LMFBR, there is a considerable fraction of the neutron flux at lower energy and the build-up of higher actinides is still not too small.

In this context, conceptual design study has been carried out on two types of minor-actinides transmutation systems which are specially designed for efficient burning of minor-actinides.

The first is the "Actinide Burning Reactor" (ABR) which is a kind of fast reactor with a very high neutron spectrum (averaged neutron energy is more than 700 MeV). The loading capacity of a 1000 MWt ABR corresponds to less than 3000 kg of minor-actinides and the ABR transmutes more than 300 kg of minor-actinides annually. The second is the proton accelerator driven minor-actinides transmutation system which is a hybrid system composed of an intense proton accelerator and a subcritical fast reactor core. Minor-actinides inventory of the system is about 3000 kg and. its minor-actinides transmutation capability corresponds to about 300 kg/GWt y.

Assuming that the deployment of these transmutation systems begin in the year 2020 and the low growth case is adopted as the nuclear power supply scenario, about 15 "units of 1000MWt grade proposed system are capable of compensating minor-actinides generation from nuclear power plants and limiting their out of pile storage capacity very much low in considerably early stage.

A double strata fuel cycle concepts shown in Fig.6 is under investigation from the view points of waste streams and disposal scenario, which consists of conventional fuel cycle for nuclear power plants and P–T fuel cycle for our proposed P–T systems.

4. Concluding Remarks

1) Our proposed partitioning process has a potential capability to separate long-lived nuclides from HLW, leaving final solution virtually free of both ultra-long-lived nuclides and high-decay heat nuclides.

2) Our proposed transmutation systems have a potential capability to remarkably reduce accumulation of minor-actinides by introducing relatively small number of systems. However, further study should be needed for other long-lived fission products than minor-actinides.

3) System study should be needed to show its effective contribution to HLW management, not only in one specific country but also in more broad area.

4) In this context, a preliminary system study should be performed under the framework of the OECD/NEA collaboration program, in order to find out our common understandings on incentives for introducing P-T and technical issues to be developed further.

References

[1] M.Kubota et al.: "Development of Partitioning Process in JAERI", Proc. the first OECD/NEA information exchange meeting on Actinide and Fission Product Separation and Transmutation, p158, Mito (1990).

[2] T.Mukaiyama et al.: "Characteristics of Minor Actinide Transmutation in Minor Actinide Burner Reactors and Power Reactors", Proc. the first OECD/NEA information exchange meeting on Actinide and Fission Product Separation and Transmutation, p326, Mito (1990).

[3] T.Takizuka et al.: "Conceptual Design Study of an Accelerator based Actinide Transmutation Plant with Sodium-cooled Solid Target/Core", Proc. the second OECD/NEA information exchange meeting on Actinide and Fission Product Separation and Transmutation, Argonne (1992).

[4] H.Katsuta et al.: "A Continuous Transmutation System for Long lived Nuclides with Accelerator-driven Fluid Targets", Proc. the second OECD/NEA information exchange meeting on Actinide and Fission Product Separation and Transmutation (1992).

[5] K.Tsuchihashi et al.: "SRAC: JAERI Thermal Reactor Standard Code System for Reactor Design and Analysis", JAERI 1302 (1986)

[6] K.Shibata et al.: "Japanese Evaluated Nuclear Data Library, Version 3, -JENDL-3-", JAERI 1319 (1990).

Table 1 Classification of TRU and FP *on* Their Half-lives

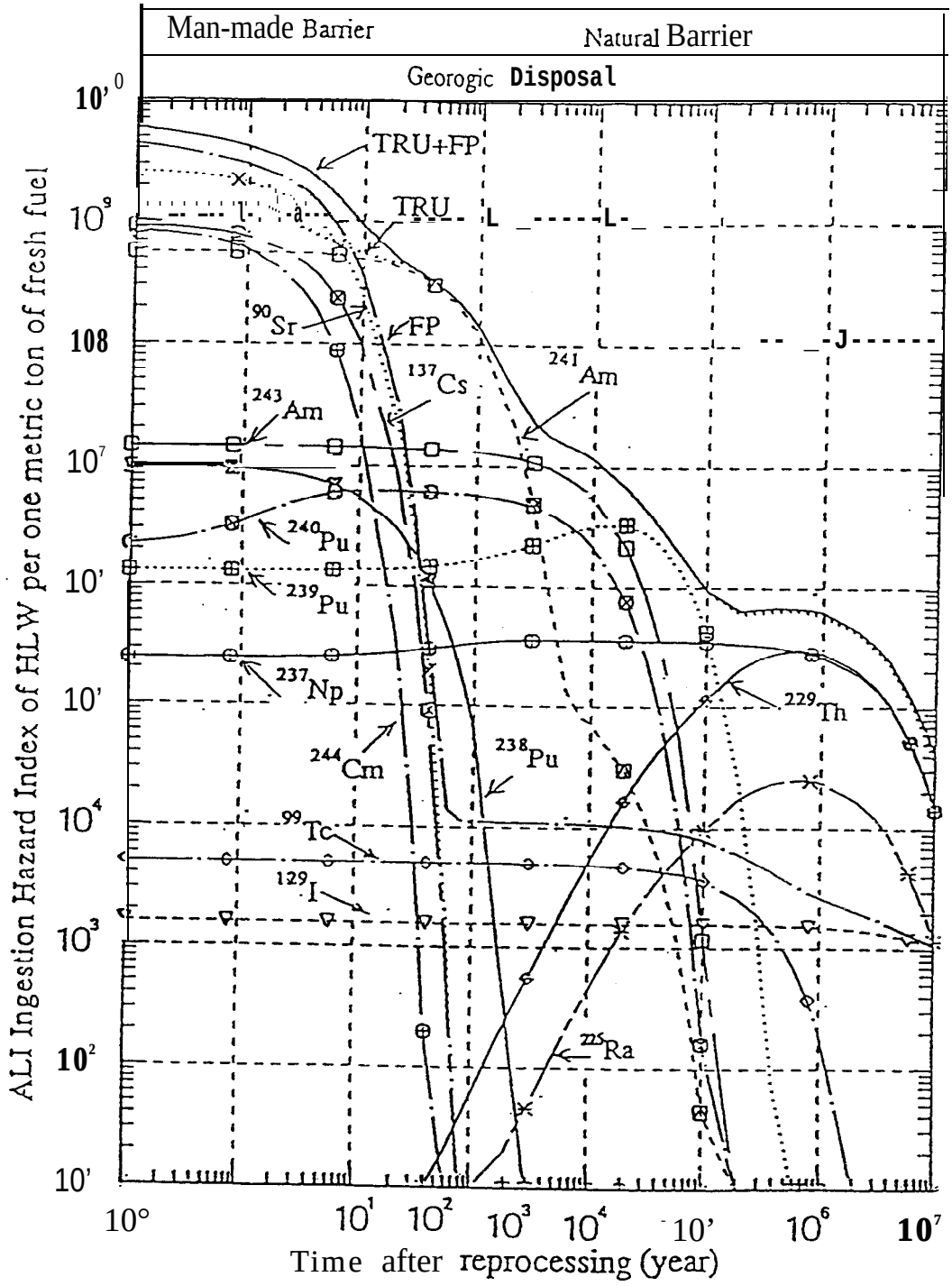
Classification	Fission-product		TRU
	Comp.(%)	Nuclide	
less than 1yr	4.8	Deleted	Deleted
1yr-10yr	1.3	Ru 106 Sb 125 Cs 134 Pm127 Eu154 Eu155	Nothing
10yr-30yr	5.3	Kr85 (11yr) Sr90 (29yr) Cs137(30yr)	Pu241(14yr) Cm243(29yr) Cm244(18yr)
30yr-100yr	0.03	Sm151	Pu238(88yr)
100yr-10^4yr	0.0	Nothing	Pu240(6.6×10^3yr) Am241(4.3×10^2yr) Am242(1.4×10^2yr) Am243(7.4×10^3yr)
10^4yr- 5×10^9yr	6.6	Se79 Zr93 Tc99(2.1×10^5yr) Pd107 Sn126 I-129(1.6×10^7yr) Cs135(2.3×10^5yr)	Np237(2.1×10^6yr) Pu239(2.4×10^4yr) Pu242(3.8×10^5yr)
more than 5×10^9yr	7.6	Rb87 In115 Ce142 Nd144 Sm147 Sm148 Sm149	Nothing
Stable	78.1	Deleted	Nothing
Total	100.0	—	

Nuclear data;JENDL-2, Burnup cal.;SRAC-FPGS code
Burnup;33,000MWD/MT, Cooling time;150 days

Table 2 Amount of Minor-actinides Annually Generated from 1000 MWe Nuclear Power Reactors

Nuclide	Cooling Time (yr)	U-LWR	MOX-LWR	LMFBR
Np-237	2	14.018	9.053	1.599
	5	14.062	9,233	1.689
	10	14.188	9.743	1.924
AM-241	2	6.539	27,949	15,323
	5	12.628	51.758	24.831
	10	20.800	83,711	37,585
Am-243	2	2.339	24.510	5.461
	5	2.338	24,504	5,459
	10	2.337	24.493	5.457
Cm-244	2	0.688	14.824	0,692
	5	0.612	13,172	0.615
	10	0.502	10.817	0.505
Cm-245	2	0.073	2,946	0.047
	5	0.073	2.946	0.047
	10	0,073	2,945	0, 047
Total	2	23,706	79,644	23.388
	5	29,749	101.843	32,875
	10	37,930	131.925	45.742

Unit;kg/GWe yr



ALI ingestion hazard index is defined as the ratio of the amount (in Bq unit) of a nuclide to ALI value of the nuclide. HL W. is recovered from the reprocessing of P WR spent fuel(burnup Of 33 GWDIMT,3 years cooling). U,Pu recovery is 99.S%.

(TRW; total transuranium elements , FP ; total fission products)

Fig.1 **Potential** Radiotoxicity of HLW from LWR Spent Fuel

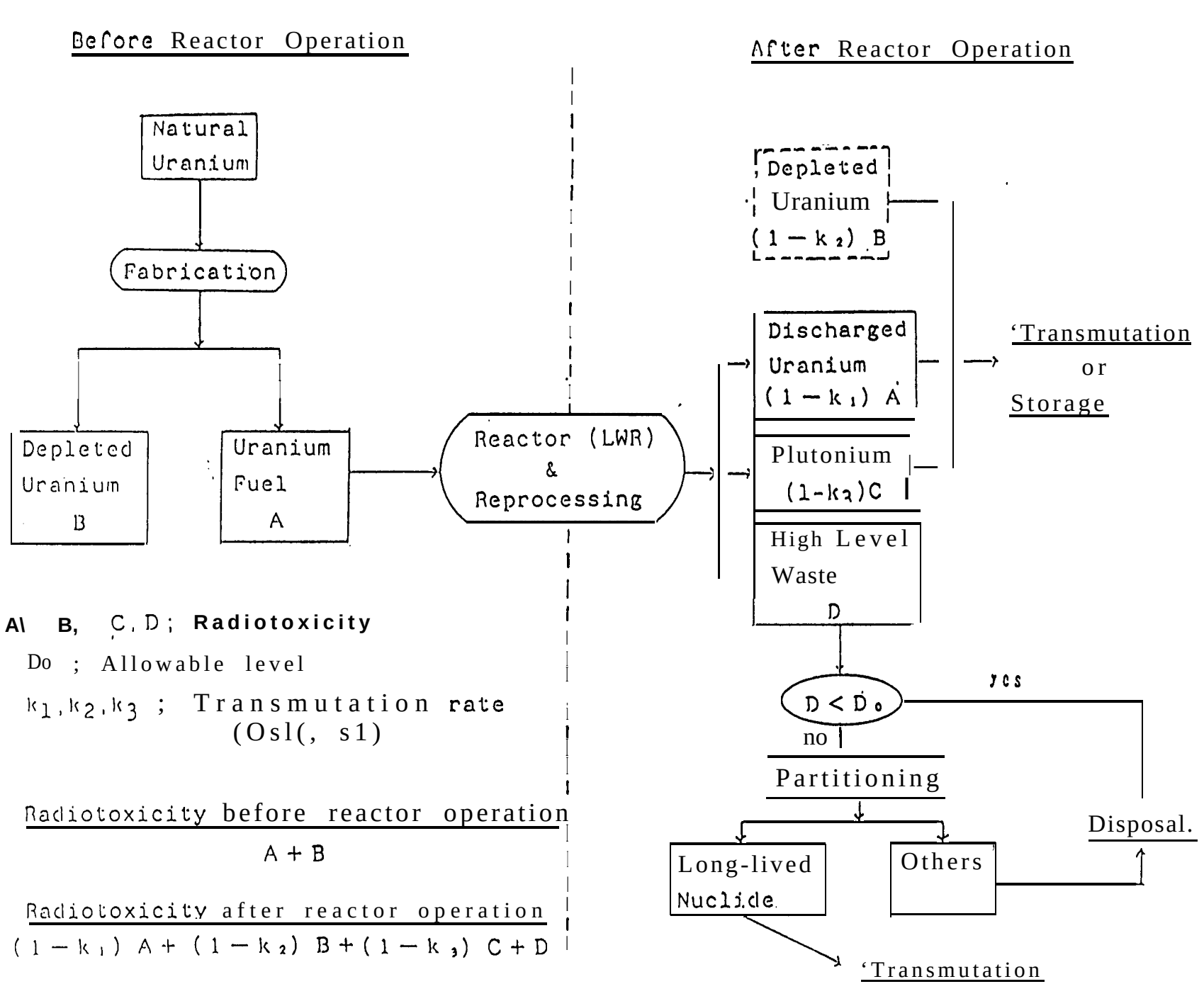


Fig.2 Flow of Materials with High Radiotoxicity in Fuel Cycle

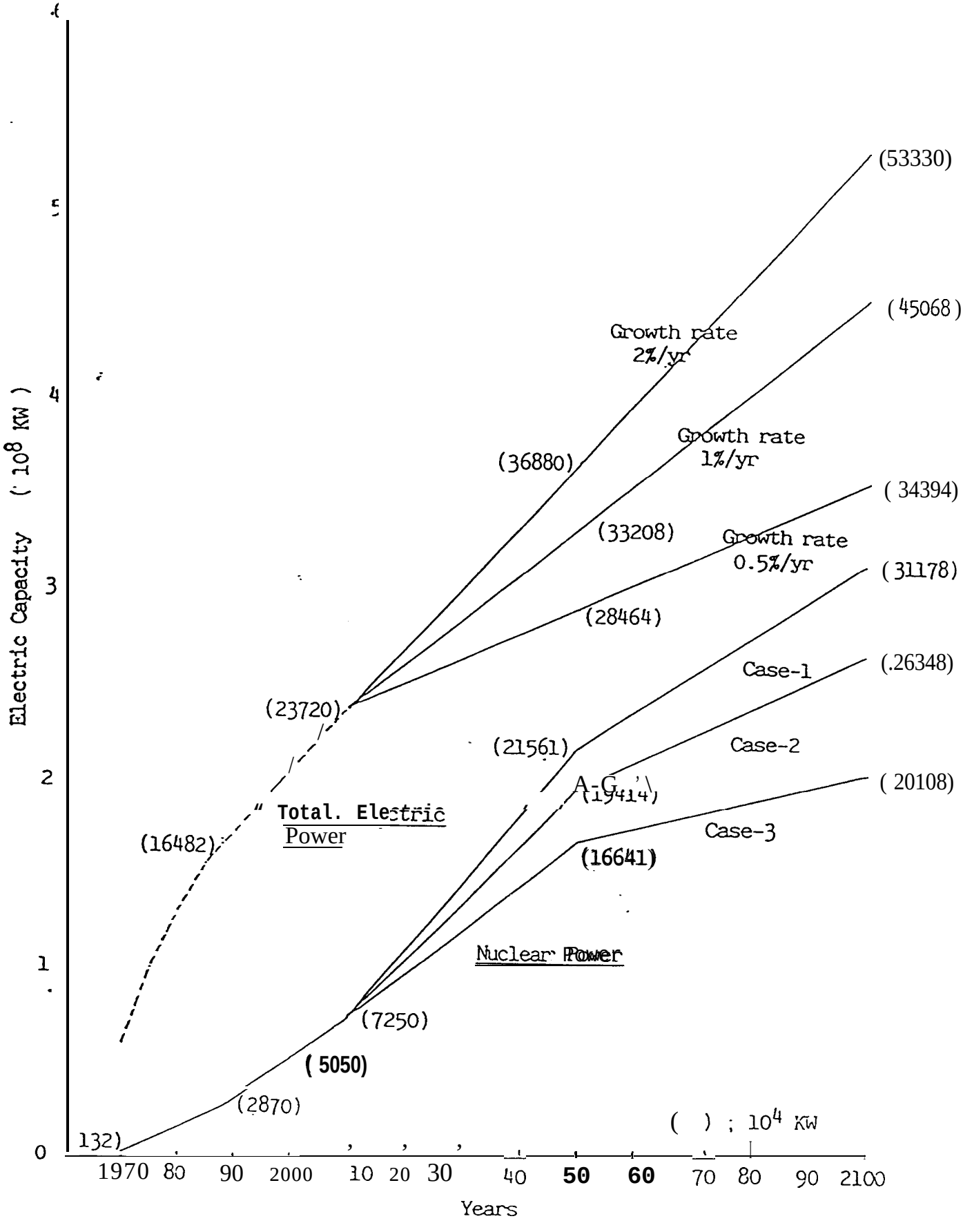


Fig.3 A Long-term Forecast of Nuclear Power Growth in Japan

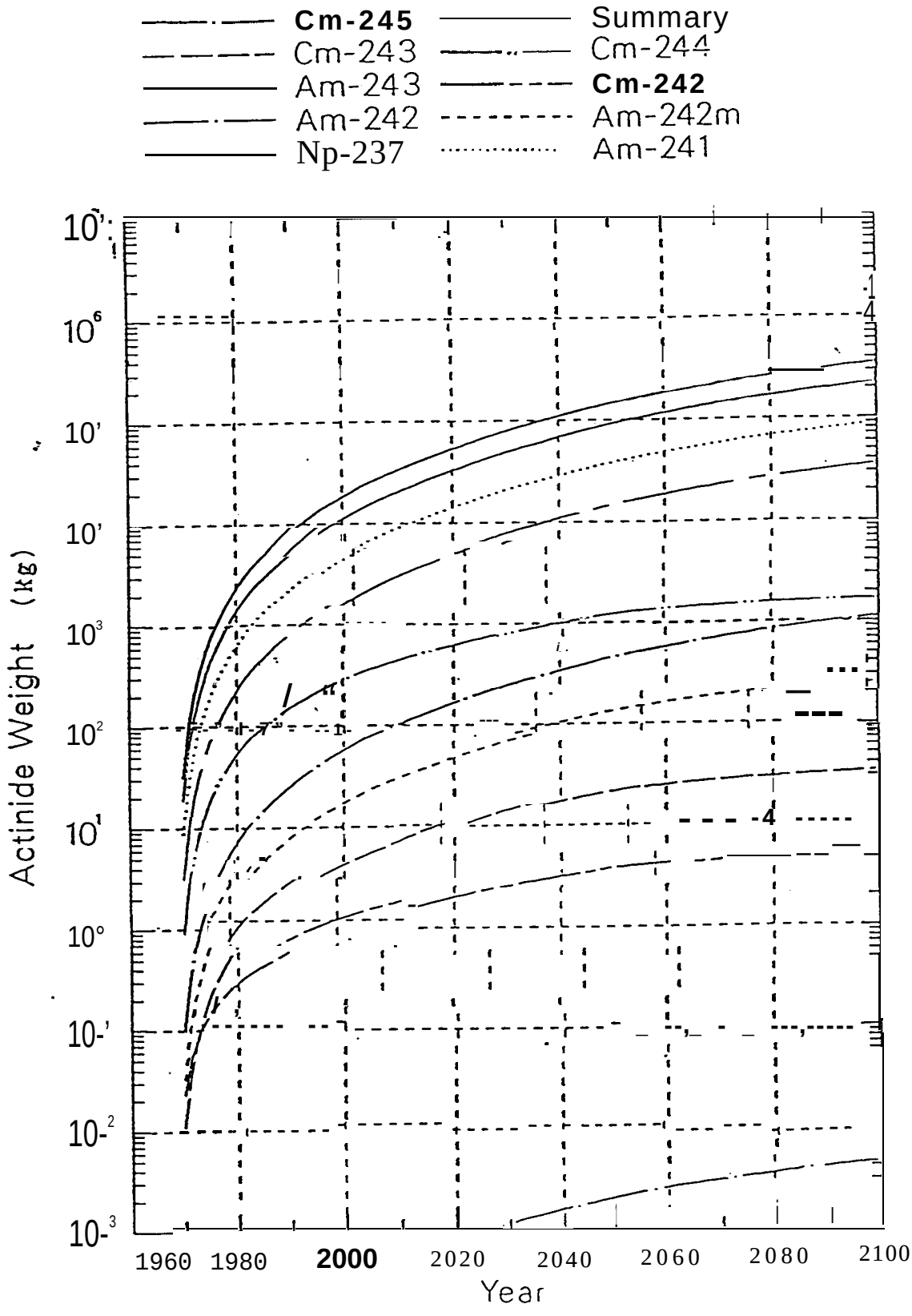


Fig.4 Accumulation of Minor -actinides without Transmutation - U-LWR, Low Growth Case -

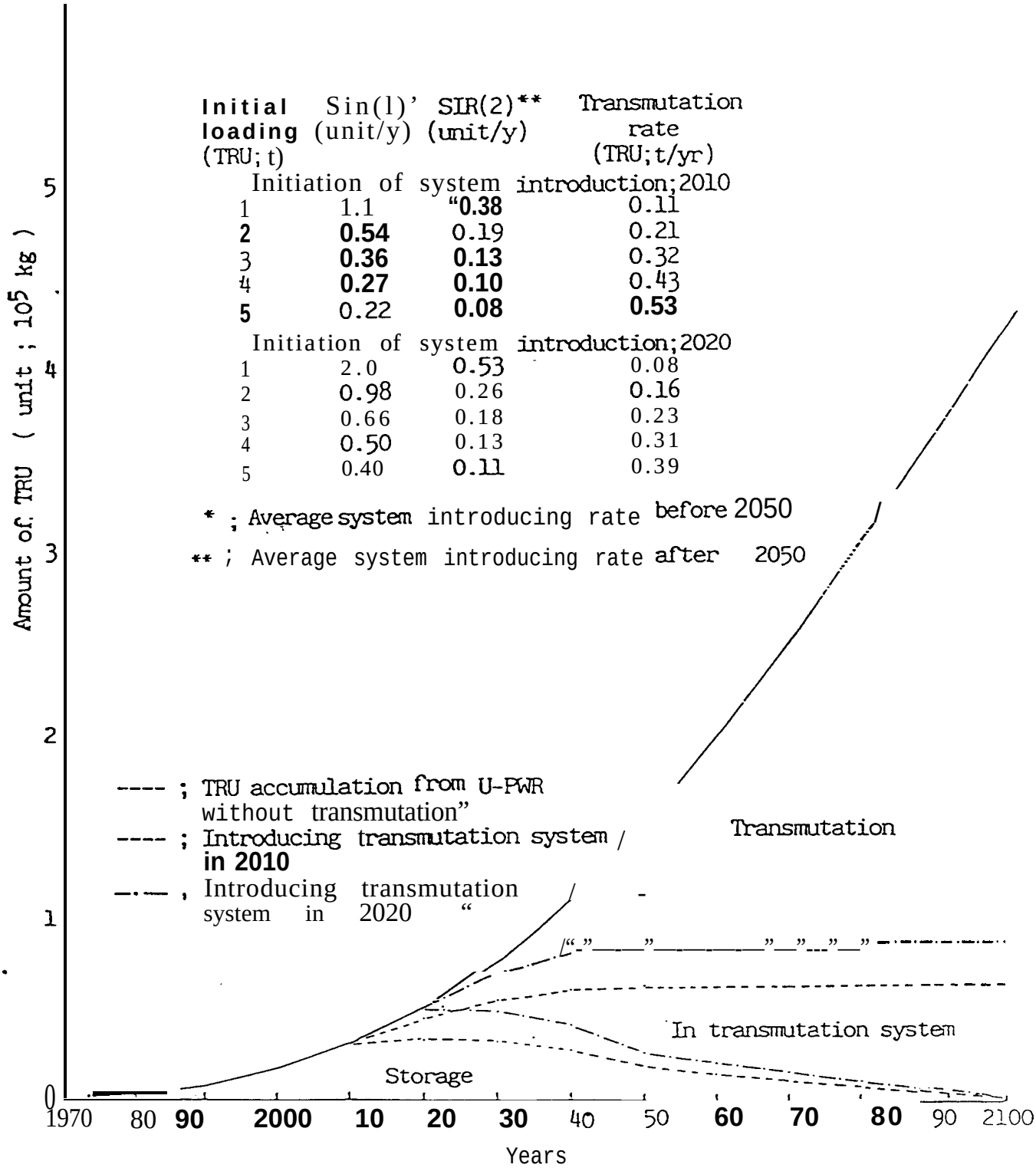


Fig.5 Effect of Transmutation on Reduction of Minor-actinides Accumulation - Low Growth Case with U-LWR -

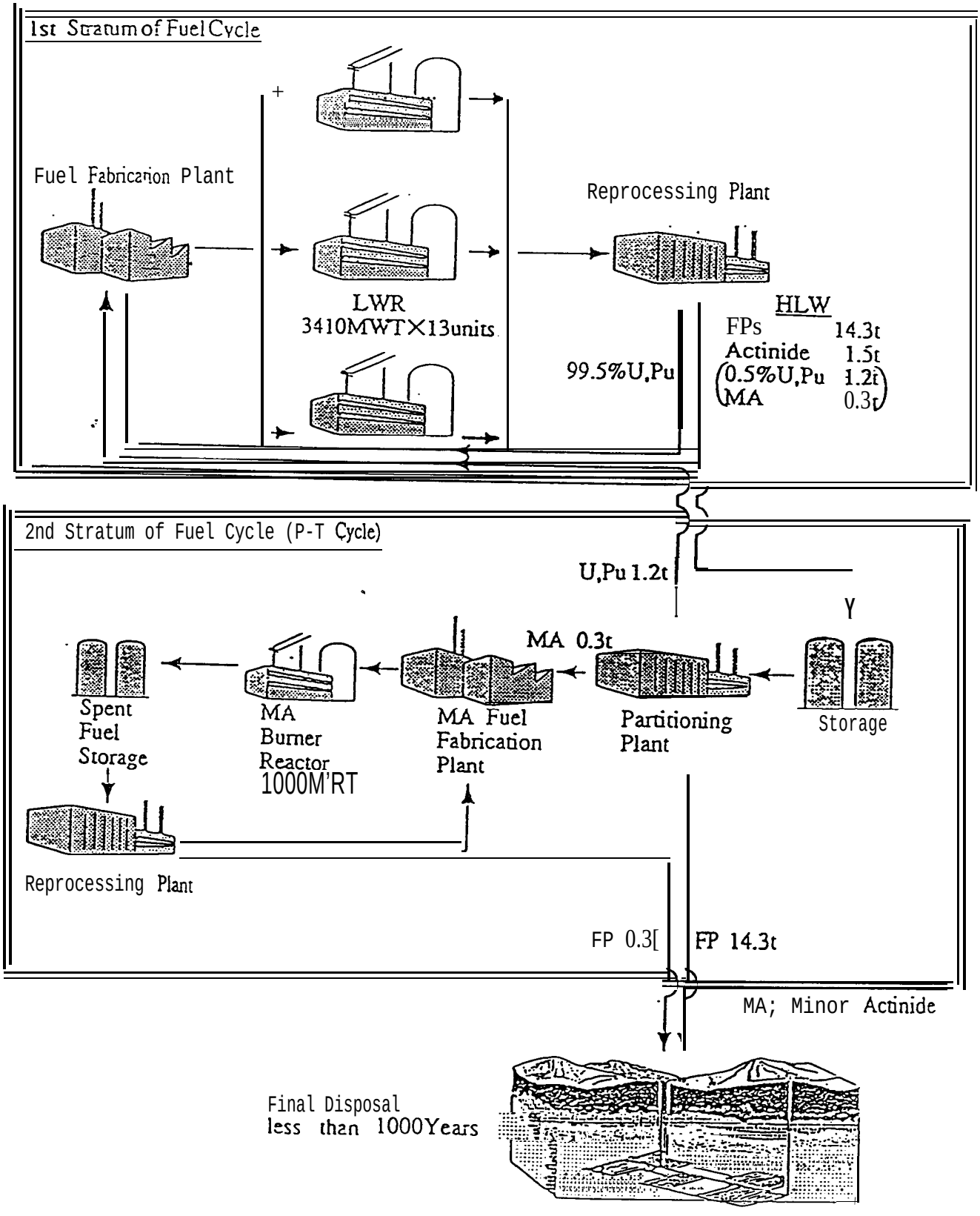


Fig.6 **Double Strata** Fuel Cycle Combined with Partitioning and Transmutation Cycle