

IMPORTANCE OF DOUBLE STRATA FUEL CYCLE FOR MINOR ACTINIDE TRANSMUTATION

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Abstract

The reactor physics characteristics of MA transmutation systems are compared between LWR, FBR and Actinide Burner Reactor (ABR). The influence of transmutation reaction products on radiation dose in fuel cycle facilities is discussed. The generation of these neutron emitters varies depending on neutron spectrum hardness of a transmutation system. The preferable feature of a dedicated MA transmutation system is compared with recycling scheme of MA in power reactor from the fuel cycle facility view point.

1. Introduction

Various systems and concepts have been proposed for transmutation of long-lived nuclides as an application of reactor technology for high-level radioactive waste (HLW) management. Recently, the application of transmutation concepts for excess Pu burning is being discussed. Pu recycling together with minor actinide (MA) has been proposed considering nuclear proliferation resistance. For the technical judgement of these concepts, most of discussions are based on consideration for such reactor engineering as fuel related matters and reactor performance. However, when the economy of these concepts is considered, fuel cycle related issues shall be taken into account.

In this paper, the influence of MA recycling in various types of reactors on fuel handling is discussed.

2. Comparison of MA transmutation from the reactor physics view points

Transmutation system concepts can be classified in two groups. The one is MA recycling in power reactors, LWR or fast reactors, and the other is MA burning in dedicated systems, actinide burner reactors or accelerator-driven hybrid systems. When MA is recycled in a power reactor, the acceptable amount of MA in power reactor fuel is limited since the addition of MA in fuel has large reactivity effect for reactor performance and major reactor parameters of power reactors are already optimized from safety and economical view points on the other hand. Therefore, addition of MA should not result in large change of these parameters. For the concepts of MA recycling in PWR, MOX-PWR (Pu usage in PWR) and MOX-FBR, the maximum allowable amount of MA will be 0.2, 0.5 and 5wt% of fuel material, respectively.

When a dedicated transmutation system becomes available, the scheme of an entire fuel cycle will be a strata structure fuel cycle. The concept of the double strata fuel cycle consisting of a conventional power reactor fuel cycle and a Partitioning-Transmutation (P-T) cycle is illustrated in Fig. 1.¹⁾ The final HLW from the double strata fuel cycle contains only short-lived and stable fission products. The separate treatment of MA from the commercial cycle will be preferable for the conventional fuel cycle because MA is a strong neutron emitter and the MA recycling in the conventional power reactor fuel cycle will introduce the problem for fuel handling.

As an example of dedicated transmutation systems, the reactor core design parameters of two types of ABRs are shown in Table 1. Fuel material of these ABRs is MA-enriched uranium nitride mixture. One of ABRs is a lead-cooled pin fuel ABR (L-ABR) and the other is a He-cooled particle fuel ABR (P-ABR).²⁾ In these ABRs, neutron energy spectrum is very hard and the core averaged neutron energy is around 720keV. These very hard neutron spectra are very effective for direct fission of MA which has fission threshold at around 600keV.

In Table 2, the transmutation characteristics are compared between P-ABR and power reactors, namely PWR, MOX-FBR and a metal fuel FBR. For the MA transmutation in power reactors, the concentration of MA is assumed as 0.2% and 5% of heavy metal for PWR and FBR, respectively.

In the table, the transmutation ratio is defined as the ratio of MA weight at the end of cycle to that of the beginning of cycle. In this definition, any nuclear reaction such as fission, neutron capture, (n, 2n), etc. can be considered as transmutation reaction and the conversion of Np-237 into Pu-238 is a part of transmutation of Np. On the other hand the MA burnup ratio is defined as the ratio of fissioned MA weight to that at the beginning of cycle. The latter is the real index of transmutation effectiveness and efficiency because only fission is a real transmutation reaction to solve the problem of long-lived MA management.

The transmutation and the burnup ratios of power reactors in Table 2 are the net ones after the MA generation in fuel being deducted. The negative value of burnup ratio of U-PWR implies that new MA generation in fuel prevails over fission of MA which is added in fuel for transmutation. The large discrepancy between the transmutation ratio and the burnup ratio in power reactors indicates the larger conversion of Np into Pu than in ABRs. The net MA burnup per 1GWt a year of ABRs is significantly larger than that of power reactors because in ABRs major fuel material is MA. The support factor which is defined as the number of power reactor units of which MA is transmuted by one unit of a transmutation system is about 10 to 15 for dedicated transmutation systems, while the support factor of FBR proposed so far is between 4 to 6.

In Table 3, the transmutation characteristics of Np-237 are compared between P-ABR, MOX-FBR and PWR. In this analysis, Np-237 is continuously irradiated with neutron flux, of which level is indicated in the table. In Table 3a, the cumulative fission fraction and the fraction of fission which undergoes as neutron capture products of Np-237 are shown as wt% of initial Np-237. In P-ABR which has very hard neutron spectrum, most of fission occurs as Np-237 or Pu-238 and total fission after 6000 days irradiation is 98.6% of initial Np-237. In MOX-FBR, fission of Np-237 is dominant as Np-237, Pu-238 and Pu-239, and total fission in 6000 days is only 75%. In PWR, Np undergoes fission as Pu-239 or 241, and total fission is 92% which is larger than that of MOX-FBR. In Table 3b, residual actinide after irradiation is shown. In PWR, generation of americium and curium from Np-237 is significant. From this table, it is evident that very hard neutron spectrum is favorable for MA transmutation. In PWR or under thermal flux, transmutation or fission of MA is slow at the beginning of irradiation but after the sufficient conversion of MA to fissionable isotopes, transmutation is very efficient. In PWR, however, generation of heavier MA is serious problem.

3. Influence of MA transmutation products on fuel handling at fuel cycle facilities

There are several criteria for engineering judgment of transmutation systems. At present, most of system evaluations are discussed from the reactor engineering view points, namely, transmutation rate, influence of MA on reactor performance, fuel related issues. The other important point for judgement is the influence of transmutation products on the fuel cycle facilities. In Table 4, the increase of heavier MA generation and neutron emission rate in spent fuel are shown for the case of 0.2 wt% MA addition to PWR fuel. Only 0.2wt% of MA addition results in 700 times increase of Cf and 7 times increase of spontaneous neutron emission thereof. To evaluate the effect of these heavier nuclides, most of which are strong neutron emitters, the increase of decay heat, neutron emission and γ -ray intensity was calculated when MA is added to power reactor fuels.

In this analysis, typical power reactors for MA recycling scenario was selected. Power reactors selected were PWR, MOX-PWR and MOX-FBR. The effect was calculated for both fresh and spent fuels of these reactors. In the present calculation, the fraction of MA in the fuels are 0.2wt% heavy metal of PWR fuel and 5 weight % of MOX-PWR and MOX-FBR fuels. The fuel burnup is 45, 45 and 85GWD/HMT, respectively. The cooling time of spent fuel is 10 years. In Table 5, the result of the analysis is summarized. The increase of decay heat and γ -ray emission are at the acceptable level but the increase of neutron emission in fresh fuels is large.

The influence of neutron emission increase on fuel cycle facilities was evaluated for the scenario of MA recycling in these power reactors. Facilities and fuel handling considered are a) fuel manufacturing, b) fresh fuel transportation, c) spent fuel storage, d) spent fuel transportation and e) reprocessing. The evaluation was made for the process where the decay heat and radiation dose are possibly severest.

a) Fuel manufacturing process

The oxide powder mixture process is the one where the quantity of fuel material will be largest throughout fuel manufacturing. The powder mixture machine was simulated as a ball of 0.5mm thick stainless steel wall. The powder density is 2 g/cm³. In the case of PWR fuel, 3 ton of UO₂ powder is contained in a mixture machine of 142 cm diameter. In the case of MOX-PWR fuel and MOX-FBR fuel, 300kg MOX powder is contained in a mixture machine of 66cm diameter. The calculation model is illustrated in Table 6.

The values in the table are the ratios of dose from fuel material powder which contains MA to that without MA. The radiation dose increase in the PWR fuel with 0.2wt% MA addition is prohibitably high as about 10⁴. In the case of MOX-PWR and MOX-FBR fuel, the increase is 50 to 100 times. The significant increase of neutron dose results in the significant reinforcement of radiation shielding in the fuel manufacturing facility for handling fuel even with very small fraction of MA addition.

b) Fresh and spent fuel transportation

Radiation dose and decay heat at a fuel cask were evaluated.

c) Spent fuel storage

Spent fuel storage facility was selected for the evaluation.

d) Reprocessing facility

Radiation dose and decay heat at a feed preparation tank was evaluate. The calculation model is shown in Table 7. The volume of the tank is 25m³ and the wight of heavy metal in liquid solution is 8.75ton which is calculated from the subcriticality density limit of 36g/l. The cooling

time of spent fuel is 4 years. The increase of the radiation dose is 1.2 to 2 times of that of normal fuel.

In Table 8, the influence of MA contained fuel on fuel cycle facilities is summarized for MA recycling in PWR, MOX-PWR and MOX-FBR. The significant radiation shielding reinforcement is required for fuel manufacturing and fresh fuel transportation. The reinforcement of radiation shielding in fuel cycle facilities will result in the increment of fuel cycle cost.

For the fuel handling in fuel cycle facilities of a ABR or an accelerator-driven system, radiation shielding and decay heat removal are much severer problem than that of MA recycling in power reactors since the concentration of MA is very high in these dedicated systems. The fuel cycle facilities for dedicated systems, however, are very compact and the required number of these facilities is small because nitride fuel can be reprocessed by pyrochemical process and the mass flow is small compared with those of conventional fuel cycle.

4. Conclusion

As a result of increase of heavier MA for the MA transmutation scheme in power reactors, the radiation shielding reinforcement will be needed for the fresh fuel handling (manufacturing and transportation). This may cause the cost increase of the of electricity generation. In the case of a dedicated system, the shielding and the decay heat removal are much severer problem than the MA transmutation in power reactors. However, the cost of construction and operation of compact facility even with heavy radiation shielding and remote handling may not be significant compared to that of large scale facility with medium radiation shielding.

The cost comparison of the fuel cycle between MA recycling in power reactors and MA recycling in a dedicated system is inevitable for judgement of transmutation system.

From the socio-techno view point, the confinement of troublesome MA in one closed site will be very important. In this sense, the double strata fuel cycle concept can provide the closed HLW management park concept.

At the time of MA transmutation system selection in the future, the criteria for the selection should be established not only from the reactor or system performance view points but also from the fuel cycle facility view points.

REFERENCES

- 1) T. Mukaiyama, et al.: "Conceptual study of actinide burner reactors", Proc. Intn'l Reactor Physics Conf. (Jackson Hole, 1988), Vol. IV, p369.
- 2) T. Mukaiyama, et al.: "Minor Actinide Transmutation in Fission Reactors and Fuel Cycle Consideration", Proc. OECD/NEA 2nd Information Exchange Meeting on P-T at ANL, NEA/P&T REPORT No. 7, p320 (OECD, 1993).

Table 1 Reactor design parameters of modified Actinide Burner Reactors

	L-ABR ¹⁾	P-ABR ²⁾
Fuel concept material	pin-bundle ($^{64}\text{NpAmCm-36U}^{3)})_{1,0}\text{N}^{4)}_{1,0}$	coated particle ($^{65}\text{NpAmCm-35U}^{3)})_{1,0}\text{N}^{4)}_{1,0}$
MA initial loading,kg	918	2870
MA/U	588/330	1865/1005
Reactor power, MWth	180	1200
Coolant material	Lead	Helium
Neutron flux, $10^{15}\text{n/cm}^2 \cdot \text{sec}$	3.1	6.6
Core averaged mean neutron energy, keV	700	700
Reactivity (% $\Delta k/k$)		
Coolant-void reactivity/core	-1.3	—
Doppler reactivity/core ($\Delta t=300^\circ\text{C}$)	-0.01	-0.01
Kinetic parameters		
β_{eff}	2.6×10^{-3}	2.6×10^{-3}
L_p , sec	1.3×10^{-7}	1.5×10^{-7}
Cycle length, full-power days	550	300
MA burnup, %/cycle	11	13

- 1) L-ABR: MA nitride fuel with lead cooling burner reactor
- 2) P-ABR: MA particle fuel burner reactor
- 3) 90% enriched uranium
- 4) ^{15}N enriched

Table 2 Comparison of MA transmutation in various reactors

	MA Burner Reactors	Power Reactors		
	P-ABR	U-PWR	MOX-FBR	LMR ¹⁾
Output (MWt)	1200	3410	2600	2632
Cycle length ²⁾ (EFPD)	300	850	1368	900
Core averaged				
Fast neutron flux ($\times 10^{15}\text{n/cm}^2 \cdot \text{s}$)	5.9	0.37	4.1(3.0)*	6.1(4.1)*
Mean neutron energy (keV)	720	thermal	480	490
MA loaded (kg)	1865	180 ³⁾	1450 ³⁾	1200 ³⁾
MA transmutation ratio ⁴⁾ (%/cycle)	18.8	9.0	33.5	29.8
MA burnup ratio ⁵⁾ (%/cycle)	13	-23.8	8.8	8.3
MA transmutation(kg/1GW · year)	292	1.7	40.9	44.5
MA burnup (kg/1GW · year)	202	-4.4	10.7	12.4

- 1) Metal fuel FBR
 - 2) Effective fuel irradiation time
 - 3) Concentration of MA in fuel ; 0.2% for U-PWR, 5% for MOX-FBR and LMR
 - 4) $\text{MA transmutation ratio} = \{ \text{MA(BOC)} - \text{MA(EOC)} \} / \text{MA(BOC)}$
 - 5) $\text{MA burnup ratio} = (\text{MA fissioned} - \text{MA generated}) / \text{MA(BOC)}$
- Values of 4) and 5) are those for the equilibrium cycle in ABRs, and average values of 1 to 10th cycle for power reactors.
- *) Values in the parentheses are for the outer core

Table 3 Comparison of transmutation of Np-237 between different neutron spectra of ABR, MOX -FBR and PWR

Table 3a ²³⁷Np Fission
(unit:% of initial ²³⁷Np)

Reactor	Flux (×10 ¹⁵)	Cycle No.	Cumulative Fission	Fissioned as					
				Np-237	Pu-238	Pu-239	Pu(0+1)	Am	Cm
P-ABR	8.4	1	13.2	10.6	2.4	0.13	<0.01	<0.01	<0.01
		10	87.1	35.0	36.9	12.1	1.01	0.02	<0.01
		20	98.6	36.1	41.0	15.5	2.20	0.09	0.02
MOX-FBR	3.3	1	3.4	2.80	0.57	0.02	<0.01	<0.01	<0.01
		10	44.0	15.1	21.4	6.24	0.22	<0.01	<0.01
		20	74.7	18.1	35.1	15.9	1.26	0.04	<0.01
U-PWR	0.37	1	1.8	0.41	0.24	0.70	0.03	<0.01	<0.01
		10	65.1	1.61	5.35	39.4	13.9	0.26	0.38
		20	91.6	1.71	6.70	51.4	21.2	0.47	2.05

Table 3b ²³⁷Np Capture
(unit:% of initial ²³⁷Np)

Reactor	Flux (×10 ¹⁵)	Cycle No.	Residual Actinide	Capture to					
				U	Np-237	Pu-238	Pu-239	Am	Cm
P-ABR	8.4	1	86.8	0.05	71.3	14.3	0.90	<0.01	<0.01
		10	12.9	1.74	3.23	4.40	2.33	0.03	<0.01
		20	1.4	0.53	0.13	0.21	0.17	0.02	<0.01
MOX-FBR	3.3	1	96.6	0.04	85.1	11.0	0.37	<0.01	<0.01
		10	56.0	5.22	20.0	22.2	7.12	0.03	<0.01
		20	25.3	6.34	4.06	7.61	4.55	0.15	0.01
U-PWR	0.37	1	98.2	0.07	75.8	19.9	1.87	<0.01	<0.01
		10	34.8	4.43	6.36	14.4	2.92	0.82	1.59
		20	8.3	2.29	0.58	1.80	0.38	0.33	1.73

One cycle ; 300 days irradiation and 3 years cooling

Table 4 Effect of MA addition to power reactor fuel handling

Reactor/Fuel	Ratio of value of MA added fuel to that of normal fuel		
	Decay heat	Neutron emission	Photon intensity
U-PWR (0.2wt%) ^a			
fresh fuel(U235:4wt%) ^b	3.6×10^3	8.3×10^4	1.3×10^3
spent fuel(45GWD/t) ^c	1.5	4	1
MOX-PWR (0.5wt%)			
fresh fuel(Pu:6.5wt%)	1.4	4.8	1.2
spent fuel(45GWD/t)	1.5	1.7	1
MOX-FBR (5wt%)			
fresh fuel(Pu:30wt%)	2.2	1.0×10^2	2.1
spent fuel(80GWD/t)	2.8	19	1

a: minor actinides(MA) fraction in fuel (HM weight %)

b: fuel enrichment

c: fuel burnup (cooling time : 10 years)

Table 5 Effect of MA addition in PWR fuel

(values : per ton of HM)

Item	Reference PWR	MA-PWR	(MA-PWR)/(Ref.PWR)
Nuclide (g)			
Np	469	918	2.0
Am	162	276	1.7
Cm	38	296	7.7
Bk	3.4×10^{-7}	1.7×10^{-4}	510
Cf	3.5×10^{-7}	2.4×10^{-4}	690
α -activity (10^5 Ci)	0.29	1.3	4.6
(α, n) (10^6 n/s)	6.3	36	5.8
Spnt. fission(10^9 n/s)	0.49	3.5	7.1

MA addition : 0.2% of HM

U enrichment : 3.2%

Burnup : 33000MWD/T

Irradiation : 847 days

Cooling : 150 days

Table 6 Effect of MA on radiation dose of fuel manufacturing facility

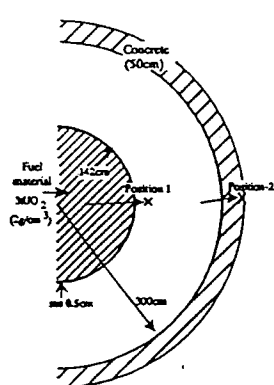
(values: ratio of dose from fuel material with MA to that without MA)

Fuel	Position of dose	Neutron	γ	Total
U-PWR	1	8.3×10^4	7.9×10^2	9.6×10^3
	2	8.3×10^4	3.3×10^3	2.8×10^4
MOX-PWR	1	48	6.5	41
	2	48	26	45
MOX-FBR	1	100	12	79
	2	97	86	96

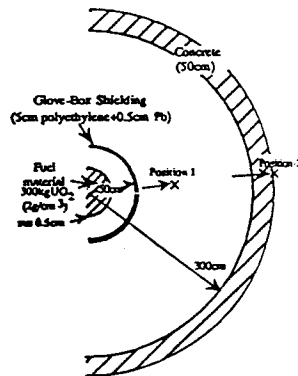
Position-1: 1m from powder mixing machine for PWR

1m from Glove Box Shielding for MOX-PWR and MOX-FBR

Position-2: Outer surface of 50cm thick concrete placed at 3m from the center



Calculation Model for PWR



Calculation Model for MOX-PWR and MOX-FBR

Table 7 Effect of MA on radiation dose at feed preparation tank of reprocessing plant
(values: ratio of dose from fuel material with MA to that without MA)

Fuel	Neutron	γ	Total
U-PWR	4.4	1.8	2.0
MOX-PWR	1.2	1.2	1.2
MOX-FBR	---	---	$\sim 2^*$

Position : Outer surface of concrete wall (2m thick)

Fuel material: 8.75MTHM (360g U+Pu/ℓ, volume 25m³)

* Estimated value from the case of MOX-PWR

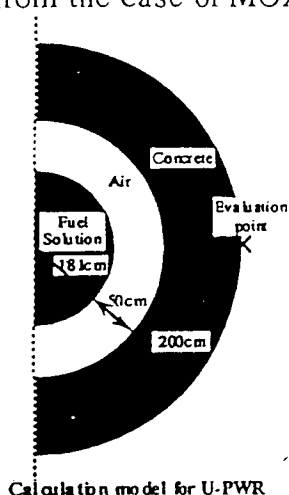


Table 8 Effect of MA on shielding design of fuel cycle facilities
when MA added to power reactor fuels

Reactor \ Process	Fuel Manufacturing	Fresh Fuel Transportation	Spent Fuel Storage	Spent Fuel Transportation	Reprocessing
U-PWR	S	S	N	M	M
MOX-PWR	S	S	N	M	M
MOX-FBR	S	S	N	M	M

S: Significant effect, radiation dose increases by order of magnitude

M: Minimum effect, radiation dose increases factor of two or less

N: Negligible effect

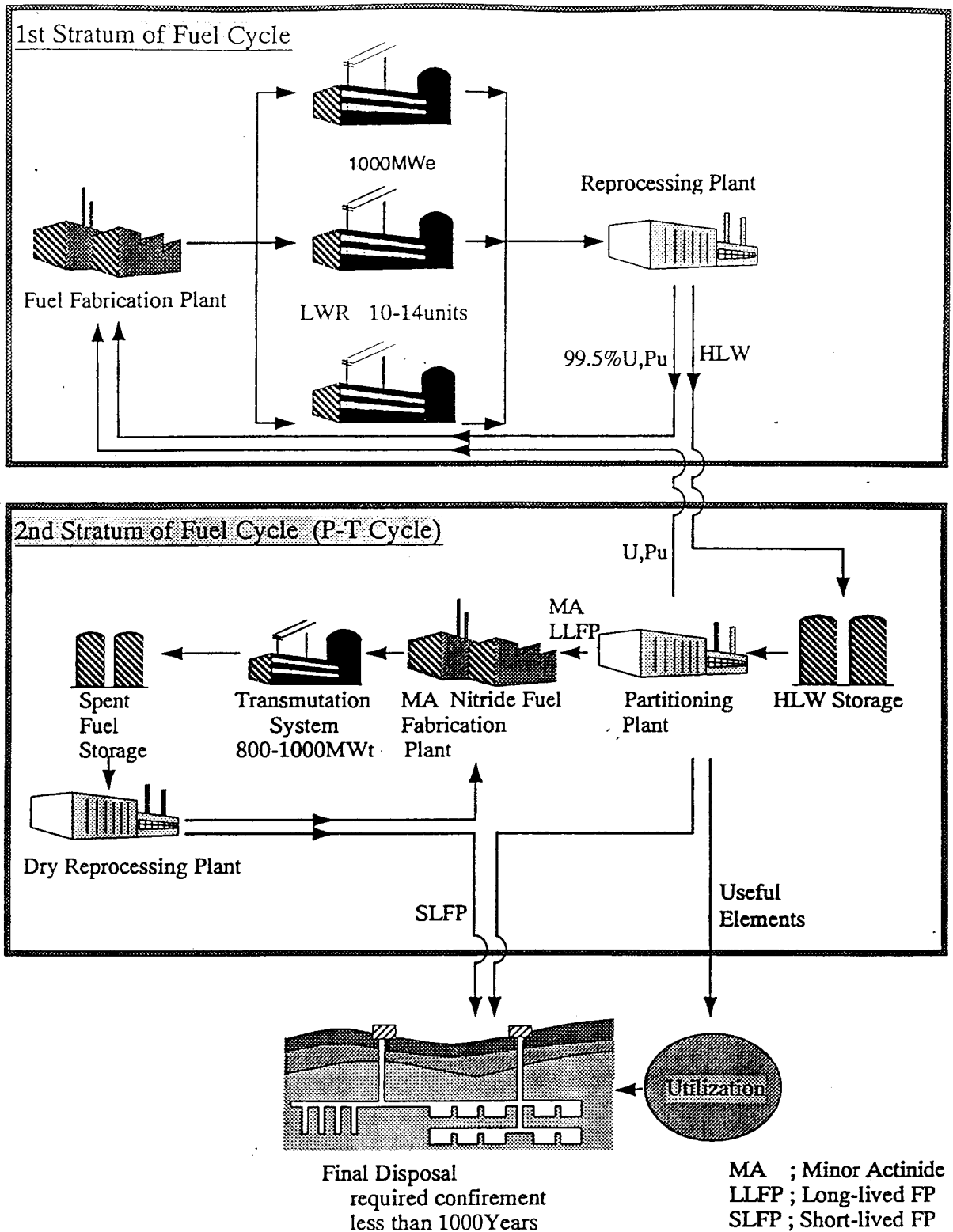


Fig. 1 JAERI's Concept of Double Strata Fuel Cycle for Complete HLW Management