MINOR ACTINIDE TRANSMUTATION IN FISSION REACTORS AND FUEL CYCLE CONSIDERATION

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Abstract

Two concepts of minor actinide burner reactors were previously proposed by the authors, one with metal fuel core and the other with nitride particle fuel tom. These reactors have very hard neutron spectrum and very high neutron flux which are effective and efficient for fissioning of minor actinides. In these burner reactors, however, β_{eff} was very small and the sodium void coefficient of a metal fuel reactor was positive and large.

In order to improve the safety characteristics, the design of these burner reactors was modified AS a part of transmutation system evaluation minor actinide transmutation by actinide burner reactors is compared with that by power reactors from both the reactor physics and fuel cycle viewpoints.

1. Introduction

Various methods of minor actinide transmutation have been proposed in order to alleviate the burden of the high-level radioactive waste(HLW) depository problem. One of the practical methods is to recycle them in fast reactors since minor actinides such as ^{237}Np, ^{241}Am, ^{243}Am (hereafter referred as MA) are fissionable with fission threshold in about 700 keV range and capture cress sections of these nuclides rapidly decrease with neutron energy higher than this threshold A special reactor with a very hard neutron energy spectrum and high neutron flux would be very efficient and effective for MA transmutation if it is technically feasible. In this context we have been developing the concept of MA burner reactors(ABR; Actinide Burner Reactor). Once these ABRs will become available, the troublesome MA from the waste management view point will be contained in a separate fuel cycle from the commercial one.

The concept of the double strata fuel cycle consisting of the commercial fuel cycle and the Partitioning-Transmutation (P-T) cycle is illustrated in Fig.1. The final HLW from this fuel cycle contains only 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 fuel cycle will introduce the problem of the radiation shielding of the fuel cycle facilities.

The results of the ABR design study was presented at the first OECD/NEA information exchange meeting on P-T held at Mito.[1)] The disadvantage of the ABRs presented were the small value of β_{eff} and the large positive sodium void coefficient. In this report the design modification of ABRs for improving the safety is discussed. Also, the efficiency of MA transmutation is compared between ABRs and power rectors (PWR and FBR). The effect of MA recycling in power reactors is discussed from the fuel cycle facility view points.

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2. Original design of ABRs

The guidelines for designing an ABR were as follows;

- **MA as the major fuel material,**
- **very hardcore averaged** neutron spectrum,
- very high neutron flux,
- pyrochemical reprocessing for compact fuel cycle facilities.

Two types of ABR design were obtained namely MA metal fuel ABR **and MA nitride particle** fuel ABR The followings are the brief description of the Original ABRs. The details of these designing are described elsewhere.[1,2,3)]

1) Na cooled MA alloy fuel ABR(M-ABR)

To design a core with a very hard neutron spectrum, a metal fuel core is the first choice. The pyrochemical reprocessing of metal fuel is also attractive from an economic view points of fuel cyck facilities because of the Compactness.

Because Np and Am are theoretically predicted not to melt each other, MA alloy has to be separated into two alloy systems, namely, Np-(Pu)-Zr and Am-Cm-(Pu)-Y. In these alloys, Pu is added because of two reasons; 1) to reduce critical mass, 2) to **compensate for reactivity gain which is caused as** a **result of conversion** of MA to fissionable **nuclides** with the **burnup** reactivity loss of Pu. The addition of Pu, however, is needed only at the initial loading and after the first loading Pu converted from ^{237}Np **plays this role.**

2) *He cooled MA particle bed ABR(P-ABR)*

Low thermal conductivity and melting point of MA metal fuel are the limiting factors for the high MA burnup in M-ABR Therefore, the particle bed reactor concept was applied as an alternative ABR, which has the high efficiency in heat transfer since small particle size produces a large heat transfer surface per volume. The bed of coated fuel particle contained in double concentric porous frits is directly cooled by helium. The fuel is a microsphere of MA nitride which is coated with a refractory material such as TiN. The kernel of the fuel particle is homogeneous mixture of Pu and MA nitride. The reprocessing of spent fuel of nitride fuel will be by **pyrochemical process.**

3) *Characteristics of ABRs*

The reactor core design parameters of M-ABR and P-ABR at their equilibrium state are shown in Tabk 1, respectively. Comparison of core averaged neutron spectra is shown in Fig.3 In this figure, neutron spectrum of MOX-LMFBR is also shown for comparison. Significantly hard neutron spectra of ABRs are obvious. In the M-ABR, the neutron flux is not so high as opposed to the initial attempt to design a very high neutron flux reactor because of low melting point and low thermal conductivity of MA fuel. In the P-ABR, contrary, the neutron flux is very high owning to the efficient heat removal characteristics of particle fuel. The MA transmutation (fission and capture) per cycle is 2S ~26%, and the MA burnup (fission) per cycle is 17 +- 18%.

In the original design of ABRs, the effective delayed neutron fraction β_{eff} and Doppler reactivity coefficient are very small and the positive sodium void coefficient is large owing to the lack of U **in** a core and also to the **very hard neutron spectrum.**

3. Modification of ABR design

To improve the safety characteristics, the design of the original ABRs was modified, 1) to increase the value of β_{eff}, and 2) to avoid the large positive sodium void coefficient. Also, in the new design, MA alloy fuel is replaced with the MA nitride fuel because of relatively low melting point and low thermal conductivity of MA alloy fuel. The other advantage of nitride fuel. is that nitride fuel can be processed with the pyrochemical reprocessing and hence, the fuel cycle facilities can be very compact

To increase /3 $_{eff}$, uranium is added as a part of fuel material because of its large delayed neutron fraction. ^{236}U has the largest β value but is not adequate as fuel material of ABR because the ABR is a system to burn MA and ^{238}U is source nuclide of MA. In the modified ABR, therefore, Pu is replaced with enriched uranium(EU, enrichment; 90%). β_{eff} at the initial cycle of the modified ABR with fuel is 0.36% and at the equilibrium cycle it is 0.2.S%. These values are significantly larger than 0.16%~0.17% of the original ABRs. The neutron generation time is also increased from $7\sim11\times10^{-8}$sec of the original ABRs to $1.3\sim1.5\times10^{-7}$sec of *new* ABRs.

In ABR, the slight hardening of neutron spectrum introduces large positive reactivity effect because fission chain reaction is maintained by fast fission of MA. It is, therefore, difficult to reduce the large positive value of sodium void reactivity worth of ABR. In the new design, sodium is replaced with liquid lead as a coolant. The lead-cooled fast reactor concept is proposed by Adamov et al') as an inherently safe fast reactor. In the lead-cooled ABR with MA nitride fuel(L-ABR), coolant void coefficient is always negative and void coefficient is negative.

The fuel concepts of L-ABR and P-ABR are shown in Fig.2. The reactor design parameters of *modified* ABRs are *summarized* in Table 2 The neutron spectra are slightly softer than those of original design.

As far as the Doppler effect is concerned, no improvement was attained because ^{238}U is not contained in the fuel and the neutron spectrum is very hard.

4. Comparison of MA transmutation in ABRs and in power reactors

In Table 3, the transmutation characteristics are compared between two types of ABRs together with thermal and fast reactors. For the MA transmutation in power reactors, the concentration of MA is assumed as 0.2% and 5% of heavy metal for U-PWR and fast reactors, respectively so that the addition of MA will not affect major reactor design parameters such as enrichment, coolant void coefficient etc.

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 arty nuclear reaction such as fission, neutron capture, (n,2n), etc. can be used for transmutation and the conversion of ^{237}Np into ^{238}Pu is a part of transmutation of Np. The MA burnup ratio is defined as the ratio of MA weight fissioned during the irradiation 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.

The transmutation and the burnup ratios of power reactors in Table 3 are the net ones after the MA generation in fuel being deducted. The negative value of burmtp mtio of U-PWR implies the more MA generation than MA burnup. The large discrepancy between transmutation ratio and burnup mtio in power reactors indicate the larger conversion of Np into Pu than in ABRs. The net MA burnup per lGWt a year of ABRs is significantly larger than that of power reactors because in ABRs major fuel material is MA.

5. Impact of MA transmutation in power reactors on fuel cycle facilities

For the MA transmutation using power reactors, not only reactor performance and fuel manufacturing but also the influence of transmutation on the fuel cycle facilities should be taken into account. The large difference between the transmutation ratio and the burnup ratio of power reactors shown in Table 3 indicates the larger conversion of MA to heavier nuclides than fission of MA.

To evaluate the effect of MA addition to the fuel, the analysis was carried out to calculate the increase of decay heat, neutron emission, and y-ray intensity. In Table 4, the effect of MA addition to the fuels of power reactors, namely, U-PWR, MOX-PWR(Pu usage in PWR) and FBR, is summarized. In this analysis, the effect of MA addition was calculated for both fresh and spent fuel. The amount of MA addition to the fuel is limited so as not to lead to significant change of major reactor parameters which are economically optimized The fraction of MA in the fuels of U-PWR, and MOX-PWR are 0.2. 0.5 and 5 weight % of heavy metal, respectively, in this calculation. The increase of decay heat is caused by the generation of ^{244}Cm and the increase of neutron emission is caused by the generation of higher Cm isotopes and ^{252}Cf.

The problem associated with ^{208}Tl build up was pointed out because of its high energy y -ray (2.6MeV).[5] ^{208}Tl builds up in the Th decay chain starting from Np-236 which is formed by (n,2n) reaction of Np-237. When 5wt% MA is added to the MOX-FBR fuel, the ^{208}Tl build up is about 50 times as much as that of without MA addition. At this level of the concentration, ^{208}Tl build up will pose a shielding problem for heavy metal handling after reprocessing of spent fuel..

As a result of increase of these Cm, Cf, for all of the MA transmutation scheme in power reactors shown in Table 4 the radiation shielding design change will be needed not only for the fresh fuel handling (manufacturing and transportation) but also for the spent fuel handling (transportation and reprocessing). This may cause the increase of cost of,* of electricity generation. In the case of ABRs, the shielding and the decay heat removal are much severer problem than the MA transmutation in power reactors since the concentration of MA is very high in ABRs. 'he fuel cycle facilities for ABRs, however, are very compact and the required number of these facilities is small because nitride fuel can be reprocessed by pyrochemical process and the amount of material to be handled is small compared with those of conventional fuel cycle. Therefore, the economy of MA transmutation maybe favorable for the ABRs even if the resources required to develop ABRs is *larger than* that for MA transmutation in power reactors.'[1]

6. Conclusions

The original ABR design was modified to improve safety characteristics. Nitride fuel of MA is used instead of alloy fuel because of relatively low melting point and thermal conductivity of MA alloy. Mixture of enriched uranium in MA fuel increases effective delayed neutron fraction and neutron generation time significantly. Sodium was replaced with liquid lead as coolant to avoid the large positive sodium void effect.

The MA burnup per 1GW per year is 30 to 40% smaller than those of the original ABRs because the contribution of ^{235}U fission is 30 to 40% of the total fission in the modified ABRs.

From the view points of fuel cycle facilities, MA transmutation in power reactors (LWRs and FBRs) will require the design change of the radiation shielding in the whole fuel cycle facilities because of the increase of strong neutron emitting nuclides as a result of neutron capture of MA even if MA content in fuel is marginally small from reactor physics viewpoints. Cost evaluation is needed to select a cost effective transmutation system based on the reliable database which is presently not available.

The ABR concept will enable the containment of troublesome MA in one closed site. From the economies and safety view point, the containment of MA in a closed site where the management will be easier may be more desirable than the nationwide spread of MA in power reactors.

Table 1 Reactor design parameters of Actinide Burner Reactors

	M-ABR[1]		P-ABR[2]
Fuel concept	pin-bundle		coated particle
material	IC[3]:Np-22Pu-20Zr OC:AmCm-35Pu-5Y		$(66NpAmCm\text{-}34Pu)_{1.0}N_{1.0}$
MA initial loading,[4] kg	666		2065
Np/Am,Cm/Pu	255/199/212		765/598/702
Reactor power, MWth	170		1200
Coolant material	sodium		Helium
velocity, m/s	8	total flow, kg/s	1088
		inlet pressure, MPa	10
		pressure drop, kPa	13
inlet temperature,℃	300		127
outlet temperature(core max),℃	IC:484 OC:440		340
Fuel temperature,℃ max[5]	IC:834 OC:809		722
Clad temperature,℃ max[6]	IC:517 OC:484	Frit temperature, max	560
Neutron flux, 10^{15}n/cm^2·sec	IC:4.1 OC:3.4		8.4
Neutron fluence (E>0.1MeV), 10^{23}n/cm^2	IC:2.2 OC:1.7		2.2
Core averaged mean neutron energy, keV	IC:766 OC:785		743
Reactivity (% Δk/k)			
Na-void reactivity/core	"2.52		
Doppler reactivity/core(Δt=300℃)	-0.01		-0.01
Kinetic parameters			
β_{eff}	1.55 X 10^{-3}		1.72X 10^{-3}
'P' sec	6.84X 10^{-8}		10.8 X 10^{-8}
Cycle length, full-power days[7]	730		300
MA transmutation %/cycle	26.0		25.3
MA burnup, %/cycle	17.8		17.3

1) M-ABR:MA metallic&1 burner reactor
2) P-ABR :MA particle fuel burner reactor
3) IC:Inner Core, OC:Outer Core
4) After 1st cycle,only Np,Am,Cm are added.
5) Predicted melting point of fuel 900℃ for MA alloy fuel of M-ABR
   Max. allowable temp. of fuel 727℃ (1/3 of M.P. 3000K) for P-ABR
6) MAJL allowable temp. of cladding/frit (HT-9) 650℃
7) Fuel irradiation time

Table 2 Reactor design parameters of modified Actinide Burner Reactors

	L-ABR[1]	P-ABR[2]
Fuel concept	pin-bundle	coated particle
material	$(64NpAmCm\text{-}36U^{3})_{1.0}N^{4}_{1.0}$	$(65NpAmCm\text{-}35U^{3})_{1.0}N^{4}_{1.0}$
MA initial loading,kg	918	2870
MA/U	588/330	1865I1005
Reactor power, MWth	180	1200
coolant material	Lead	Helium
Neutron flux, 10^{15}n/cm^2 · sec	3.1	6.6
Core averaged mean neutron energy, keV	700	700
Reactivity (% Δk/k)		
"coolant-void reactivity/core	-1.3	
Doppler reactivity/core(Δt=300℃)	-0.01	-0.01
Kinetic parameters		
β_{eff}	2.6X 10^{-3}	2.6X 10^{-3}
L_p, sec	1.3 x 10^{-7}	1.5 x 10^{-7}
Cycle length, full-power days	550	300
MA bump, %/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 3 Comparison of MA transmutation in various reactors

	MA Burner Reactors		Power Reactors		
	L-ABR	P-ABR	U-PWR	MOX-FBR	LMR[1]
Output (MWt)	180	1200	3410	2600	2632
Cycle length[2] (EFPD)	550	300	850	1368	900
Core averaged					
Fast neutron flux ($\times 10^{15}$ n/cm^2 . s)	3.1	6.6	0.37	4.1(3.0)*	6.1(4.1) "
Mean neutron energy (keV)	700	700	thermal	480	490
MA loaded (kg)	918	2870	180[3]	1450[3]	1200[3]
MA transmutation ratio[4] (%/cycle)	16.0	18.8	9.0	33.5	29.8
MA burnup ratio[5] (%/cycle)	8.5	11.6	-23.8	8.8	8.3
MA transmutation (kg/1GW . year)	315	349	1.7	40.9	44.5
MA burnup (kg/1GW . year)	168	215	-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) MA transmutation ratio={MA(BOC) - MA(EOC)-(MA generated)**}/ MA(BOC)

5) MA burnup ratio=(MA fissioned-MA generated) / 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 for the outer core

**) MA generated in the fuel during irradiation

Table 4 Effect of MA addtion on power reactor fuel

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]	3.6X 10^3	8.3X10^4	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)

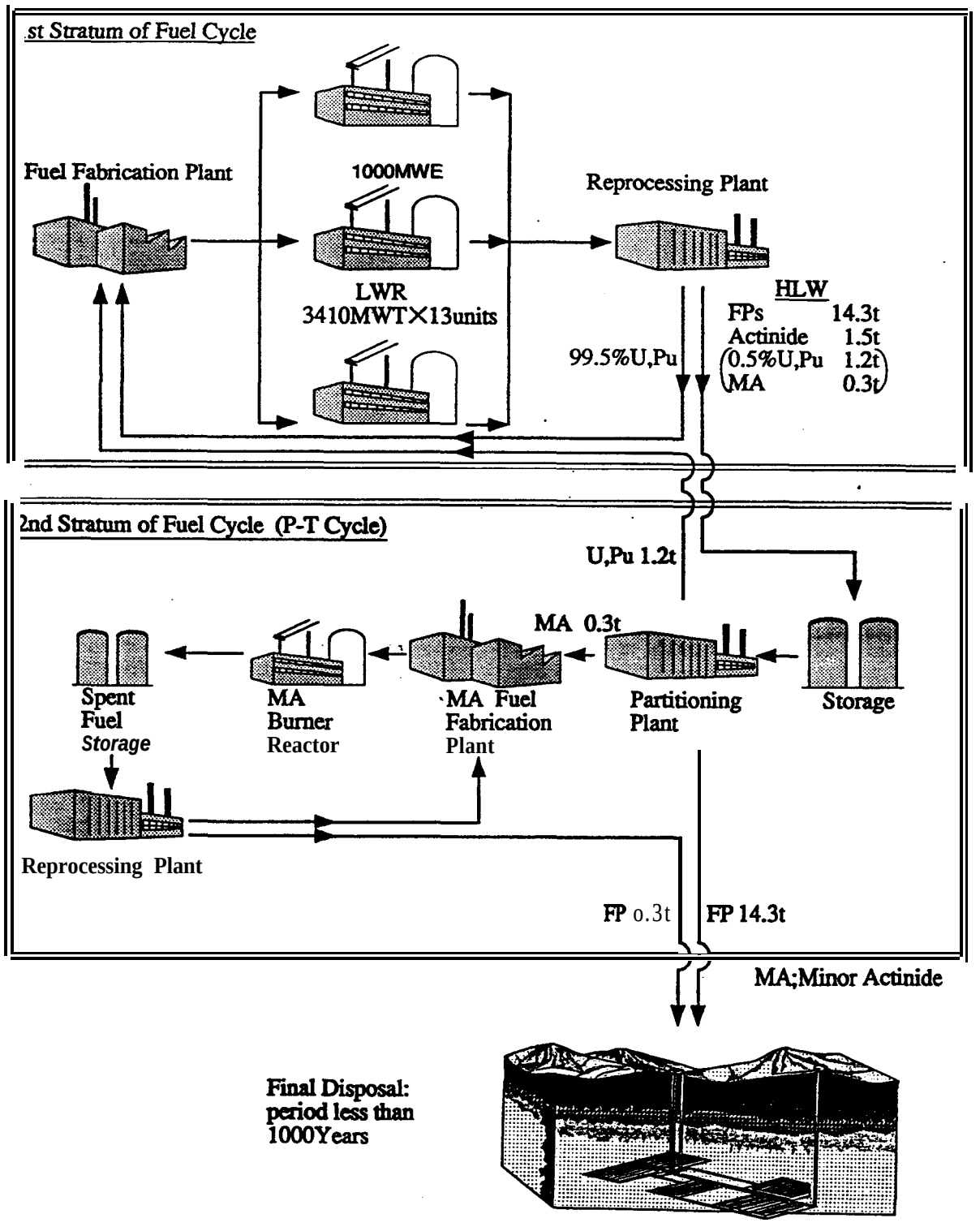

Fig. 1 Flow of high-level **radioactive waste per year** through double strata **fuel cycle combined with partioning and transmutation** (MA burner reactor) cycle ,

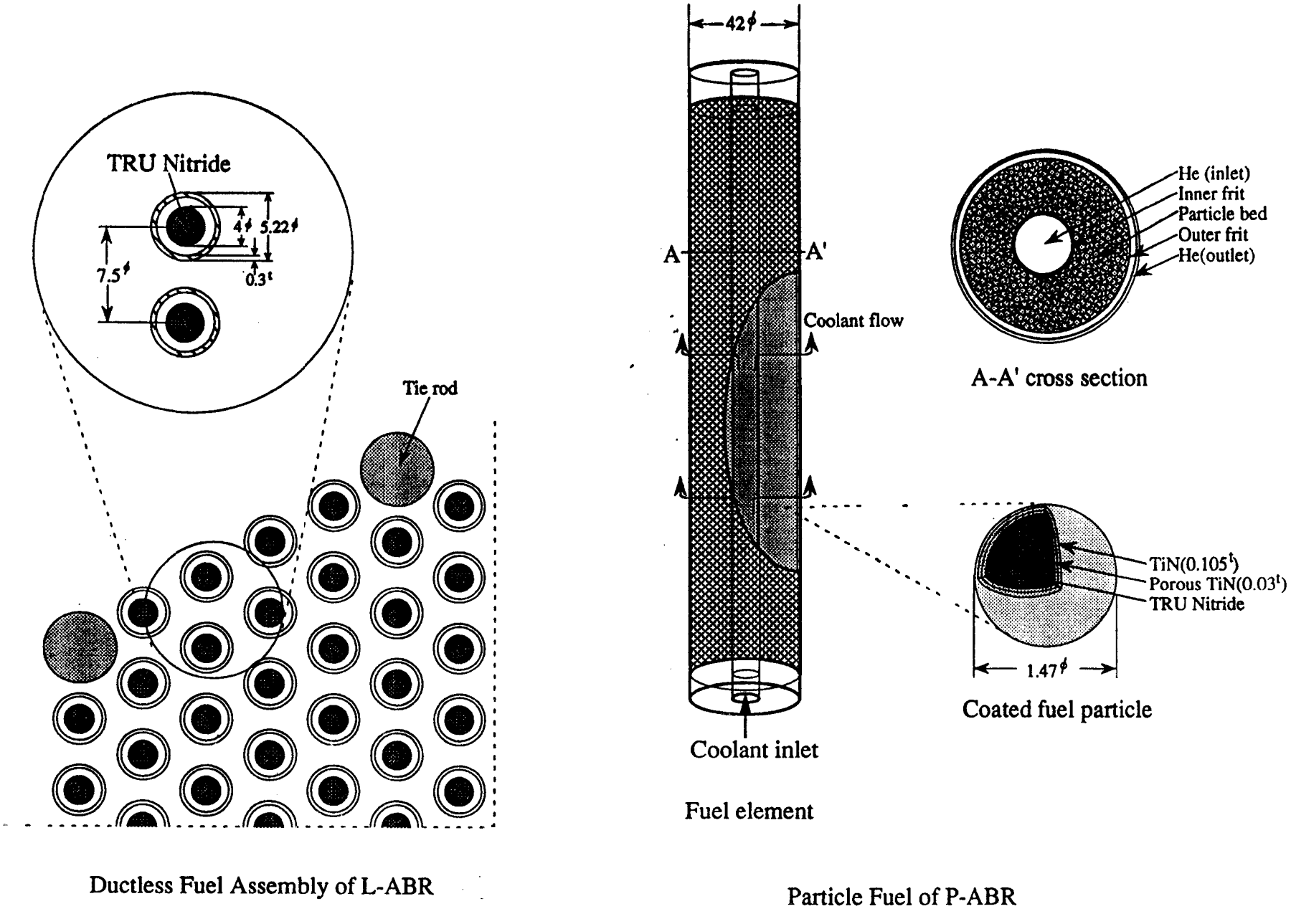

Fig. 2 Fuel Concept of ABR (unit mm)

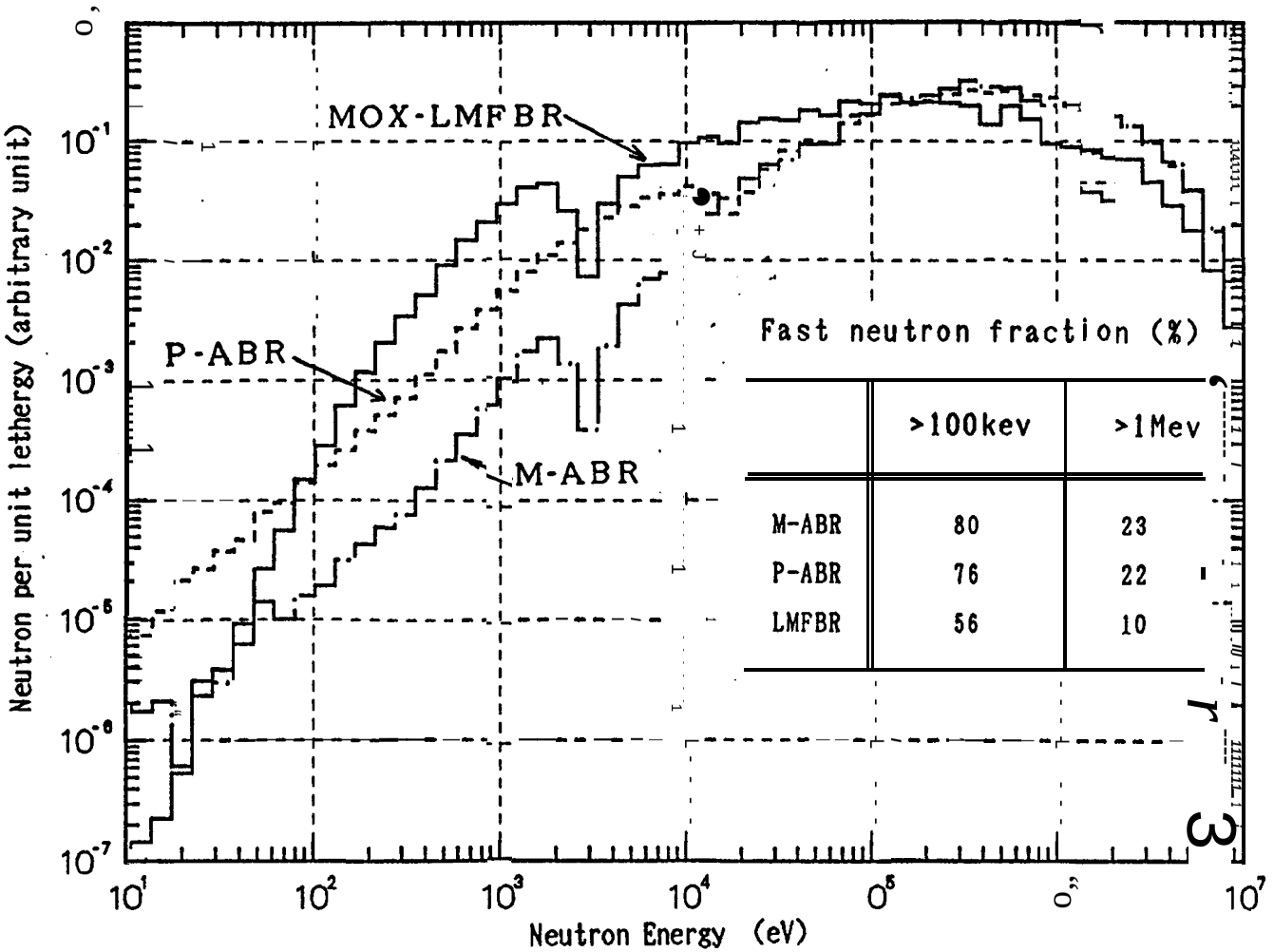

Fig.3 Comparison of core averaged neutron spectra of minor actinide burner reactors and MOX-FBR