FEASIBILITY STUDIES ON MA AND FP TRANSMUTATION IN FAST REACTORS

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Abstract

Feasibility studies have been performed to develop an optimized fast reactor core for reducing long-term radiotoxicity of nuclear waste by minor actinide(MA) and long-lived fission product(FP) transmutation , taking into consideration fuel cycle technology. Systematic parameter survey calculations were implemented to investigate the basic characteristics of MA and FP transmutation in a fast reactor core . The hybrid MA-loading method , where Np nuclide is dispersed uniformly in the core and target subassemblies containing Am, Cm and rare earth nuclides are loaded into the blanket region , has the potential to achieve the maximum transmutation of MA with no special fuel design considerations. The introduction of target subassemblies using duplex pellets - a moderator annulus surrounding a ^{99}Tc core - has a great potential to transmute long-lived fission products in the radial blanket region of the fast reactor core .

1. INTRODUCTION

One of the distinctive features of a fast reactor is its good neutron economy. Utilizing the excess of neutrons enables us to construct flexible cores such that they breed or burn plutonium in consideration of plutonium stockpile balance, and also incinerate minor actinides(MA) and long-lived fission products to reduce radiotoxicity.

Some of the MA nuclides (Np,Am,Cm) and fission products(^{99}Tc,^{129}I etc.) contained in residual waste from reprocessing have extremely long-term radiotoxicity. Partitioning and transmutation of the MA and fission products are attracting considerable attention at present as an option to reduce the long-term radiological hazard of the high-level nuclear waste. There is general agreement that the implementation of partitioning and transmutation in waste management is technically feasible.

Means of reducing the radiotoxicity of the MA nuclides are presently under investigation. The MA nuclides could produce useful energy if converted into short-lived fission products by neutron bombardment. From this standpoint, a nuclear reactor provides the obvious means for transmutation of MA nuclides. Among the various nuclear reactors, a fast reactor is considered to have the greatest potential to transmute MA effectively , because of its hard neutron spectrum(1)-(5).

The beta-emitting fission products technetium(^{99}Tc, half-life 2.13×10^5 year) and iodine(^{129}I, half-life 1.57x107 year) are among the important long-lived nuclides in high-level waste, they dominate the beta radiotoxicity for more than a million years. Transmutation of ^{99}Tc and ^{129}I by neutron capture as a result of irradiation in nuclear reactors will yield the stable isotopes ^{100}Ru and ^{130}Xe, respectively. However, due to the small neutron cross sections , the transmutation efficiency in LWRs is low. Moderated subassemblies in fast reactors are more appropriate devices for the transmutation of the fission products.

Feasibility studies have been performed to investigate the basic transmutation characteristics of MA and long- lived fission products in the fast reactor(6,7).

2. MA Transmutation

Systematic parameter survey calculations were implemented to investigate the basic characteristics (transmutation rate , burnup reactivity, Doppler coefficient, sodium void reactivity , maximum linear heat rate, etc.) of a fast reactor core with MA transmutation, the following items were considered:

(1)Study on loading method of MA in the core (homogeneous , heterogeneous , hybrid , blanket, etc.)

(2)Selection of fuel material for MA transmutation (oxide ,inert matrices such as Al_2O_3 , CeO_2 , etc.),

(3)Study on the maximum tolerable amount of rare earth (RE) nuclides ,

(4)Effect of MA recycling on core characteristics and fuel cycle system.

(1) Study on MA Loading method

Since MA loading considerably affects not only core characteristics but also fuel material properties, it is necessary to investigate MA loading methods taking into account this influence upon core characteristics and fuel material properties. Possible MA loading methods (homogeneous , heterogeneous , hybrid , blanket , etc.) were investigated for fast reactor cores with no special design adaptation for MA loading. The MA is dispersed uniformly throughout all the fuel in the core in the homogeneous method . In the heterogeneous method , a small number of fuel subassemblies with concentrated MA (target S/As) are loaded into the core. The hybrid MA loading method is a combination of the homogeneous and heterogeneous methods : the Np nuclide is loaded in the core region uniformly and a small number of subassemblies containing Am ,Cm and RE nuclides are loaded into the blanket region.

The comparison of core performance for various MA loading methods is shown in Table 1. The MA transmutation in a fast reactor core has no serious drawbacks in terms of core performance, provided that the homogeneous loading method can be employed with a small ratio of MA to fuel (~5wt%). Since a 1000MWe-class LWR produces about 26 kg of MA per year, a fast reactor with 5%wt MA loading can transmute the MA mass from six LWRs .

The heterogeneous MA loading method can be made feasible by optimizing the fuel design, loading pattern and the coolant flow of the MA-loaded fuel subassemblies. The reduction of the fuel pin diameter and the Pu enrichment is essential to reduce the power of MA-loaded fuel in the heterogeneous MA loading method.

The hybrid MA loading method can transmute a large amount of MA without serious drawbacks in terms of core performance. The transmuted mass of MA is about 530kg/cycle, which is almost 16 times the mass produced by an LWR of the same power output.

The MA loading in the blanket region causes no problems from the viewpoint of core performance. Minor actinides are transmuted at a rate of 6% per cycle in the axial and radial blanket regions.

It was found that the hybrid MA loading method , where the Np nuclide is dispersed uniformly in the core and target subassemblies containing Am, Cm and rare earth nuclides are loaded into the blanket region , has the potential to achieve the maximum transmutation of MA with no special design considerations.

(2)Selection of fuel material for MA transmutation

Different types of inert matrices , instead of UO_2 , for the heterogeneous MA-loading method have been investigated , they avoid the buildup of higher actinides via ^{238}U and achieve a high MA transmutation rate. Inert matrices of Al_2O_3 and CeO_2 were examined in this study. The MA transmutation rate of the target subassembly using inert matrices is larger than that of the target subassembly using UO_2 . The use of inert matrices in the target subassembly effectively increases the MA transmutation rate.

(3) Study on the permissible RE level in homogeneously loaded MA

Systematic parameter survey calculations were performed to investigate the basic characteristics of a fast reactor core loaded homogeneously with MA which contains RE , and also to establish a MA and RE loading method which has no serious influence on the core design. The homogeneous loading of MA and RE has no serious effects on the reactor core performance , provided that the amounts of MA and RE in the fuel are less than 5 and 10wt% respectively . In the case of adding Am,Cm and RE in the radial blanket region , it is possible , from the viewpoint of core performance , to insert ~50wt% of Am and Cm, and ~50wt% of RE in the target assemblies , as is shown in Table 1.

(4)Effect of MA recycling on core characteristics and fuel cycle system.

The effects of MA recycling on the core characteristics and the fuel cycle system in the homogeneous loading method were evaluated . The absolute value of the Doppler coefficient is increased by MA recycling , the value at the 8th recycle is ~14% larger in comparison with that in the initial core , as is shown in Table 2 . This is caused by the reduction in Pu enrichment with MA recycling , this increases the resonance absorption of ^{238}U. Sodium void reactivity decreases with MA recycling , and the value at the 8th recycle is ~7% smaller than that in the initial core. The recycling of MA in a fast reactor is feasible from neutronic and thermal-hydraulic points of view . However, during multi-recycling the Np fraction is significantly reduced compared to the unirradiated feed, and the fraction of Cm is greatly increased because of neutron capture in Am. The accumulation of Cm as a result of the MA recycling will bring about some problems concerning fuel handling and reprocessing , because of an increase in both the decay heat and the neutron emission rate from ^{244}Cm .

3. FP Transmutation

To calculate the transmutation rate of ^{99}Tc in a neutron flux spectrum it is insufficient to account for the thermal neutron capture only ; the epithermal part of the neutron spectrum also has a contribution. There is a large resonance peak at 5.6eV and a series of minor resonances between 10 and 100eV. This suggests that a neutron spectrum where there is more absorption in the resonance region than in the thermal region is advantageous in order to increase the absorption rate of ^{99}Tc. This is because such a spectrum helps to suppress absorption by structural materials. Therefore , the appropriate loading mass of moderator depends upon its moderating power.

Systematic parameter survey calculations were performed to investigate the basic characteristics of FP(^{99}Tc) transmutation in the blanket region of a fast reactor. A moderated target subassembly was used for ^{99}Tc transmutation. The subassembly consists of moderator pins containing $ZrH_{1.7}$ and ^{99}Tc target pins distributed between the moderator pins. The moderated target subassemblies were loaded in the radial blanket region of the fast reactor core. The arrangement of the moderator and the target pins in the subassembly , the volume ratio of target to moderator and the moderator materials were selected as parameters in the present study. A new concept of duplex pellet - a moderator annulus surrounding a ^{99}Tc core - for ^{99}Tc transmutation was also adopted in the present study to get a better transmutation performance.

The core configuration and main parameters of the 600MWe-class fast reactor core used in the study are shown in Fig. 1. The arrangements of the moderator pins and the FP pins in the subassembly are shown in Fig. 2. The Monte Carlo computer code for neutron photon transport (MVP)(8) was adopted for FP transmutation calculations in the moderated target subassembly because of its versatility and comprehensive geometry features.

The dependence of ^{99}Tc transmutation performance on the number of FP pins in the 127-pin target assembly is shown in Table 3. The transmutation rate of ^{99}Tc increases as the number of FP pins in the target subassembly decreases , because the moderating power increases.

The sensitivity of the ^{99}Tc transmutation performance to the neutron spectrum in the target subassemblies was calculated by changing the moderator materials($ZrH_{1.7}$, BeO , Al_2O_3 , SiC). Table 4 shows the results of ^{99}Tc transmutation performance for various moderator materials. Neutron spectra and capture reaction rate distributions of ^{99}Tc in the FP pin region for various moderator materials are shown in Fig. 3 and 4 , respectively. It was found that the moderating power of $ZrH_{1.7}$ and of BeO is better than that of Al_2O_3 or SiC for ^{99}Tc transmutation.

The FP transmutation performance of a new target subassembly concept using duplex pellets consisted of ^{99}Tc and $ZrH_{1.7}$ was investigated. Configurations of moderated target subassemblies using duplex pellets are shown in Fig. 5. The results of the calculations are shown in Table 5. The transmutation rate of ^{99}Tc in the new target subassembly is larger than that in the subassembly consisting of separate ZrH1.7 moderator pins and ^{99}Tc target pins as shown in Fig. 2. As a result of the present study , a maximum ^{99}Tc transmutation rate of about 10%/year was obtained by using the new target subassembly loaded in the blanket region of the fast reactor . The new target subassembly can achieve the optimum transmutation performance by adjusting the volume ratio of ZrH1.7 and ^{99}Tc in the duplex pellet.

The effects of loading target subassemblies on main core characteristics were also analyzed. It was found that the power density of the core fuel adjacent to the target is rather high and is about the same as the maximum in the core. However , the power spike is much mitigated compared to the case of loading target subassemblies in the core region.

4. CONCLUSION

Feasibility studies have been performed to investigate the basic characteristics of MA and FP transmutation in fast reactors , and also to clarify the feasibility of MA and FP transmutation .

MA transmutation in a fast reactor core has no serious drawbacks in terms of core performance, provided that the homogeneous loading method can be employed with a small fraction of MA in the fuel (~5wt%). The hybrid MA-loading method , where Np nuclide is dispersed uniformly in the core and target subassemblies containing Am, Cm and rare earth nuclides are loaded into the blanket region , has the potential to achieve the maximum transmutation of MA with no special fuel design considerations.

The introduction of target subassemblies using duplex pellets - a moderator annulus surrounding a ^{99}Tc core - gives the maximum transmutation rate of Tc-99 in the radial blanket region of the fast reactor core.

It was found that the fast reactors have an excellent potential for transmutating MA and FP effectively. However, much research needs still to be done to improve the partitioning of the radionuclides as well as to resolve the technological problems of the transmutation, to make the process industrially attractive.

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Table 1 Comparison of Core Performance for Various Ma Loading Methods

Item	Reference (No MA)	Homo. Loading	Hetero. Loading	Homo. Loading	Hybrid Loading	Hybrid Loading
MA and RE Loaded in the Core Region	-	Np,Am,Cm :5% RE : 0%	Np,Am,Cm :49% RE : 0% (Number of target S/A:39)	Np,Am,Cm :5% RE : 10%	Np : 9.8% RE : 0%	Np : 9.8% RE : 0%
MA and RE Loaded in the Blanket Region(Target)	-	-	-	-	Am,Cm: 46% RE : 46% (Number of target S/A:72)	Am,Cm: 46% RE : 46% (Number of target S/A:72)
Matrix of Target			UO_2		UO_2	Al_2O_3
Pu Enrichment (wt%)(IC/OC)	15.4 /18.6	16.6 /20.1	15.4 /18.6	20.0 24.2	19.0 23.4	19.0 23.4
Burnup Reactivity (%Δk/kk')	3.31	2.12	1.83	3.71	0.87	0.90
Max.Linear Heat Rate(w/cm) (Driver/Target)	420	407	439 /309	413	405 /217	406 /174
Na Void Reactivity	1.0	1.3*	1.3*	1.4*	1.5*	1.5*
Doppler Coefficient	[illegible]	0.6*	0.7*	0.5*	0.45*	0.45*
MA Tran						
Amount(kg/cycle)	-	172	186	164	514	529
Rate(Core)(%/cycle)	-	10.9	11.3	10.3	13.9	14.0
Rate(Blanket) (%/cycle)	-	-	-	-	2.9	3.3

* Relative Value

Table 2 Effect of MA Recycling on Core Performance (Homogeneous Loading Method)

Item	Reference (InitialCore)	4th Recycle	8th Recycle
MA(wt%)	5	5	5
Pu Enrichment (wt%)	17.8(Inner) 21.6(Outer)	17.6(Inner) 21.3(Outer)	16.9(Inner) 20.4(Outer)
Burnup Reactivity (%Δk/kk')	1.6	0.4	0.5
Na Void Reactivity	1.0 (Ref.)	0.96 (Relative Value)	0.93 (Relative Value)
Doppler Coefficient	1.0 (Ref.)	1.08 (Relative Value)	1.14 (Relative Value)
MA Transmutation Rate(%)	10.3	10.5	10.1

Table 3 Dependence of ^{99}Tc transmutation performance on the number of FP pins in the 127-pin target assembly.

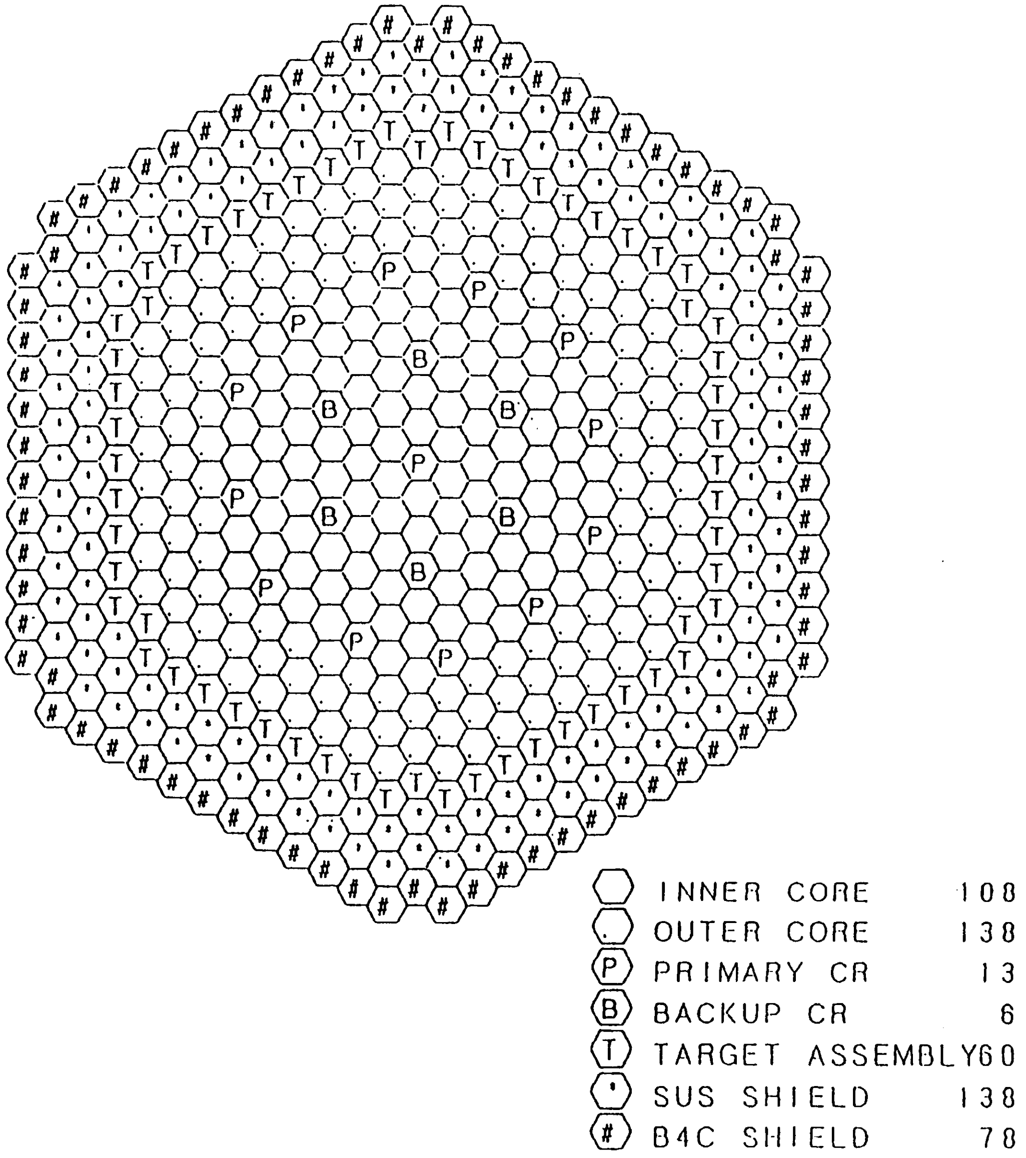
No.of FP pins	Radius of FP pins (cm)	V_{fp}/V_{mod}	Transmutation Rate (%/y)	Transmuted ^{99}Tc amounts(kg/y)	Loaded ^{99}Tc amounts (kg)
52	0.5	0.69	1.5	47.0	3170
37	0.5	0.41	1.8	41.1	2250
27	0.5	0.27	2.1	34.4	1640
22	0.5	0.17	2.5	27.2	1100

Table 4 Difference of ^{99}Tc transmutation performance on moderator material

Moderator	Transmutation Rate(%/y)	Transmuted FP amounts(kg/y)
$ZrH_{1.7}$	2.1	34.4
BeO	2.2	36.0
Al_2O_3	1.7	28.0
SiC	1.6	27.0

Table 5 Results of parameter survey calculations on ^{99}Tc transmutation performance using duplex moderator pins.

No.of duplex pins	FP pin Radius (mm)	Transmutation Rate (%/y)	Transmuted amounts(kg/y)	Loaded amounts(kg)
127	2.00	3.5	38.1	1101
127	0.63	9.8	10.8	110
217	2.00	2.5	46.7	1882
217	0.63	9.1	17.1	188
37	5.30	1.9	41.8	2253
37	0.63	10.1	2.9	29

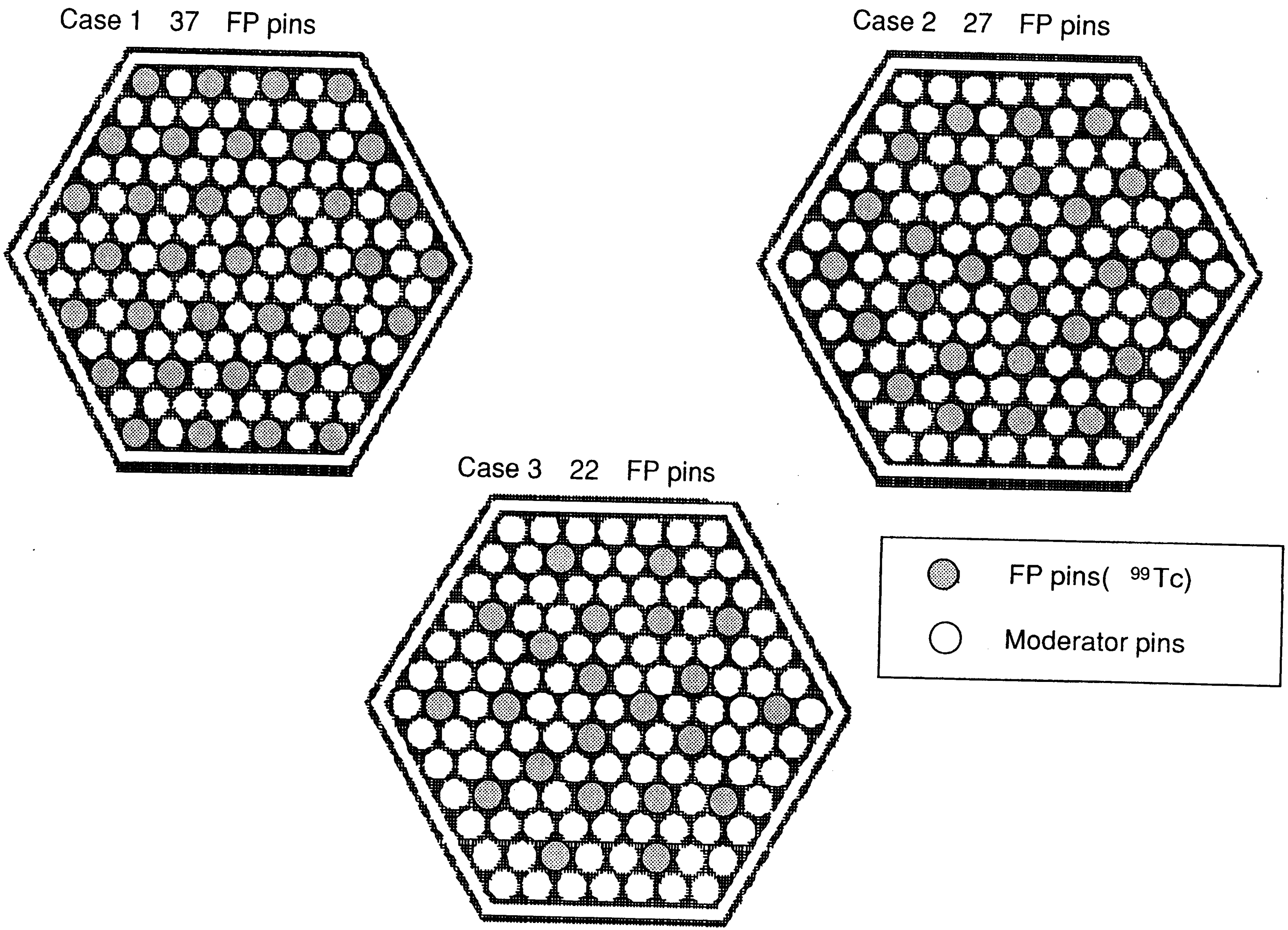


Reactor Power	1600MWth
Core Height	100cm
Axial branket	35cm
Core Diameter	275m
Fuel Type	MOX
Pu Enrichment	15.30/18.90
Pu composition 8/9/0/1/2	3/53/25/12/7
Core Pitch	160.7mm

Volume Fraction

	Fuel	Gap	SUS	Sodium	Absober
Core	38.2	5.1	22.4	34.3	-
PCR	-	5.2	15.6	45.6	33.6
BCR	-	5.2	15.6	15.6	33.6
Shielding	-	-	80.0	20.0	

Fig. 1 Core layout and main parameters



Case 1 37 FP pins

Case 2 27 FP pins

Case 3 22 FP pins

Fig.2 ^{99}Tc pin arrangements in Target Assembly (based on 127-pin assembly)

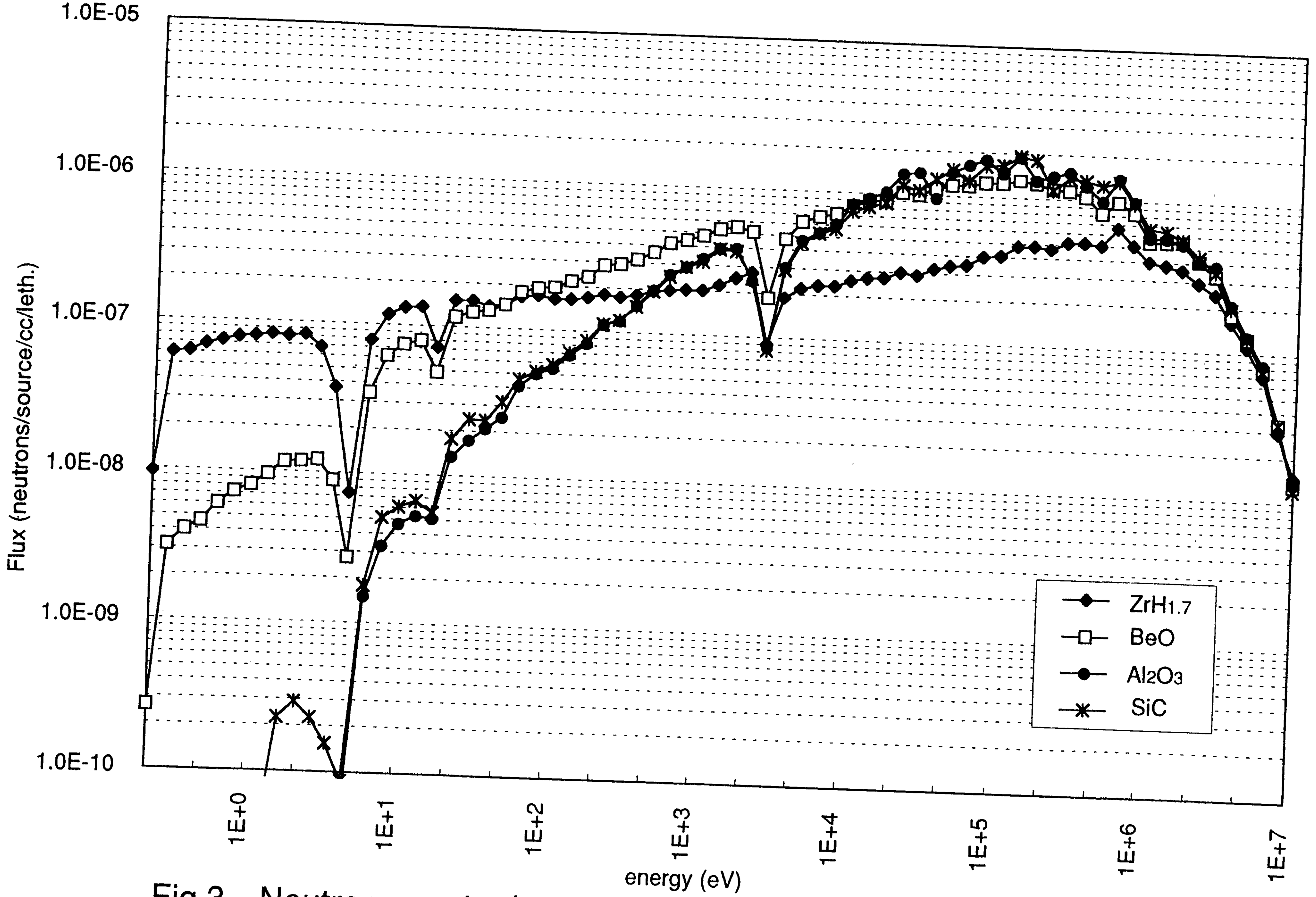


Fig.3 Neutron spectra in ^{99}Tc region for various moderator materials (27 FP pins in 127 pins model)

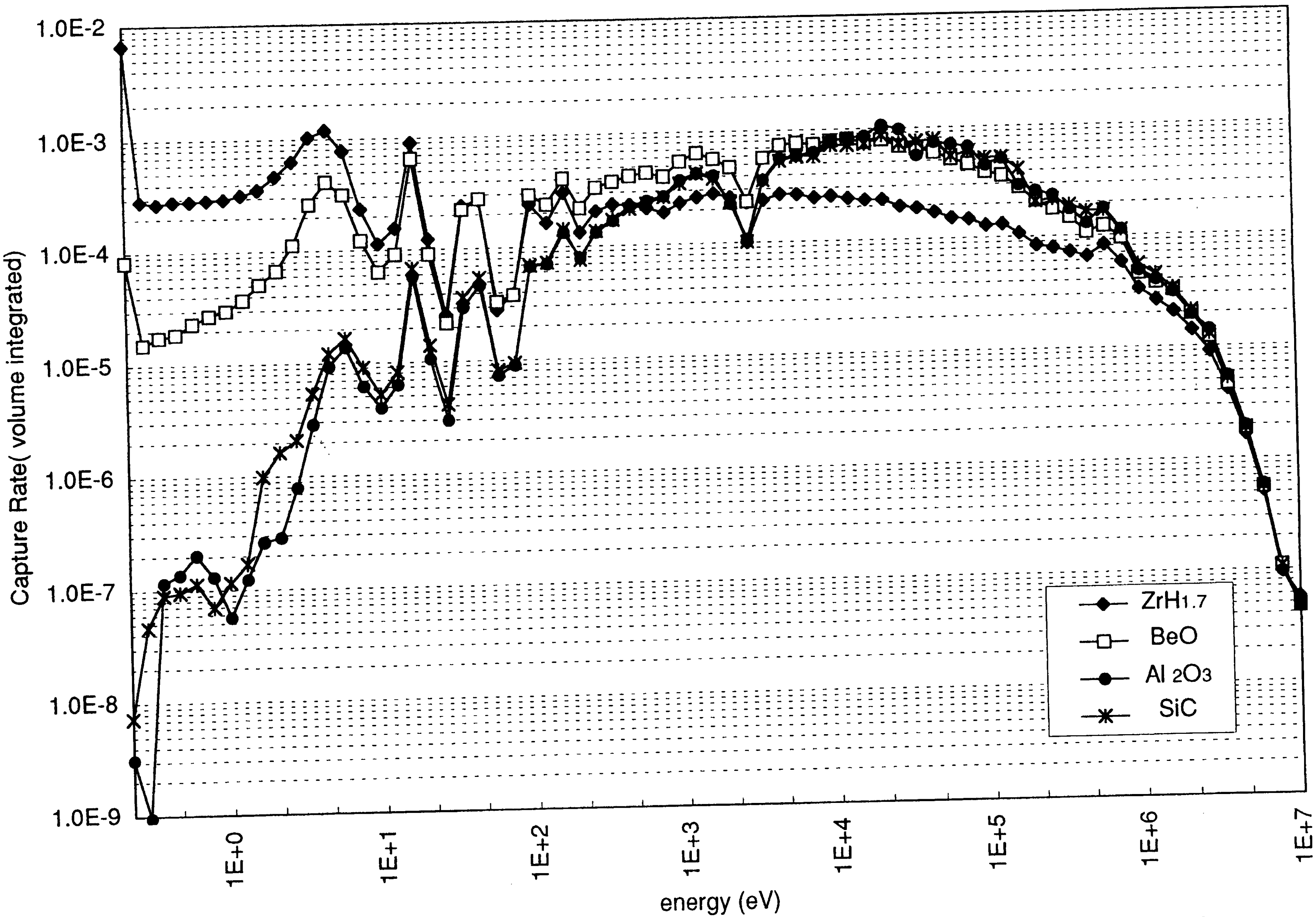
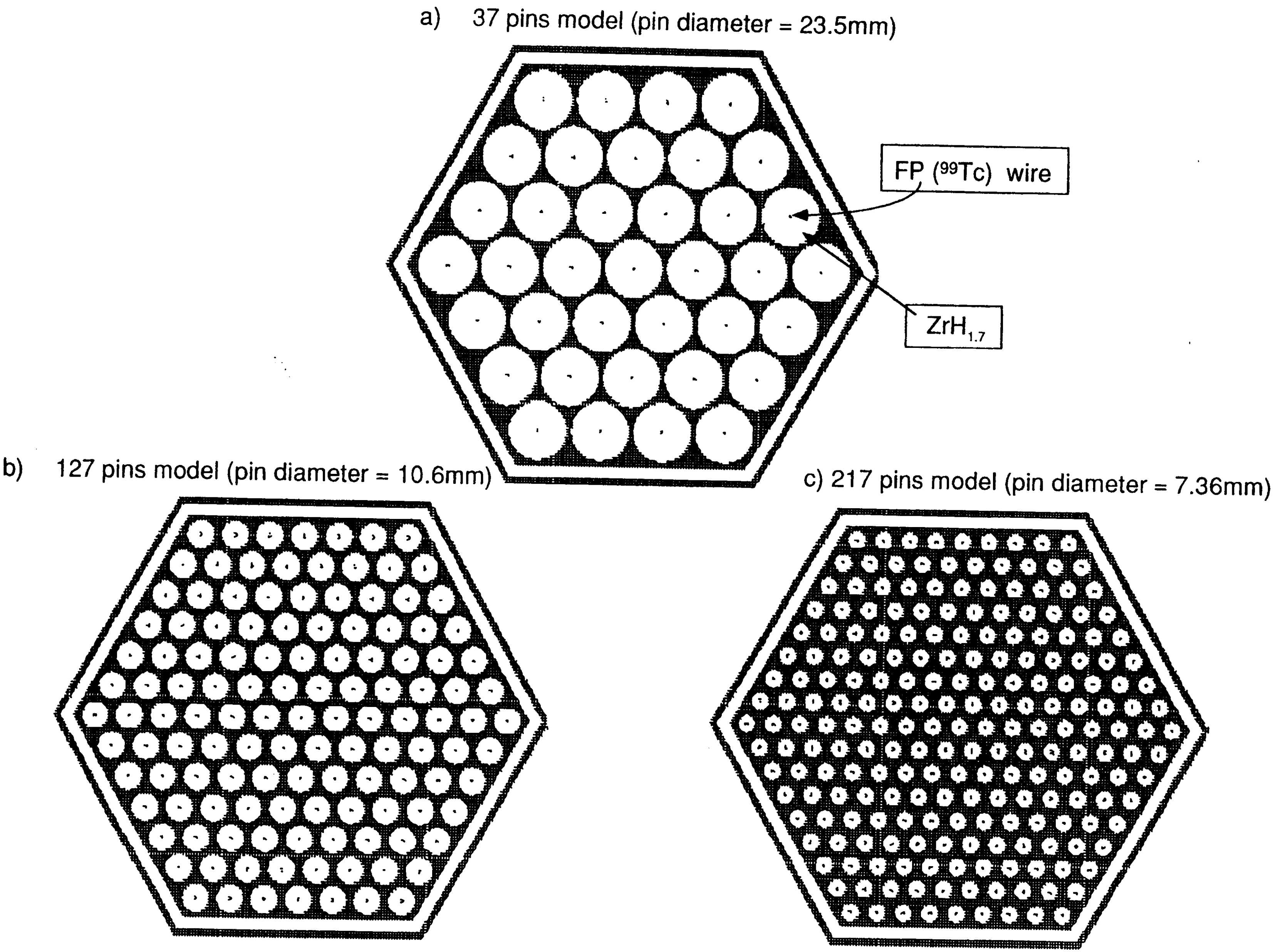


Fig. 4 Capture Rates in ^{99}Tc region for varions moderator materials
(27 FP pins in 127 pins model)



a) 37 pins model (pin diameter = 23.5mm)

b) 127 pins model (pin diameter = 10.6mm)

c) 217 pins model (pin diameter = 7.36mm)

Fig. 5 Duplex pin arrangements in Target assembly (Radius of FP(^{99}Tc)= 0.6mm or 2.0mm)