A Study on the Transmutation Capability of Accelerator Driven System

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A preliminary investigation was performed to evaluate the capability of accelerator driven thermal neutron system for the transmutation of minor actinides and fission products. Although the accelerator driven thermal neutron system was introduced long before by LANL, there has not been that much of research on minor actinide and fission product burning. LANL has focused its research mainly on the plutonium destruction.
For the analysis, 1GeV, 20mA proton linac was employed and the LAHET code developed by LANL was adopted for the simulation. The systems are molten salt fluoride with graphite moderator and in order to keep the system subcriticality 0.95, the ratio of plutonium to MA was adjusted. For the depletion calculation, ORIGEN2 code was used and systemwise one group cross section was generated by HMCNP.
The system was believed to have the neutronic characteristics something between LWR and FBR. The system was found to have about 14 %/yr transmutation rate for MA with a capacity factor 0.8. However it showed negligible capability for FP.

I. Introduction

Korea Atomic Energy Research Institute (KAERI) has performed a transmutation research since the middle of 1992. It had finished its first stage research in July of 1996 and obtained the tentative conclusion on its future transmutation research direction.

KAERI tried to estimate which option would be the optimal transmutation technology among LWR, FBR, accelerator-driven subcritical reactor concepts. The estimation works were done by evaluating the transmutation capability, the technical feasibility, the resistance to NPT(the possibility of commercialization), the safety, the economical advantages of each concept. A spent fuel problem is not the problem of one or two countries any more. It is the problem of world because nuclear power has been a worldwide energy source. In order to solve that worldwide problem, the technology to be developed should have something that any country can access and employ. From those points of view, weightings were assigned to the NPT resistance, transmutation capability, safety, technical feasibility, economical benefits in descending order. As results, the optimal concept was determined to be an accelerator driven subcritical reactor.

The detail neutronic analysis for an accelerator driven system will be done throughout the second stage research period from 1996 to 1999. No specific design parameters were determined yet. As a starting point, the thermal neutron system with molten salt fuel was selected. In this paper, some basic results obtained from the preliminary studies on the selected system was presented.

II. System Model Description

A program called KOMAC(KOrea Multipurpose linear ACcelerator) is under planning to build 1.0 GeV, 20mA linear proton accelerator in KAERI. KOMAC will be developed in a way to be applicable to transmutation. The proton beam of 1GeV, 20 mA was assumed for the spallation calculation. Basic concepts for the thermal system were derived from the ATW designed by LANL. Fig. 1 shows the schematic layout of the subcritical blanket and Table 1 represents the material specifications of the system. Lead target with the diameter 75cm and the height 100 cm was employed to get the neutrons induced by spallation reaction and graphite was adopted for neutron moderation. Because of very low solubility of TRU in molten fluoride, 1.0 mol% of TRU was assumed and the chemical form of fuel was $66LiF\text{-}33BeF_2\text{-}1(TRU)F_4$. The reason using fluoride instead of chloride is to enhance the neutron moderation. The operating temperature was assumed to be 650°C and the physical density of molten salt was at the operating temperature. Because a thermal neutron is the most effective tool for fission product burning, a couple of sites were assigned for fission product incineration in the graphite region.

III. Calculational Methodology

LCS(LAHET code system) developed by LANL was employed to perform the neutronic analysis in the sample model. LAHET and HMCNP are the main codes in LCS.[1] Fig. 2 shows the input-output flow between codes. By adjusting the amount of Pu in total TRU, the subcriticality was kept to be 0.95. LAHET code analyzes the spallation reactions and deals with high energy neutrons(>20MeV).[2] LAHET provides HMCNP with a sort of fixed external neutron sources(energy and location). HMCNP performs the calculation of neutron flux and thermal power in the blanket.[3] HMCNP generated one group, corewise cross sections to be used for the calculation of transmutation capability using ORIGEN2.[5]

IV. Results

By using the kcode function in MCNP code, the nuclide composition of TRU to keep the system subcriticality 0.95 was searched. In that condition, the nuclide composition of Pu and MA was found to be 72:28 and the loading amount of MA was 775 kg.

The isotope composition of lead target was that of natural lead(Pb-204:Pb-206:Pb-207:Pb-208=2:24:22;52). The neutron production rate depends on the isotope composition and the size of the target. The more stable the nuclide is, the less the neutron production rate is. The neutron production rate was 34n/p in the proposed system and Fig. 3 shows the number of neutrons produced by the spallation reaction versus its energy. The figure shows that the most of spallation neutrons have the energy ranging from 2MeV to 10 MeV which is much higher than the fission neutron energy. A considerable amount of heat is generated by spallation reaction. In the proposed model, 1.3MW was deposited in the target.

As it is expected, maximum flux occurs at the target region. The average neutron flux in the target is about 80 ~ 90 times higher than that of blanket while the neutron flux of the reflector is 10 times less than that of blanket. Table 2 gives the neutronic parameters generated. For a given condition, the system produced the thermal power of 967 MW. Fig.4, 5 show the neutron energy spectrum in the blanket, and graphite region, respectively. In order to evaluate the transmutation capability, one group cross sections for 21 actinides and 17 fission product nuclides were produced using the average neutron energy spectrum in the blanket and reflector region, respectively. Table 3, 4 represent the transmutation capability of the system for actinides and fission products, respectively. The proposed system is supposed to transmute MA with the rate of 14%/yr when a capacity factor is assumed to be 0.8. On the other hand, it has negligible capability for fission product burning.

V. Discussion and Conclusion

A preliminary calculation was done to evaluate the neutronic performance of the accelerator driven thermal neutron system. Because the criticality of the system was 0.95, about 5% of neutrons were come from the spallation reaction. Therefore the average source neutron energy is supposed to be somewhat higher than the fission neutron energy. In addition, the moderator was positioned in a way to surround the blanket like a reflector. Such an array can not moderator the high energy neutron efficiently. As results, the neutron energy spectrum of the system was found to be something between LWR and FBR. Epithermal neutron can be suggested to be more effective for the transmutation. It might be true for some nuclides but not for some others. Table 5 shows one group absorption cross sections of some important long lived fission products. This table tells that the proposed system is more efficient for burning I-129 while LWR is for Tc-99. The system was believed to have a considerable capability for MA transmutation. On the other hand, it had negligible for fission products. The main reason for that was due to the difference in the neutron flux. Therefore FP burning sites should be moved to somewhere a high neutron flux can be obtained. As it is known, a thermal neutron has an advantage of higher MA transmutation capability and a disadvantages of producing lots of higher actinides while a fast neutron has a reverse characteristics. Further detail studies should be done to decide which neutron is more efficient for the transmutation. Also some attention has to be paid on the definition of the transmutation when only MA is considered to be transmuted. In other words, it has to be answered whether it is transmutation or not when Np-237 is transformed to Pu by the successive absorption.

References

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[3] Judith F. Briesmeister, MCNP-A General Monte Carlo N-Particle Transport Code Version 4A, LA-12625-M.(1993)

[4] Carl A. Beard et al., Parameteric System Studies of the Aqueous-based (slurry) Balnket Concept for Accelerator Transmutation of Waste, Nucl. Tech. Vol. 110 (1995)

[5] A. G. Groff, A User Manual for the ORIGEN2 Computer Code, ORNL/TM-7175, ORNL(1980)

[6] Judith A. Benzdecny et al., Preliminary Analysis of the Induced Structural Radioactivity Inventory of the Base-case Aqueous Accelerator Transmutation of Waste Reactor Concept, Nuclear Technology, Vol 110(1995)

Table 1 Material Specifications for the proposed system

Component	Specifications
Target	• Material : Lead • Density : 11.34 g/cm^3 • Nuclide Composition : Pb-204:206:207:208=2:24:22:52
Fuel	• Chemical Form : Molten Salt (66LiF-33BeF$_2$-(TRU)F$_4$) • Density : 2.06 g/cm^3 • TRU Composition : Pu/MA = 72/28
Moderator	• Material : Natural Graphite • Density : 2.25g/cm^3
Target and Blanket Container	• Material : HT-9 • Density : 7.75 g/cm^3 • Composition : Fe: Ni : Cr: Mo: Mn = 67.50:11.25:17.0:2.25:2.0 (at./wt.%)

Table 2 Some Neutronic Parameters

Parameter	Target Region	Blanket Region	Graphite Region
Spallation Neutron	34 n/p	negligible	negligible
Avg. Flux(10^{14}n/cm^2-sec)	656.0	8.2	0.88
Thermal Power	4.8	966.6	0.05

Table 3 Transmutation Rate for MA

(unit : kg , kg/yr)

Nuclide	Loading Amount	After-30-days	Amount[a)] (kg/yr)	Rate[b)] (%/yr)
^{237}Np	3.419E+02	3.296E+02	-1.476E+02	-43
^{238}Np	0.000E+00	1.231E+00	1.477E+01	-
^{238}Pu	3.062E+01	4.163E+01	1.321E+02	431
^{239}Pu	1.155E+03	1.113E+03	-5.040E+02	-44
^{240}Pu	5.269E+02	5.335E+02	7.920E+01	15
^{241}Pu	1.701E+02	1.715E+02	1.680E+01	10
^{242}Pu	9.945E+01	1.016E+02	2.580E+01	26
^{241}Am	3.710E+02	3.473E+02	-2.844E+02	-77
^{242m}Am	0.000E+00	9.869E+00	1.184E+02	-
^{242}Am	0.000E+00	4.311E-01	5.173E+00	-
^{243}Am	6.119E+01	5.492E+01	-7.524E+01	-123
^{242}Cm	0.000E+00	1.038E+01	1.246E+02	-
^{243}Cm	1.950E-01	2.144E-01	2.328E-01	119
^{244}Cm	1.147E+01	1.959E+01	9.744E+01	850
^{245}Cm	6.290E-01	1.062E+00	5.196E+00	826
U	0.000E+00	3.045E-02	3.654E-01	-
Np	3.419E+02	3.308E+02	-1.332E+02	-39
Pu	1.982E+03	1.961E+03	-2.520E+02	-13
Am	4.321E+02	4.126E+02	-2.340E+02	-54
Cm	1.230E+01	3.125E+01	2.274E+02	1849
MA*	7.863E+02	7.747E+02	-1.398E+02	-18
TRU**	2.768E+03	2.736E+03	-3.918E+02	-14
ACT***	2.768E+03	2.736E+03	-3.914E+02	-14

* Total minor actinide nuclides, ** Total transuranium nuclides, *** Total actinide nuclides.

a) (loading amount - after-30day amount)*365/30.

b) (transmutation amount/loading amount) *100.

Table 4 Transmutation Rate of Fission Products

(unit : kg, kg/yr)

Nuclide	Loading Amount	After-1-year Amount	Amount[a)] (kg/year)	Rate[b)] (%/year)
^{99}Tc	2.60E+02	2.59E+02	-6.00E-01	-0.23
^{100}Ru	0.00E+00	5.64E-01	5.64E-01	-
^{127}I	1.51E+01	1.51E+01	-2.00E-02	-0.13
^{128}Xe	0.00E+00	1.63E-02	1.63E-02	-
^{129}I	4.87E+01	4.86E+01	-6.00E-02	-0.12
^{130}Xe	0.00E+00	6.07E-02	6.07E-02	-
^{133}Cs	3.14E+01	3.13E+01	-9.00E-02	-0.29
^{134}Cs	0.00E+00	7.43E-02	7.43E-02	-
^{134}Ba	0.00E+00	1.32E-02	1.32E-02	-
^{135}Cs	8.36E+00	8.36E+00	-4.00E-03	-0.05
^{136}Ba	0.00E+00	4.49E-03	4.49E-03	-
^{137}Cs	2.65E+01	2.59E+01	-6.00E-01	-2.27
Tc	2.60E+02	2.59E+02	-6.00E-01	-0.23
Ru	0.00E+00	5.65E-01	5.65E-01	-
I	6.38E+01	6.37E+01	-8.00E-02	-0.13
Xe	0.00E+00	7.70E-02	7.70E-02	-
Cs	6.62E+01	6.56E+01	-6.20E-01	-0.94
Ba	0.00E+00	6.22E-01	6.22E-01	-
Total LLFP*	3.90E+02	3.89E+02	-1.30E+00	-0.33
Total FP*	3.90E+02	3.90E+02	-3.62E-02	-0.01

* Total amount of long-lived fission products.

** Total amount of fission products.

a) (loading amount - after-30day amount)*365/30.

b) (transmutation amount/loading amount) *100.

Table 5 Absorption Cross Section Comparison for Major FPs

(unit : barn)

Nuclide	Absorption Cross Section		
	Proposed System	PWR	FBR
^{99}Tc	6.910E+00	9.136E+00	4.767E-01
^{127}I	5.984E+00	4.846E+00	5.450E-01
^{128}Xe	2.547E+00	6.541E-01	1.709E-01
^{129}I	7.140E+00	3.225E+00	3.757E-01
^{130}Xe	1.518E+00	6.260E-01	1.074E-01
^{131}Xe	4.881E+01	3.046E+01	2.130E-01
^{133}Cs	1.296E+01	1.072E+01	4.845E-01
^{134}Cs	3.330E+01	1.675E+01	5.366E-01
^{135}Cs	4.085E+00	2.391E+00	7.307E-02
^{137}Cs	3.843E-02	2.559E-02	1.303E-02

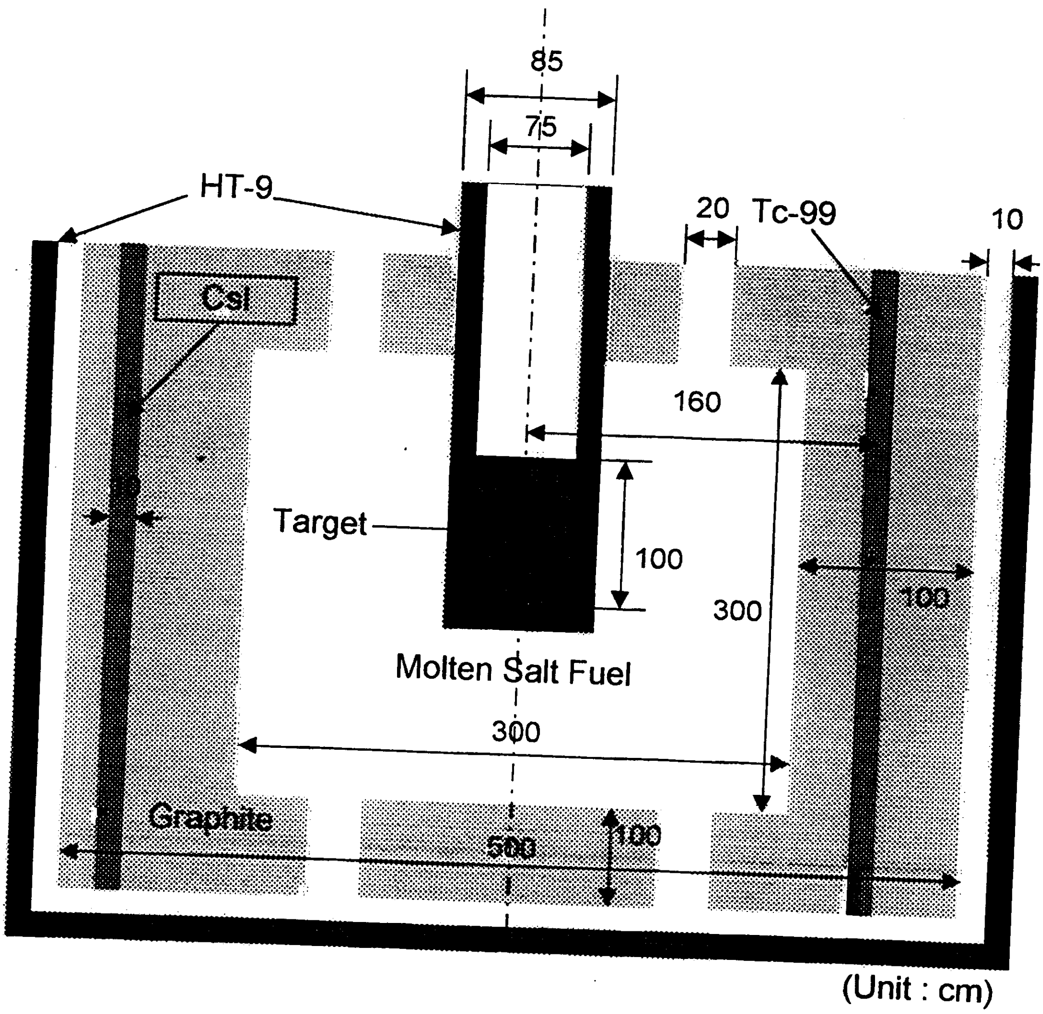

Fig. 1 Schematic Layout of the System.

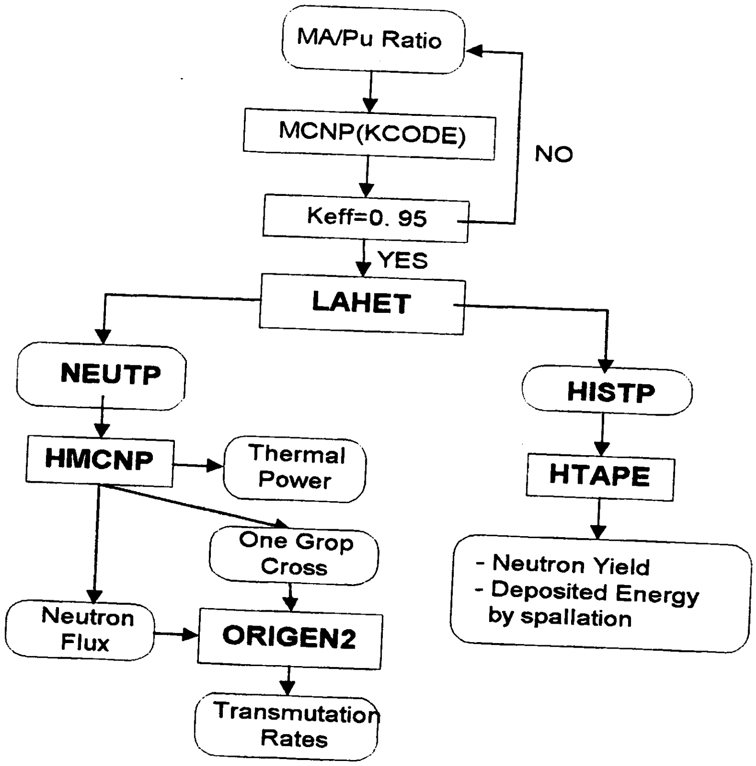

Fig.2 Data Flow between Codes

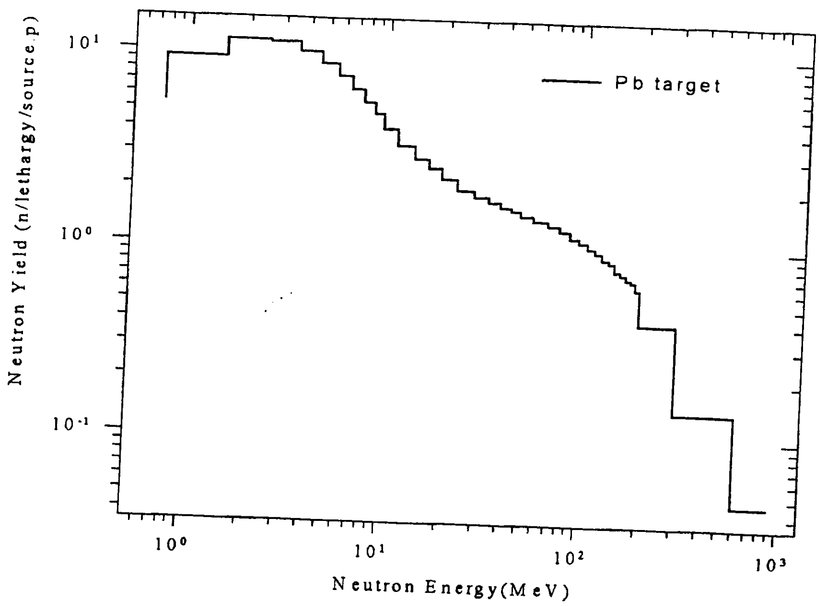

Fig.3 Spallation Neutron Energy Spectrum

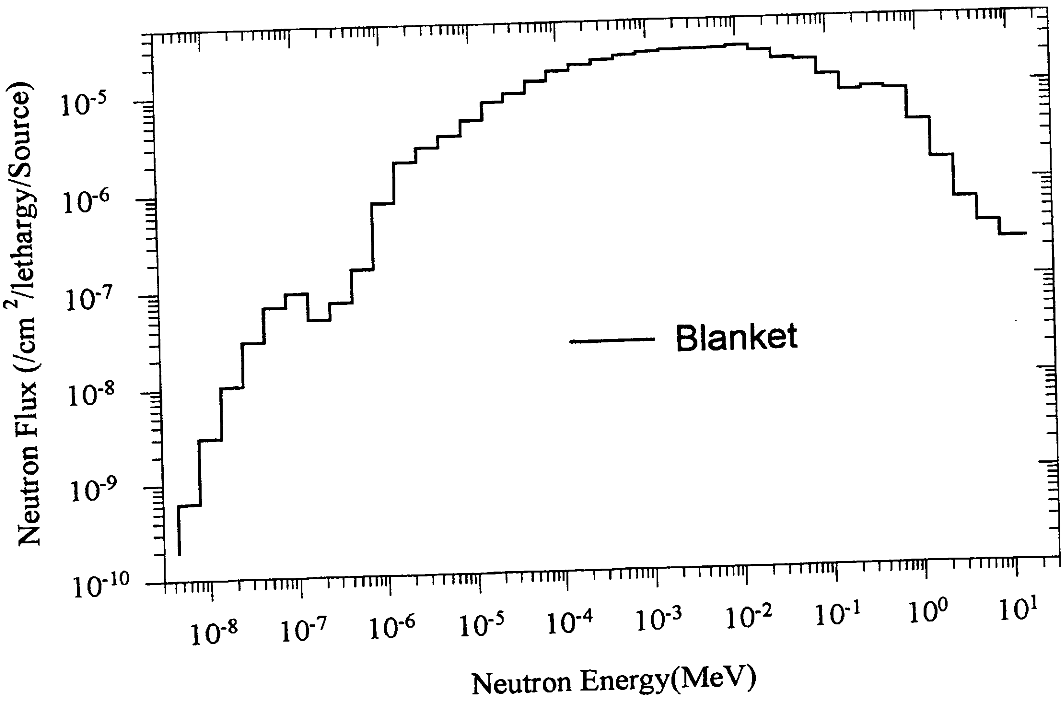

Fig. 4 Neutron Energy Spectrum in Blanket

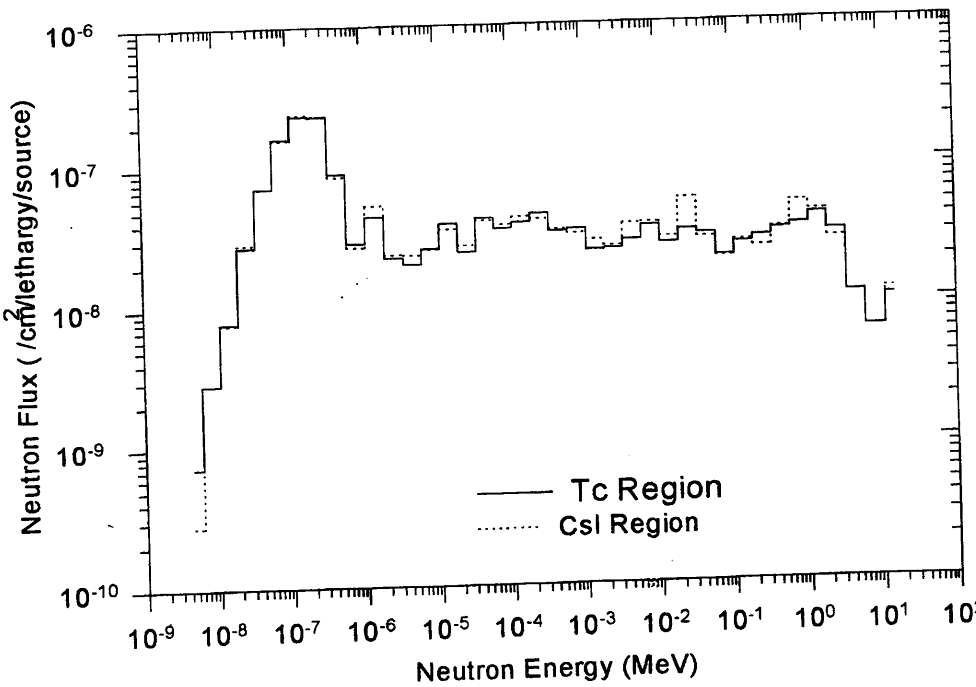

Fig. 5 Neutron Energy Spectrum in FP Burning Region