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CORE DESIGN STUDY FOR HYBRID TYPE TRANSURANIUM
NUCLIDES INCINERATION PLANT PART I. CONCEPT

August 1990

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Core Design Study for Hybrid Type Transuranium
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Part 1. Concept

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It is foreseen that much efforts are required to get social understanding that the nuclear energy is clean from the view point of preservation of natural environment. The efforts should be directed to two targets. One is the development of safer nuclear reactors compared with the existing ones. The other is the establishment of the radio-active waste management technology. In the spent fuel of the nuclear power reactor, are included the transuranium nuclides (TRU) such as Np, Am and Cm. They must be kept far away from the human living area for very long time, because some TRU have very long lives exceeding 10^4 years. But, it is not an easy task. The application of the intense proton accelerator is one of the possible solutions, because the TRU nuclides can be transmuted to stable or short life ones through the proton induced spallation and the fission reactions by resultant neutrons. The subcritical fast reactor driven by an intense proton accelerator with energy of 1.5 GeV and current of 10 mA class has the potential to incinerate the amount of TRU nuclides, which are produced in 10 units of the 1000 MWe light water power reactor. The subcritical fast reactor is loaded with the TRU metal fuel and cooled by liquid sodium. The incineration plant can be built satisfactorily safe, because its power output can be promptly shut down by merely switching off the proton beam. In order to realize such an incineration plant, are

required the development of a large scale accelerator, which has not ever been experienced and also that of wide range technologies related to the subcritical reactor.

In this context, was made a core design study for transuranium nuclides incineration plant driven by the intense proton linear accelerator. The reference core comprising the bundle pin type metal fuels of Am-Cm-Pu-Y and Np-Pu-Zr and the tungsten target, both of which are cooled by liquid sodium, is annually able to incinerate the transuranium nuclides produced in 7.6 units of the 1000 MWe LWR power stations. Under the condition that the fuel elements can withstand the burnup of 100000 MWD/ton the reference core can transmute 36 percents of the initially loaded ^{237}Np to shorter life nuclides during the operation period. The reactivity swing remains within 5.3 % $\Delta k/k$ and then the core can be kept subcritical.

Keywords: Transuranium Nuclides, Proton Accelerator, Transmutation, Spallation Reaction, Fission Reaction, Subcritical Fast Reactor Incineration Plant, Metal Fuel

ハイブリッド型超ウラン元素消滅プラントの炉心設計研究

第I部 概 念

日本原子力研究所東海研究所原子炉工学部

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(1990年7月13日受理)

原子力エネルギーが環境保全の観点からクリーンであるという社会的理解を得るには、今後さらに多くの努力が必要である。努力を傾注すべきターゲットは二つある。一つは、在来の炉に比較してずっと安全である炉を開発することであり、もう一つは放射性廃棄物処理の技術を確立することであると考えられる。使用済燃料にはNp, Am, 及びCm等の超ウラン元素が含まれているが、これは非常に長い半減期をもっており、一万年を超えてもなお残存することになるので、人の生活圏から遠ざける必要がある。強力な陽子加速器の応用が有力な解答を与える可能性がある。というのは、以上の超ウラン核種は核破砕反応及びこの反応により生成される中性子による核分裂反応によって安定ないし短半減期の核種に変換できるからである。加速エネルギー1.5 GeVで電流が10 mA級の大強度陽子加速器は100万キロワットの電気出力の軽水発電炉10基からの超ウラン元素を処理し得る潜在力があると予測される。

ここでは、大強度陽子加速器により駆動される超ウラン元素消滅プラントの炉心設計研究を行った。標準炉心はAm-Cm-Pu-YとNp-Pu-Zrの金属燃料とタングステンターゲットより成り、液体ナトリウムで冷却されている。この炉心では、100万キロワット電気出力の軽水炉7.6基分の超ウラン元素を消滅する能力があることが示された。また、燃料体が100000 MWD/tonの燃焼度に耐えると仮定すると、初期炉心に装荷した ^{237}Np の中、36%が消滅されることになる。この間の反応度の変動は初期値に比べ正側に5.3% $\Delta k/k$ であり、十分臨界未満に保たれる。

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1. INTRODUCTION

Much efforts are required to get the social understanding that nuclear energy is clean from the view point of preservation of natural environment. The social understanding is one of the premises for that the nuclear power generation is continuously accepted for future like it was in the past. It is indicated in Fig. 1 that share of the nuclear power in the whole primary energy supply has to be much enlarged if we want to lower the emission of CO₂ to an acceptable level while keeping the economic growth rate to the level being attained at the present.⁽¹⁾ Figure 2 shows that the emission of CO₂ can be reduced to the acceptable level which is by a few tens percents smaller than the present one, if the use of the nuclear power is extended as scheduled in Fig. 1.

The efforts related to the social understanding should be directed to the following two targets. One is the development of safer nuclear reactor compared with the existing ones, for which the occurrence probability of the large scale accidents followed by the considerable level discharge of radioactive substances such as the case of the Chernobyl reactor can be thought to be effectively zero. The other is the establishment of the radio-active waste management technology.

In the spent fuel of the nuclear power reactor, are included the transuranium nuclides (TRU) such as Np, Am and Cm. Therefore, they must be kept away from the human living area for very long time, because some TRU have long lives exceeding 10⁴ years. The situation is explained in Fig. 3, where the decay of toxicity of nuclear wastes with time is predicted. The geological deposit is widely accepted in most advanced countries. But, it is not an easy task. If the TRU nuclides are transmuted to stable or short life ones, the technological burdens placed on the geological deposit can be substantially mitigated.

The Japanese Atomic Energy Commission concluded in Autumn 1988 to strengthen the research and development on partitioning and incineration of nuclear wastes. The Science and Technology Agency formulated the OMEGA project. The fuel cycle committee of OECD/NEA has decided to initiate an international collaboration frame work for the subject.

Incineration of nuclear wastes by use of accelerators is defined as one of the important tasks in the OMEGA program. The Japan Atomic Energy Research Institute (JAERI) has been engaged for several years in the basic studies on incineration technology with use of the intense proton linear accelerator. The intense proton accelerator program intends to

provide a large scale proton linear accelerator named as "The Engineering Test Accelerator". The principal purpose of the accelerator is to develop the incineration technology of the nuclear wastes. The accelerator is intended to be used also in the other industrial application and also in applied science.

2. CONCEPT OF INCINERATION OF NUCLEAR WASTES

2.1 Nuclear Reaction

The following nuclear reactions are usable in principle for incineration,

- (1) Nuclear fission by neutrons
- (2) Spallation by proton
- (3) (n,γ) , (γ,n) and photofission.

In JAERI, is considered the reaction of the nuclear fission by neutron to be the most feasible one from the view points of higher transmutation efficiency and more profitable energy balance. It is noted that only the fast neutrons can cause the fission reactions against ^{237}Np as indicated in Fig. 4, which are the major components of TRU from light water reactor (LWR).

Spallation reaction is suitable for generation of the source neutrons to start the fission chain reactions, because the energy required to produce single neutron is relatively small. The high energy protons constitute cascade in the targets made of the heavy metal, as illustrated in Fig. 5. Several tens neutrons evaporate from an excited nucleus and after that spallation products are left. Incineration of TRU nuclides can be done with spallation reactions, but the energy balance is not so profitable compared with the fission reactions. The development of the computer code NMTC/JAERI has been done in order to describe the spallation process.⁽²⁾ Figures 6 and 7 show the calculated results for proton energy dependence of the number of spallated ^{237}Np nuclei and half life distribution of the spallation products due to a single proton, respectively.⁽³⁾ Dependence of the number of neutrons from the ^{237}Np target on proton energy is given in Fig. 8.

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2.2 Concept of Incineration Plant

Conceptual design studies have been performed on the plants aiming at incineration of TRU nuclides. The amount of TRU produced in a 1000 MWe LWR is about 26 kg per year as given in Table 1. A double layer fuel cycle such as illustrated in Fig. 9 was proposed in JAERI.⁽⁴⁾ First layer is usual energy production cycle. In addition to it the second layer is allocated to incinerate the TRU nuclides by use of a special burner reactor or a subcritical reactor driven by an accelerator. The former plant is such reactors as loaded with fuels containing the TRU nuclides. The latter plant comprises the intensive proton accelerator and the subcritical core surrounded with blanket. The target generating neutrons is likely made of heavy metals, the function of which is sometimes substituted by the TRU fuel assemblies themselves for simplification of the plant. The core is loaded with the fuels containing the TRU nuclides and is cooled by liquid metals. This hybrid type plant is not simpler than the special burner reactor in the sense that a big accelerator is added as an extra burden.

The hybrid type plant, however, has the following superior properties:

- (1) The fission reactions in the plant can be promptly shut off only by switching off the beam current of the accelerator.
- (2) The plant is always operated in a far subcritical state. Therefore, control and safety rods are not required; the core structure becomes simple.
- (3) The burn-up of the TRU fuels is not constrained from the condition of keeping the criticality of the core.
- (4) The core design is free from the safety related restrictions such as the non-positive Na void coefficient and does not place any strict requirement on the informations of the isotopic abundances in fuel compositions. The very small values of delayed neutron fraction of TRU fuels give rise no problem.

3. CONCEPTUAL CORE DESIGN AND ANALYSIS OF CHARACTERISTICS

At first, JAERI conceived an incineration plant to destruct the TRU nuclides mainly through the spallation reactions, using a 1.5 GeV - 300 mA class proton linear accelerator. The incineration rate was estimated about 100 kg per year which is equal to the TRU production rate from the

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four units of 1000 MWe LWR. The problem of the plant is that the scale of the accelerator is probably large from the view point of cost-benefit analysis.

At second, a different design study is made on a plant of the hybrid type driven by a smaller size machine of 1.5 GeV - 10 mA class, which seems to be realized by use of continuous wave mode. Core configuration of the hybrid plant is shown in Fig. 10.⁽⁵⁾ Core of the plant is loaded with the bundle pin type metal fuels of Am-Cm-Pu-Y and Np-Pu-Zr and cooled by liquid sodium. The fuel assembly in the core is similar to that commonly employed in LMFBR design. The fuel pin cell geometrically selected for the target is shown in Fig. 11. The reason for adoption of the metal fuels is to make the neutron spectrum so hard that the average neutron energy exceeds the fission threshold energy of ^{237}Np , 0.6 MeV. Tungsten target is located so as to generate spallation neutrons in the central region of the core. Generation of ~ 30 neutrons per proton is expected in Fig. 8, which initiate the fission chains and are able to transmute ~ 380 TRU nuclei in total. The effective multiplication factor of the core k_{eff} is around 0.92. The core and target design parameters are shown in Table 2. Predicted performance of the hybrid plant is given in Table 3. It is shown there that the TRU nuclides of about 200 kg can be incinerated, the amount is equivalent to the annual product from about 7.6 units of 1000 MWe LWR.

The power distributions in the incineration core are shown in Fig. 12. The power peaking is not so strong as to give rise a severe problem for the heat removal. The maximum fuel meat temperature can be kept below 900°C that is the estimated melting point of the Np-Pu-Zr alloy. The coolant temperature can be maintained far below the boiling point of Sodium. The coolant velocity through the core was chosen below 8 m/s to prevent the vibration of the fuel assembly. It is also estimated that the core is not damaged in such an accidental case that the coolant flow by the forced circulation is lost. The decay heat can be removed from the core through the natural circulation.

The calculated performance of the plant indicated in Table 3 is that at the initial stage of operation. The change of k_{eff} with burn-up is estimated as shown in Fig. 13 for the reference core, the value of which increases at the beginning from 0.92 up to 0.97 in 900 day's operation and then decreases down to 0.90 in 2000 days. The inventory of the TRU nuclides changes as illustrated in Fig. 14. Under the condition that the fuel elements can withstand the burn-up of 630 days (~ 100000 MW/ton,

3.5×10^{23} n/cm²), the reference core can transmute 36 percents of the initially loaded ²³⁷Np to shorter life nuclides. In the case of the 2000 day's burnup, about 97 percents of the initially loaded ²³⁷Np nuclei have been incinerated there, although it is not clear at present whether the fuel elements can resist against such a high burn up. It seems that the window material can likely withstand the proton beam bombardment, because the window area is expanded and the fraction of the proton's energy deposit through it is small compared with those in the target area. Design studies are extended to some versions of the above mentioned reference core. Their core constitutions are indicated in Table 3. The version-1 core has the tungsten target like the reference core, but it is cooled by lead-bismuth. The version-2 and 3 cores utilize the TRU fuel elements as the target generating neutrons and are cooled by sodium and lead-bismuth respectively. The performances of the various versions have also been investigated and summarized in Table 3, together with that of the reference core. The performances of the version-2 and 3 with respect to the number of the LWR units are poorer than those of the reference core. This trend is, however, to be somewhat improved by optimizing the lattice pitch of the fuel elements located in such area where most of the neutron generations due to the proton beam bombardments occur.

4. SUMMARY

A core design study for the transuranium nuclides incineration plant driven by the large scale proton linear accelerator was performed. It was assumed that the energy and beam current are 1.5 GeV and a few tens mA, considering the present status of the accelerator technology. The reference core comprising the bundle pin type metal fuels of Am-Cm-Pu-Y and Np-Pu-Zr and the tungsten target, both of which are cooled by liquid sodium, is annually able to incinerate the transuranium nuclides produced in 7.6 units of the 1000 MWe LWR power stations when the beam current is estimated 23 mA. Under the condition that the fuel elements can withstand and burnup of 100000 MWD/ton ($\sim 3.5 \times 10^{23}$ n/cm²) the reference core can transmute 36 percents of the initially loaded ²³⁷Np to shorter life nuclides during the operation period. The reactivity swing remains within 5.3 %Δk/k, and then the core can be kept subcritical.

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Table 1 TRU Production per Year from 1000 MWe PWR

Nuclide	Weight (kg)	Fraction (%)
^{237}Np	14.5	56.2
^{241}Am	6.82	26.4
^{243}Am	3.1	12.0
^{243}Cm	0.0078	0.03
^{244}Cm	1.32	5.1
^{245}Cm	0.072	0.3
Total	25.8 (kg)	100.0

Fuel Burn Up : 33,000 MWD/T
 Cooling Time before Reprocessing : 3 years
 Cooling Time before Partitioning : 5 years
 Collection Rate of U and Pu : 100%

Table 2 Core design parameters

Coolant	Na/Pb-Bi
Proton energy	1.5 GeV
Core	200~260 cm
Length	100 cm
Height	100 cm
Width	100 cm
Target	
Length	60 cm
Height	100 cm
Width	10 cm
Reflector	Stainless steel
Composition	20 cm
Thickness	
Fuel	Np-15Pu-30Zr
Composition	AmCm-35Pu-10Y
Bond	Na
Clad	HT-9 steel
Fuel slug diameter	4.00 mm
Clad outside diameter	5.22 mm
Clad thickness	0.3 mm
Pin length	1000 mm

Table 3 Performance of Incineration Plant

Core type	Reference	Version-1	Version-2	Version-3
Target	tungsten	tungsten	TRU	TRU
Coolant	Na	Pb-Bi	Na	Pb-Bi
Effective multiplication factor	0.92	0.86	0.94	0.95
Pin pitch (mm)	9.5	10.5	10.5	12.0
Actinide loading (kg)	2866	2013	2682	1584
Beam current (mA)	22.6	7.5	18.2	5.4
Neutrons per proton	38.1	52.8	35.3	55.1
Fissions per proton (> 15 MeV)	0.67	0.24	0.64	0.42
(< 15 MeV)	150.6	171.3	108.0	147.4
Average neutron energy (keV)	739	629	774	626
Average neutron flux ($\times 10^{15}$ n/cm ² ·s)	4.6	6.6	2.0	1.9
High energy component (> 1.0 MeV)	20%	18%	20%	17%
(< 0.1 MeV)	72%	78%	71%	77%
Operation time (days)	270	270	270	270
Burnup rate (%)	7.0	6.9	4.3	2.7
weight (kg)	202	139	114	42
Unit of 1000 MWe LWR*	7.6	5.3	4.3	1.8
Burnup reactivity swing (%Δk/k)	3.8	2.9	2.7	2.1

* Numbers of units produce the amount of the TRU nuclides which can be incinerated in the plant.

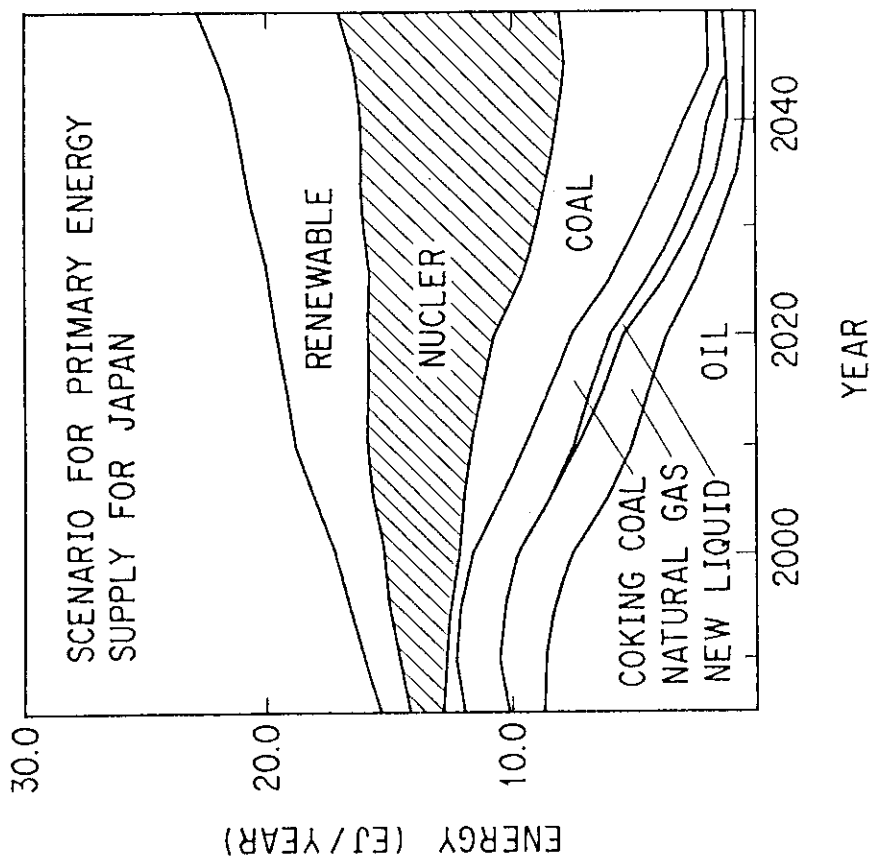


Fig. 1 Primary Energy Supply under Constraint for CO₂ Emission.

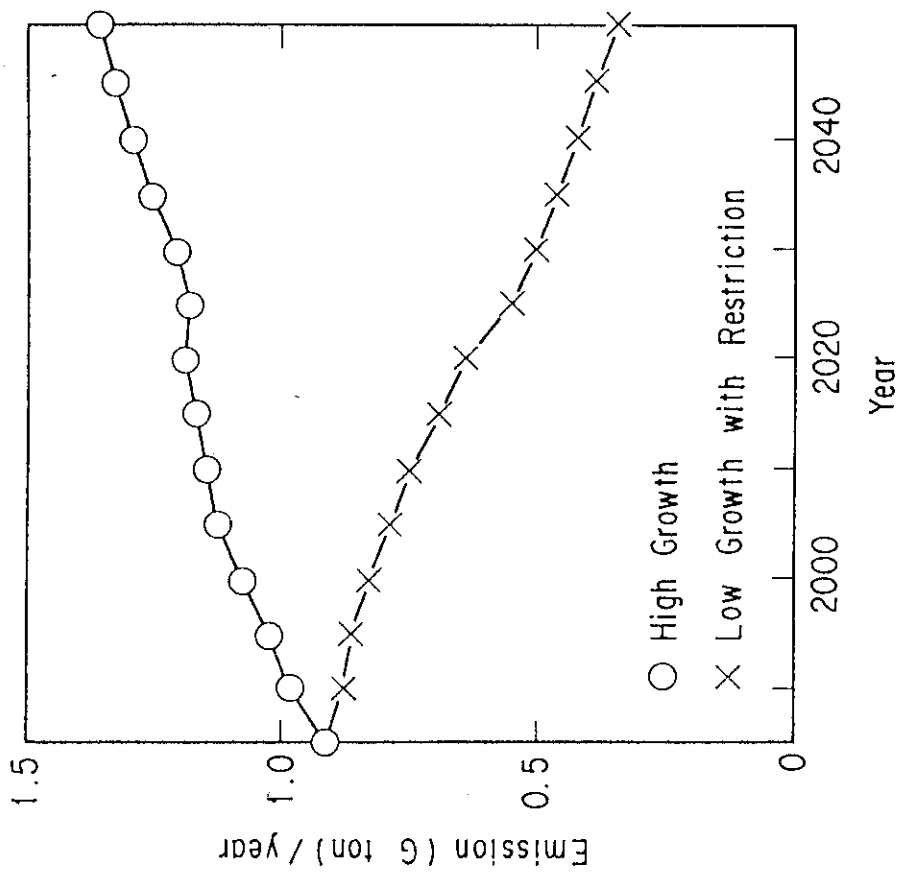
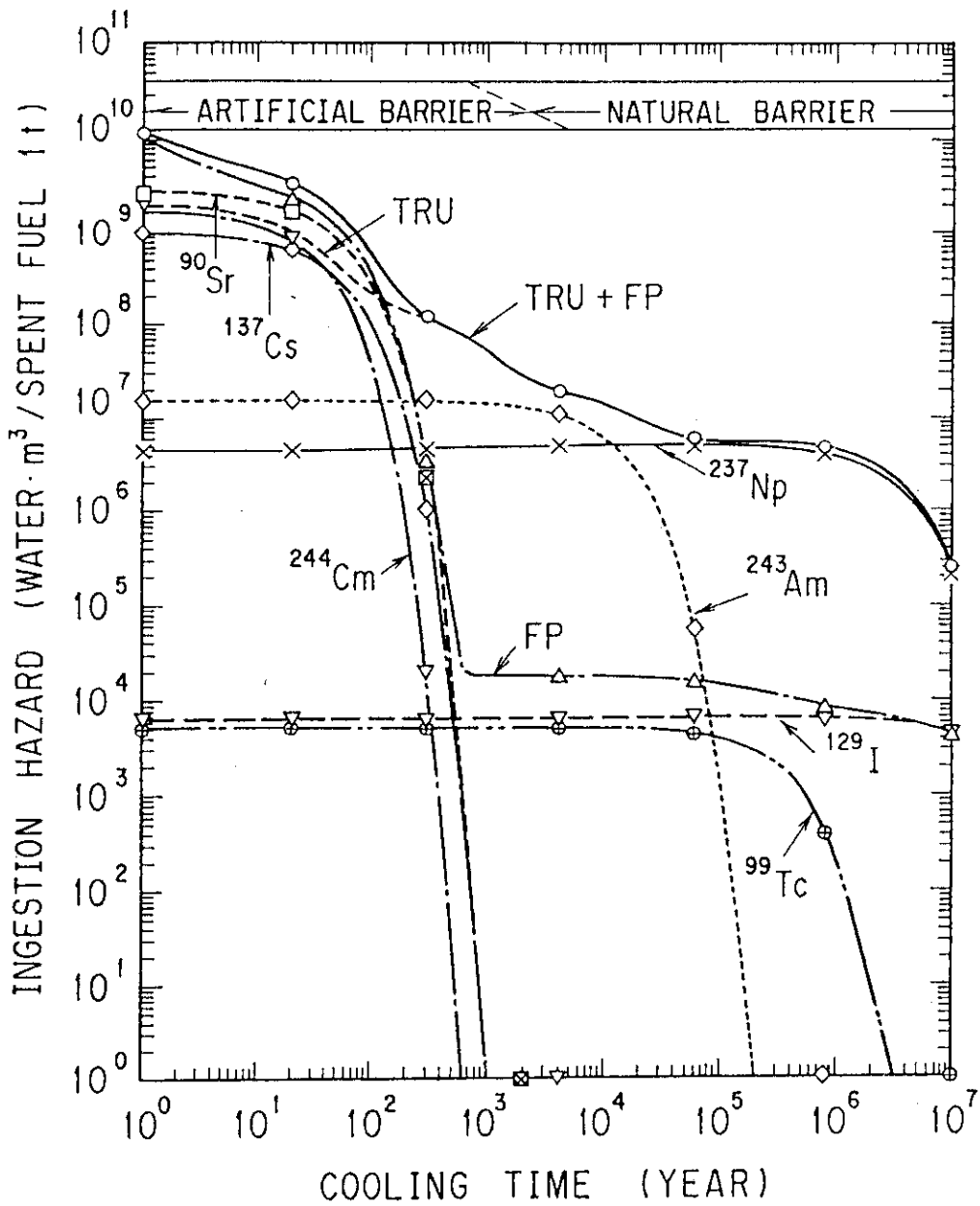


Fig. 2 Future Reduction of CO₂ Emission.



Ingestion Hazard is Indicated by Water Volume
 Required to Dilute Radioactive Waste Down to
 Acceptable Level

Collection Rate of U and Pu : 99.5 %

Burn-up : 33000 MW_td

Fig. 3 Change of Radiological Hazard of Nuclear Waste
 from PWR Spent Fuel.

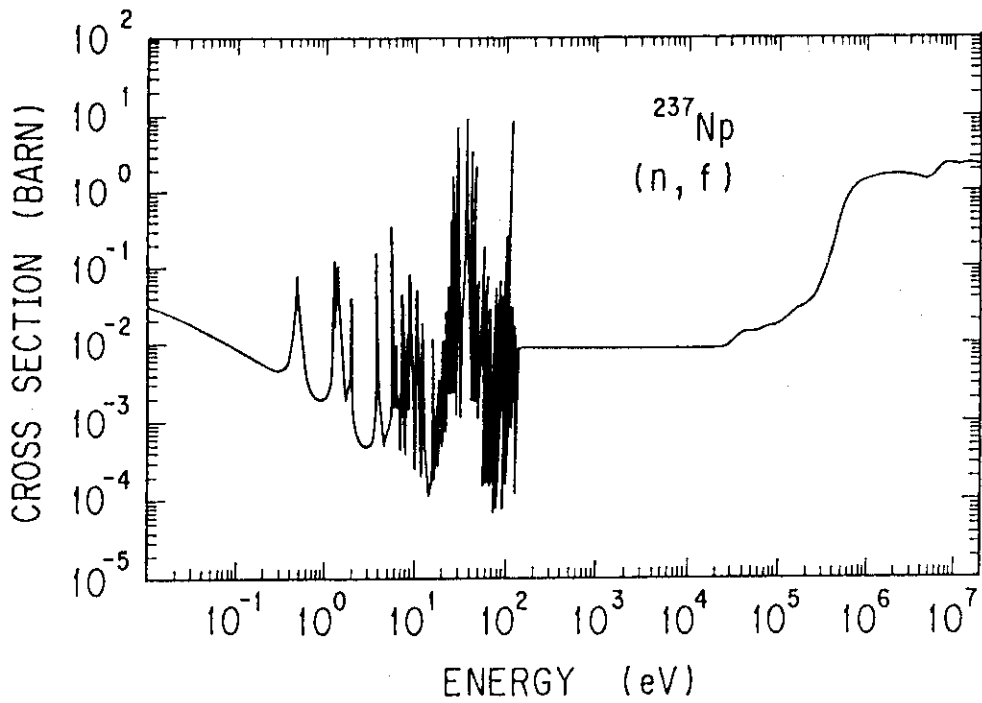
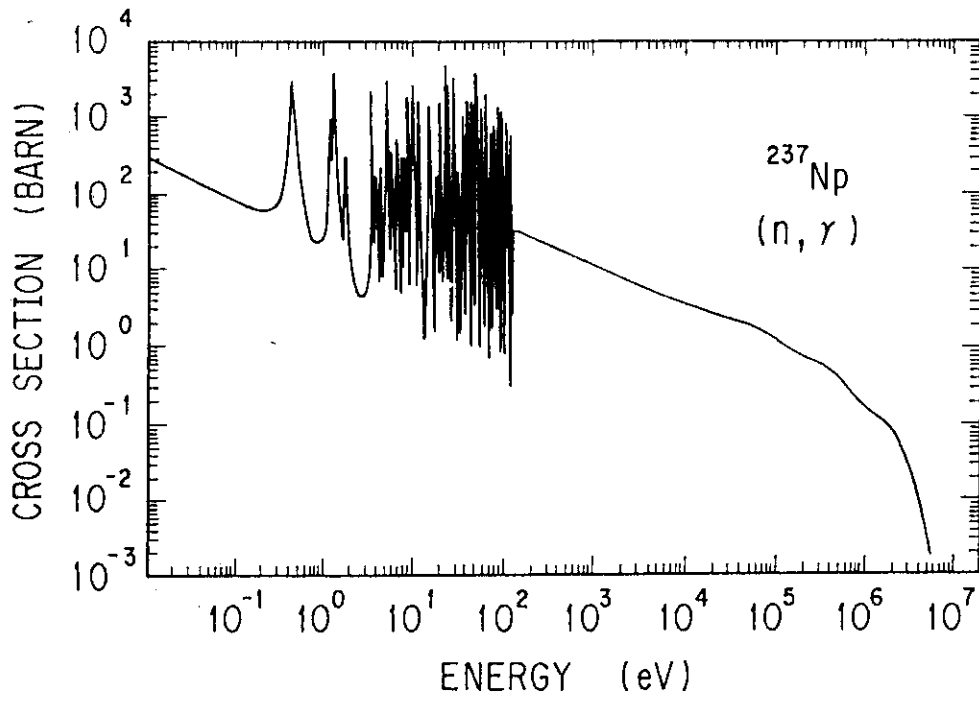


Fig. 4 Neutron Cross Section of ^{234}Np .

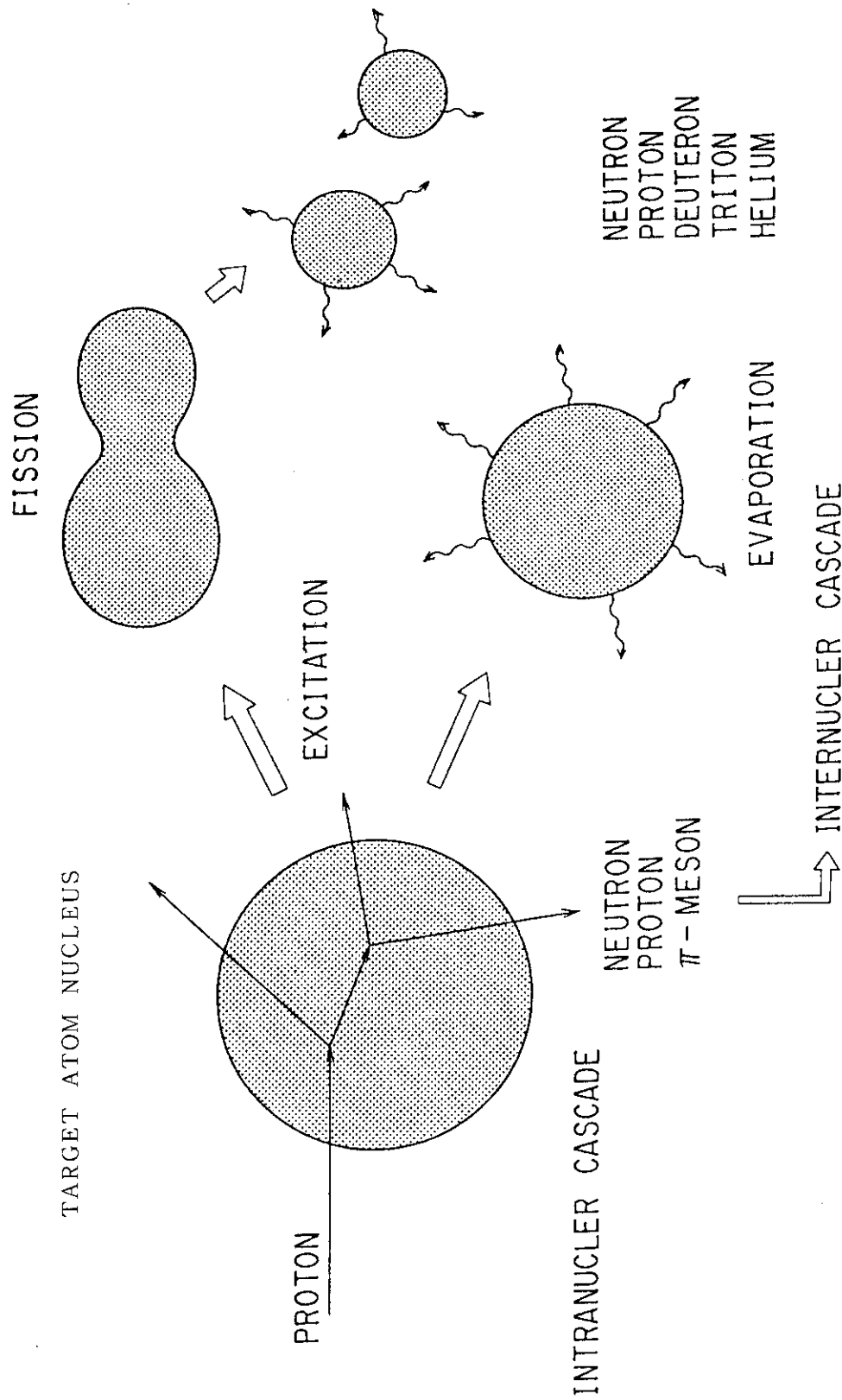


Fig. 5 Spallation Reaction by High Energy Proton.

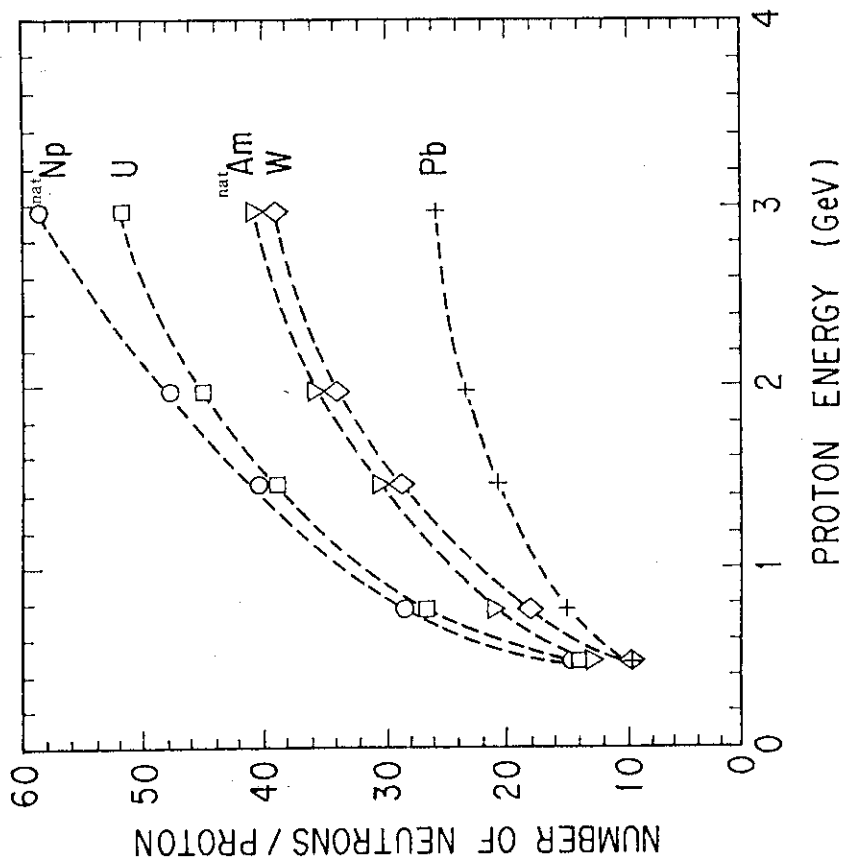


Fig. 8 Energy Dependence of Number of Neutrons Generated by Spallation Reaction.

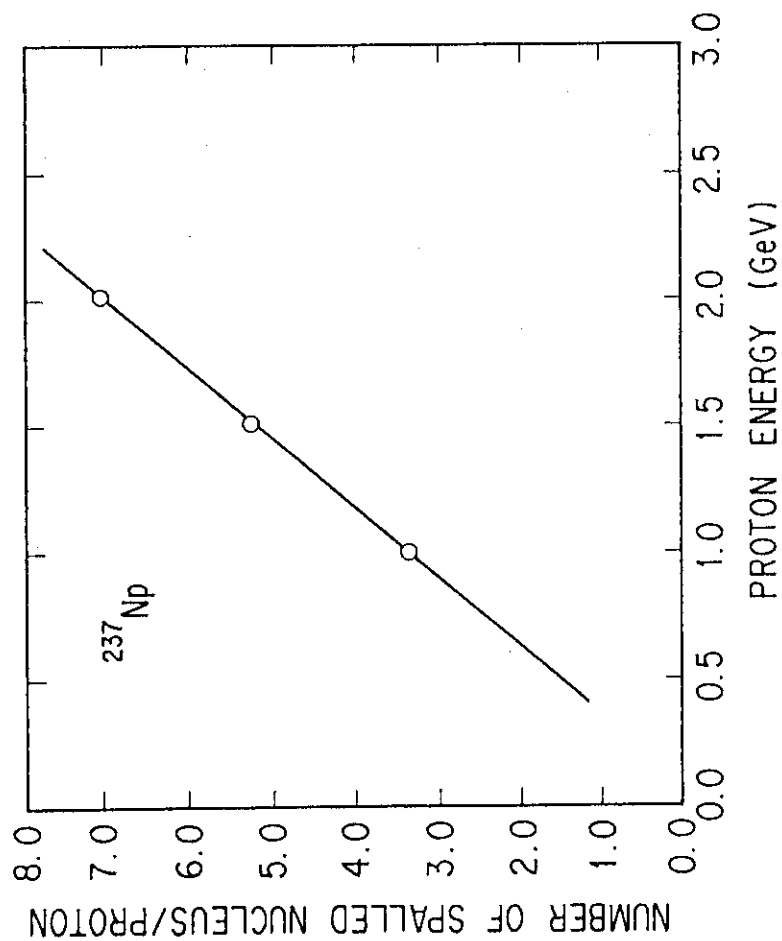
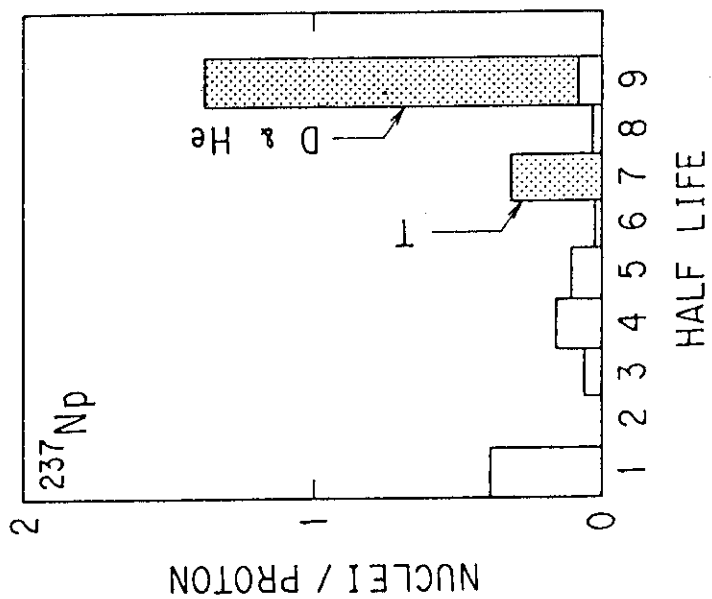
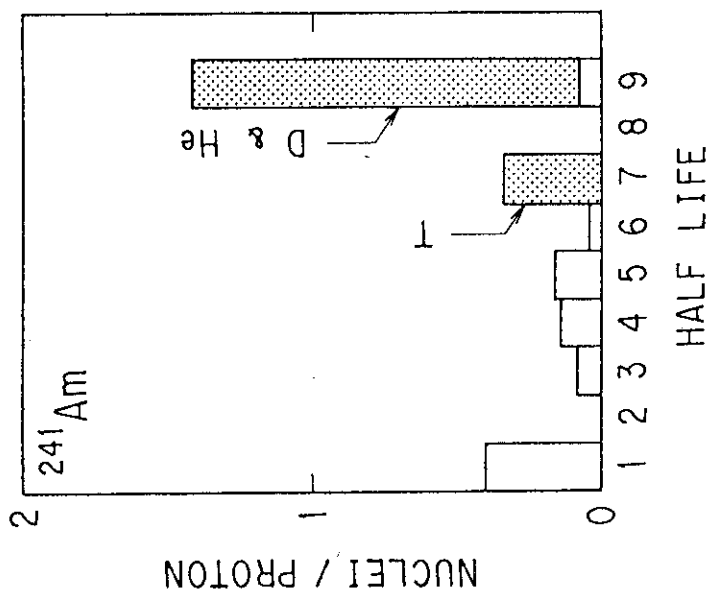


Fig. 6 Energy Dependence of Number of Destroyed Nucle due to Spallation.



Classification by Half Life $T_{1/2}$

1 : $0s < T_{1/2} < 10^{-3}s$	6 : $5d < T_{1/2} < 1y$
2 : $10^{-3}s < T_{1/2} < 1s$	7 : $1y < T_{1/2} < 100y$
3 : $1s < T_{1/2} < 1m$	8 : $100y < T_{1/2} < 1 \times 10^8 y$
4 : $1m < T_{1/2} < 1h$	9 : Stable Nuclides
5 : $1h < T_{1/2} < 5d$	

Fig. 7 Half Life Distribution of Spallation Products due to 2GeV Proton.

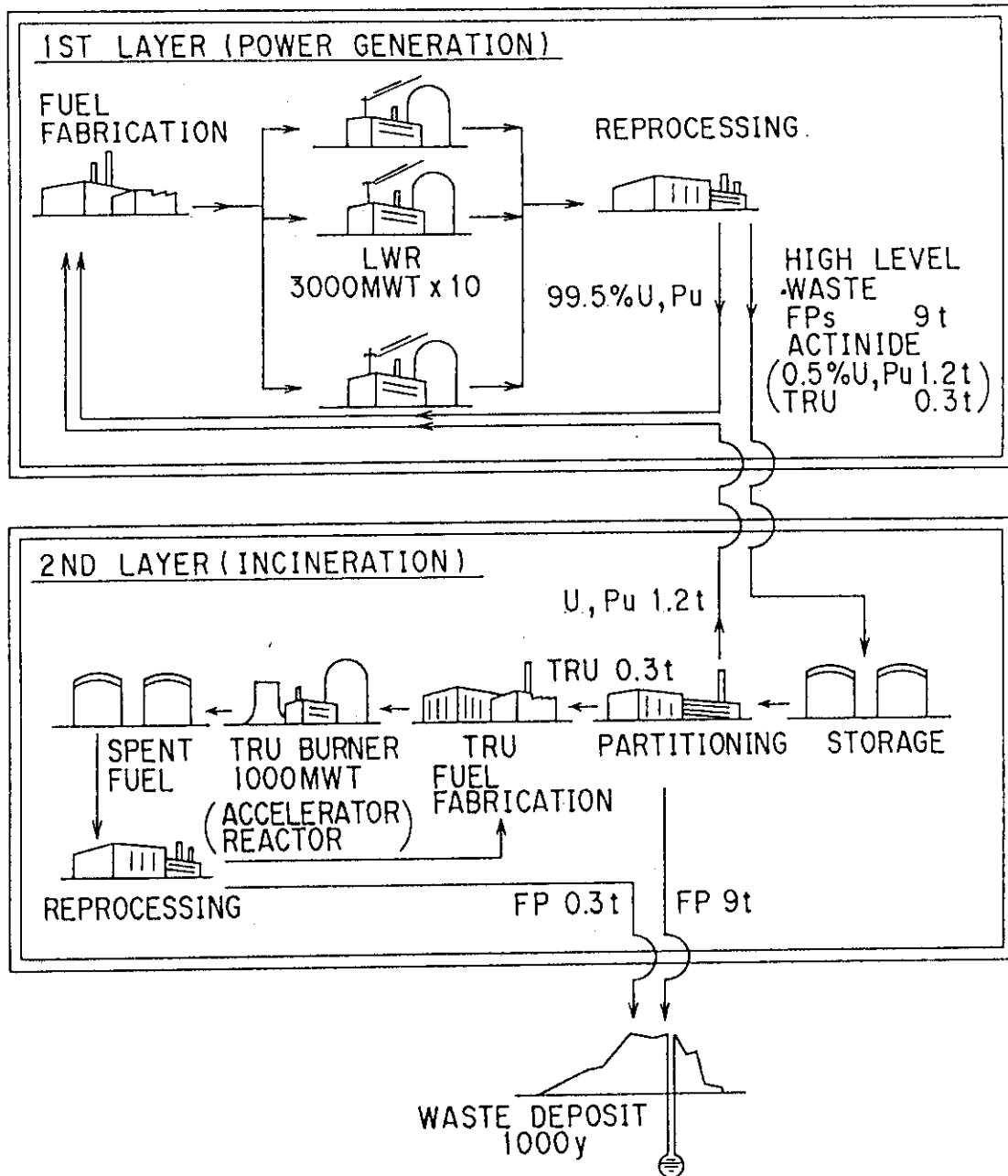


Fig. 9 Dual Layer Type Nuclear Fuel Cycle with Use of Reactors for Incineration of TRU from LWR.

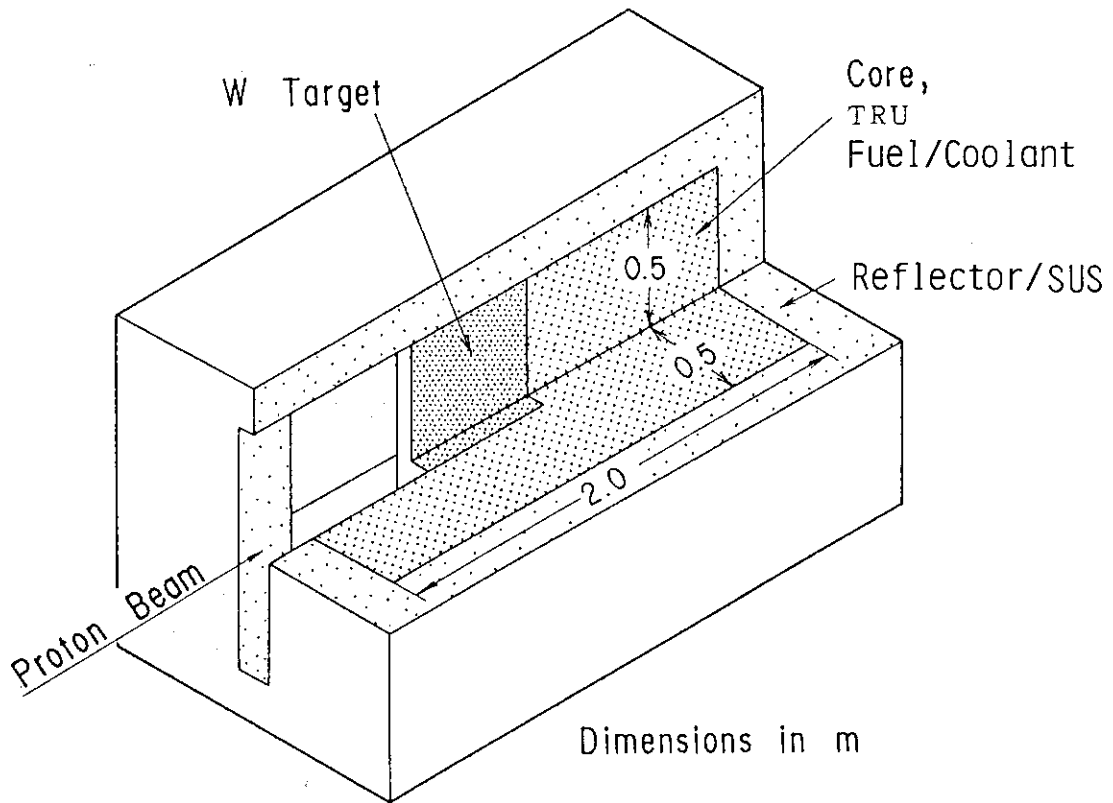


Fig. 10 Core Configuration of Hybrid Plant (Reference Core)

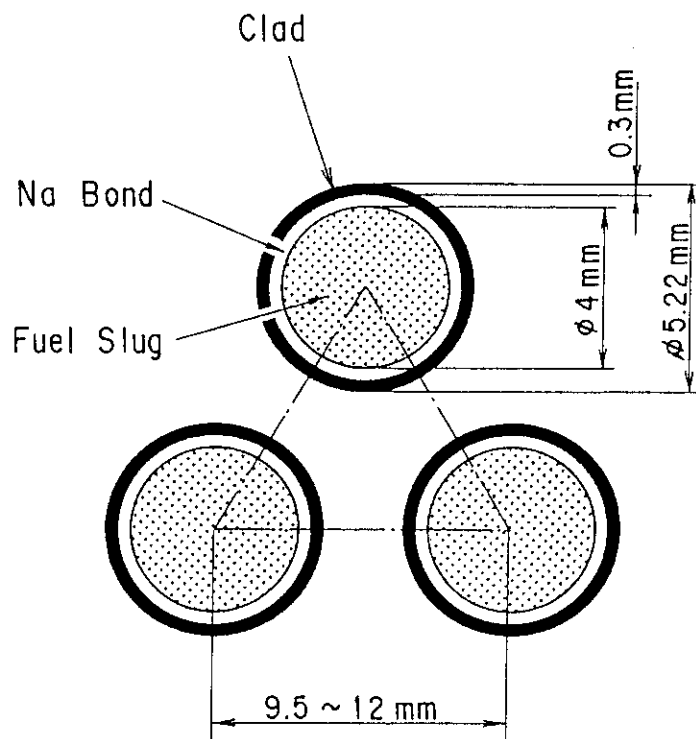


Fig. 11 Fuel Pin Geometry.

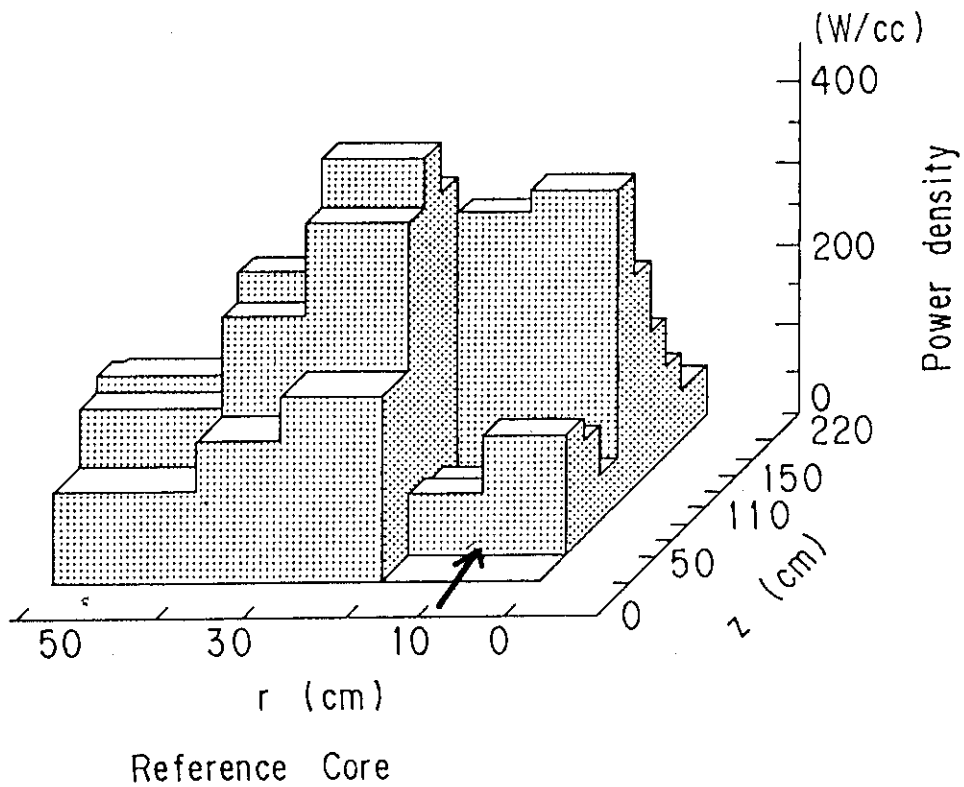
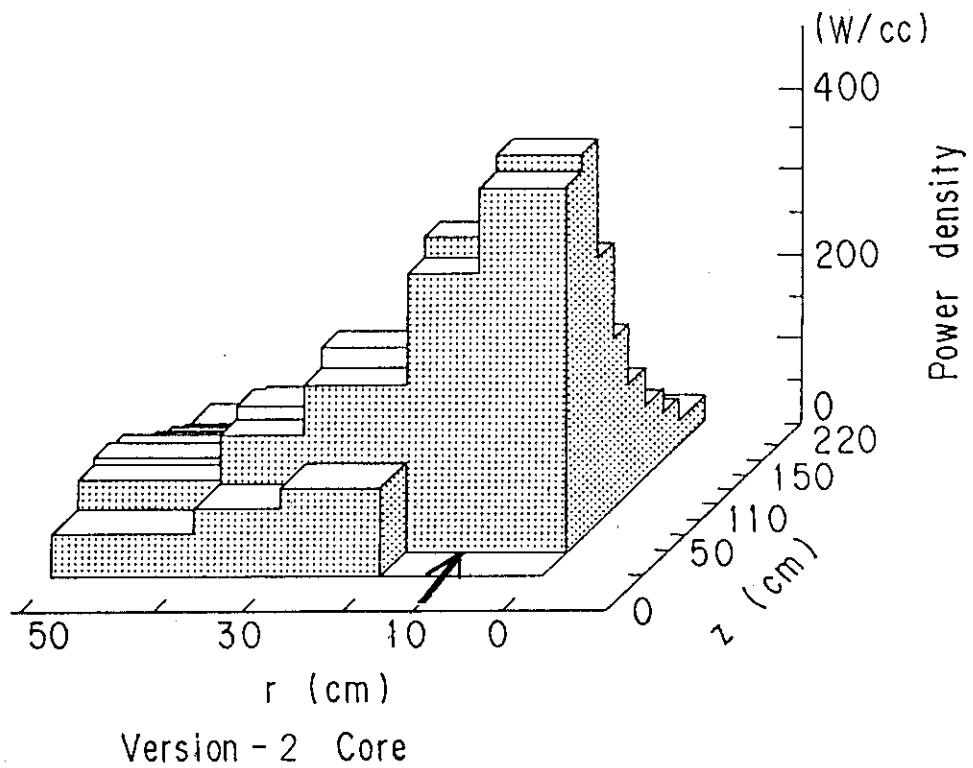


Fig. 12 Power Distribution in Hybrid Plant.

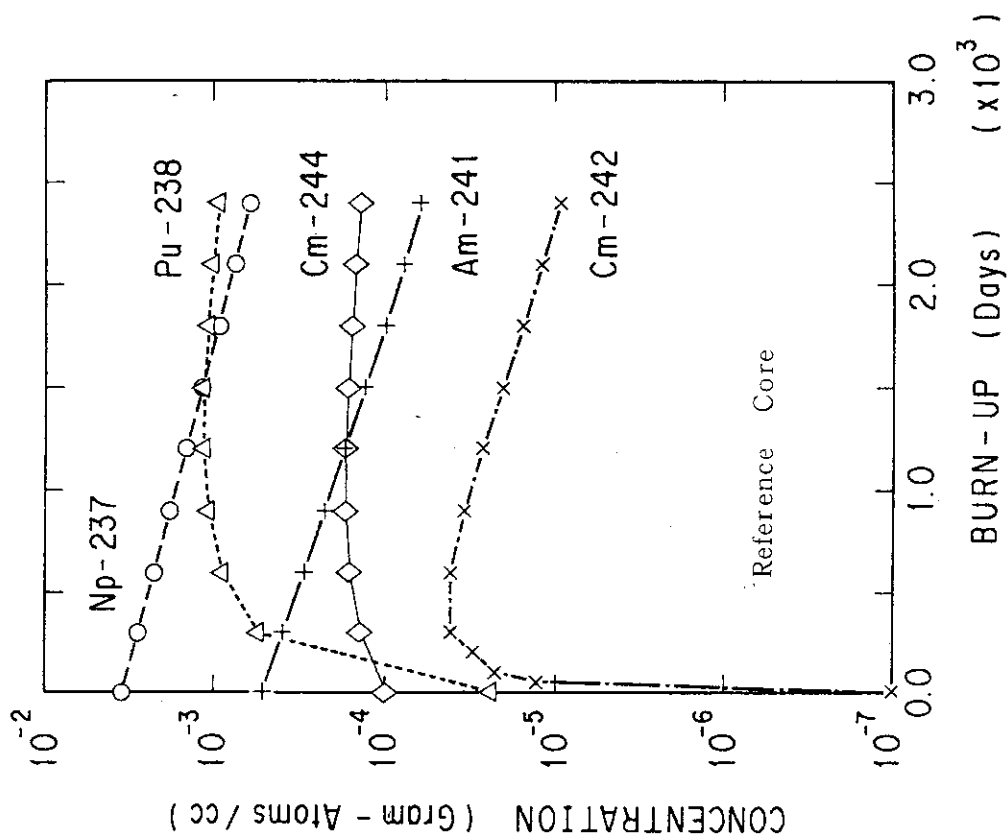


Fig. 14 Change of Transuranium Inventory with Burn-Up.

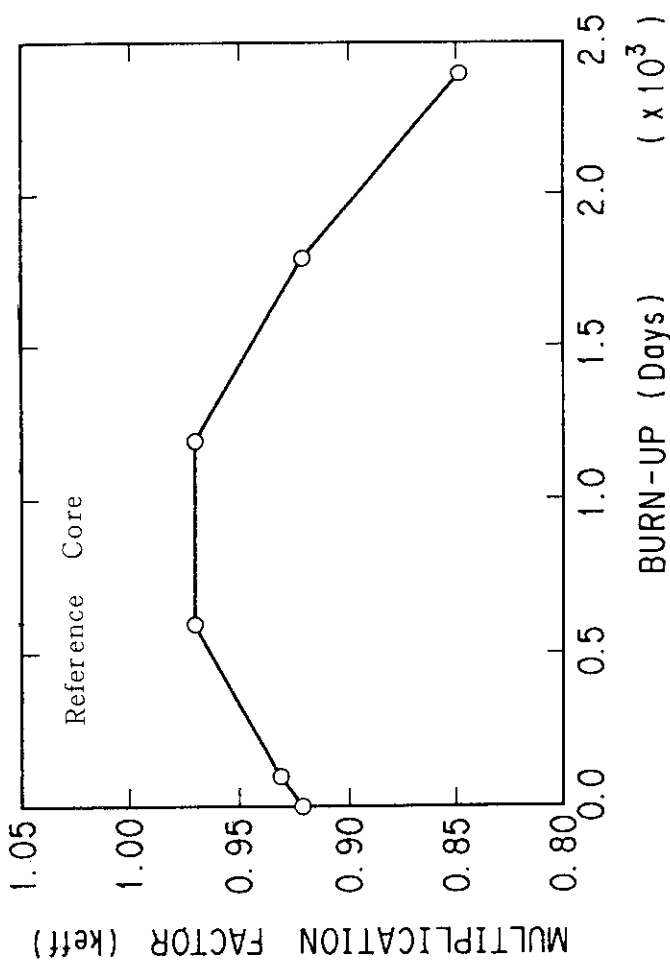


Fig. 13 Change of k_{eff} with Burn-Up for Reference Core.