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STUDY OF TRU TRANSMUTAION SYSTEMWITHA PROTON ACCELERATOR

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(Abstract)

Researches on the TRU transmutation with a proton accelerator, which were performed at JAERI in these years, have been promoted as one of main themes in the newly started national project OMEGA, which aims to establish new safer technologies to process long-lived radio-active wastes. Conceptual design studies of the transmutation plant have been made from thenucleonics and hydraulics analysis points of view. The present transmutation systemis a hybrid one of an intense proton accelerator, a tungsten target cooled with sodium and a subcritical core loaded with the TRU alloy fuel. In this system the transmutation rate of about 200 kgTRU a year (generated from about 8 units of 1 GWe PWR) are attainable and the marginal electricity more than one needed to drive the accelerator can be produced.

With the aim to make assessment of the system design and to upgrade the computer codes for simulating nuclear spallation processes in the transmutation system, the integral spallation experiment has been planned. The lead target has been set up along the 500 MeV beam line of a proton synchrotrons booster at KEK. The first irradiation experiment has started Autumnin 1990.

1. Introduction

The management of minor actinides and fission products in the high level wastes is an important, hazardous problem due to their strong radio activities. In particular, transuranium nuclides(TRU) have a very long half lifetime of millions of years. Most of countries promoting the nuclear power generation have developed the vitrification and geological disposal techniques for managing these wastes. This subject, however, should be re-examined from the view piont of applying new, advanced technologies at present. By establishing new transmutation technologies the upgrade of safety assurance in the waste managnent will be achieved.

In Japan the OMEGA project(Option Making Extra Gains from Acitinides and fission products) started to research and develop the new technologies on nuclear waste partitioning and transmutation as the long term one. As a part of the project, Japan Atomic Energy Research Institute has set up the R & D plans mainly on

1) advanced partitioning technology,
2) TRU transmutation in burner and power reactors and
3) TRU transmutation with proton accelerators. "
4) basic researches for TRU technology.

The research for the item (3) at JAERI is being promoted as following items (a) ~ (d),

a) development of the basic simulation code system,
b) conceptual study of the TRU transmutation system,
c) spallation integral experiment,
d) development of an intense proton beam accelerator.

As for the item (d) the detailed description is given in another proceeding paper by Dr. Mizumoto. In the present paper brief descriptions are given for the spallation reaction & the particle transport simulation codes, some results in design studies of TRU transmutation target-core system driven by proton accelerator and spallation experiment. Tree structure illustration of R & D Items for the TRU Transmutation Plant and High Intensity Proton Beam Accelerator is shown in Fig.1.

2. Research of transmutation system driven with proton accelerator

2.1 Nuclear spallation reaction occured by high energy protons

Figure 2 shows the schematic illustration of main nuclear reactions and the particle transport process generated in the transmutation target system discussed in the present paper. Above the cutoff energy of 15 MeV the intranuclear and internuclear cascade processes with the spallation, evaporation and high energy fission reactions occure from the proton bombardment. Through these processes many light particles such as neutron and helium are emitted. Most of evaporated neutrons transport in the target after slowing down into the energy range below 15 MeV.

The flow sheet in Fig.3 shows the mutual relation among the simulation code systems prepared at JAERI. The High Energy Nuclear Reactions and Nucleon-Meson Transport CodeNMTC/JAERI1[2] is main code in this code system. The NMTC/JAERI-NMTA and NUCLEUS[3] codes included in the right side of vertical dot-line simulate the high energy nuclear reactions above 15 MeV. MORSE-DD[4] and TWOTRAN codes in the other side carry out the neutron transport calculation below 15 MeV. SPCHAIN and ORIGEN-2 calculate the time evolution process in the energy range larger and smaller than the cutoff energy respectively.

The results computed by these spallation codes presents us the important, basic data[5],[6] for researching the feasibility ofTRU transmutation, as represented in the following figures. As seen in Fig. 4thebirdeye's views of yields of products are drawn in log scale for the cases of 1 GeV proton impinging on a) uranium and b) lead nuclei. The yield consists of three components of the hill of spallation product, the spire of evaporated particles and the valley of high energy fission products between them. It is apparent that the yield of SP is more by two orders than FP. Table 1 summarizes the number of produced light particles such as proton, neutron and so on. They are emitted from a uranium target nucleus bombarded by protons with the energies of0.5 GeV to 3 GeV. The number of emitted particles has the maximum value around 2 GeV, where the neutrons more than 17 are emitted per one uranium nucleus. The results show that the incident energy has the optimum value around 2 GeV for the neutron production. Figure 5 shows the histogram of half-life distributions of products whena2 GeV proton bombarding on ^{237}Np and ^{241}Am target nuclei. The shaded portions in the calssifications of7 and 9 represent toritons and deuterons & heliums respectively. The nuclides included in the classification 8 are a few. These nuclides are considered to be most harmful from the hazardous point of view. Most ofSP in this case have the halflife shorter than one year except toriton and deutron. High energy nucleons generated in the spallation can transmute TRU nuclides through the cascade processes. Figure 6 shows the dependence of the number of spallated nuclides on the incident proton energy when the proton injects on the ^{237}Np metal target (20 cm ϕ x 60 cm).

The number of nuclides transmutated at 1.5 GeV is about 5 per incident proton. The rough estimation shows that it is too small to process TRU wastes in the commercial base unless the proton beam can have current higher than 300 mA. The heat generation is not sufficient to drive the intense accelerator. However it is noted that several tens neutrons with hard spectrum similar to the one in the fast reactor are emitted in the spallation process. The computer simulation result shows that the number of spallation neutrons generated in the targets of actinides such as U, Np and Am, and heavy elements such as Pb and W increases monotonously when the proton energy increases up, as shown in Fig. 7. For the case of ^{237}Np target bombarded by a 1.5 GeV proton the neutron number is ~40. As seen in Fig.8 7) the (n, f) cross section of ^{237}Np is larger by a factor of two or three than the (n, 7) cross section of ^{237}Np in the energy range above 0.5 MeV. These facts suggests that the application of these neutrons to the TRU transmutation system is very promising.

The recent advance made in accelerator technology during the past decade has given the high possibility of providing the intense proton beam to the present transmutation system.

2-2. Basic design study of the TRU transmutation system

At the present stage the accelerator-driven transmutation system mainly utilizing the fission reactions induced by spallation neutrons has been studied as the type of transmutation plant because of high transmutation rate and good energy balance. We have been promoting the conceptual studies on the TRU transmutation in the target-core system driven by an accelerated proton beam.7),8) The basic conditions settled for the system design are (1) high transmutation rate of TRU, the goal line of which is the transmutation of amount of TRU produced in about ten commercial 1 GWe PWR plants, and (2) good energy balance, in which it can generate enough electricity to operate the accelerator at least. The yields of TRU produced per year from 1 GWe PWR are summarized in Table 2. Total amount of TRU is about 26 kg/y, 56 % of which is ^{237}Np.

Therefore it is considered to be advantageous to adopt high energy proton-induced spallation and the secondary neutron causing fission reactions as a means of the nuclear transmutation of TRU. Figure 9 shows the target-core of hybrid plant driven by high power proton beam with the energy of 1.5 GeV and the current of several tens of mA. The core design parameters are summarized in Table 3. The tungsten target is 60 cm long in the direction of the incident beam, 1 m high and 10 cm wide and is installed in an TRU-fueled subcritical core (keff: 0.9- 0. 94). The core has dimensions of 2- 2.6 m length, 1 m height and 1 m width, surrounded by the HT-9 steel container with thickness of 20 cm. A beam window is located at a depth of 0.7 m from the front face and has a rectangular cross section with dimensions of 1 m high and 0.1 m wide. The heat generated in the TRU fuel is removed by the forced circulation of liquid metal coolants Na/Pb-Bi. The heat removal performance is one of the major factors to determine the rate of TRU transmutation in the system. The core consists of metallic alloy fuel of TRU and provides considerably harder neutron spectrum than the other types of fuels. The fuel consists of two types of alloys, Np-22Pu-20Zr and AmCm-35Pu-5Y and has the sufficiently high phase stability. The fuel pin cell geometry is shown in Fig- 10, with a diameter of 4 mm cladded with HT-9 steel. The pin pitches has been adjusted to be 8 mm and 10 mm for Na and Pb-Bi cooled cores, respectively, to keep k_{eff} around 0.86 ~ 0.95. Here Pu is added initially to the fuel in order to suppress the reactivity swing within an acceptable burnup range. With addition of 20 wt% of Zr, the melting point of Np is supposed to increase from 640 "C UP to about 900 "C. The fuel assembly in the core is similar to that employed in a TRU burner reactor design. ″

For neutronics calculations the target-core system is approximated by an axially symmetric cylinder with the same volume as the original system for the efficient computation. A circular beam window located at the center line has a diameter of 0.36 m and the maximum beam diameter is 0.2 m. The nuclear spallation processes above the cutoff energy of 15 MeV were calculated by NMTC/JAERI code. For the reaction below 15MeV the Monte Carlo transport code MORSE-DD was used with 52 neutron group constants edited from JENDL-2 and ENDF-B4, where spallation neutrons were treated as the source. The results of the neutronics calculations, for four cases of the system cooled by Na and Pb-Bi, with and without the tungsten target, are summarized in Table 4. The number of TRU nuclei disintergated in fast fission reactions is much larger than that in the spallation reaction for each case. Profiles of the two-dimensional power distribution for these cases are shown in Fig. 11 (a) to (d), respectively. It is apparent that the power peaking which occurs just behind the beam window is lower in the system with the tungsten target than in the one without it due to the flattening effect for cases of both coolants and the flatter power distribution increases the number of transmuted nuclides. The maximum transmutation rate is 202 kg/year in the core with the target cooled byNa (reference system).

Thermal hydraulics calculations for the system were done to obtain the maximum achievable thermal power within the maximum temperature limits of fuel and cladding. Maximum temperatures in the TRUfuel and theHT-9 cladding tube are limited to 900 °C and 650 °C, respectively, where the temperature at the inlet of coolant is set to 300 °C. The temperature distributions along the hottest fuel pins cooled by Nainthe core with the tungsten target are shown in Fig. 12. The maximum thermal power is limited by the maximum allowable fuel temperature of900 °C. The operating conditions of the target-core system are summarized in Table 5. In the case ofNa cooling and the tungsten target the maximum thermal power is 691 MW with the maximum and average power densities of 889 W/Cc and 307 W/cc. The thermal power is sufficiently large to supply the electric power to the accelerator while the beam current required for the power is 22.6 mA. Without tungsten target the thermal power is 405 MW with the maximum and average power densities of 776 W/cc and 159 MW/cc and its peaking factor is larger bya facter of 1.7 than the case with the tungsten target. The maximum powers of Pb-Bi cooled core with and without the target are considerably lower than those of the Na cooled one and the beam current required is less than 8 mA. This is attributed mainlyto the lower thermal conductivity of the coolant and the wider fuel pin pitch than in the case ofNa cooling. The variation of multiplication factor k_{eff} with burnup days was calculated as shown in Fig. 13. The increase tendency of k_{eff} at the initial stage turns the decrease around 1000 days and the subcritical operation of the system can be kept during the burning time. The changes of concentrations of some minor actinides with burn-up days in the reference system(Na cooling, with tungsten target) were also calculated, as shown in Fig.14, using the burnup code ORIGEN2. The amounts of ^{237}Np and ^{241}Am at 1500 burning days become one half of their initial inventories, while ^{238}Pu and ^{242}Cm, which are not contained in the initial loading, build up.

The hybrid transmutation system of accelerator-target-core hasthe additional merits :

(1) The system can be quickly shut down only by switching off the beam current of proton accelerator.

(2) Since the hybrid target-core is always operated in a subcritical state, it can have a simpler structure without safety and control rods than the reactor.

(3) The higher burnup rate is expected for the TRUfuel in this system

with no constraints for the criticality. In this case the main limitation is the lifetime of fuel and structual material under the irradiation conditions.

(4) The target-core designing is flexible because it is free from the safety requirements of non-positve Na void coefficients and the poisoning effect due to variation of isotope abundances in the fuel composition as the fuel is burning.

However there are technological items requiring further researches and developments:

(a) an intense proton beam accelerator (1.5 GeV, ~10 MA),
(b) TRU technologies,
(c) high energy radiation shielding.

Moreover the spallation target system can be used for other applications such as the breeding of fissile nuclides and the creation of very intense neutron sources. The useful nuclides or short-life RI's used for special purposes can be produced from residual nuclides after transmutation and nuclide partitioning processes also.

2-3. Spallation intergal experiment

More accurate experimental data for the spallation reaction in the energy range of ~ 100 MeV to ~ 1 GeV are needed to examine the actual efficiency of TRU transmutation by spallation reaction9),10) and to upgrade the simulation code system for the TRU transmutation processes. The research plan of spallation integral experiment by using the high energy proton beam has started. The lead cylinder system for the experiment has been set up last March near the dump of beam line connected to the proton synchrotrons booster at KEK. Figure 15 shows the lead cylinder installed in a Stainless Steel container with 100 cm length and 60 cm diameter. This has several small holes parallel to the central axis, which are pluged by specimen wires such as Ni, Au, Cu and Fe. Reaction products in these specimen by irradiation of 500 MeV protons are identified from their γ-ray emissions measured by a Ge(Li) detector. The energy of spallation neutron can be known from the activity of specimen foils with the threshold energy of neutron emission, imbedded in the holes in the cylinder. The safety analysis for spallation experiment has been made to know whether the activity in the irradiated specimen and the dose rate are lower than the values restricted by the law when they are transported in the cask to be measured at JAERI. The switching magnet has been newly equipped on the booster beam line to control the intensity of irradiating beam by adjusting the number of pulses in current. The irradiation experiment for the lead system has started November this year. In the next step, a tungsten or a depleted uranium target, which is inserted in the central region of the lead cylinder, will be used to simulate the TRU target spallation experiment.

3. Summary and Conclusion

Finaly the report is summarized as follows.

The computer code system has been developed and upgraded for simulating the spallation reaction induced hy high energy protons and designing the TRU transmutation system .

The conceptual design studies have been made for comparison of the accelerator-driven TRU transmutation systems with and without the tungsten target. When the Na cooled TRU metal fuelled core with tungsten target is operated at the thermal power of 691 MW and the beam current of 23 mA, this system can transmute about 200 kg TRU per year. In the case of the Pb-Bi cooled system at the thermal power of 342 MW and the beam current of 7.5 mA, it can transmute 140 kgTRU annually. Improvement and optimization of target-core design will be carried out also in more detail through the plant design studies. The performance of transmutation plant of the type of molten TRU is examined as the next step.

The spallation integral experiment for lead target will start soon, using 500 MeV proton beam at the KEK booster facility .

References

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Table 1 Number of particles emitted from a spallated nucleus

Emitted particle	Energy of incident proton (MeV)			
	380	1000	2000	2900
Proton	0.994	2.924	3.697	3.276
Neutron	12.085	16.050	170319	15.243
Deutron	0.1249	0.7063	0.9108	0.7729
Triton	0.0576	0“.2719	0.3407	0.2956
Helium 3	0.0010	0.0258	0$0411	0.0361
Alpha	0.0732	0.2777	003079	0.2588

Table 2 TRU production per Year from 1 GWe PWR

Nuclide	Weight (kg)	Fraction (O/J
^{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 (l(g)	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 3 Target-core design parameters

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

Table 4 Target-core operating condition

		Reference	Version-1	Version-2	Version-3
Pins		TRU+W	TRU+W	TRU	TRU
Coolant		Na	Pb-Bi	Na	Pb-Bi
Proton Beam Current(mA)		22.6	7.5	18.1	5.4
Thermal Power (MW)		691	484	405	163
Power Density (W/cc)	refix.	889	S23	776	425
	ave.	307	246	159	83
Linear Power Rating (W/cm)	max.	695	499	713	530
	ave.	240	235	146	103
Coolant Temperature (°C)	outlet	389	451	352	377
Clad Temperature (°C)	max.	492	610	481	589
Fuel Temperature (°C)	max.	900	900	900	900
Coolant Velocity [m/s]	max.	8	2.35	8	2.35
Pressure Drop (kPa)		78	67	62	48

Table 5 Performance of this transmutation system

Target system	Reference	Version-1	Version-2	Version-3
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 > 5MeV	0.67	0.24	0.64	0.42
(< 15MeV)	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 · sec	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 3000 MWt LWR	7.6	5.3	4.3	1.8
Burnup reactivity swing (% Δk/k	3.8	2.9	2.7	2.1

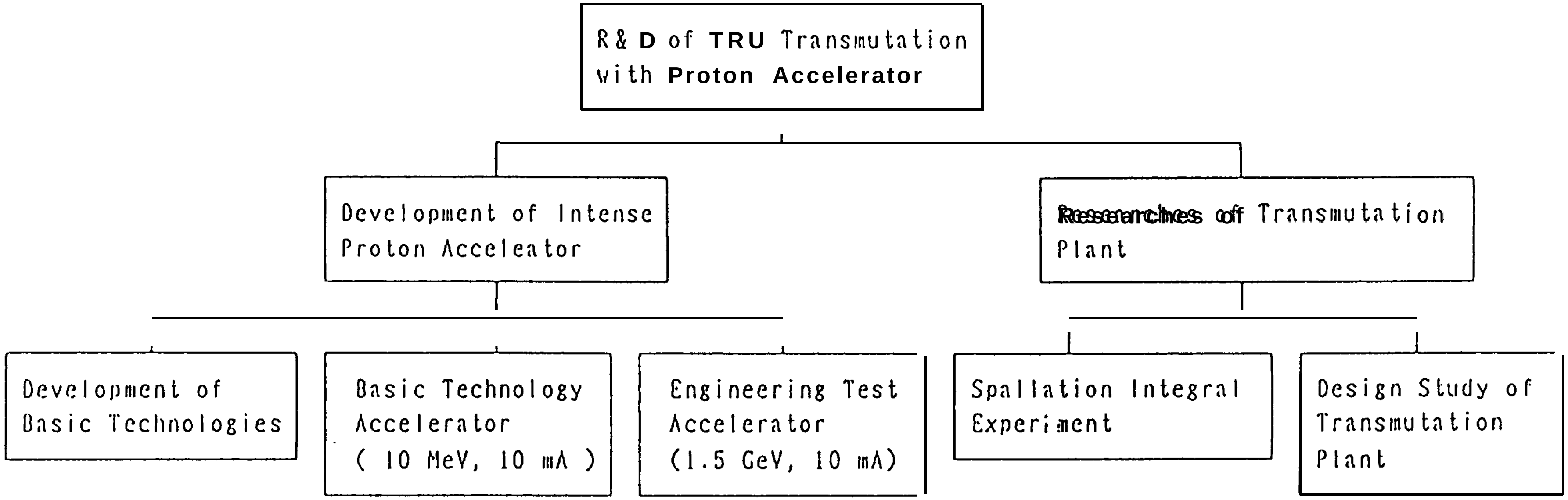


Fig. 1 Tree Structure of R & D Items for TRU Incineration Plant Driven by High Intensity Proton Accelerator

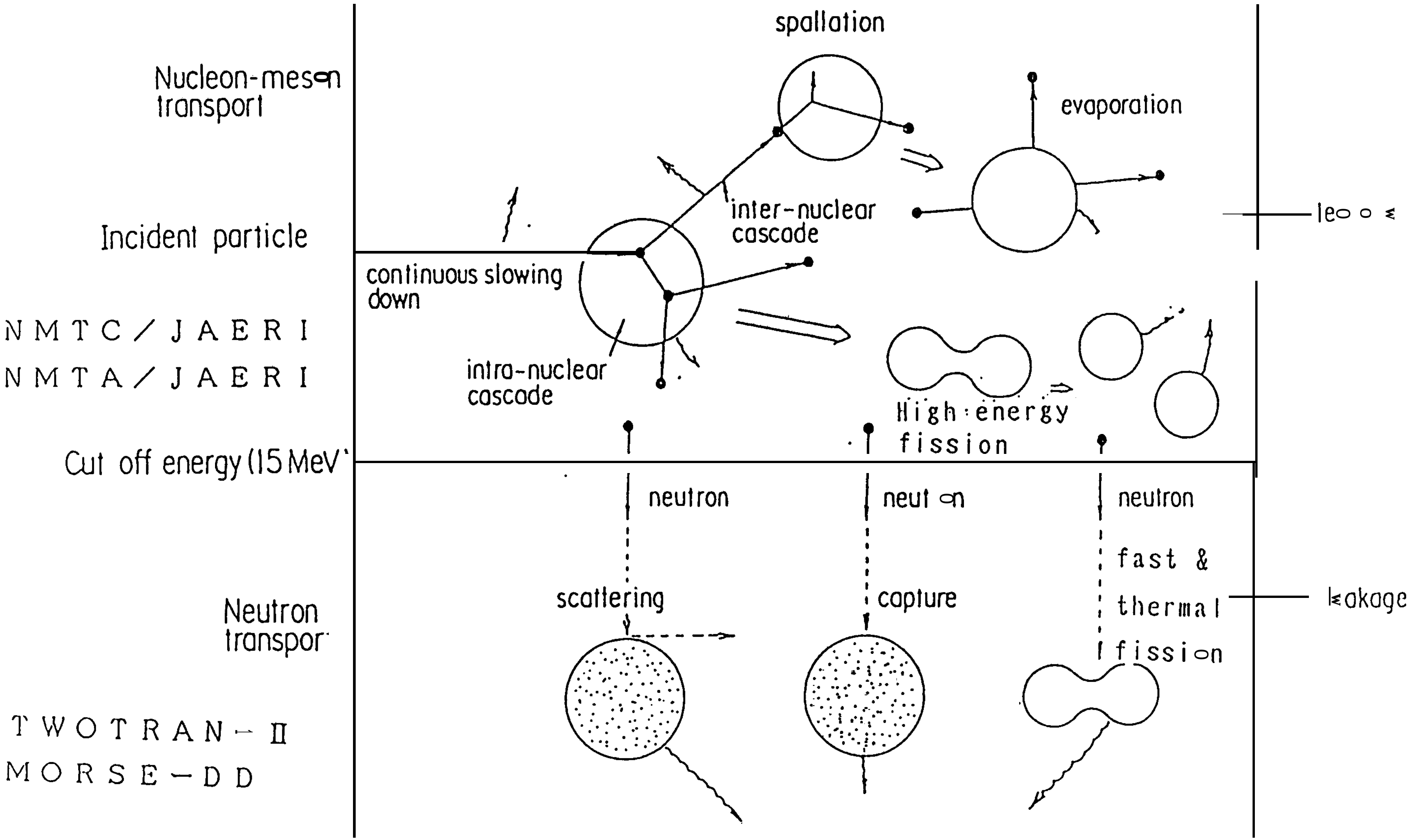


Fig. 2 Spallation process in the target injected by high energy protons

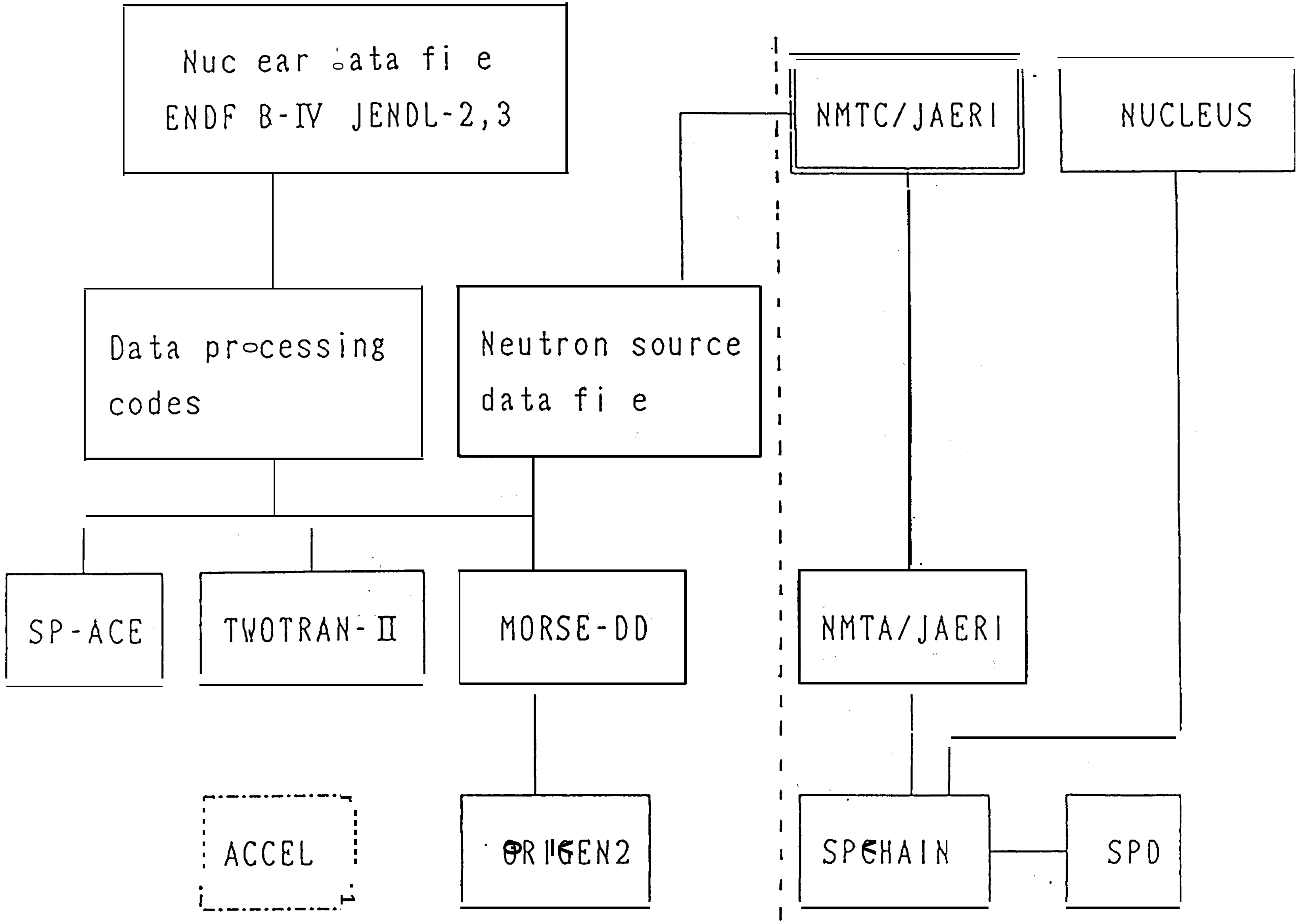


Fig. 3 Simulation code system for the accelerator transmutation system

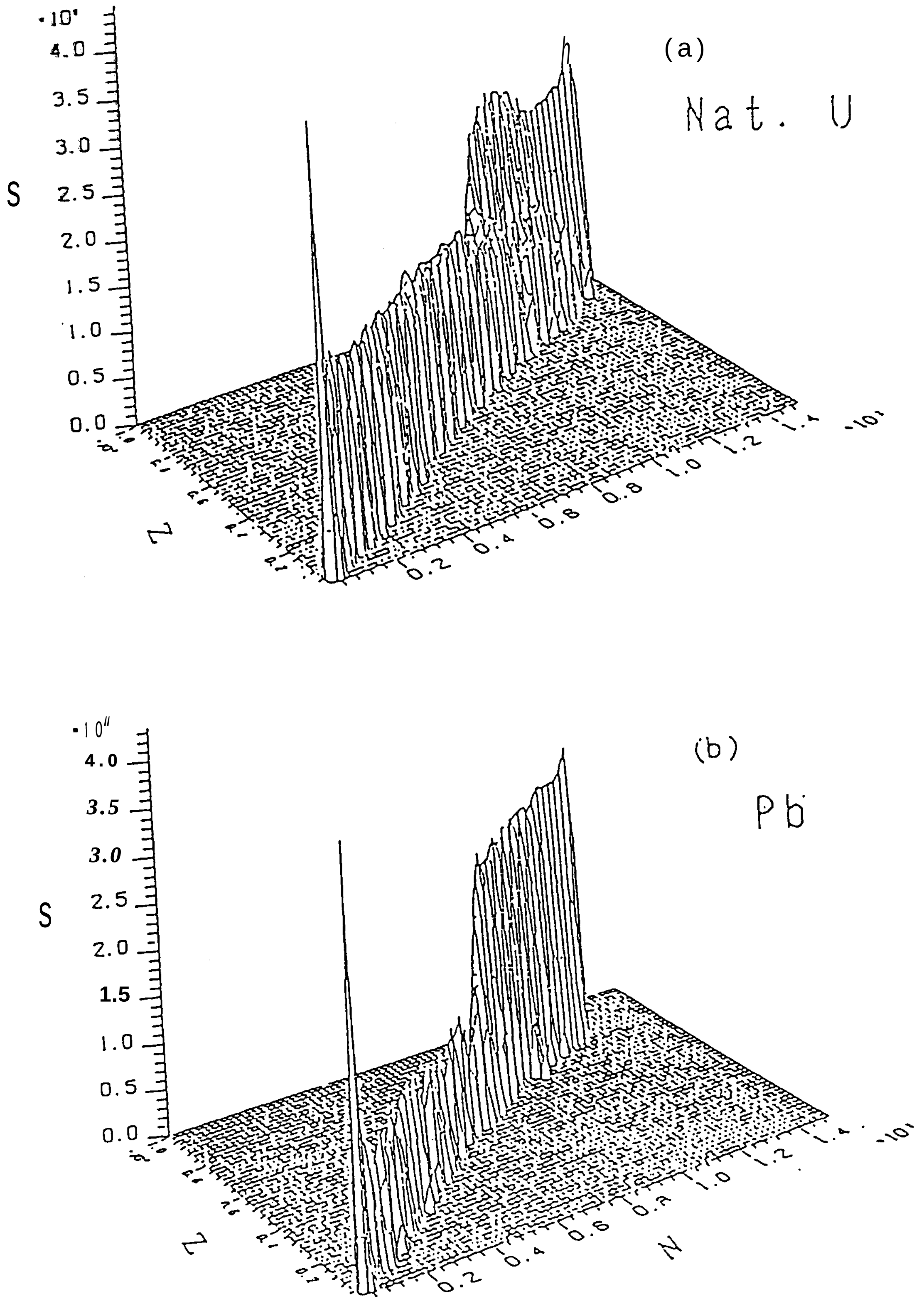
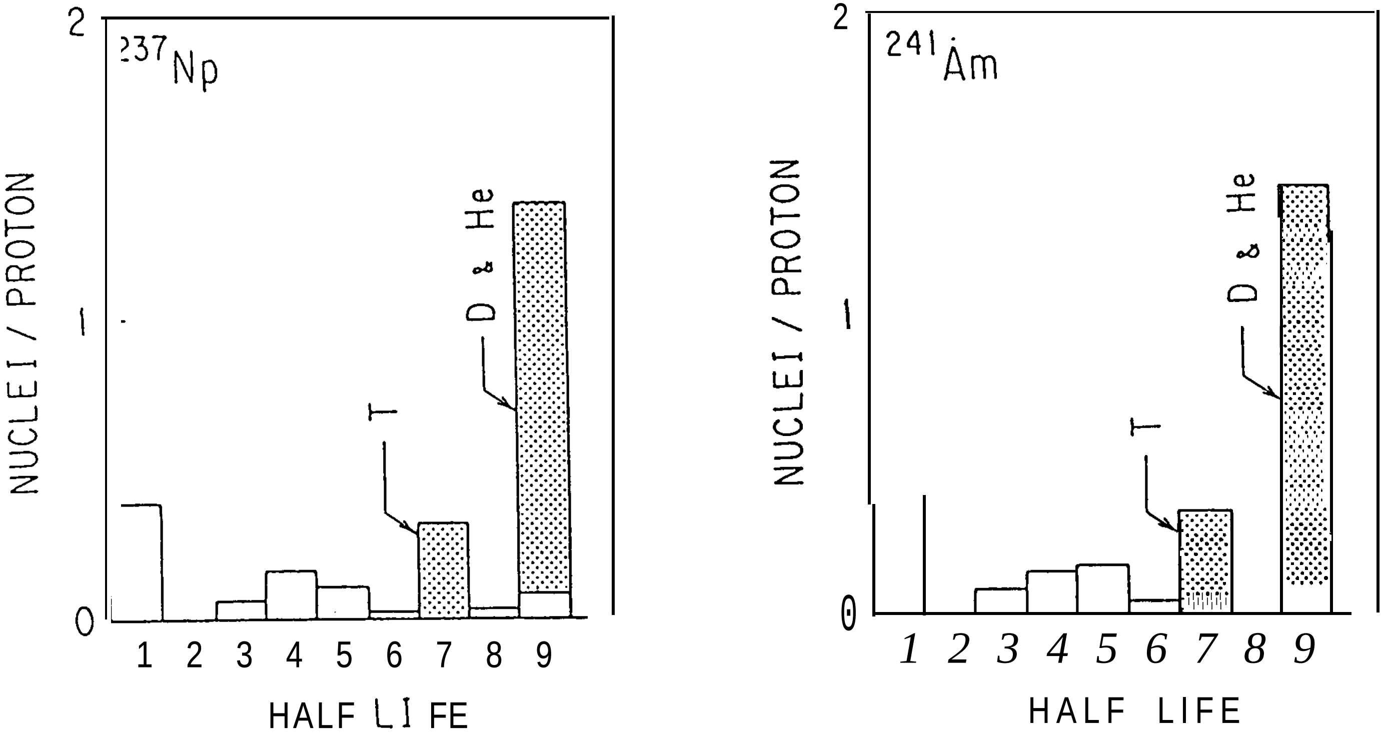


Fig. 4 Yields of reaction products from a) U and b) Pb nuclei bombarded by a 1 GeV proton



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

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

Fig.5 Halflife distribution of products in the spallation reaction due to 2 GeV protons

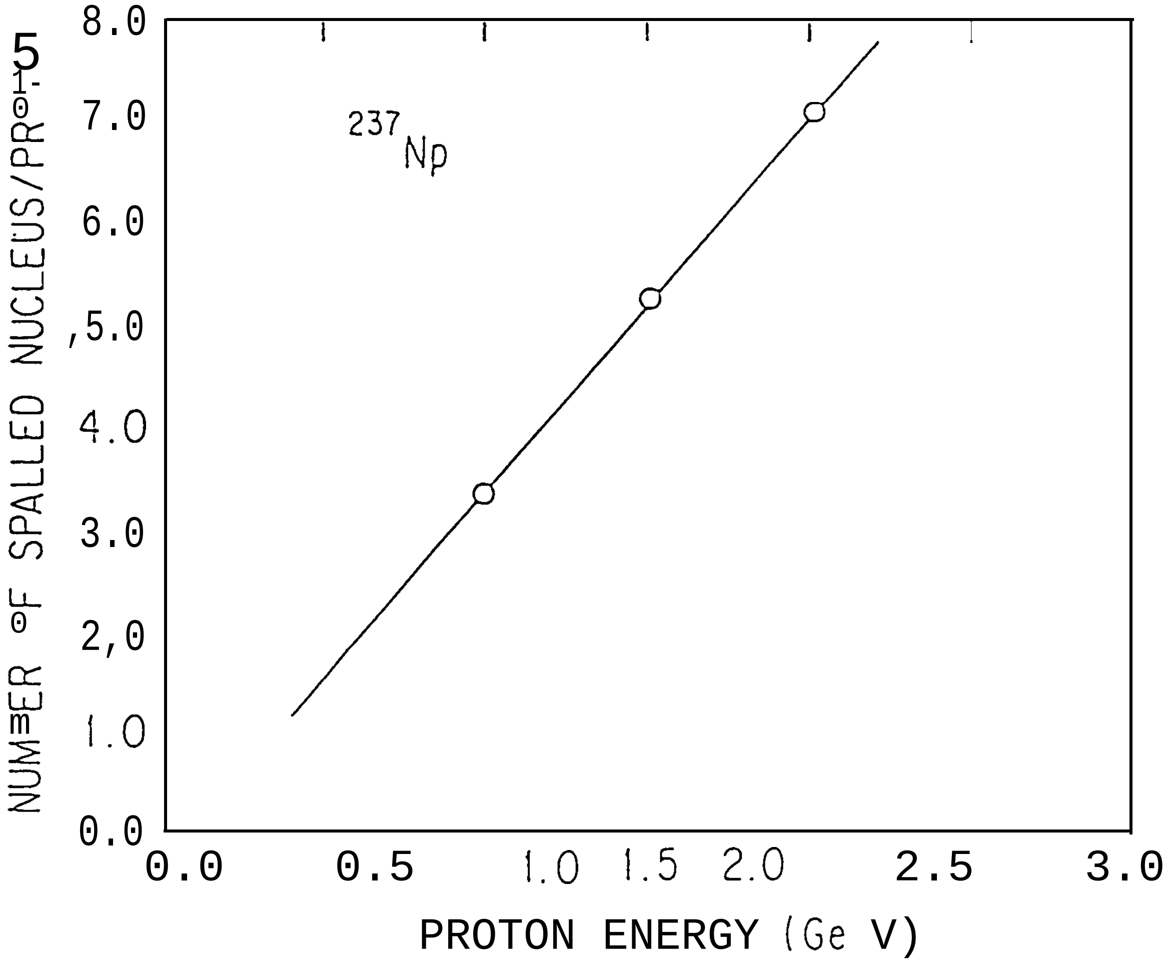


Fig. 6 Energy dependence on number of nucleus destructed due to spallation reaction

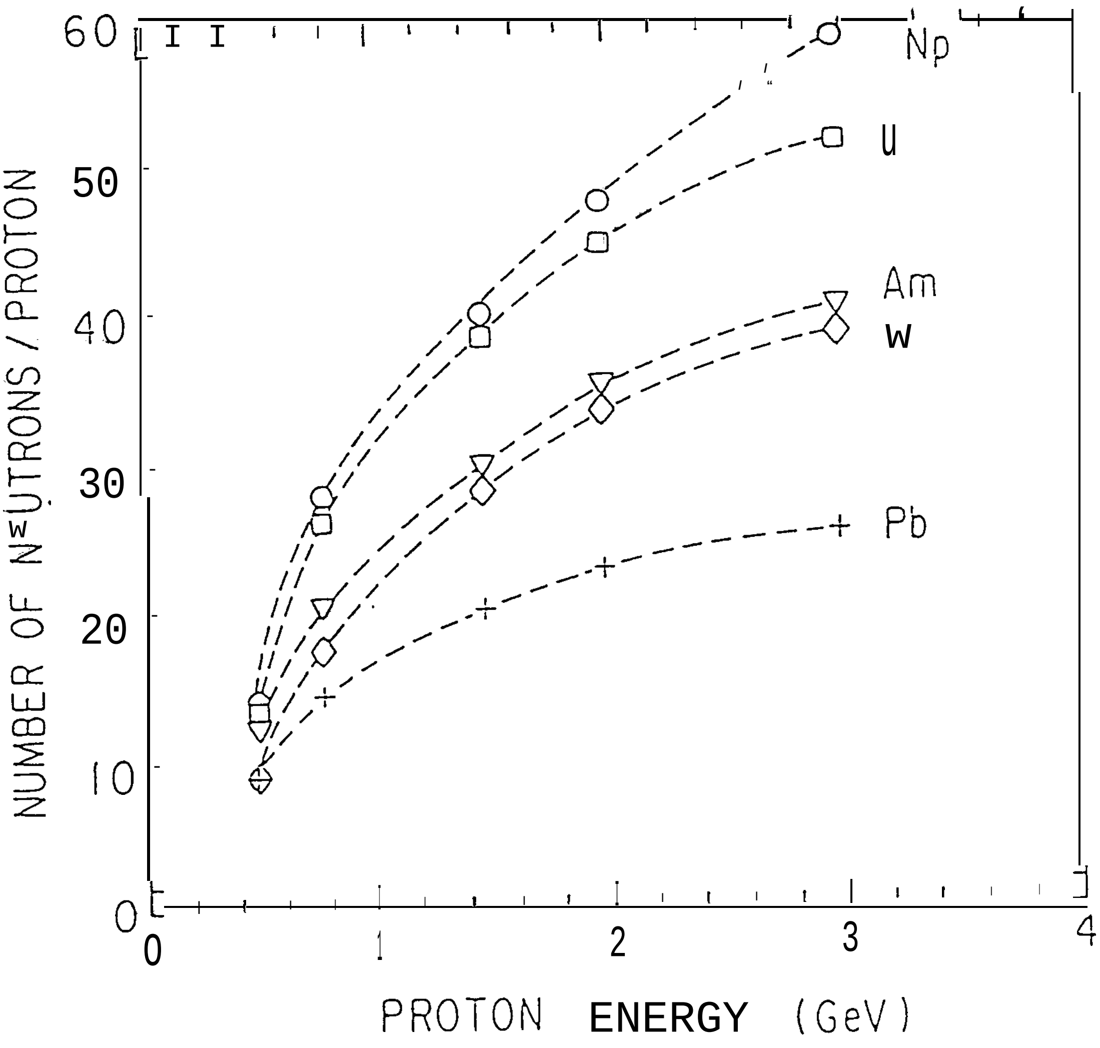


Fig. 7

Energy dependence on number of neutrons generated by spallation reaction

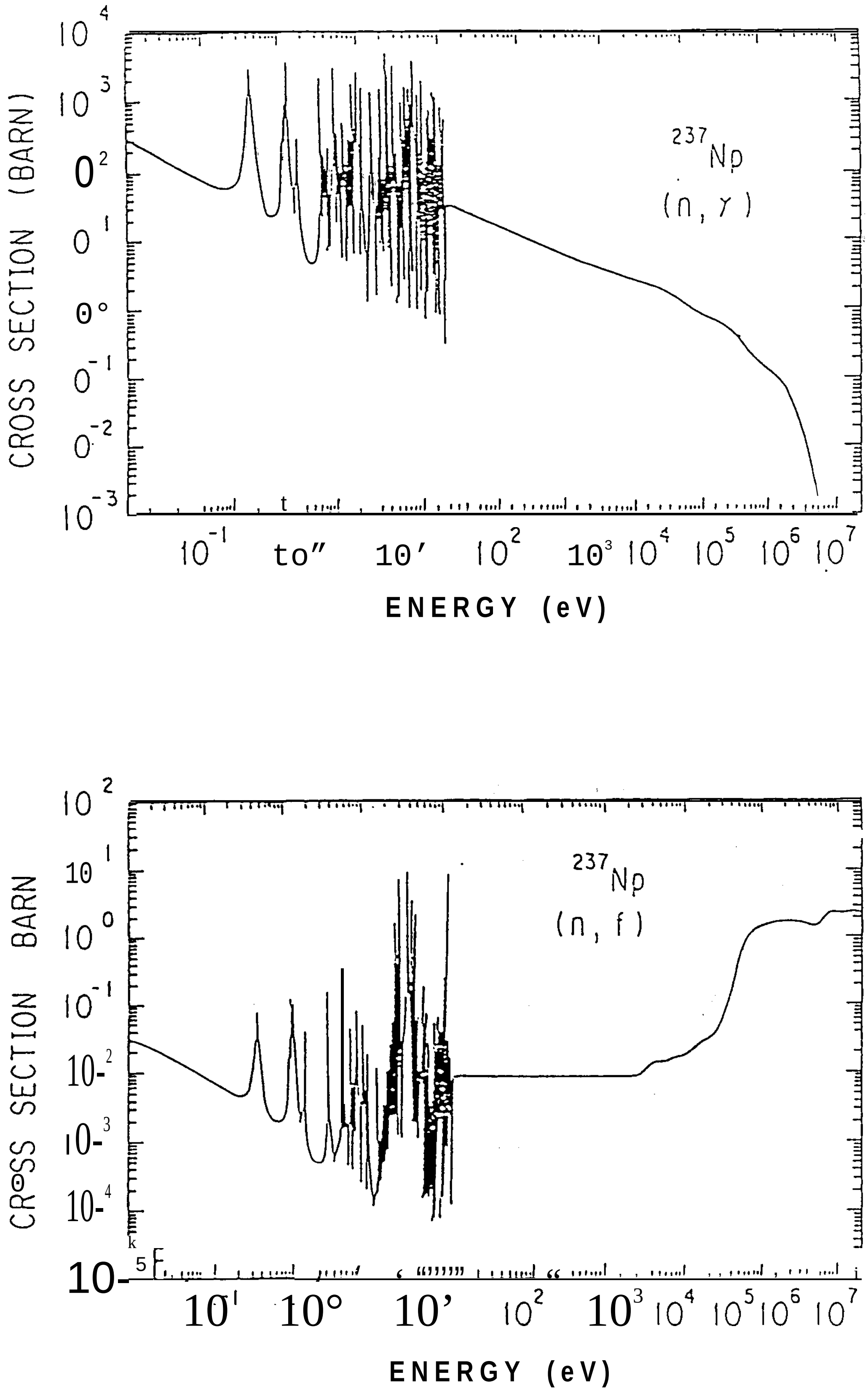


Fig. 8 Neutron cross section of ^{237}Np

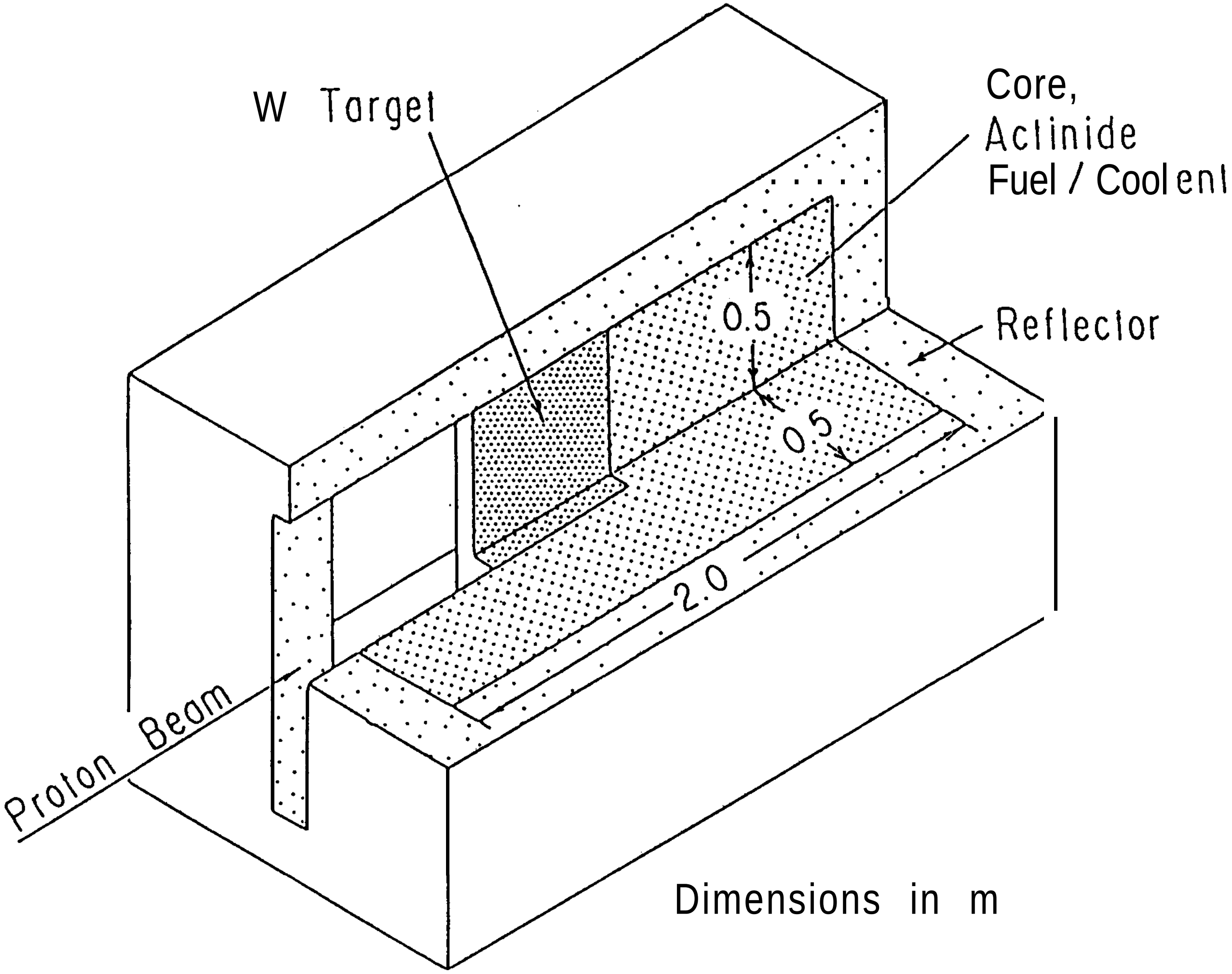


Fig. 9 Target-core configuration of hybrid plant(refernece system)

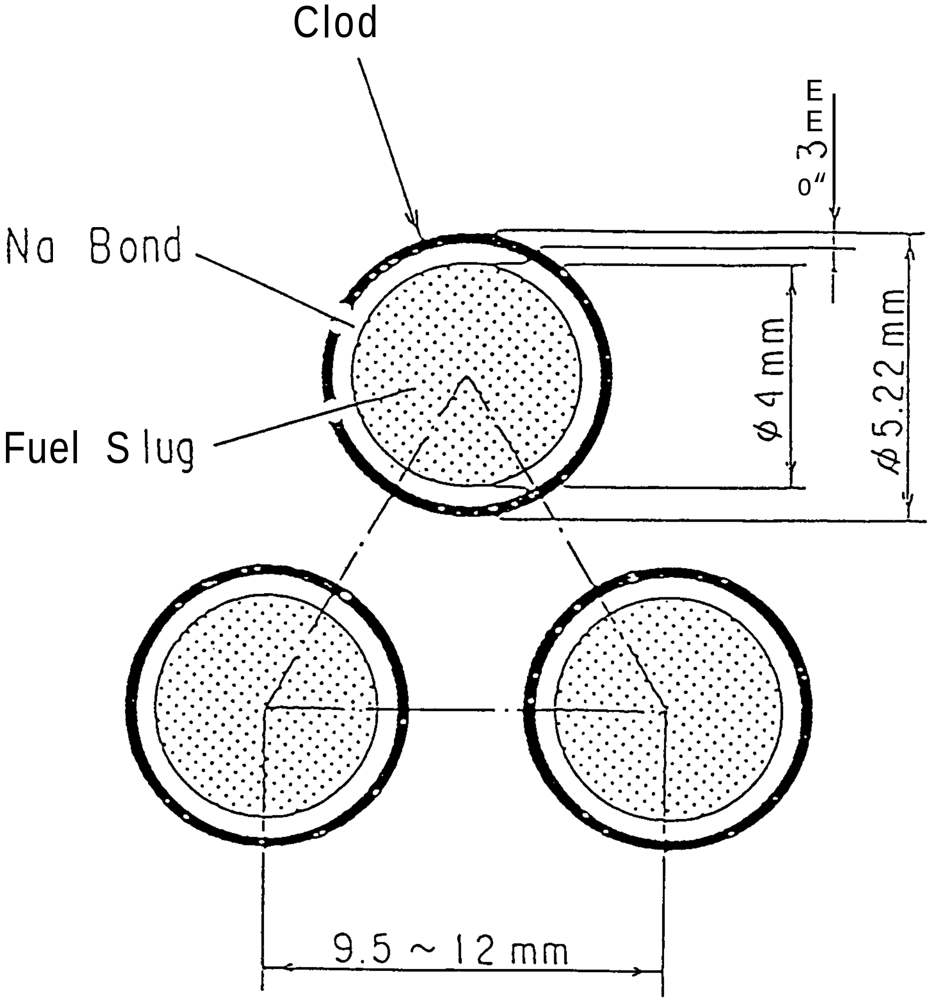
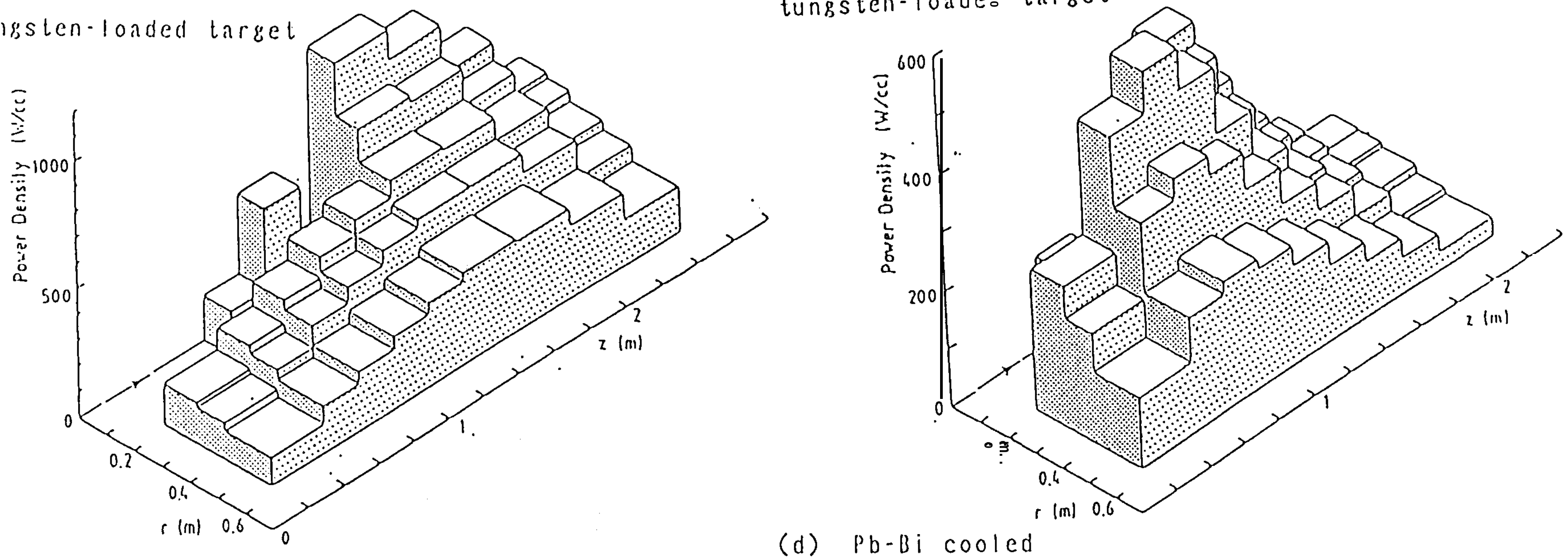
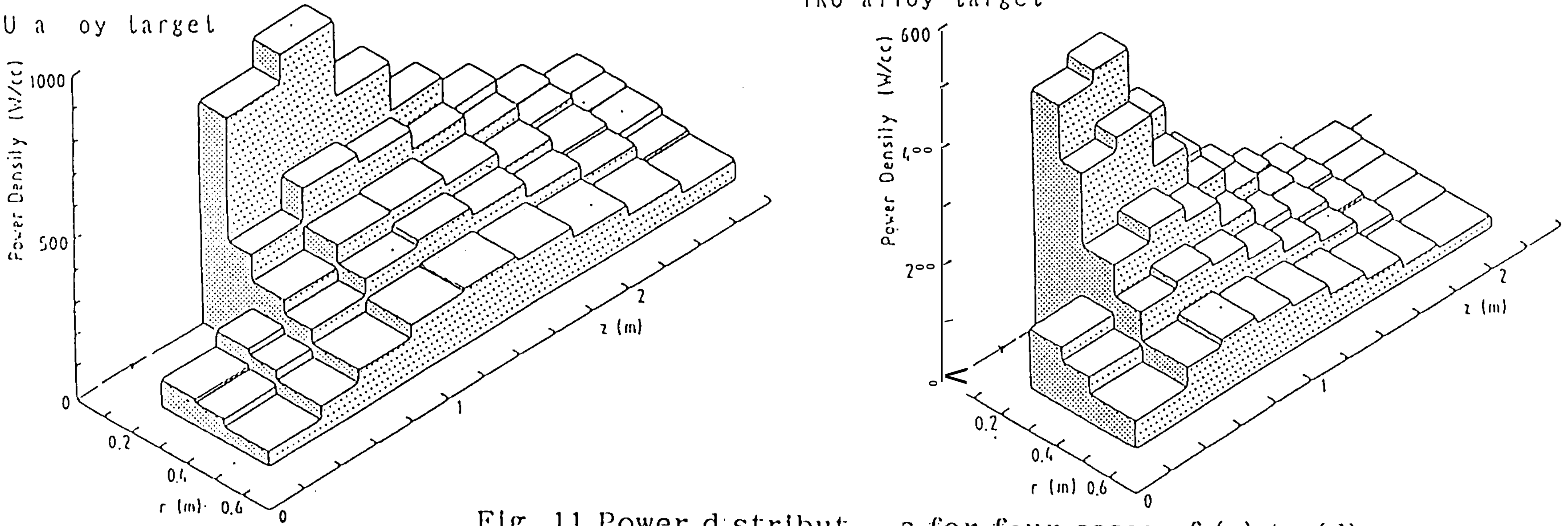


Fig. 10 TRU alloy fuel pin geometry


(a) Na cooled
tungsten-loaded target

tungsten-loaded target


(c) Na cooled
TRU alloy target

(d) Pb-Bi cooled
TRU alloy target

Fig 11 Power distributions for four cases of (a) to (d)

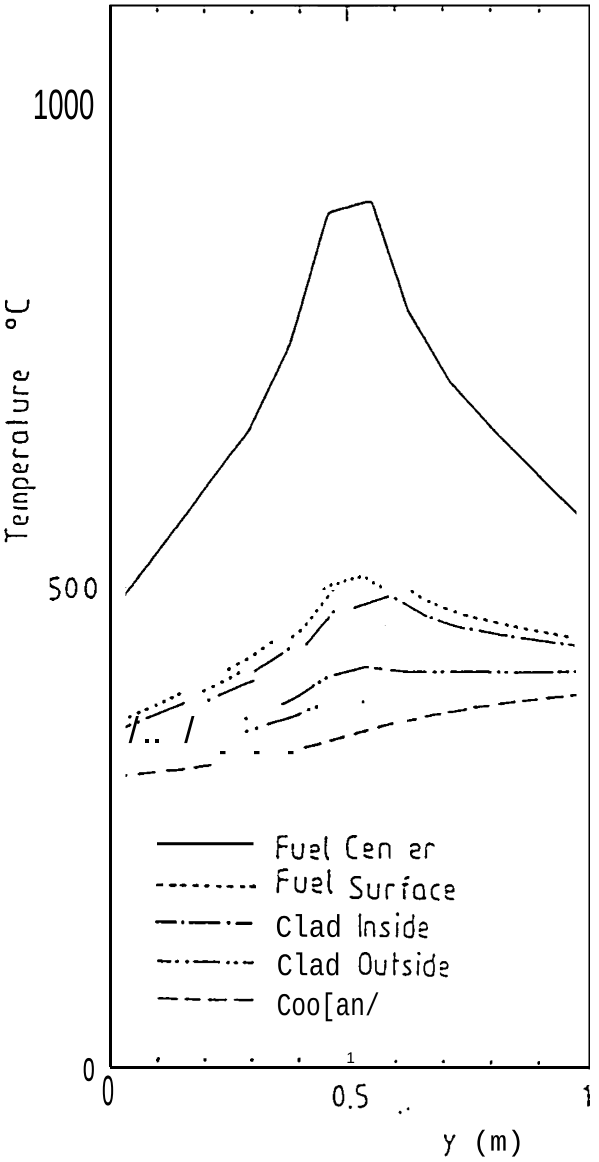
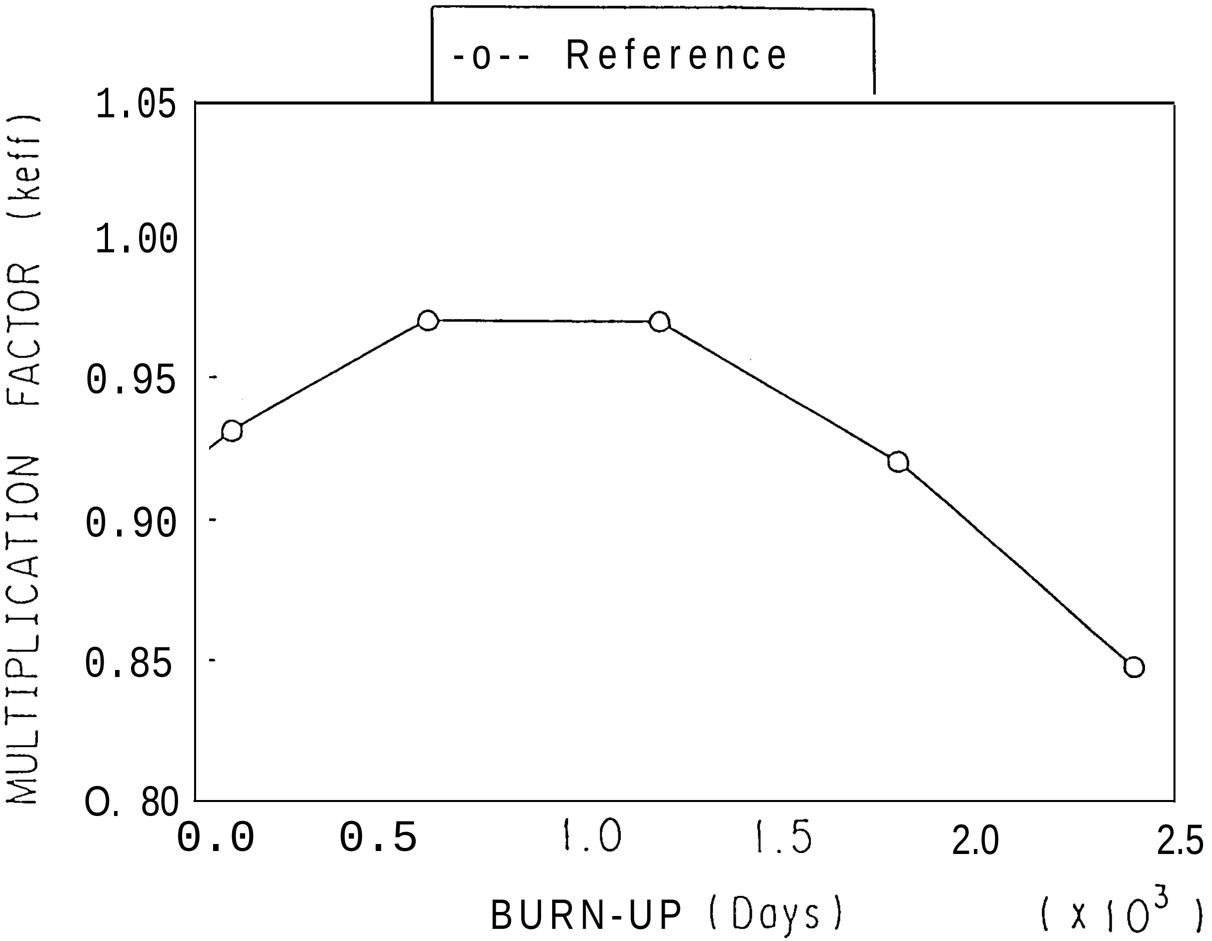


Fig. 12 Temperature distribution in the reference core



* 1,000 burnup days correspond to --105 MWD/ ton

Fig. 13 Change of k_{eff} with burn-up

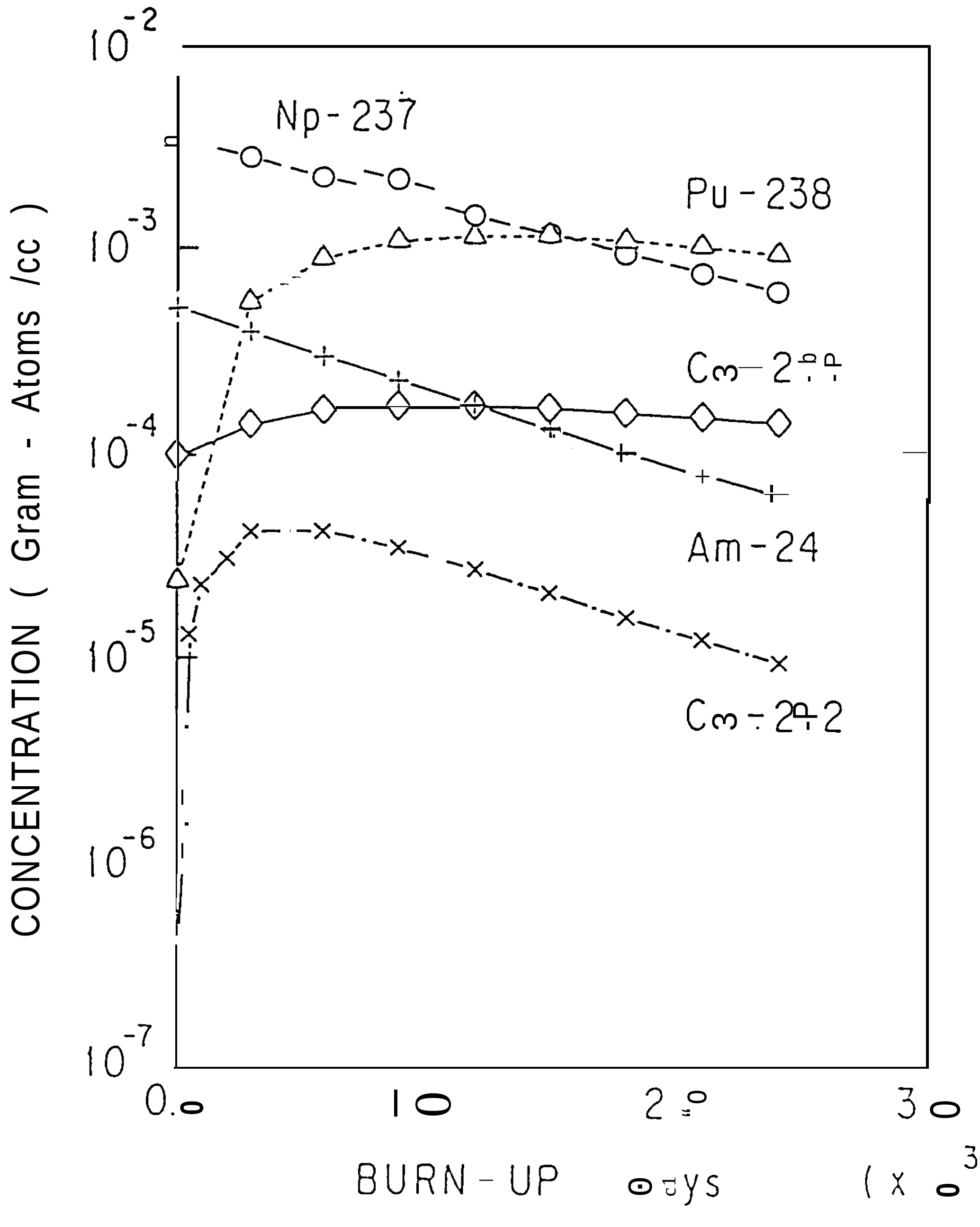


Fig. 14 Change of trans-uranium inventory with burn-up

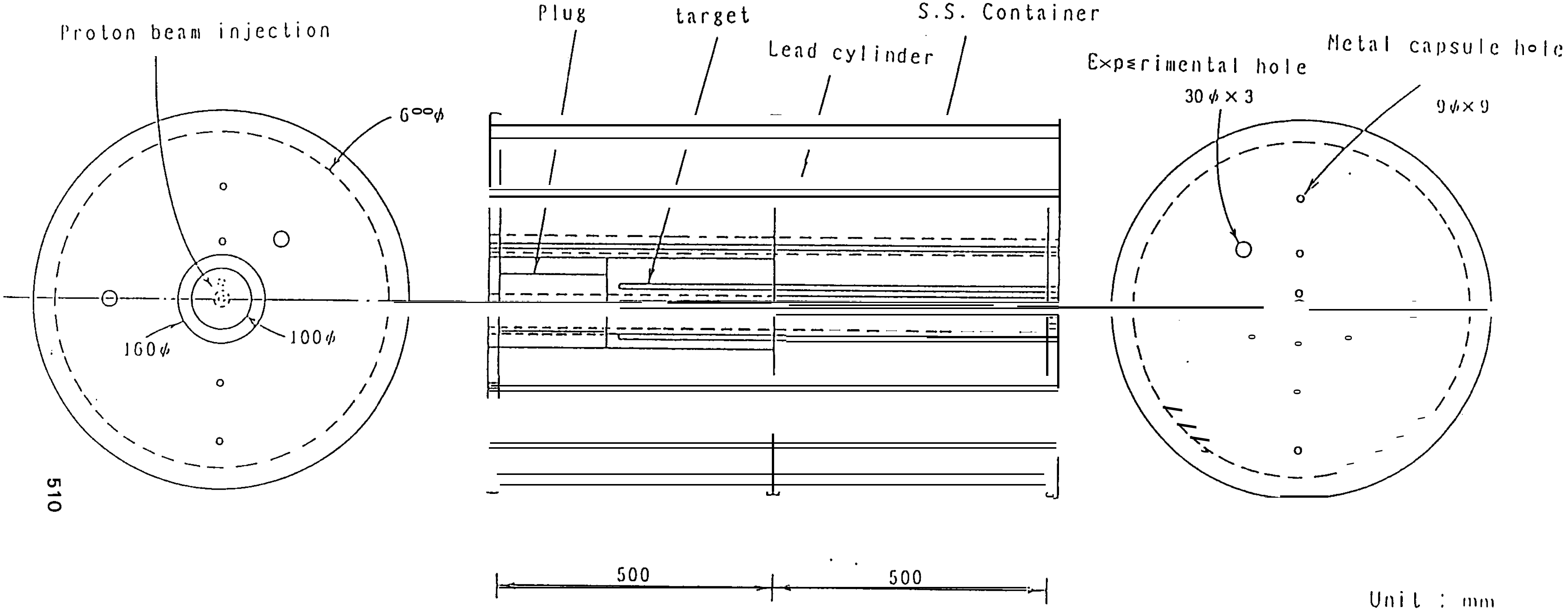


Fig. 15 Lead cylinder target for the spallation experiment