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STUDY OF TRU TRANSMUTATION PLANT WITH A 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 the nucleonics and hydraulics analysis points of view. The proposed transmutation plant is a hybrid system of an intense proton accelerator, a tungsten target cooled with sodium and a subcritical core loaded with the TRU metal fuel. In this plant the transmutation rate of about 200 kg TRU a year (generated from about 8 units of 1 GWe PWR) are attainable and the marginal electricity more than one needed to drive the acclerator can be produced.

With the aim to make assessment of the plant design and to upgrade the computer codes for simulating nuclear spallation processes in the transmutation plant, the integral spallation experiment has been planned. The lead target has been set up along the 500 MeV beam line of a proton synchrotron booster at KEK. The first irradiation experiment is scheduled to start this Autumn.

The R & D schedules of the Engineering Test proton Accelerator(1.5 GeV, ~ 10 mA) have been made to ascertain the engineering feasibility of the transmutation plant. As the first step the Basic Technology proton Accelerator(10 MeV, 10mA) is to be constructed to develop the advanced technologies of higher beam intensity, high quality beam loading with low emittance, high efficient RF power, etc.

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. [1]

The recent advance made in accelerator technology during the past decade has given the high possibility of providing the intense proton beam to the proposed transmutation system. Most of the products in the transmutation by using only the spallation reaction have halflives shorter than most of fission products. The nuclear spallation reaction between high energy protons (above 1 GeV) and heavy metal such as TRU generates many neutrons with the hard spectrum likely in a fast reactor. These facts makes the use of an proton accelerator attractive as a means of nuclear transmutation of TRU. 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. The hybrid transmutation system of accelerator-target-core has the 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 TRU fuel 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.

In the present paper the present status of the research at JAERI is described as following items,

1) development of the basic simulation code system,
2) conceptual study of the TRU transmutation plant,
3) spallation integral experiment,
4) development of a intense proton beam accelerator.

Tree structure illustration of R & D Items for the TRU Transmutation Plant and High Intensity Proton Beam Accelerator is shown in Fig.1.

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. Research of transmutation system driven with proton accelerator

2-1. Development of simulation codes

The knowledge of residual nuclides accumulated in the target spallated by the proton beam irradiation is very improtant for the feasibility estimation of proton-induced TRU transmutation. Actually, however, it is almost impossible to obtain exactly the time evolution of yields of all the nuclides in the target due to enormous computing time. The SPCHAIN code has been developed to compute approximately the buidup and decay of spallation products (SP) by expanding the depletion code DCHAIN2 2) for fission products. By assuming that any complicated decay process of nuclides is disintegrated to linearized decay chains, the equation can be solved by the Bateman method. The new data of decay types, decay constants, branching ratios and decay schemes have been compiled in the SPCHAIN data library for about 1100 nuclides needed for the TRU spallation calculation. These data were mainly collected from Table of Isotope (7th version) and the data stored in the DCHAIN2 library. Figure 2 represents the nuclide distribution of both data sources in the (A,Z) plane. When the halflife time of a nuclide was not obtained by surveying the data, it was calculated by using the decay calculation program 3) or guessed from the trend of data of nuclides located in the neighborhood of the nuclide in the Nuclear Chart. The yields of SP in the spallation reaction were computed by using the code NUCLEUS. 4) As a preliminary analysis the yields and radioactivities of residual nuclides in a ^{241}Am target irradiated by 1 GeV protons has been calculated. Figure 3 shows the activity rate distribution of buildup elements at the time stage of one year cooling after irradiation of ten hours.

The target-core design code system SP-ACE with a deterministic calculation method is being developed to simulate the transmutation process of TRU wastes in a subcritical system in reasonable computing time and precision. Figure 4 shows the main flow chart of this system. The neutron transport code RABBLE-THERMOS computes the region-wise neutron flux, using ultra fine group constants and the distribution of spallation neutron source, which can be obtained from Monte Carlo calculations of high energy nuclear reactions and particle cascades in NMTC/JAERI. Using these fluxes the routine COLAPU gives the effective cross section and the average flux in each region for the burnup code COMRAD to calculate the yields of nuclides, heat generation and γ-ray intensitiy in the transmutation system.

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

We have been promoting the conceptual studies on the TRU transmutation in the target-core system driven by an accelerated proton beam. 5),6) 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 1. Total amount of TRU is about 26 kg/y, 56 % of which is ^{237}Np.

High energy nucleons generated in the spallation can transmute TRU nuclides through the cascade processes. Figure 5 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 but it is too small to process TRU wastes in the commercial base unless the proton beam can have high current more 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. 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. 6. For the case of ^{237}Np target bombarded by a 1.5 GeV proton the neutron number is ~40. As seen in Fig.7 6) the (n, f) cross section of ^{237}Np is larger by a factor of two or three than the (n, γ) cross section of ^{237}Np in the energy range above 0.5 MeV.

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 8 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 mA. The core design parameters are summarized in Table 2. 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 (k_{eff}: 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 stability⁶⁾. The fuel pin cell geometry is shown in Fig. 9, 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 ℃ up to about 900 ℃. The fuel assembly in the core is similar to that employed in a TRU burner reactor design. ⁷⁾

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 ⁸⁾. For the reaction below 15

MeV the Monte Carlo transport code MORSE-DD [9] 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 3. 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.10(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 flatterning effect for cases of both coolants and the flatterned power distribution increases the number of transmuted nuclides. The maximum transmutation rate is 202 kg/year in the core with the target cooled by Na.

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 TRU fuel and the HT-9 cladding tube are limited to 900 ℃ and 650 ℃, respectively, where the temperature at the inlet of coolant is set to 300 ℃. The temperature distributions along the hottest fuel pins cooled by Na in the core with the tungsten target are shown in Fig. 11. The maximum thermal power is limited by the maximum allowable fuel temperature of 900 ℃. The operating conditions of the target-core system are summarized in Table 4. In the case of Na 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 sufficietly 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 by a 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 mainly to the lower thermal conductivity of the coolant and the wider fuel pin pitch than in the case of Na cooling.

The variation of multiplication factor k_{eff} with burnup days was calculated as shown in Fig. 12. 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.13, using the burnup code ORIGEN2. The amounts of ^{237}Np and ^{241}Am at 1500 burning days become one half of their initial inventries, while ^{238}Pu and ^{242}Cm, which are not contained in the initial loading, build up.

2-3. Spallation intergal experiment

More accurate experimental data for the spallation reaction in the energy range of ~1 GeV to ~100 MeV are needed to examine the actual efficiency of TRU transmutation by spallation reaction[10),11)] 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 synchrotron booster at KEK. Figure 14 shows the lead cylider installed in a SS 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 irradiation experiment for the lead system will be started this Autumun according to the machine schedule. The switching magnet will be newly equipped in the near future on the booster beam line to control the intensity of irradiating beam by adjusting the number of pulses in current. 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.

2-4. Development of intense high energy proton accelerator

The basic concept of the Engineering Test Accelerator with a beam energy of 1.5 GeV and a current of 10 mA was proposed for the TRU transmutation as described at the last NEA/CRP meeting. The accelerator has a large scale compared with the conventional ones which are used for basic nuclear physics experiments. In particular, the proton beam is nearly 50 times more intense than that for exisiting machines. Only a linac can satisfy the requirements of such a high beam current with higher efficiency of beam extraction than other circular accelerators. As the first step of the development, the low energy portion of the accelerator structure will be carefully studied, since the beam quality determined at the low energy part is a key factor of dominating the beam efficiency in the high β accelating system. Therefore the smaller size proton linac, Basic Technology Accelerator, with current of 10 mA is going to be constructed to develop the element technology. Figure 15 shows an illustration of the proposed arragement of BTA, which consists of (a)ion source, (b) Radio Frequency Quadrupole linac, (c) Drift Tube Linac and (d) high energy beam transport & dump system, with the output beam energies of 100 keV, 2 MeV, 10 MeV for (a) to (c), respectively. The final beam energy of BTA is chosen to be 10～20 MeV.

Main items of the development of BTA are listed as

① Ion source and its power supply,
② Radio-frequency quadrupole (RFQ) linac,
③ Drift tube linac (DTL),

④ RF power supply system,

⑤ Beam control technology.

The basic experiments of the negative ion source has started to increase the output beam current with high emittance using Neutral Beam Injection test equipment for the fusion experiment at JT-60. The preliminary calculations of beam transport in RFQ linac was made using the computer codes SUPERFISH and PARMTEQ . The trade-off study of ETA system will start this Autumn in coopration with LANL to optimize the concept of ETA arrangment. The input and output energy, the emittance and the acceptance for the various components such as ion source, RFQ and DTL have to be determined carefully and systematically. The high energy portion of the accelerator(high β structure, finally in CW operation) will be also studied in advance of the second stage development. As the opertion mode of the accelerator in the first step, the low duty operation will be appropriate to adjust the various parameters so that the adequate parameters will be surveyed.

3. Summary and Conclusion

The computer code solving the time evolution equation has been developed to calculate the yield of decay and buildup nuclides in the spallation reaction.

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 kg TRU 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 lead target has been set up along the 500 MeV beam line of a proton synchrotron booster at KEK. The first irradiation experiment is scheduled to start this Autumn.

The R & D scheduele of a high intense Engineering Test Accelerator (1.5 GeV,~ 10 mA) is being planned for examining the engineering feasibility of the transmutation system. As the first step basic researches of Basic Technology Accelerator(proton linac: 10~20 MeV, 10 mA) have started to obtain the advanced accelerator technologies.

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Table 1 TRU production per Year from 1 GWe 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 Plant 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 AmCm-35Pu-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 3 Performance of the transmutation plant

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 ($>$ 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 · 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 (% $\Delta k/k$)	3.8	2.9	2.7	2.1

Table 4 Plant operation conditions

		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]	max.	889	523	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

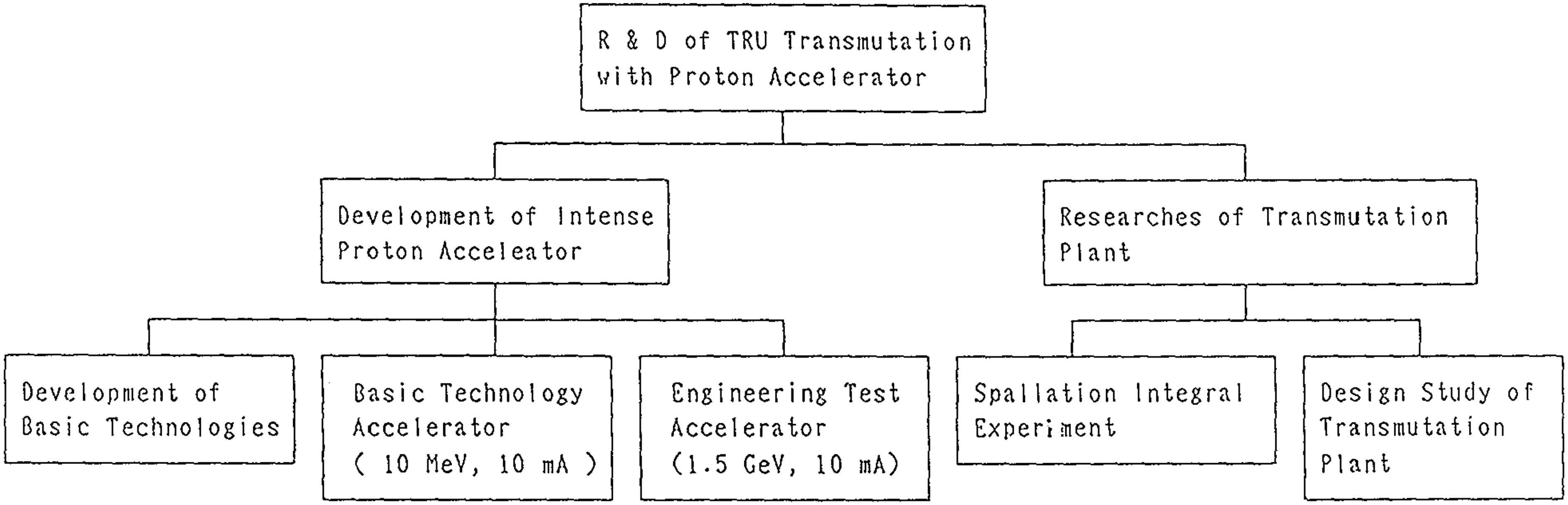


Fig. 1 Tree Structure of R & D Items for TRU Inceneration Plant Driven by High Intensity Proton Accelerator

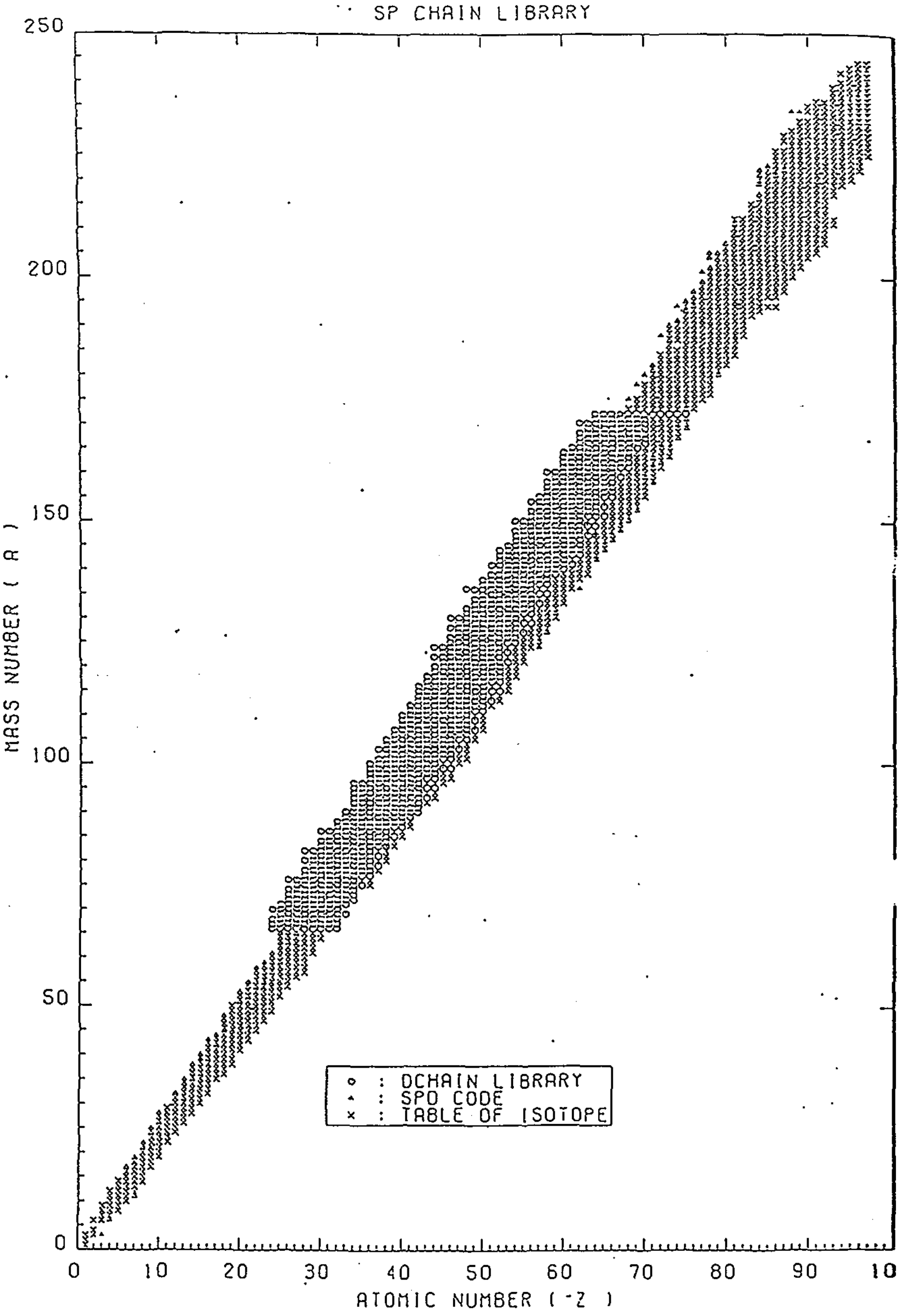


Fig. 2 Distribution of nuclides compiled in the SPCHAIN data file

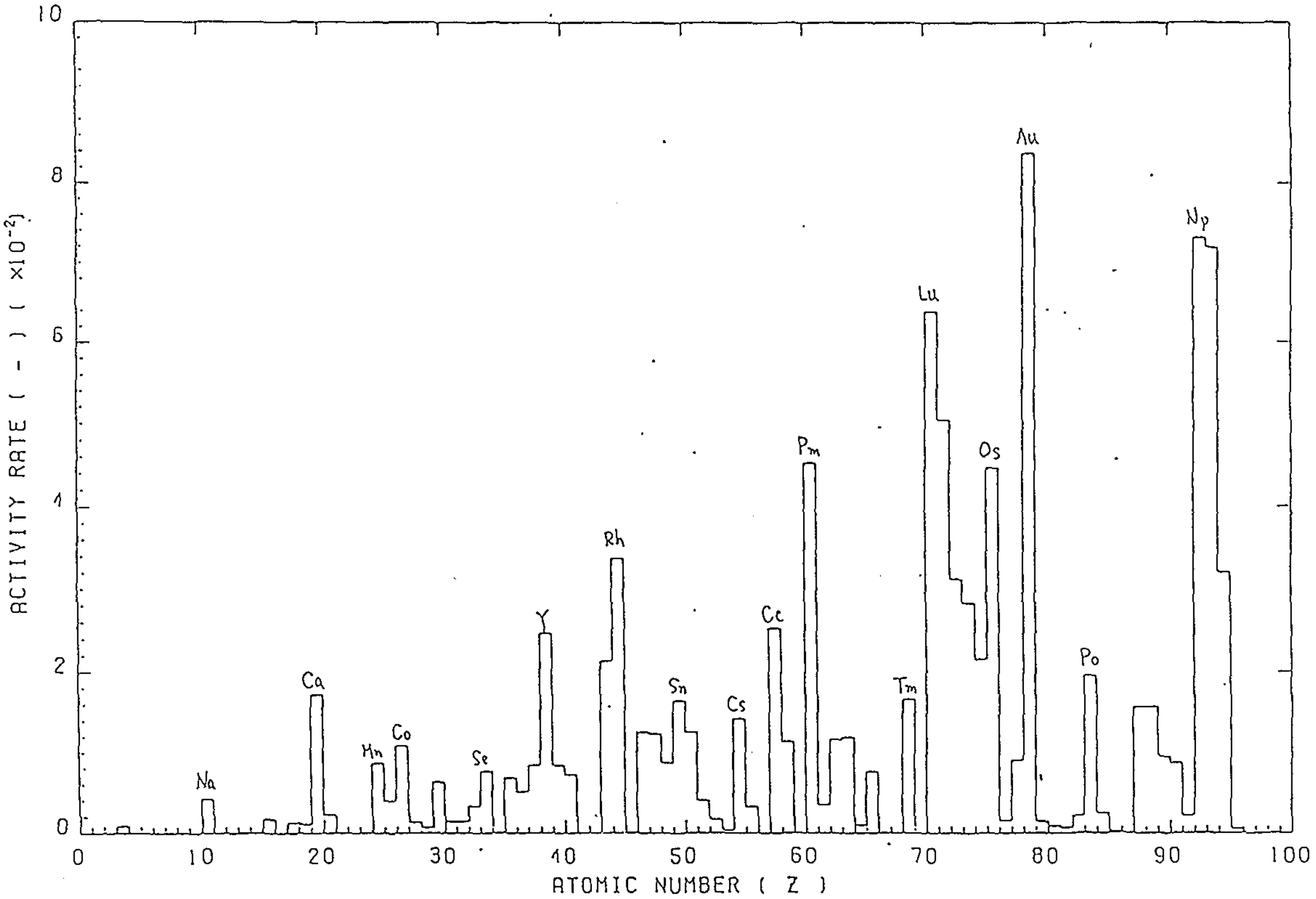


Fig. 3 Activity rate distribution of resibual elements in a ^{241}Am target at the time stage of one year cooling after irradiation of ten hours of 1[illegible]eV protons

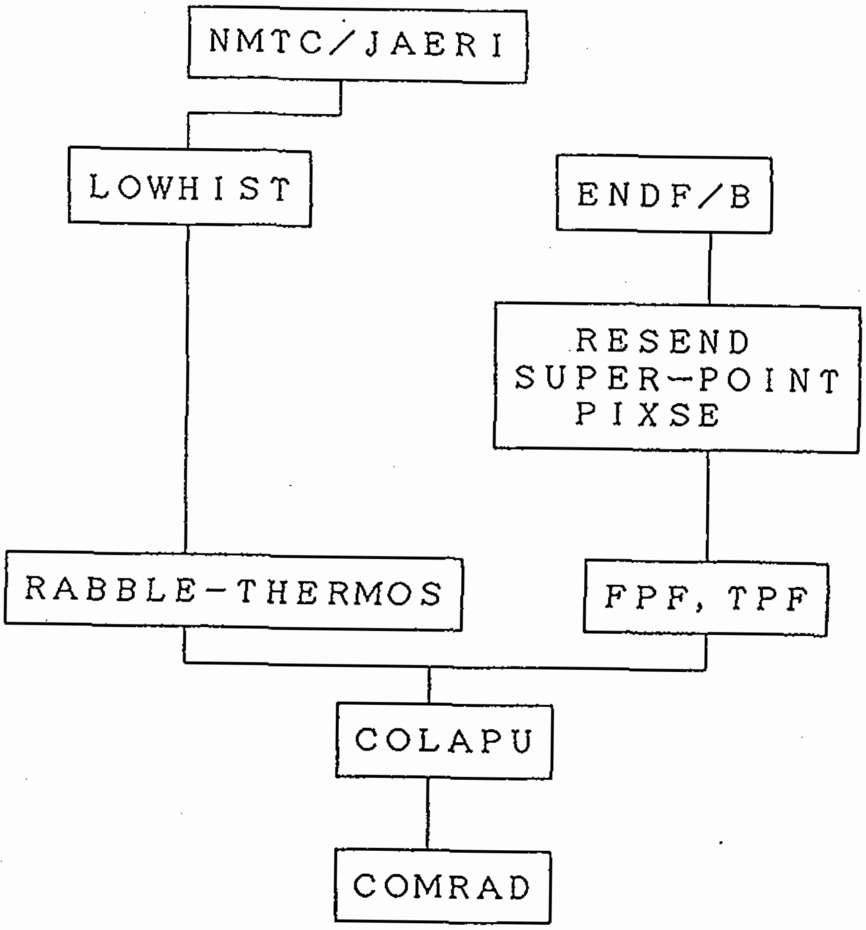


Fig. 4 SP-ACE code system for designing the transmutation core driven by a proton accelerator

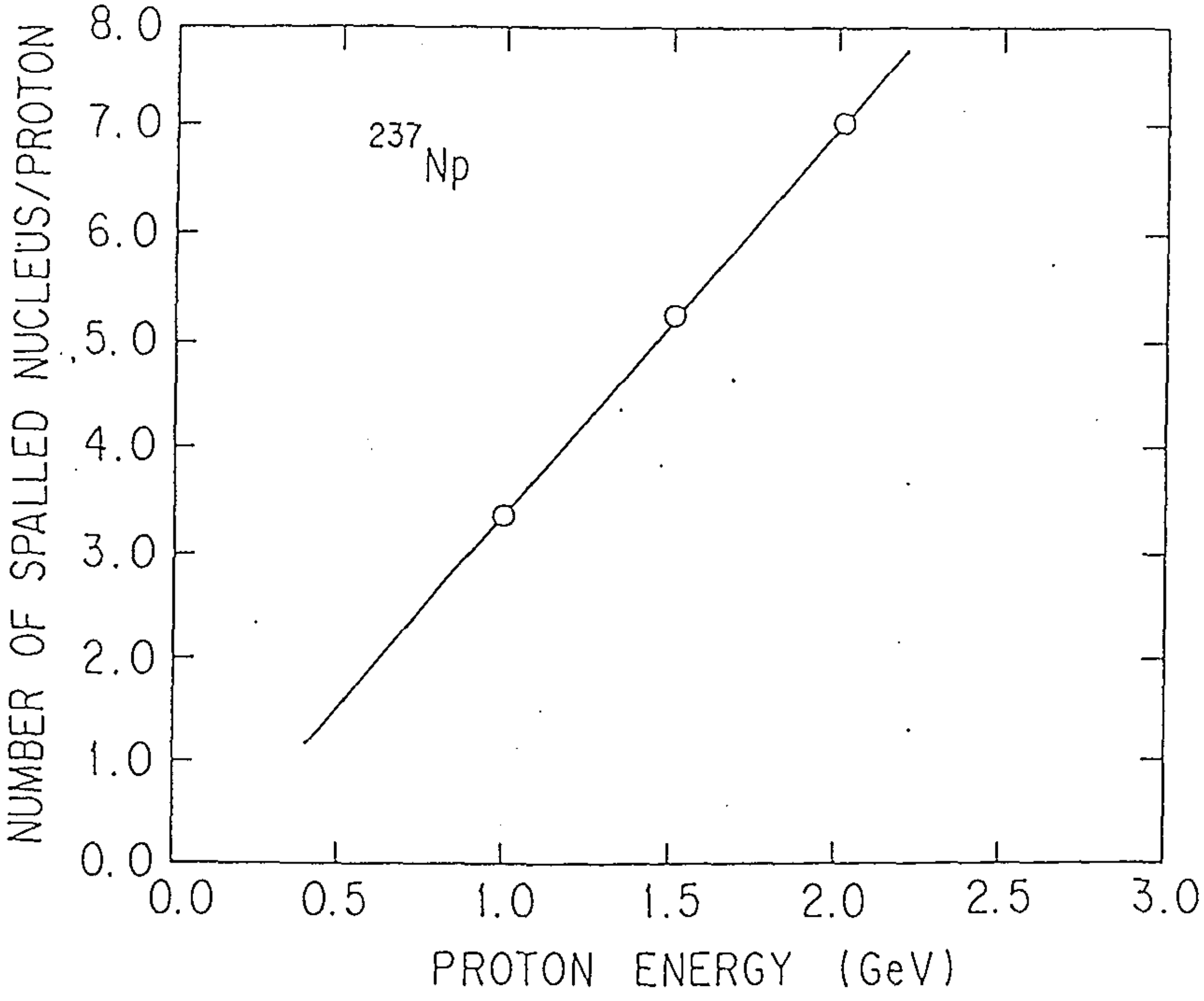


Fig. 5 Energy dependence on number of nuclei destructed due to spallation reaction

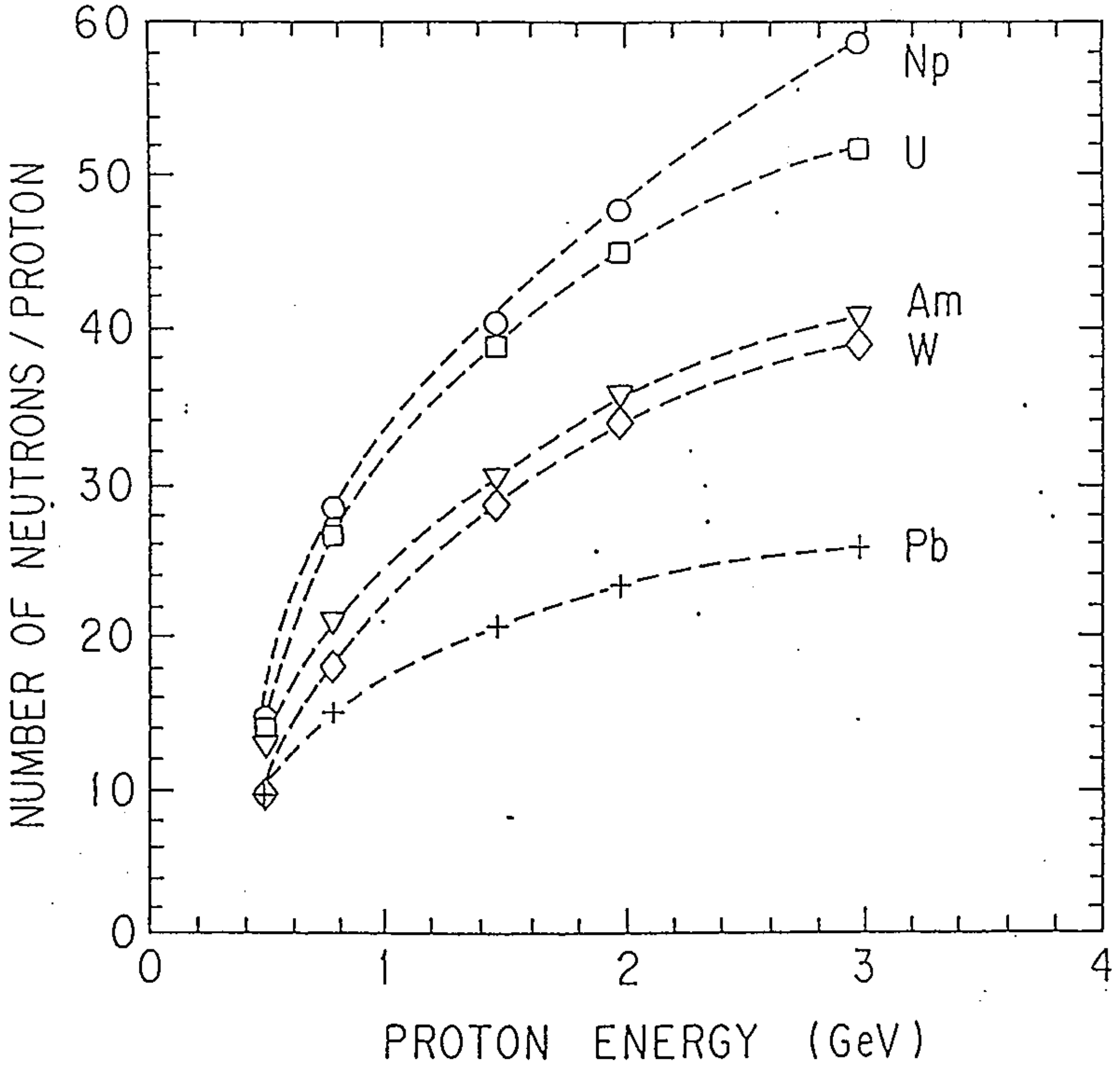


Fig. 6 Energy dependence on number of neutrons generated by spallation reaction

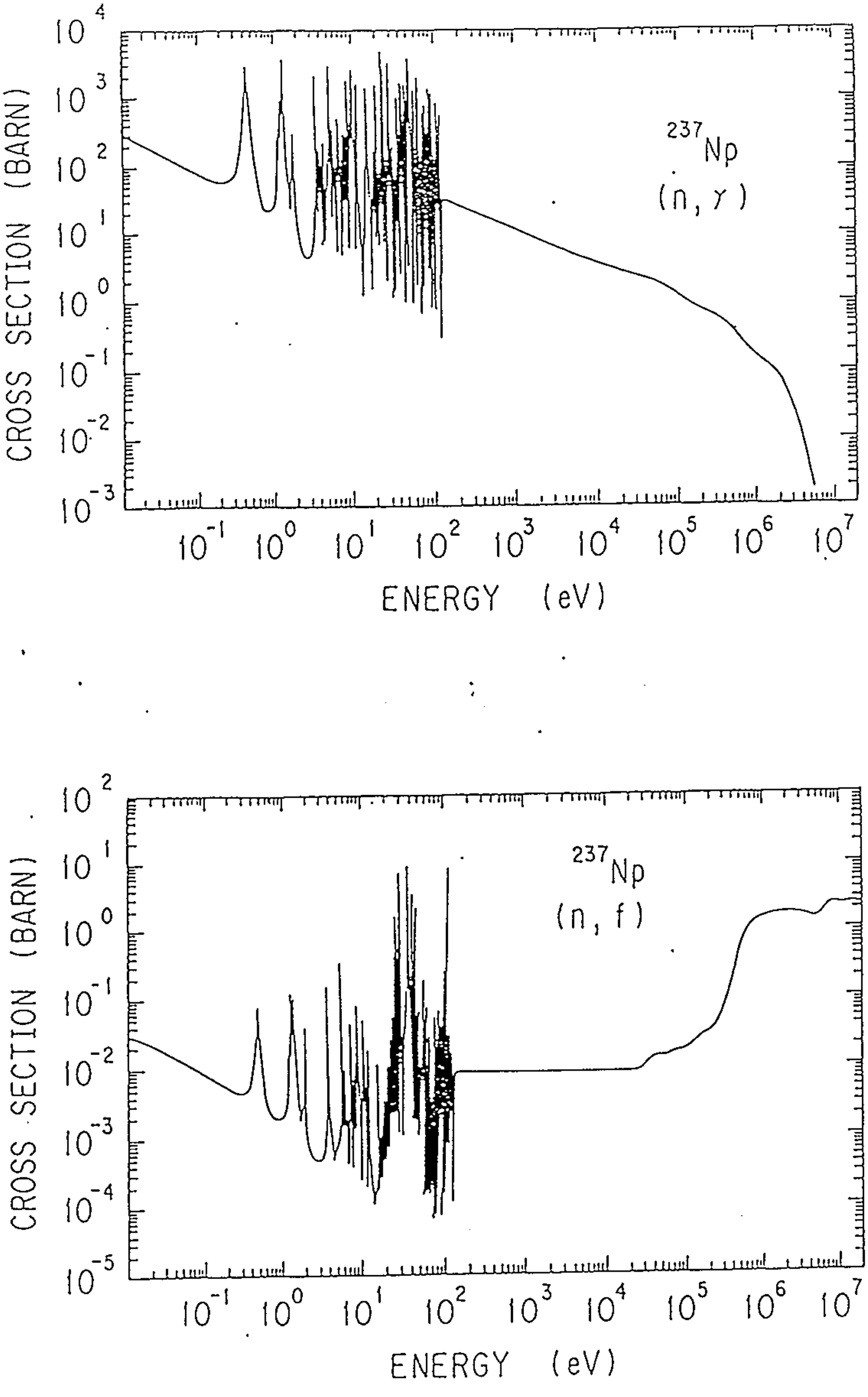


Fig. 7 Neutron cross section of ^{237}Np

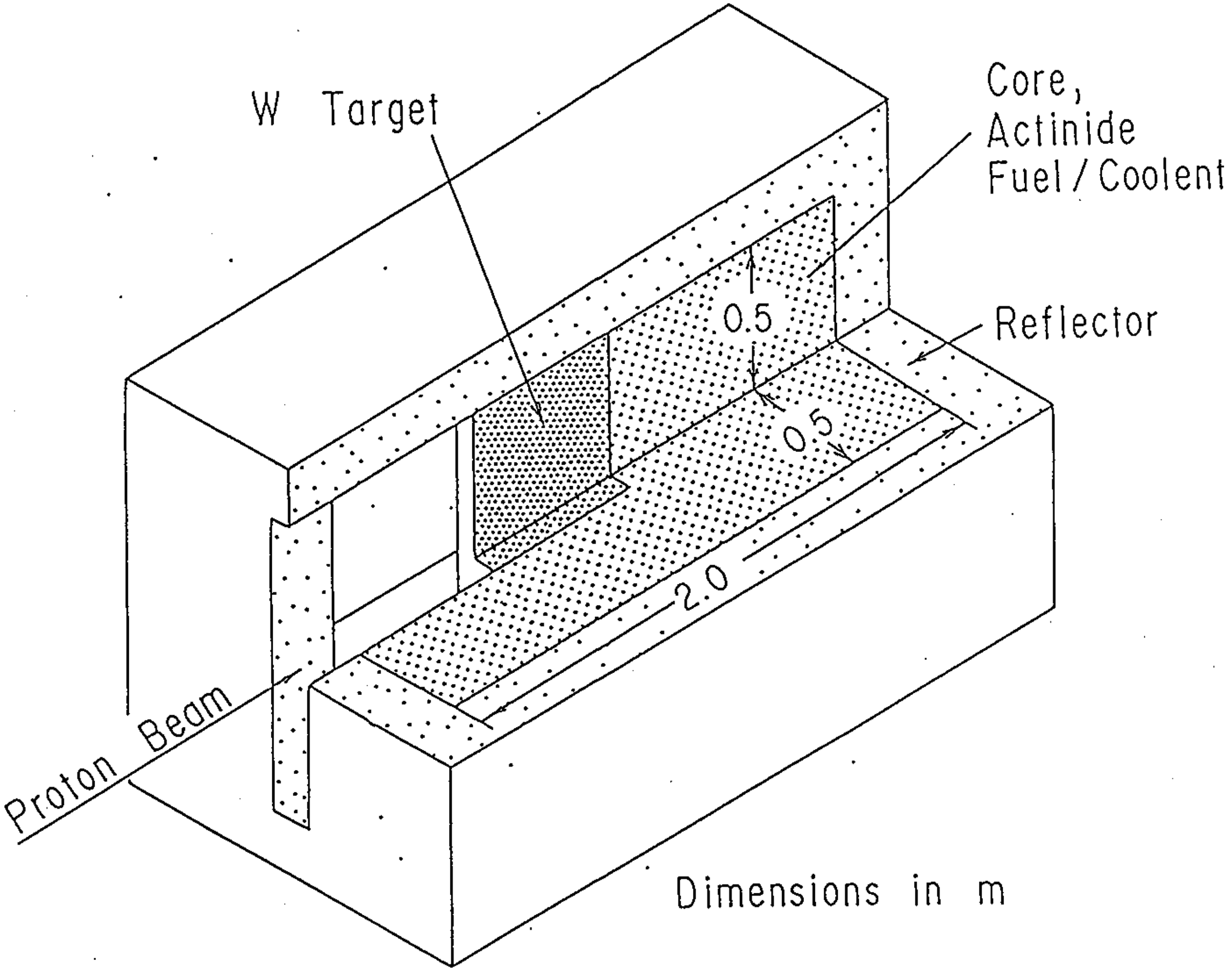


Fig. 8 Target-core configuration of hybrid plant(refernece system)

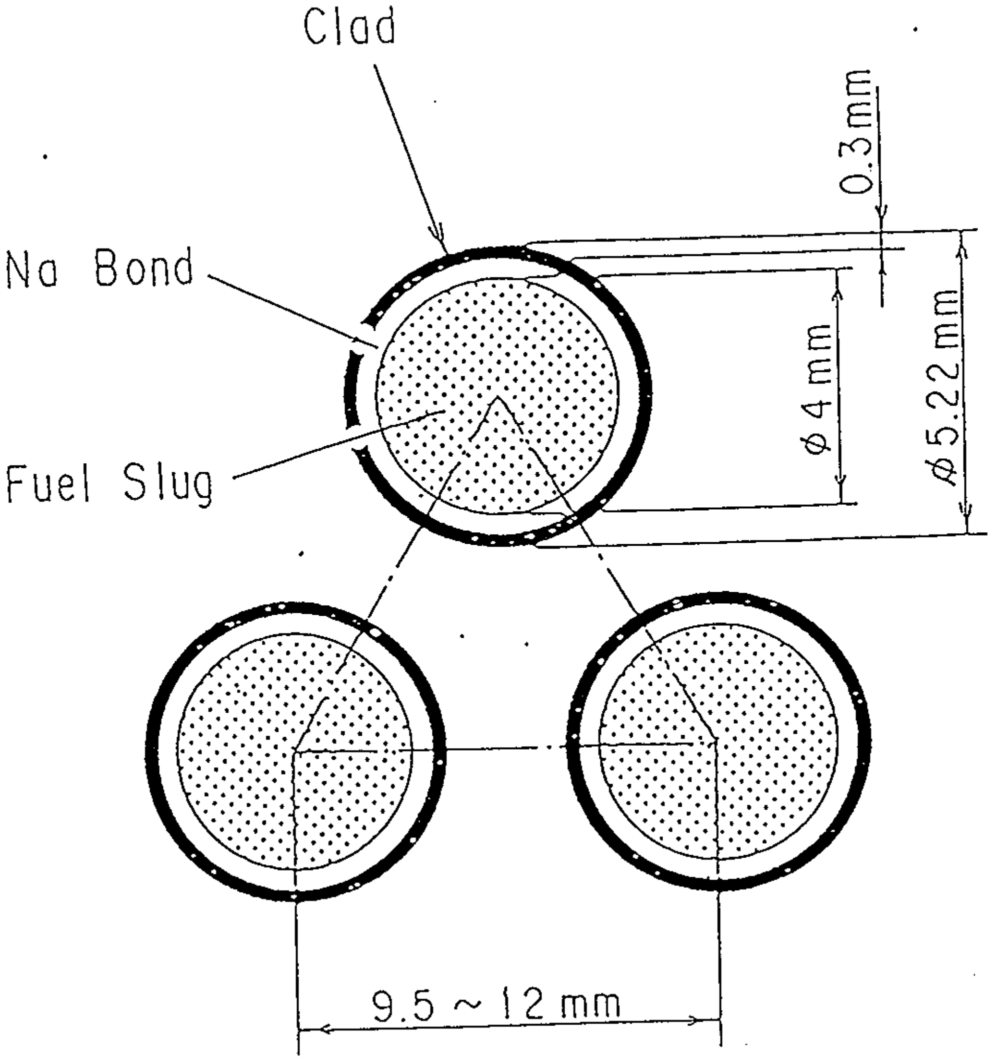
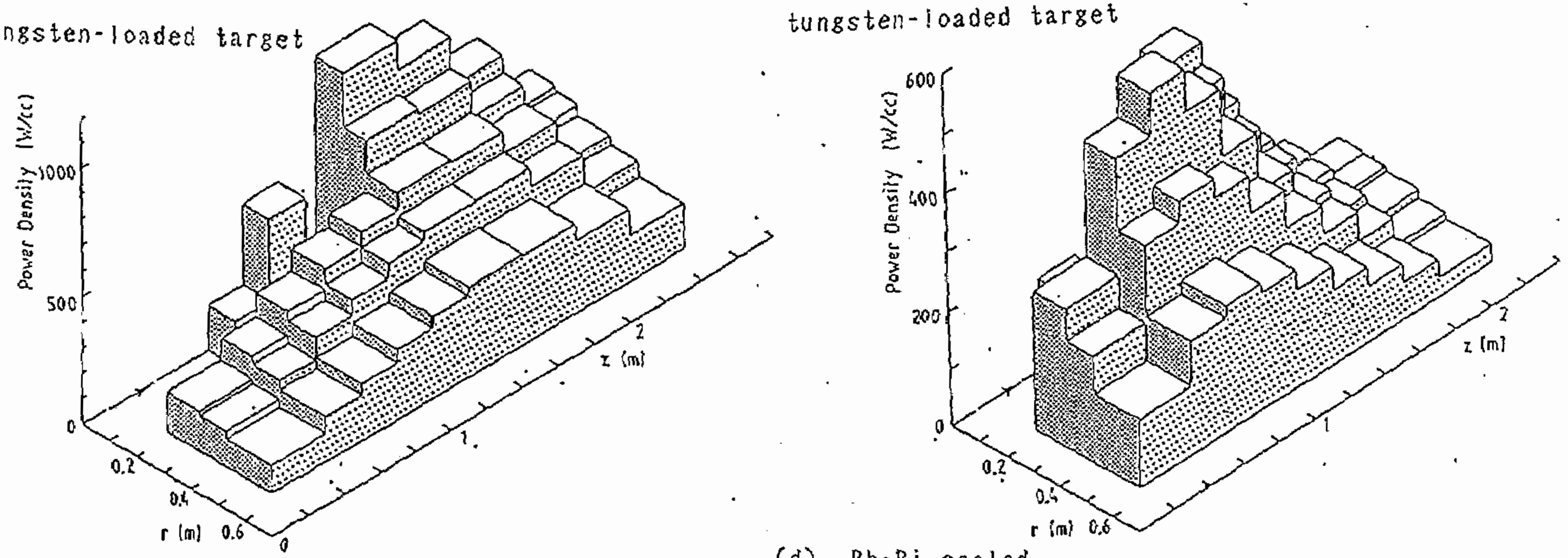


Fig. 9 Actinide fuel pin geometry

(a) Na cooled
tungsten-loaded target

(b) Pb-Bi cooled
tungsten-loaded target



(c) Na cooled
TRU alloy target

(d) Pb-Bi cooled
TRU alloy target

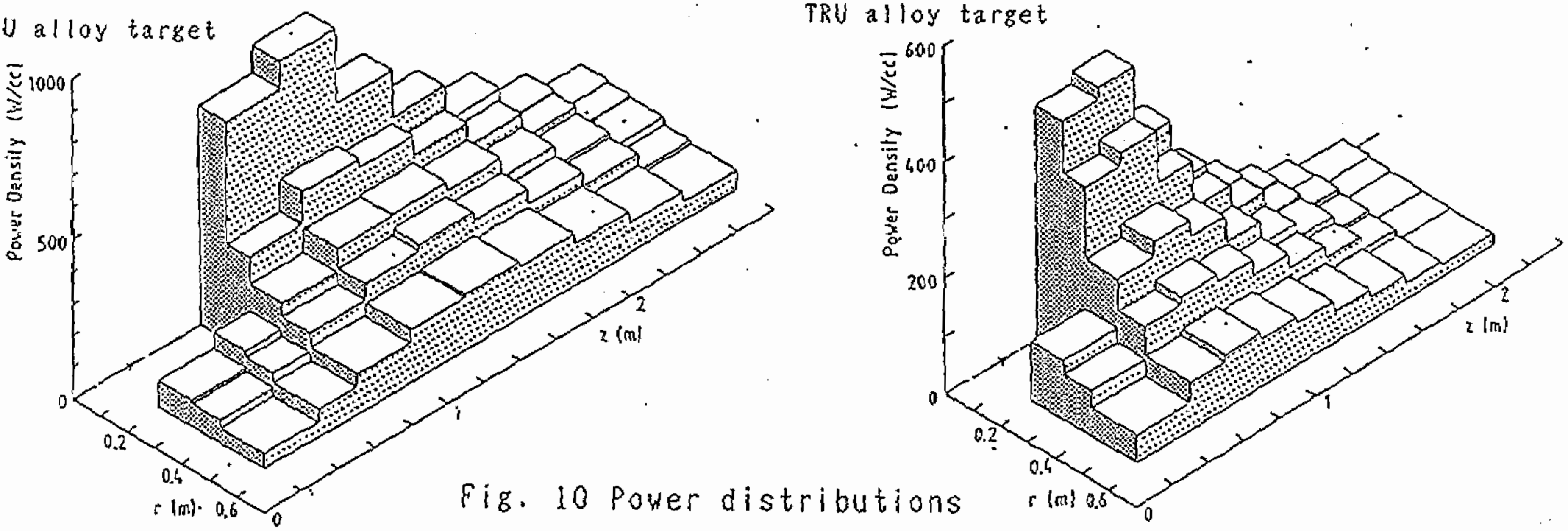


Fig. 10 Power distributions

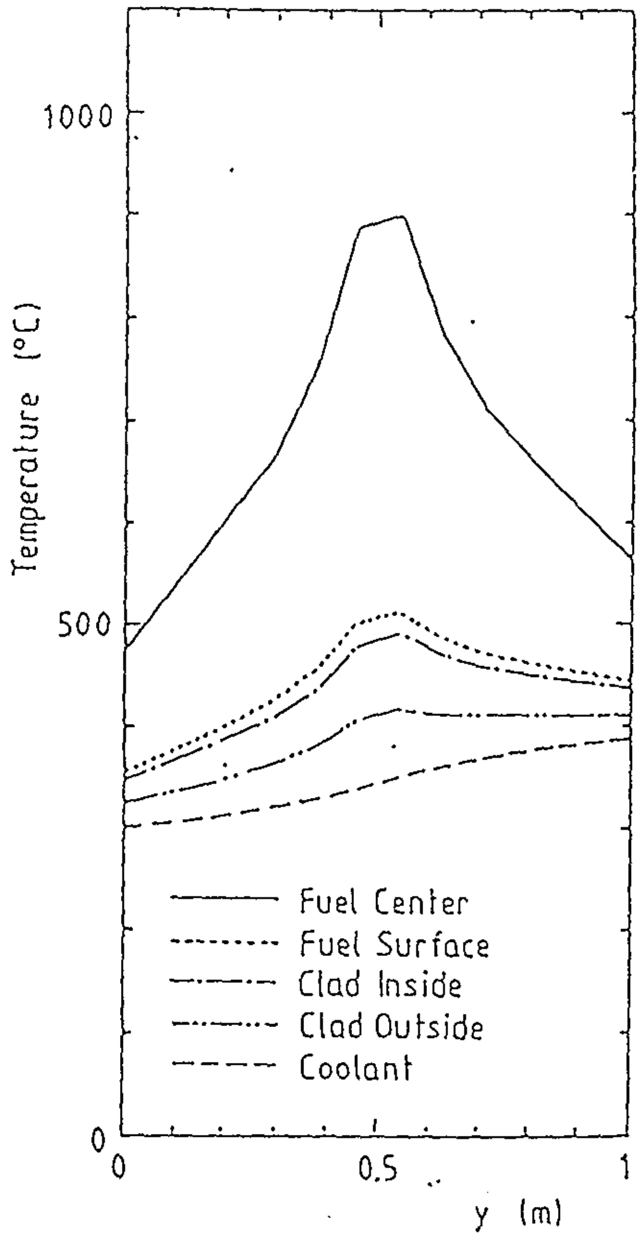
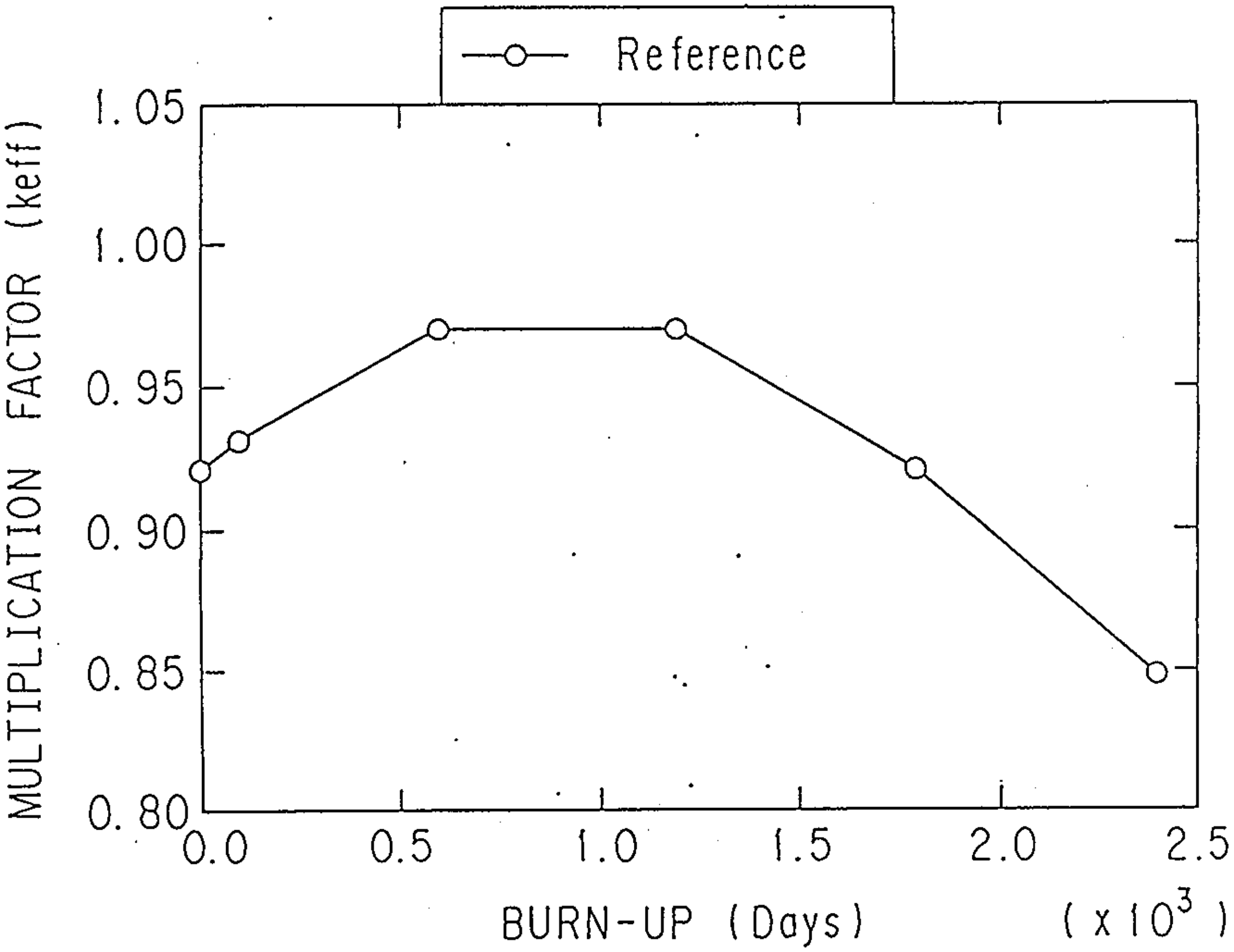


Fig. 11 Temperature distribution in the reference core



* 1,000 burnup days correspond to ~10^5 MWD/ton

Fig. 12 Change of k_{eff} with burn-up

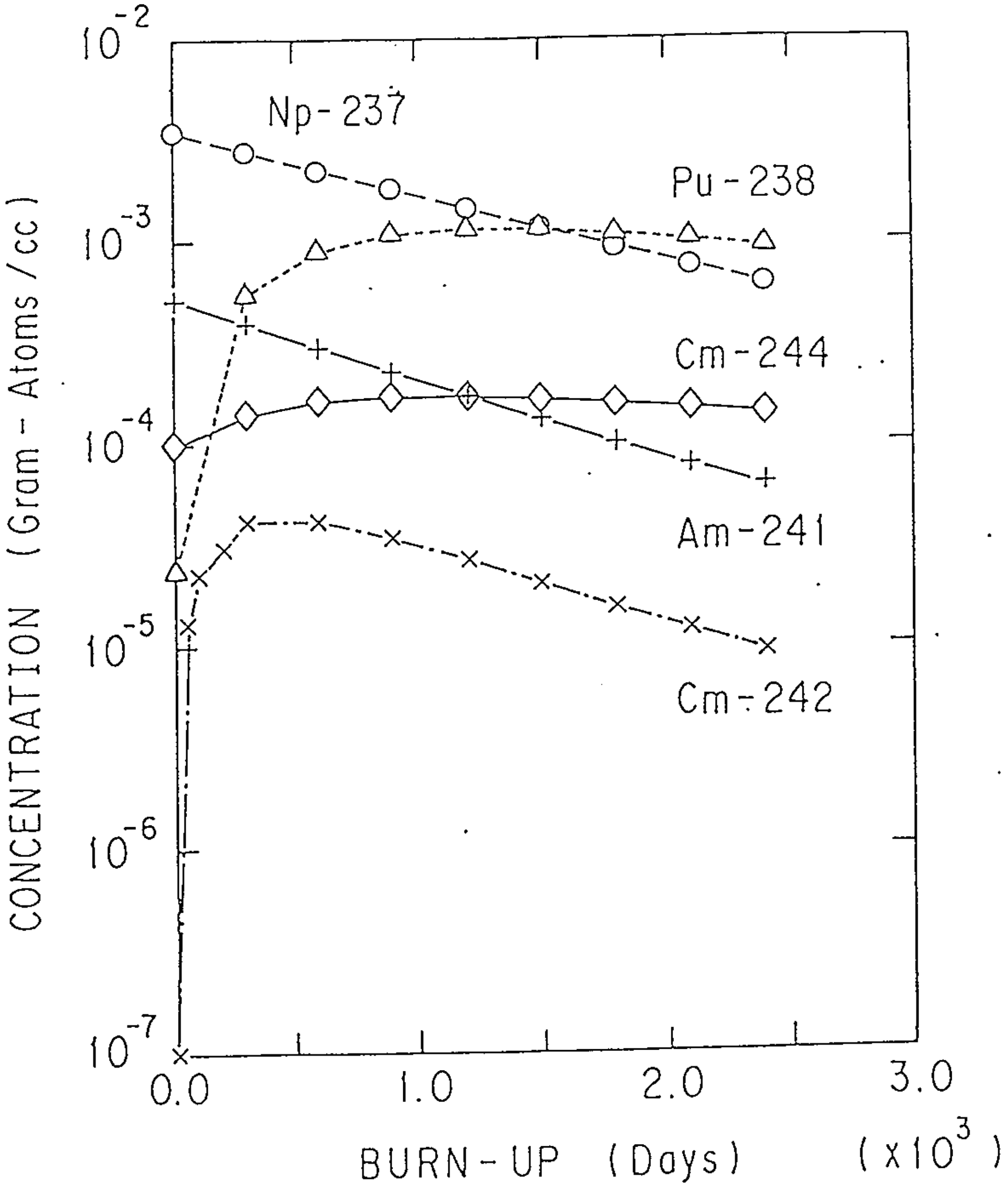


Fig. 13 Change of trans-uranium inventory with burn-up

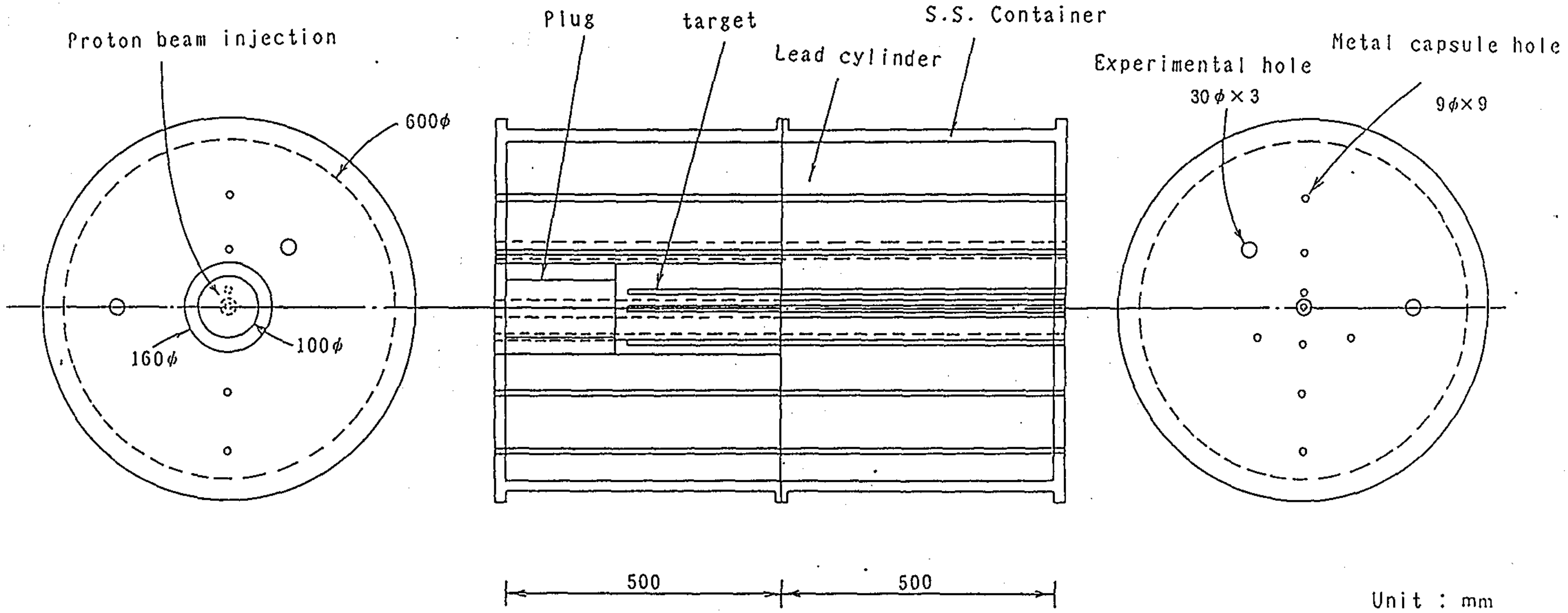
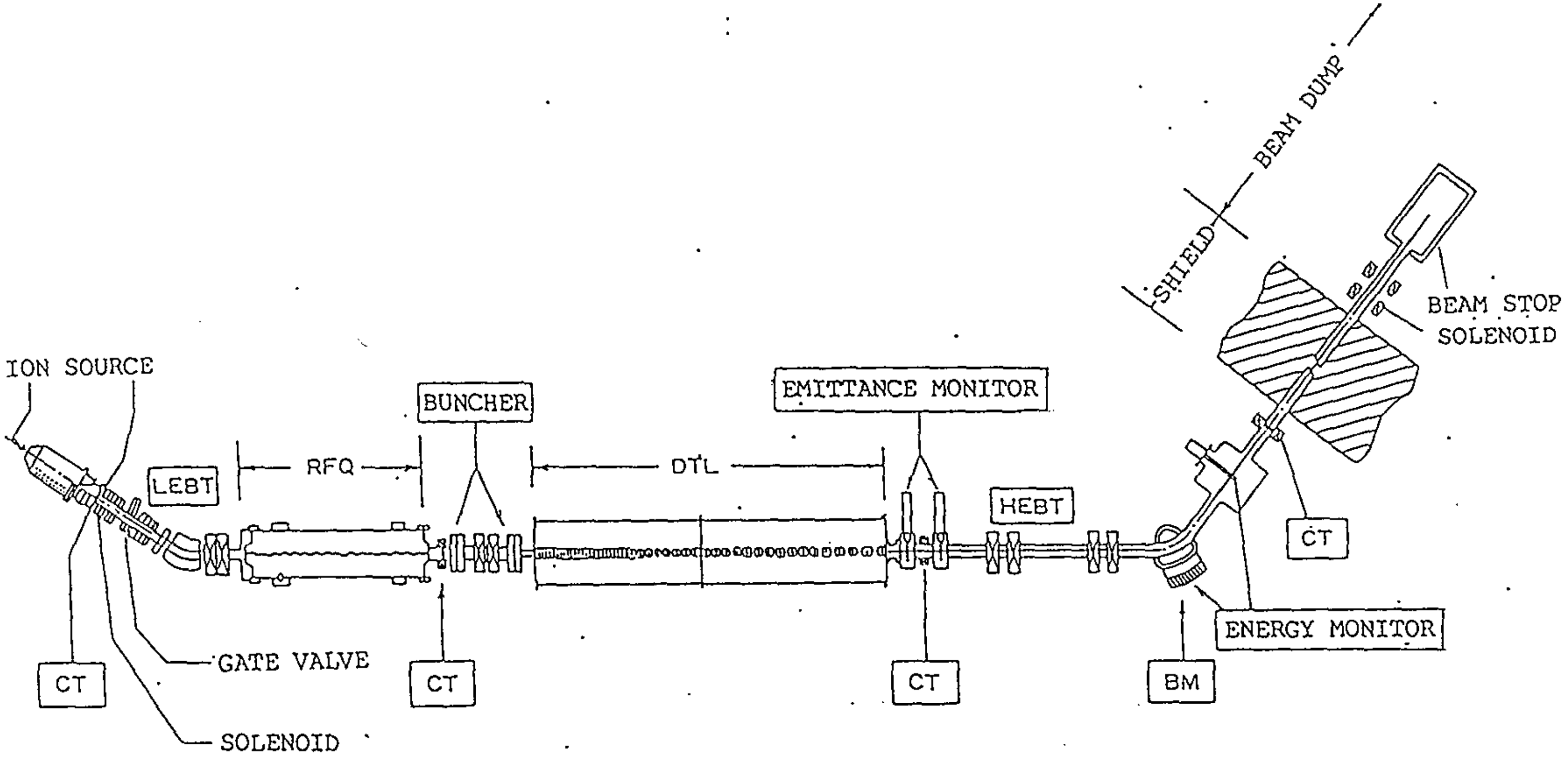


Fig. 14 Lead target-cylinder system for the spallation experiment



BM : BENDING MAGNET
CT : CURRENT TRANSFORMER
LEBT: LOW ENERGY BEAM TRANSPORT
HEBT: HIGH ENERGY BEAM TRANSPORT

Fig. 15 Configuration of Basic Technology Accelerator (10 MeV, 10 mA)