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Calculation code system for fission and spallation products

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ABSTRACT

It is important to use computer codes with high precision for analyzing the physical process in the intense neutron source induced by the high energy proton beam. A brief description is given for the spallation and fission reactions and the particle transport codes which have been developed and upgraded at JAERI.

The simulation code for High Energy Nuclear Reaction Process in the energy range of 15 MeV to 3 GeV, NMTC/JAERI, is a JAERI version of NMTC which was developed at ORNL and revised later at LANL and BNL. In this version the high energy fission reaction can be calculated, using the statistical model, in competition with the particle evaporation reaction. The mass number A of nuclides which can be dealt in the spallation calculation has been extended from [A=1;8< A≤ 239] to [A=1; 6< A < 250]. A simulation code NUCLEUS for the spallation reaction of one nucleus without taking into account the internuclear cascade, was also developed to evaluate the computational model for spallation reaction and analyze effectively the data measured in thin foil experiments. For the whole energy range less than 3 GeV, two simulation code systems had been developed by connecting NMTC/JAERI with the neutron transport codes MORSE-DD and TWOTRAN-II respectively. The code SPCHAIN for calculating the time evolution process of spallation products has been developed on the base of the depletion code DCHAIN2, which had been made for the calculations of decay and built-up of fission products in a nuclear reactor. The new nuclear data have been compiled in SPCHAIN data file for about 1100 nuclides needed for TRU spallation calculation. These codes are used to calculate reaction products measured in the spallation experiment using 500 MeV protons in the booster facility of the synchrotron at KEK and to perform the conceputual design study of TRU transmutation system driven by an intense proton accelerator.

I. INTRODUCTION

It is important to use computer codes with high precision for analyzing the physical processes in an intense neutron source induced by the high energy proton beam. In this report brief descriptions are given for the spallation reaction and the particle transport codes, which have been developed or upgraded at JAERI and some results computed by using these codes are presented. Our main purposes are to perform the design study of transuranium nuclides (TRU) transmutation target-core system driven by a proton accelerator and analyze the data measured in spallation experiments. Analyses of the beam window on the spallation target and accelerator strucutral materials irradiated by high energy particle beam will be also carried out.

II. SPALLATION ANALYSIS AND TRANSMUTATION SYSTEM DESIGNING CODE SYSTEMS DEVELOPED AND UPGRADED AT JAERI

As well known, the high energy protons injecting on a target cause more complicate nuclear reactions than neutrons transporting in a reactor core. Figure 1 shows the schematic illustration of nuclear reactions and particle transport processes in a heavy metal target irradiated by the high energy proton beam. In the energy range above ~15 MeV, the intranuclear and internuclear cascades with the spallation (particle knock-on), evaporation and high energy fission reactions are induced by the proton bombardment. Through these processes many neutrons and some light particles such as proton, deuteron, triton, helium-3, α particle are emitted. Most of these spallation neutrons, which have the energy spectrum similar to one in a fast reactor, transport in the target after slowing down into the energy range lower than ~15 MeV. As nuclear reaction simulation codes, NMTC/JAERI - NMTA and NUCLEUS codes are prepared for the energy range above 15 MeV and MORSE-DD , SP-ACE and TWOTRAN-II codes for the range below 15 MeV at JAERI. ORIGEN-2 and SPCHAIN calculate the time evolution process of TRU transmutation products in the lower and upper energy ranges, respectively. The flow chart in Fig.2 shows the mutual relations among the simulation code systems prepared at JAERI. The High Energy Nuclear Reactions and Nucleon-Meson Transport Code

NMTC/JAERI is the main code in this code system. The codes included in the left side of this figure simulate the high energy nuclear reaction process above 15 MeV and ones in the other side carry out the neutron transport calculation below 15 MeV.

(1) NMTC/JAERI[1]

The code NMTC/JAERI is used for the Monte Carlo simulations of nuclear spallation induced by incident particles(proton, neutron, pion) from an external source in a heterogeneous medium. The subsequent internuclear transport processes is also calculated in the energy range of 15 MeV to 3 GeV. In the JAERI version, the fission process has been incorporated as a competing process with particle evaporation. The range of mass number A of nuclides in the target has been extended from[A=1;8< A< 239] to [A=1; 6< A < 250]. The major part of NMTC is almost the same as the old version of HETC. The detailed descriptions about NMTC/JAERI had been given by Nakahara at the former conference ICANS-IV.[2] So in this report only two graphs computed using NMTC/JAERI for the analysis of transmutation system are presented.

Figure 3 shows the dependence of the number of spallated nuclides on the incident proton energy when the ^{237}Np target(20 cm in diameter, 60 cm in length) is bombarded by protons. The number of nuclides transmutated by one proton with 1.5 GeV in energy is about 5 but it is too small to manage TRU wastes in the commercial base. It must be noted that several tens of neutrons of hard energy spectrum are emitted in the spallation reaction. The computer simulation results show 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 seen in Fig. 4. For the case of ^{237}Np target(20 cm in diameter, 37 cm in length) bombarded by a 1.5 GeV proton the neutron number is $\sim$40. The application of these neutrons to the transmutation system is described later.

(2) NMTA/JAERI[3]

In the improved version of NMTA, which is named to the statistical routines analyzing the Monte Carlo events in the NMTC/JAERI computation, the new subroutine HEATDP has been installed. HEATDP was developed at JAERI to calculate the energy deposition and its spatial distribution for each component such as ionization loss energy and recoiling energy of fission and spallation products without γ -ray heating of excited residual nuclei in the high energy range. Figure 5 represents the dependece of total heat deposition in a metal target (20 cm in diameter, 60 cm in length) as a function of the injected proton enenrgy. $H_U^T$and H_U^I denote total heat deposition and ionization loss energy in a natural uranium target, respectively and H_{Pb}^T is total heat deposition in a lead target. The difference between H_U^T and H_U^I corresponds to the deposition of heat generated from the high energy fisson. The fraction of proton energy lost through the inonization decreases when the incident proton energy increases. The ionization loss becomes $\sim$ 50 % of total heat deposition at 900 MeV. Figure 6 (a) and (b) shows the spatial distributions due to ionization loss energy and recoiling energy of SPs and FPs, respectively, for the 900 MeV proton injecting on the uranium target with 20 cm diameter and 36 cm length.

(3)NUCLEUS[4),5),6)]

The code NUCLEUS has been also developed at JAERI by modifying and combining the Monte Carlo codes NMTC/JAERI and a routine in NMTA/JAERI. The NUCLEUS can simulate the nuclear spallation reaction between a single target nucleus and a projectile without the internuclear nucleon transport process, in order to make direct evaluations of physical and computational models efficiently. The results computed by this code can also be compared directly with the data of thin foil spallation experiment, in which the internuclear multiple scattering have little effects. In NUCLEUS the counting region of reaction products was extended to avoid the count loss due to the dimension limitation in the computer program. As for a mass formula the Uno & Yamada' mass formula[7)] has been optionally adopted to upgrade the particle evaporation calculation. New several plotter routines were also provided for rapid processing of a huge amount of output data. Figure 7 compares the original calculational data with the upgraded ones for mass number distributions of reaction products with Z from 92 to 82 from a uranium nucleus spallated by a 1 GeV proton. In the central case with the extended counting region, the counting loss is recovered and the left tail of peaks appears in the reasonable form in comparison to the left figure for the original region. By the use of Uno & Yamada's mass formula, the corrected peaks in the right figure have become wider than the central case.

On the other hand the NUCLEUS computation presents the important, basic data for researching the feasibility of TRU transmutation induced by the spallation process. In Fig. 8 the bird eye'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, namely, the hill of spallation products, the spire of evaporated particles and the valley of high energy fission product between them. It is apparent that the yield of SP is greater than FP by two orders. Table 1 summarizes the number of produced light particle such as proton, neutron, deutron, triton, helium 3 and alpha. They are emitted from a uranium target nucleus bombarded by protons with the energy of 380 MeV to 2.9 GeV. The number of emitted particles has the maximum value around 2 GeV, where the neutrons more than 17 are emitted from one uranium nucleus. The result suggests that the incident proton energy has the optimum value around 2 GeV for neutron production. Figure 9 shows the histogram of half life distributions of products when a 2 GeV proton bombarding on ^{237}Np and ^{241}Am target nuclei. The shaded portions in the half life classifications of 7 and 9 represent tritons 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 of SP in this case have the halflife shorter than one year.

(4) SPCHAIN, SPD

The composition and amount of residual nuclides accumulated in the target spallated by the proton beam is very improtant for the feasibility study 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. A code for calculating approximately the process of spallation products, SPCHAIN, has been made on the base of the depletion code DCHAIN2,[8] which was developed for the calculations of decay and build up of fission products in a nuclear reactor. We have modified the one point depletion equation to include the transmutation, decay and build up processes of TRU and spallation products(SP) in the following,

$$\begin{aligned} dX_i(t)/dt = & \Sigma l_{i,j}\ \lambda_j X_j(t) + \Sigma k_{i,j}\ \Phi \sigma_{c,j} X_j(t) \\ & - (\lambda_i + \Phi \sigma_{c,i}) X_i(t) + \gamma_i F(t) \\ & + \Sigma \alpha_{i,j}\ \Phi_P \sigma_{s,j} X_j(t) - \Phi_P \sigma_{s,i} X_i(t), \end{aligned}$$

where the last two terms due to the spallation have newly been added to four original terms used for the reactor burnup calculation. α, σ_s and Φ_P represent the production rate of SP, the spallation cross section and high energy particle flux, respectively. The equation can be solved analytically by the Bateman method. As shown in Figure 10, some necessary data are feeded from the surrounding codes. When the halflife time of a nuclide was not obtained in the prepared data, it was calculated by using the β decay calculation program SPD[9] or guessed from the trend of data of nuclides, which are located in the neighbor of the nuclide in the Nuclear Chart. The production rate α of SP was computed by using the code NUCLEUS. The spallation cross section has been interpolated or extrapolated from the measured data[10] given for proton energy and target nucleus. 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.[11] The the preliminary result in SPCHAIN calculations is shown. Figure 11 shows the activity rate distribution of residual elements in a ^{241}Am target after one year cooling after 10 hours irradiation of 1 GeV protons with 10 mA. The high activity peaks around Z = 80 just after irradiation disappear due to the short halflives of SP and the activities buildup to Au, Os and Lu isotopes, whereas the higher actinide activity such as Np and Pu increases. So they should be transmuted through the continued spallation process. Activities due to fission products are a few.

(6) Other codes (ACCEL[12], SP-ACE)

A simulation code system ACCEL had been developed by connecting NMTC/JAERI with the neutron transport Sn code TWOTRAN-II through the neutron source file, accompanying the nuclear data files. This system can calculate all the reaction processes occured in the target for the whole energy range less than 3 GeV as one computing job as shown in Fig. 12.

The SP-ACE code system is being developed for designing the transmutation core system driven by proton accelerator in the energy range below 15 MeV in reasonable computing time and precision. The neutron transport code RABBLE-THERMOS computes the region-wise neutron flux, using ultra fine energy group constants and the distribution of spallation neutron source, which can be obtained from Monte Carlo calculation in NMTC/JAERI. These fluxes and nuclear data are utilized to calculate the yield of products, heat generation and gamma ray intensity in each region by the burnup code COMRAD.

Ⅲ. CONCEPUTUAL DESIGN STUDY OF ACCELERATOR TRANSMUTATION SYSTEM

We have been promoting the conceptual studies on the TRU transmutation in the target-core system driven by an accelerated proton beam.[13),14),15)] For the reaction below 15MeV the Monte Carlo neutron transport code MORSE-DD [16)] was used with 52 neutron group constants edited from JENDL-2 and ENDF-B4, where spallation neutrons calculated by NMTC/JAERI code were treated as the source.

The basic conditions for the system design are (1) transmutation of 260 kg of TRU in a year, which is produced from ten commercial reactors of 1GWe and (2) good energy balance, in which it can generate enough electricity to operate the accelerator at least. Here total amount of TRU produced per year from 1 GWe PWR is about 26 kg/y, 56 % of which is ^{237}Np. Figure 13 is the target-core configuration of hybrid transmutation system driven by high power proton beam with the energy of 1.5 GeV and the current of several tens of mA. The tungsten target is installed in an TRU-fueled subcritical core surrounded by the HT-9 steel container. The system has dimensions given in the figure. A beam window is located at a depth of 0.7 m from the front face. The heat generated in the TRU fuel is removed by the forced circulation of liquid metal coolants Na/Pb-Bi. The core consists of metallic alloy fuel of TRU and provides considerably harder neutron spectrum than the other types of fuels. The fuel pin cell geometry is shown in Fig.14 with a diameter of 4 mm cladded with HT-9 steel.

Profiles of the two-dimensional power distribution for four cases of the system cooled by Na and Pb-Bi, with and without the tungsten target, were calculated as shown in Figures 15 (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. The flattened power distribution allows higher beam current and increases the average number of transmuted nuclides. The system performance for these cases was summarized in Table 2. In the case of Na cooling and the tungsten target the maximum thermal power is 691 MW and 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 W/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 are considerably lower than those of Na cooled one, while the beam current required is less than 8 mA. The maximum transmutation rate, 202 kg/year can be achieved for the core with the target cooled by Na. The changes of concentrations of some minor actinides with burn-up days in the reference system(Na cooling, with tungsten target) have been also calculated, as shown in Fig.16, 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 build up, which are not contained in the initial loading.

Ⅳ. SUMMARY

Finaly we summarize the outline of the present report as following items,

(1) Developments of NUCLEUS and SPCHAIN codes and upgrade of simulatin codes,

(2) Analysis of the spallation reaction & cascade process in the high energy range for TRU transmutation and

(3) Conceputual design study of hybrid transmutation system (

TRU alloy fuelled core type) driven by an intense proton accelerator. In the present design concept the amount of TRU produced from about 8 units of 1GWe PWR can be transmuted for the case of the core with the tungusten target cooled by Na and with the proton beam of energy 1.5 GeV and current 23 mA.

Furthermore we have the research items in the near future as

(a) Evaluation and upgrades of simulation codes based on new evaluated data and data measured in the spallation experiments which are being carried at KEK,

(b) Conceputual design study of hybrid transmutation system (TRU molten salt fuelled core type) and

(c) Analyses of irradiation tests for accelerator structural materials and the beam window in the target-core.

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Table 1 Number of light particles emitted from a uranium target nucleus bombarded by protons with the energies of 380 MeV to 2.9 GeV

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	17.319	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	0.3079	0.2588

Table 2 Performance of the TRU transmutation system

Core system	with W target	with W target	without W target	without W target
Coolant	Na	Pb-Bi	Na	Pb-Bi
k-effective	0.92	0.86	0.94	0.95
Beam current(mA)	22.6	7.5	18.4	5.4
Thermal power(MW)	691	484	405	163
Power density(W/cc)max	889	523	776	425
ave	307	246	159	83
Burnup rate of TRU (kg / year)	202	139	114	42
Unit of 3 GWt LWR	7.6	5.3	4.3	1.8

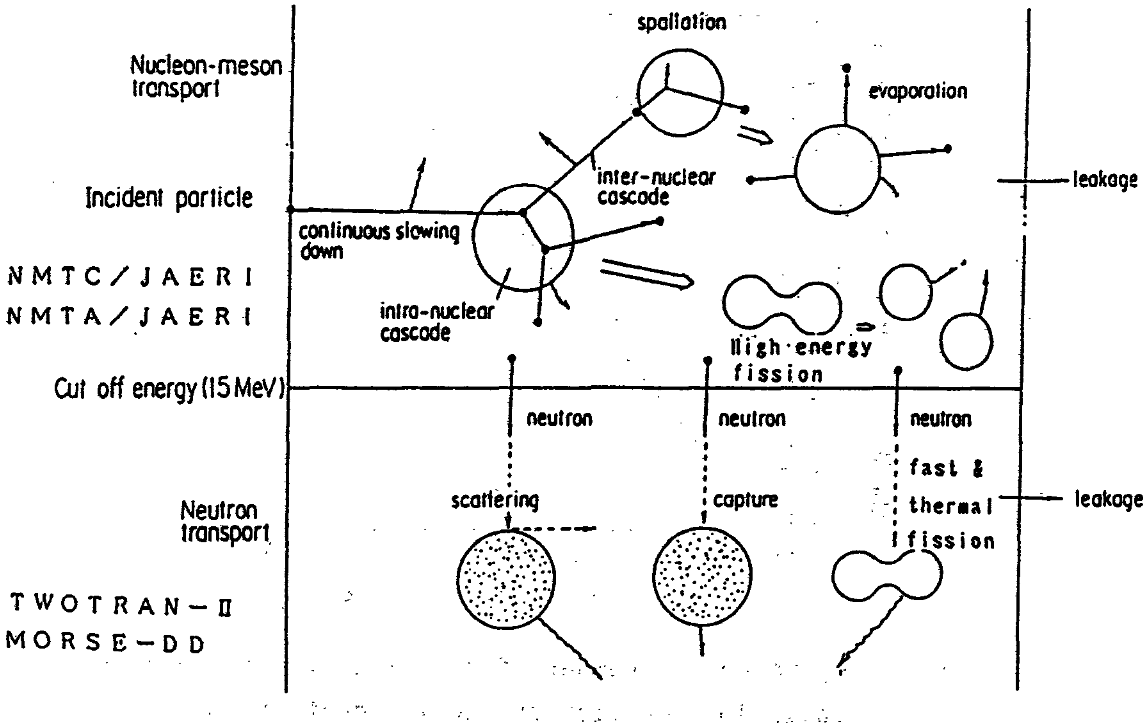


Fig. 1 Schematic illustration of nuclear reaction and nucleon transport process in a heavy metal target irradiated by the high energy proton beam.

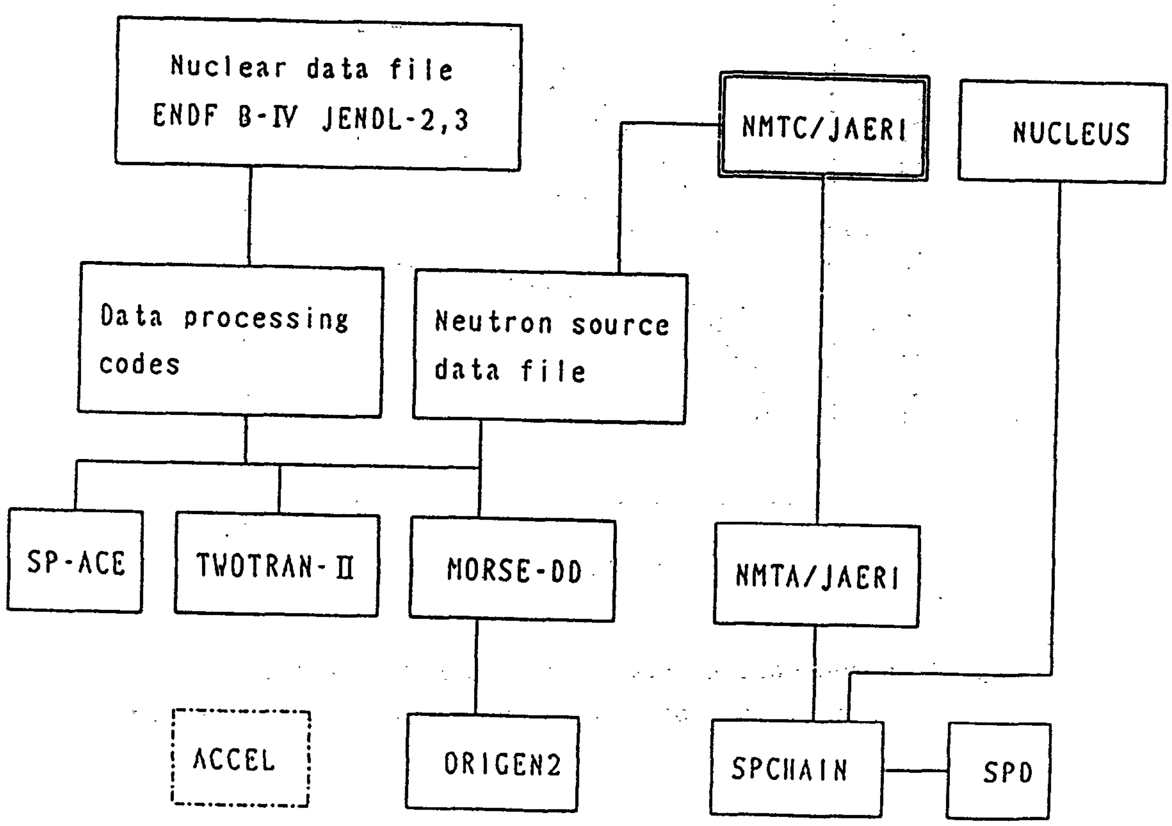


Fig. 2 JAERI code system for the spallation transmutation

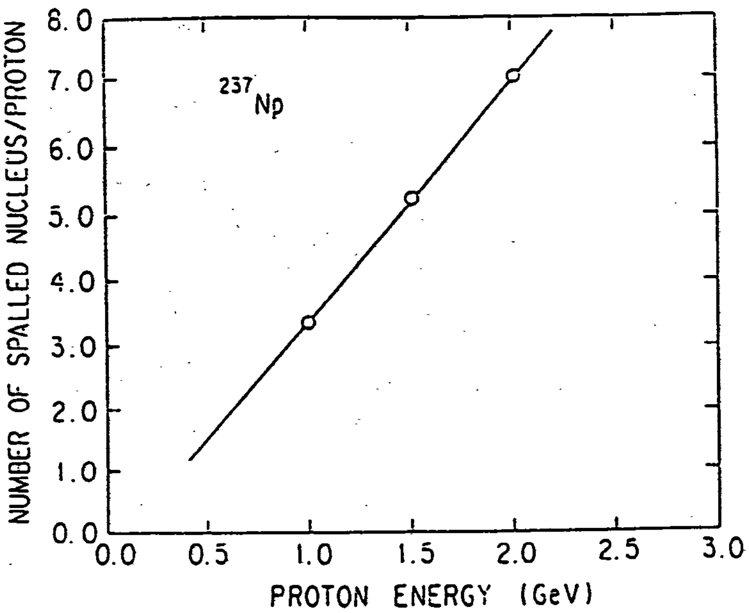


Fig. 3 Dependence of number of nuclei destructed due to spallation reaction on the incident proton energy

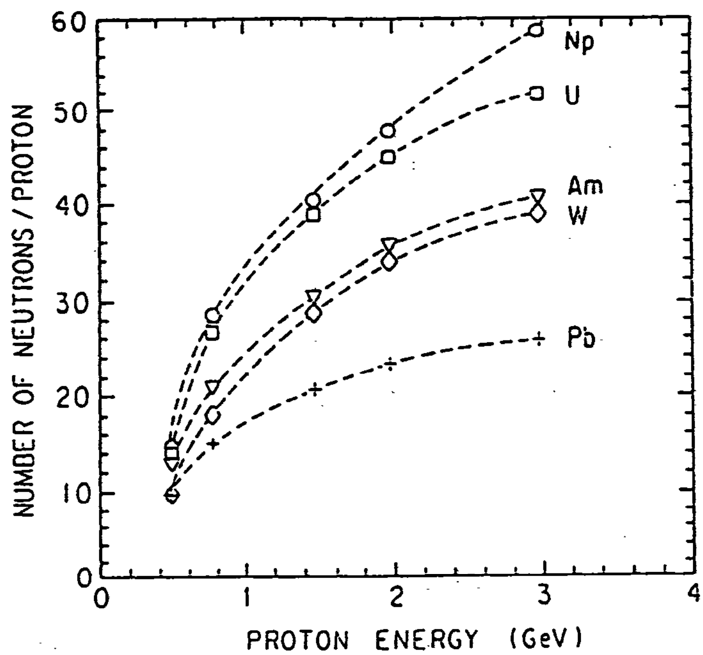


Fig. 4 Dependence of number of neutron generated in spallation reaction on the incident proton energy

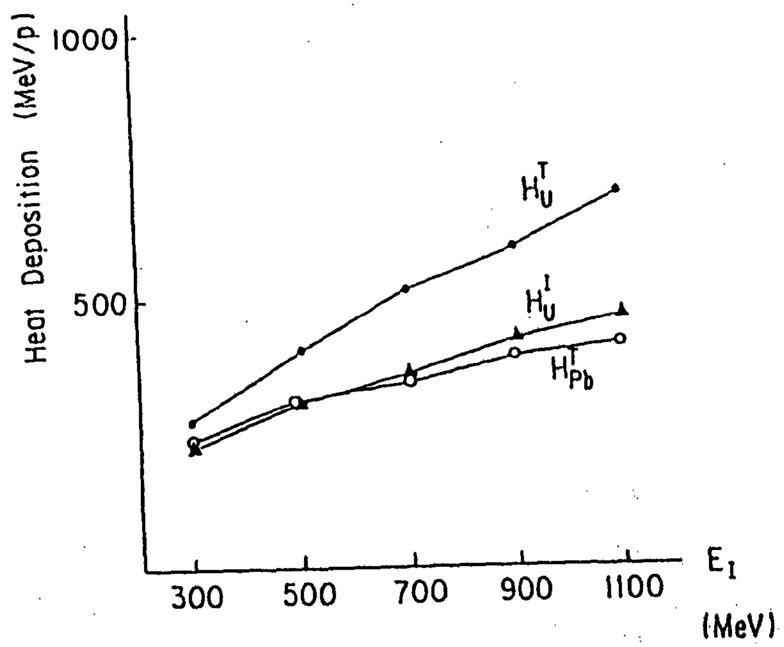


Fig. 5 Variation of total heat deposition as a function of the injected proton enenrgy

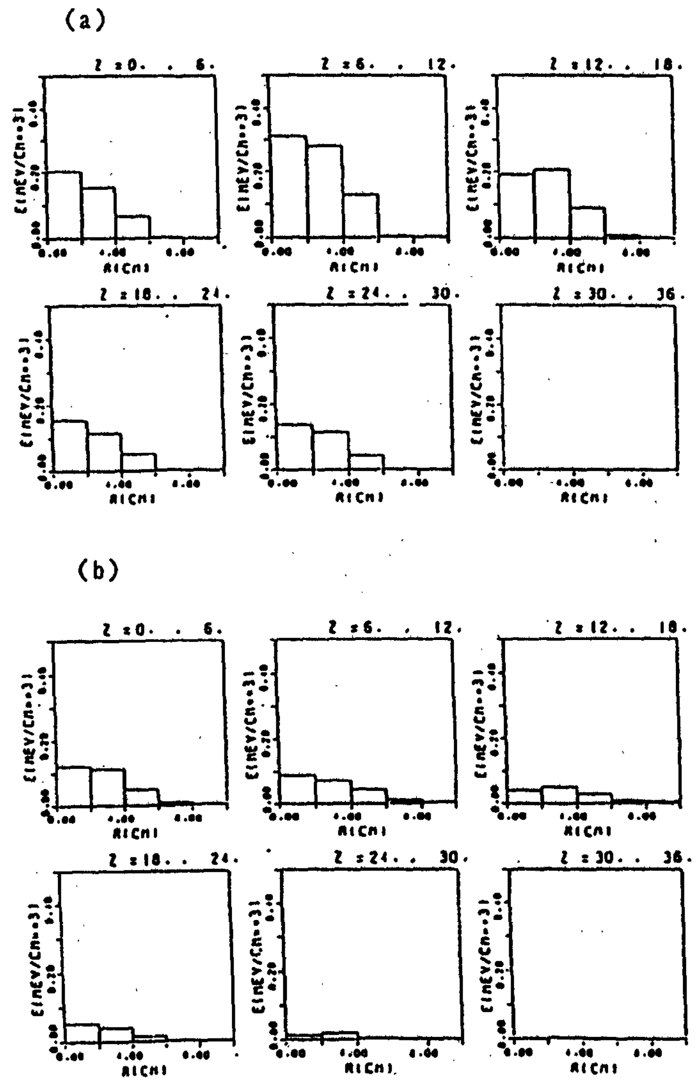


Fig. 6 Spatial distributions due to (a) ionization loss energy and (b) recoiling energy of SPs and FPs for the 900 MeV proton injecting on the uranium target

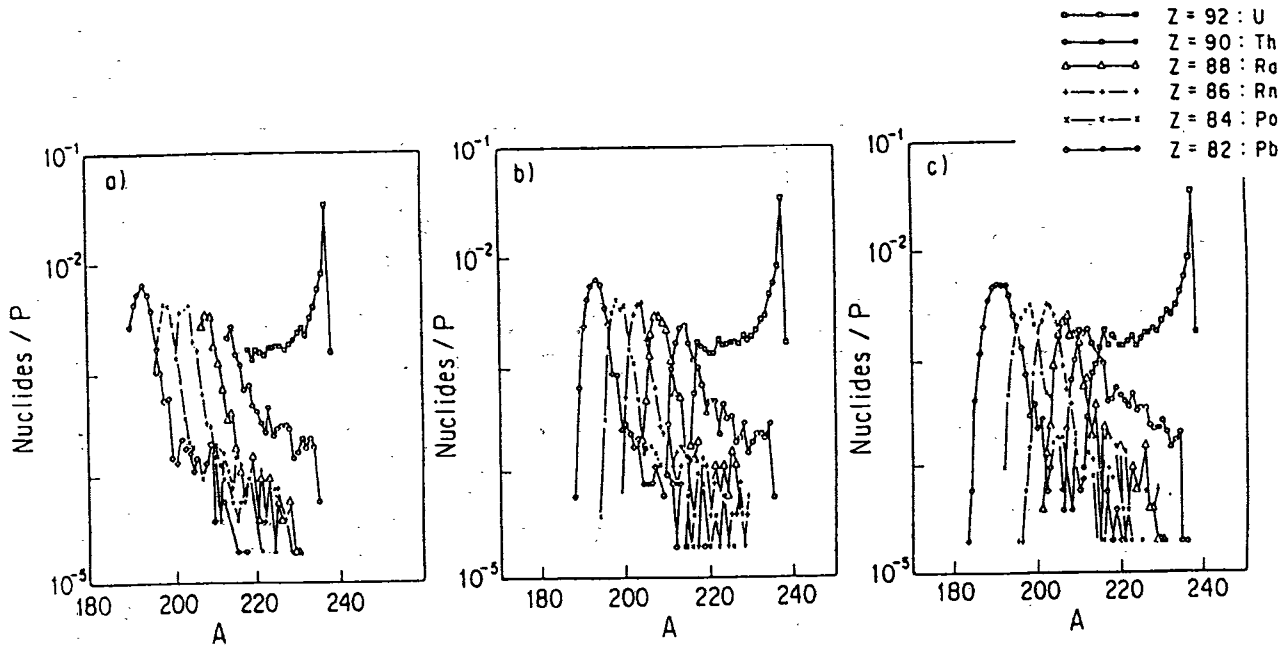


Fig. 7 Mass number distributions of products with Z from 92 to 82 from a uranium nucleus spallated by 1 GeV protons for cases of a) original version, b) extended region and c) extended region & Uno Yamada mass formula

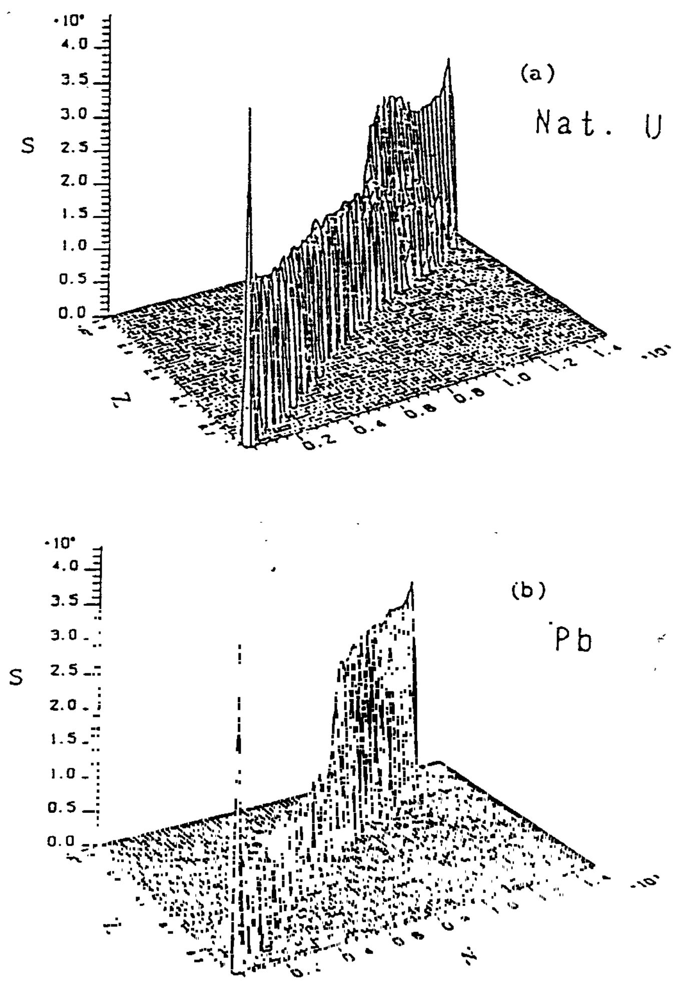
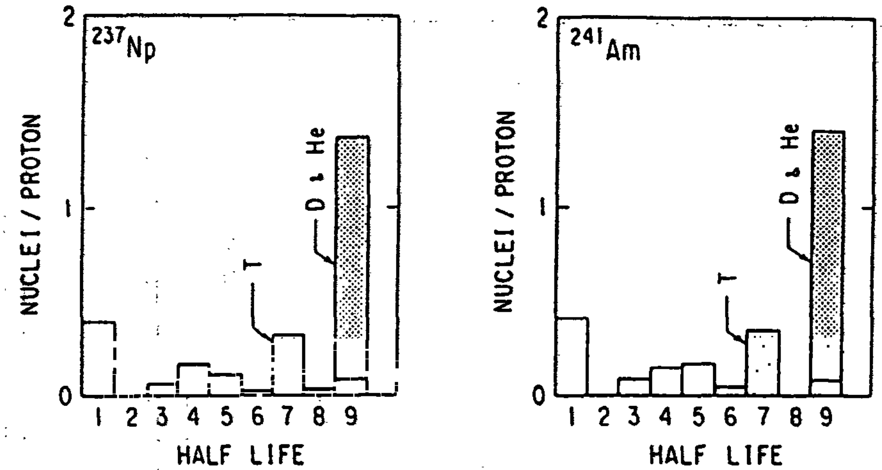


Fig. 8 Bird eye's views of yields of products for the cases of 1 GeV proton impinging on a) uranium and b) lead nuclei



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. 9 Histogram of half-life distributions of products when 2 GeV protons bombards on ^{237}Np and ^{241}Am target nuclei

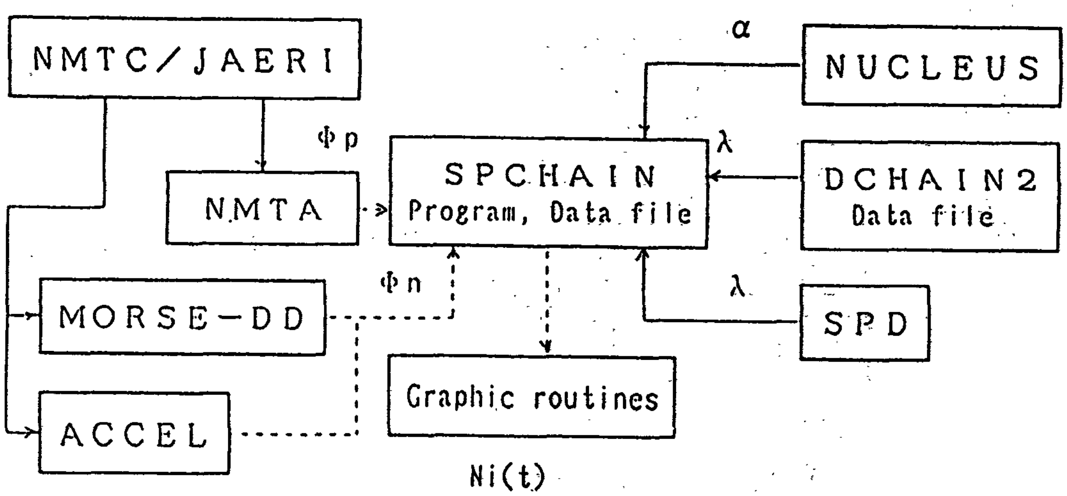


Fig. 10 Codes surrouding the SPCHAIN code

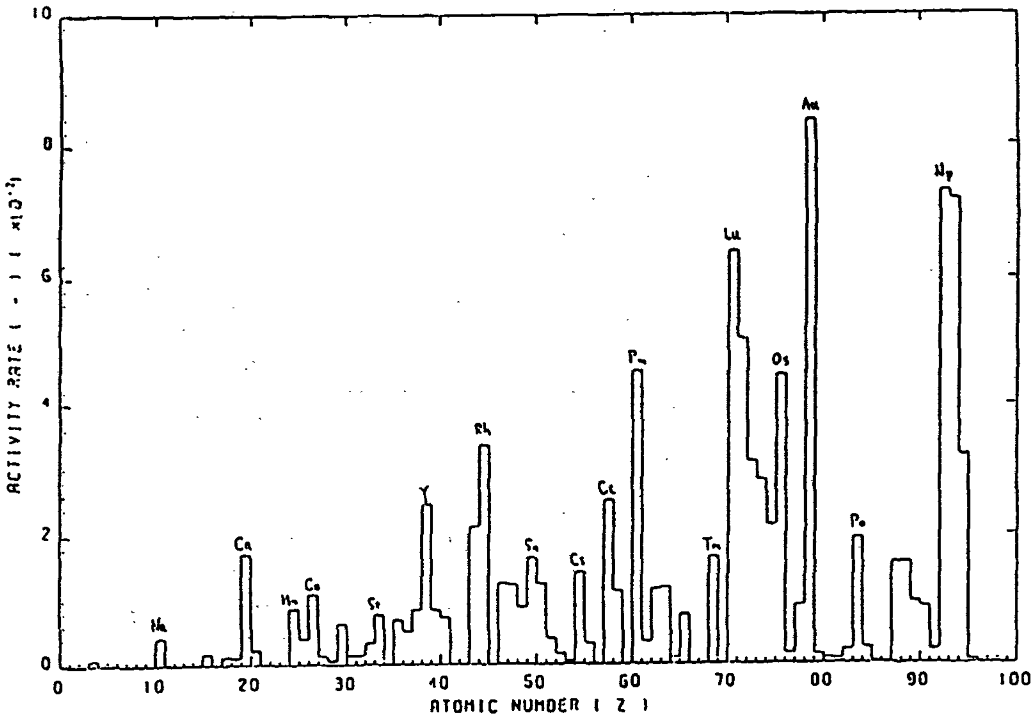


Fig. 11 Activity rate distribution of residual elements in a ^{241}Am target at the time stage of one year cooling after irradiation of ten hours of 1 GeV protons

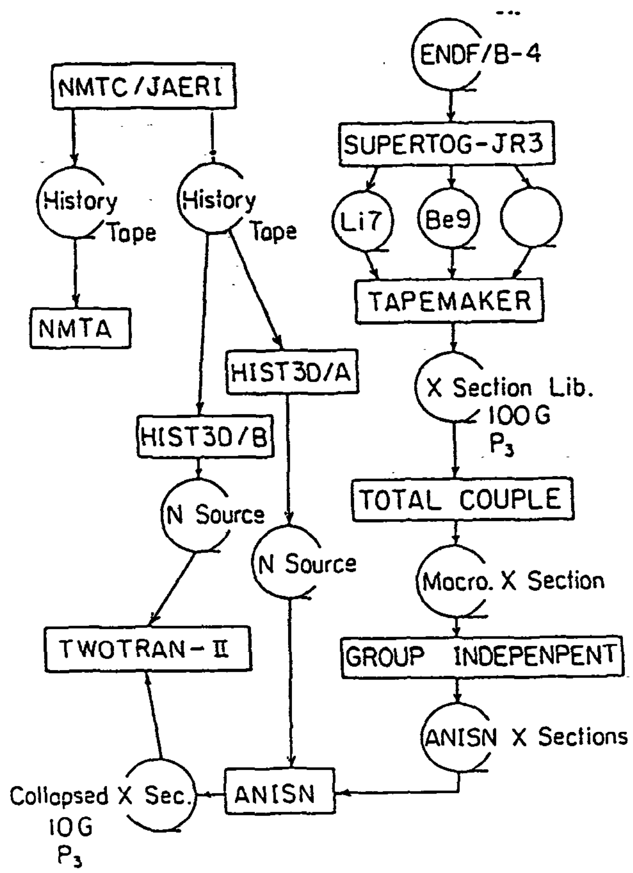


Fig. 12 ACCEL code system

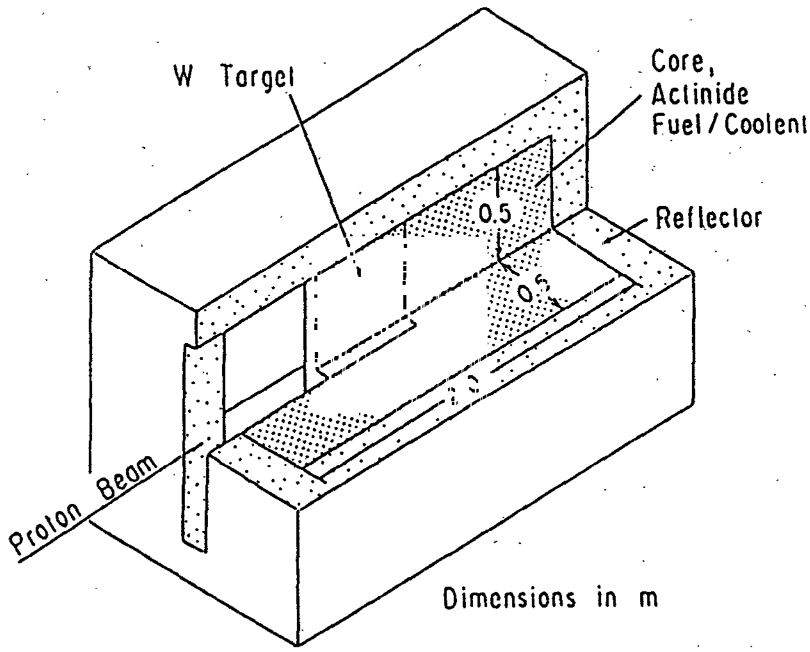


Fig. 13 Target-core configuration of hybrid transmutation system

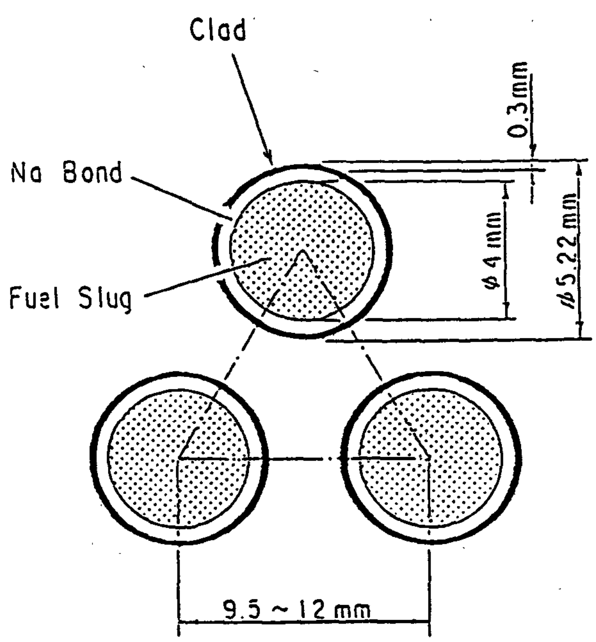


Fig. 14 TRU fuel pin geometry

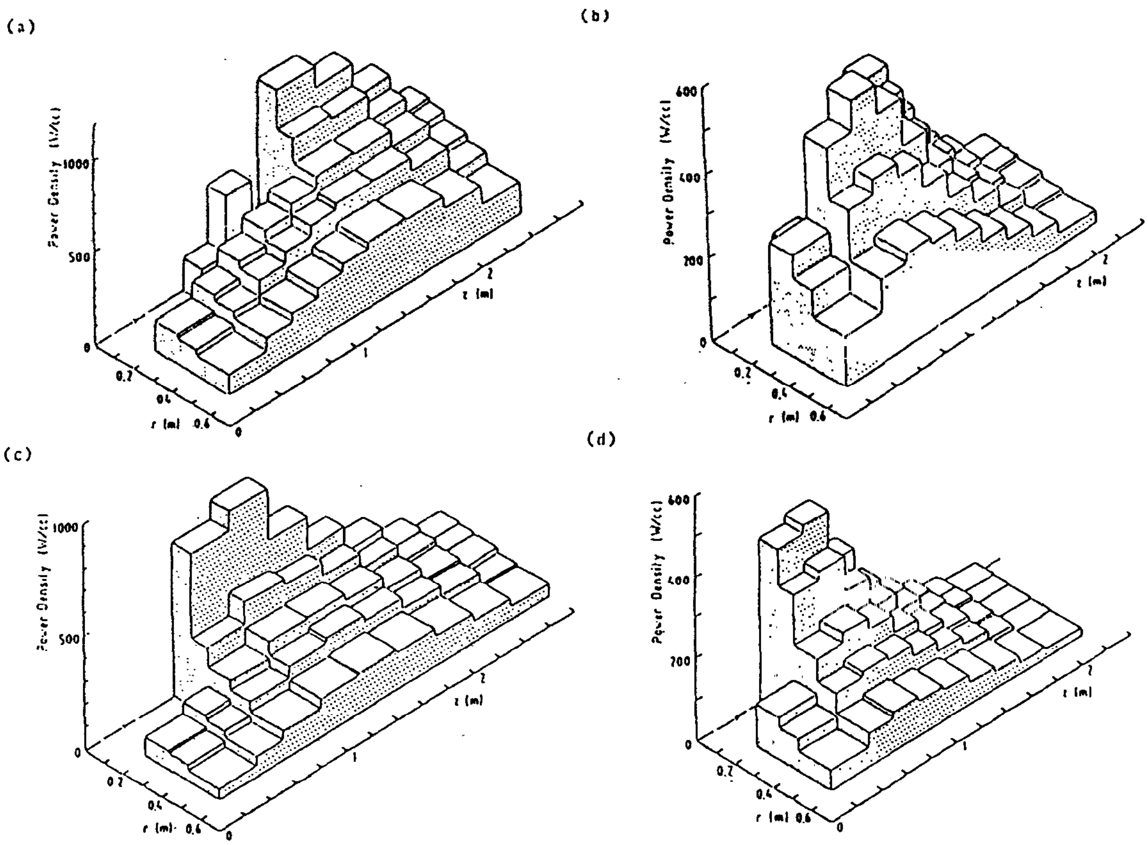


Fig. 15 Power distributions : (a) Na cooled , tungsten-loaded target, (b) Pb-Bi cooled , tungsten-loaded target, (c) Na cooled, TRU alloy target, (d) Pb-Bi cooled, TRU alloy target

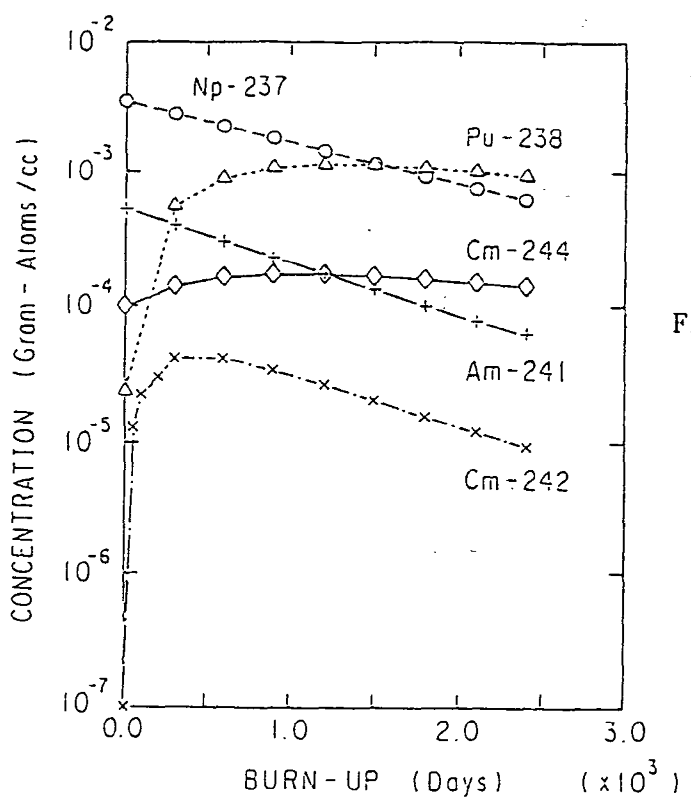


Fig. 16 Change of trans-uranium inventory with burn-up in the system