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TRANSMUTATION OF TRANSURANIC WASTES BY PROTON-INDUCED NUCLEAR SPALLATION REACTIONS

T. Nishida, Y. Nakahara, I. Kanno, H. Takada and Y. Kaneko

Japan Atomic Energy Research Institute
Tokai Research Establishment
Tokai-mura, Naka-gun Ibaraki-ken, Japan

Abstruct

The present research has been made with the aim of invetigating the feasibility of the idea that long life radioisotopes can be incinerated in a short time by the intense proton beam with high energy around 1 GeV. The outline and scope of the transmutation study of transuranic wastes (TRUW) by using the proton-induced nuclear spallation reaction are described. A Monte Carlo simulation codes NMTC/JAERI and NUCLEUS have been developed at JAERI to calculate the nuclear spallation reaction & particle transport in a target. Mass, isotope and halflife distributions of reaction products have been mainly calculated for the spallation reactions of some TRU nuclides such as Np and Am with a proton of the energy from 0.5 to 3 GeV. After spallation reactions, a variety of nuclei, especially many neutron-deficient nuclides, are produced as the results of many particle emissions. From our calculated results that most of them have halflives less than one year, it seems that this transmutation method is promising and has additional merits due to the neutron production also. Some plans in the near future are briefly described for improving the present simulation method and preparing the nuclear spallation experiments using a high energy proton beam.

(transmutation, transuranic waste, spallation, intranuclear cascade, evaporation)

Introduction

For the general acceptance of nuclear energy, the assurance to the risk of abundant radioactive wastes with long half-lives produced in a nuclear plant is one of the most important problems, espetially in the country which has a narrow land and a lot of population such as Japan. It is known that the annual TRU(Trans Uranic) Waste production rate is about 250 kg/y in ten units of 1000 MWe LWR. The protection for these products is currently achieved by reducing their volumes and cooling them for long times in the spots apart from our life environment, such as in a deep stratum. It seems that the saving of expense for these safeguards by using more effective, new methods contributes economically to the nuclear fuel cycle also. Recently the new methods, which transmute long-life wastes to shorter-life or stable nuclides in short proess time by irradiating a radioactive beam, have been actively investigated. These proposals are given in the following,

i) Transmutation of TRUW in the proton-induced spallation reaction,
ii) Transmutation of waste actinides in Actinide Burning Fast Reactor
iii) Disintegration of FP or TRU in the photo-nuclear reaction induced by high energy electrons.

Since TRU nuclides, generally, have cross sections larger than fission fragments, the feasibility of their transmutation seems promising.

Our basic idea is to transmute TRUW in a target directly by the proton beam and the subsequent cascade nucleons with the high energy. Since many high energy neutrons are simultaneously emitted as byproducts in this transmutation process, they can be used for other applications such as the breeding of fissile nuclides and the creation of very intense neutron source. It may be also pointed out that useful nuclide or short-life RI used for special purposes can be produced from residual nuclides after transmutation and group separation process. From the points of view of reactor physices and reactor engineering, new interesting pionts of this method are pointed out as follows,

a) TRU transmutation in the proton-induced spallatin reaction,
b) requirement to nuclear data of proton and neutron in the energy range above 100 MeV,
c) generation of many spallation neutrons as byproducts,
d) development of a high power accelerator applicable to the field of nuclear engineering,
e) R & D of accelarator-driven non-critical facility.

Figure 1 shows a flow chart of the nuclear fuel cycle in the thermal reactor system connected with the accelerator transmuter/breeder.

A computational study program has been carried out at JAERI to develop this idea to a realistic engineering concept since 1980. In order to make feasibility studies of the concept, we have been developing the Monte Carlo Simulation Codes for calculating the nuclear spallation reaction with a nucleus & the particle transport process in a

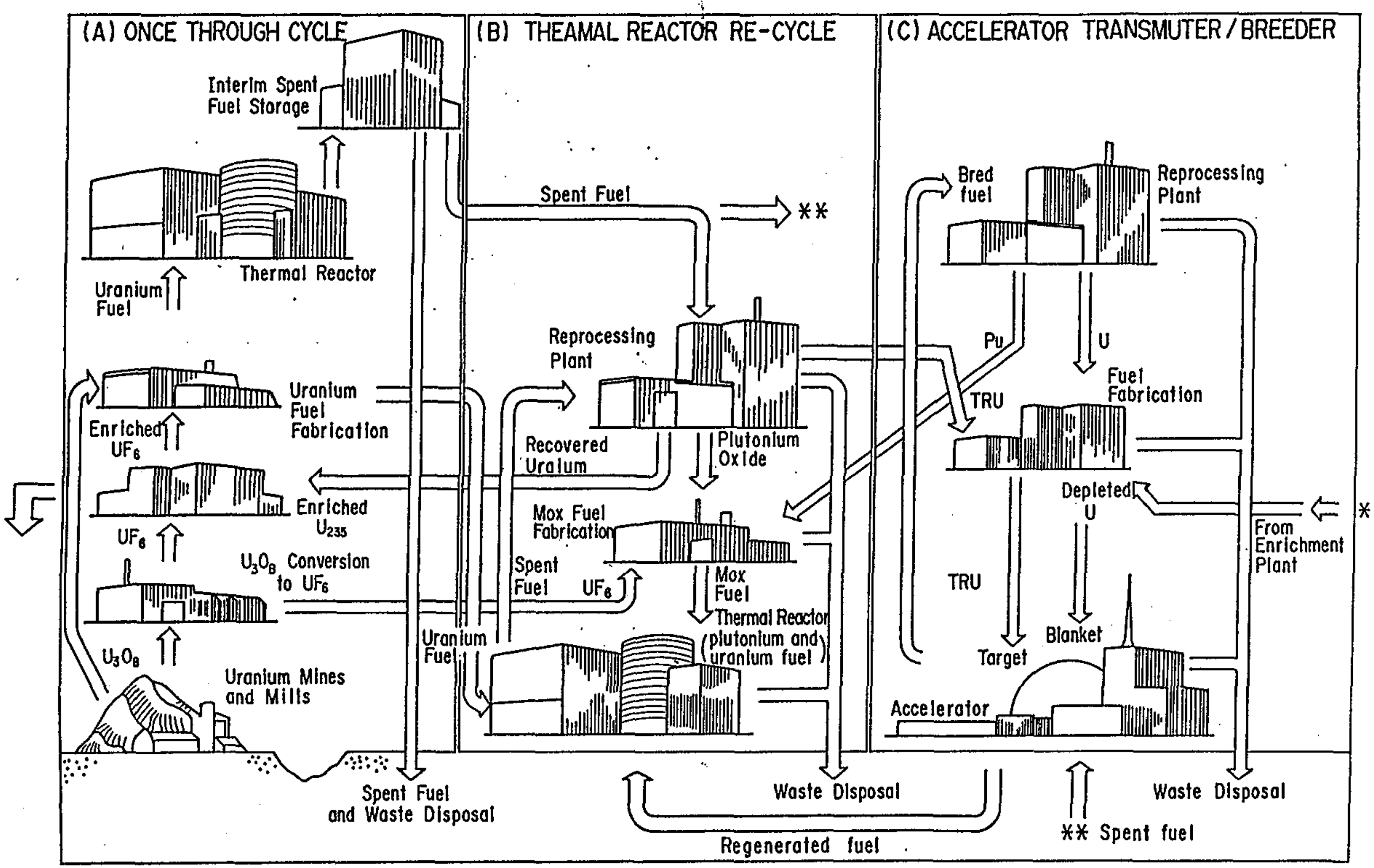


Fig.1 A new flow chart of the nuclear fuel cycle

target-blanket system. The extension and upgrade of these codes will be continued to design the TRU transmutation plant in the future. We have planning a series of proton-induced spallation integral experiments on lead and depleted uranium targets to examine the characteristics of the TRU transmutation plant.

Theoretical Study of Nuclear Spallation Process

Calculational Model

A nucleus bombarded by a high energy particle, such as a proton with the energy of hundreds to thousands MeV, undergoes a complicated destruction process, i.e., the so-called spallation. We use the two step model which consists of intranuclear cascades and competing decays through the high energy fission and particle evaporation. After a high energy particle has bombarded on a heavy nucleus, the intranuclear cascade of nucleons, pions and knocked-on particles are computed as the fast step of the nuclear reaction. In the present model a nucleus is assumed to be a sphere of a degenerate Fermi gas, in which the two body collision model [1]gives a good approximation to the collision process in the intranuclear cascade in the energy range higher than 100 MeV. At each nucleon-nucleon collision event the relativitistic conservations in particle's energy and momentum are checked and it is examined also if Pauli's exclusion principle admits the scattered Fermions. Pion production cross sections are calculated using the Isobar model. [2]

In the slow step after the intranuclear cascade has ceased, a highly excited compound nucleus selects the path to the particle evaporation or the nuclear fission as the subsequent process according to the fission probability based on Bohr-Wheeler theory with the level density parameters [3] fitted to Il'inov's experimental data.[4]. A semi-empirical combination of the Gaussian and folded - Gaussian distributions are used to determine masses of fission fragments, while their charges are selected from the Pik - Pichak & Strutinskii distribution.[5] The evaporation process is computed for neutron, proton, deuteron, triton, helium 3 and alpha particle by the Weisskopf statistical model.

With the aim of improving the computational accuracy we have developed a Monte Carlo code EXCITON[6] based on the Griffin's model[7] to simulate preequilibrium nuclear decay processes. The preequilibrium decay step is to be inserted in between the two steps mentioned above to make a complete simulation of an entire reaction proceess.

Simulation Codes

We have been developing a computer code system, the principal code of which is the Neutron Meson Transport Monte Carlo Simulation Code NMTC/JAERI [8], revised at JAERI to incorporate nuclear fissions from highly excited states into the computational flow. In order to examine in detail the computational model of the nuclear spallation reaction the Monte Carlo simulation code NUCLEUS [9] has been developed at JAERI for calculating only the nuclear spallation reaction. The theoretical models used in NUCLEUS are essentially the same as those in NMTC [10]and HETC [11] except the fission model. As the results calculated by this code can be compared directly with nuclear data measured by thin foil experiments, the code is useful for improving the computational methods and upgrading the values of nuclear parameters currently used in the calculations. Some results of evaluation studies were compared already with experimental data [12].

For the TRUW (Trans Uranic Waste) incineration process the time evolution calculation of biuldup and decay of all the nuclides produced in the target irradiated continuously by proton beams is necessary. In the actual computation, however, it is impossible to solve exactly the time evolution equation, due to the enormous consumption of computing time and computer memory and the insufficient compilation of nuclear data in the energy range higher than 20 MeV for nuclides. At present we have no useful means to solve the problem except computing the approximate rate equation. The one-point depletion code DCHAIN [13], which had been developed at JAERI based on the Bateman method for the calculations of decay and buildup of fission products, will be extended to the form including the spallation products in near future.

Cross section data for charged particles and neutrons in the intermediate and high energy range up to 3 GeV are important in the evaluation of our computational results. Although these data, especially experimental data, are few, the compilation project of them on ENDF/A-6 format up to 1000 MeV has been actively promoted by MENDWG (Medium Energy Nuclear Data Working Group), which has main office at BNL since 1986.

Calculated Results and Discussion

From the transmutation point of view, the reliable estimate of spallation ptoduct yield and the determination of its half-life distribution are very important. In the present paper we discuss the feasibility of spallation transmutation of TRU mainly based on some computational results obtained by NUCLEUS.

All the residual nuclides and particles produced from a ^{237}Np nucleus bombarded by protons with energies from 0.5 to 3.0 GeV are calculated. Figure 2 (a) shows the mass yield distribution of residual nuclei for the incident proton energy of 500 MeV. The first peak near the target nucleus and the second peak around A = 200 correspond to residual products, which are characterized mainly neutron-deficient nuclides just after intranuclear cascade and non-fission evaporation. Three spires in the light mass region correspond to the evaporated α, $^3He + t$ and d respectively. A flat region between them represents products due to the high energy fission. As the proton energy increases from 0.5 GeV to higher energy, the hill of non-fission product yield (A = 190~240) transforms to the one with a milder slope. The mass distributions of reaction products were examined in some detail for the nuclear spallation reactions of other nuclei, such as Nat. U, ^{241}Am, Pb and Ag in the same energy range. For a target nucleus with the mass number less than 200 such as Pb and Ag, scarce fission products appeared in the mass distribution. These results showed that the distribution of reaction products ceases to change its form and the yields decrease slightly as the proton energy increases over about 2 GeV, smilarly in the case of ^{237}Np. The bird-eye's view in the right upper figure represents the distribution of products from a uranium nucleus bombarded by 1 GeV protons in log-scale on the (N,Z) plane. It is apparent that non-fission products (mass number >180) are larger by two orders than fission ones.

The same tendency is seen in the energy variation of the number of particles emitted from a nucleus. Table 1 summarizes the number of emitted neutrons per incident proton and nucleus . It is apparent that the maximum yield of neutrons emitted from transuranic nuclides, Nat. U, ^{237} Np, ^{241} Am, is about 17 at 2 GeV, while it decreases also slightly over 2 GeV probably because of an increase of nuclear transparency for incident protons. But we can expect larger neutron yields in a bulk target due to a subsequent internuclear cascade as described later. The same tendency is seen in cases of lead and silver nuclei.

In the spallation reaction a variety of nuclei, especially many neutron-deficient nuclides with nuclear charges not far from the one of a target nucleus, are produced. In Fig. 3 mass distributions of isotope products in the nuclear spallation reaction by 1 GeV protons

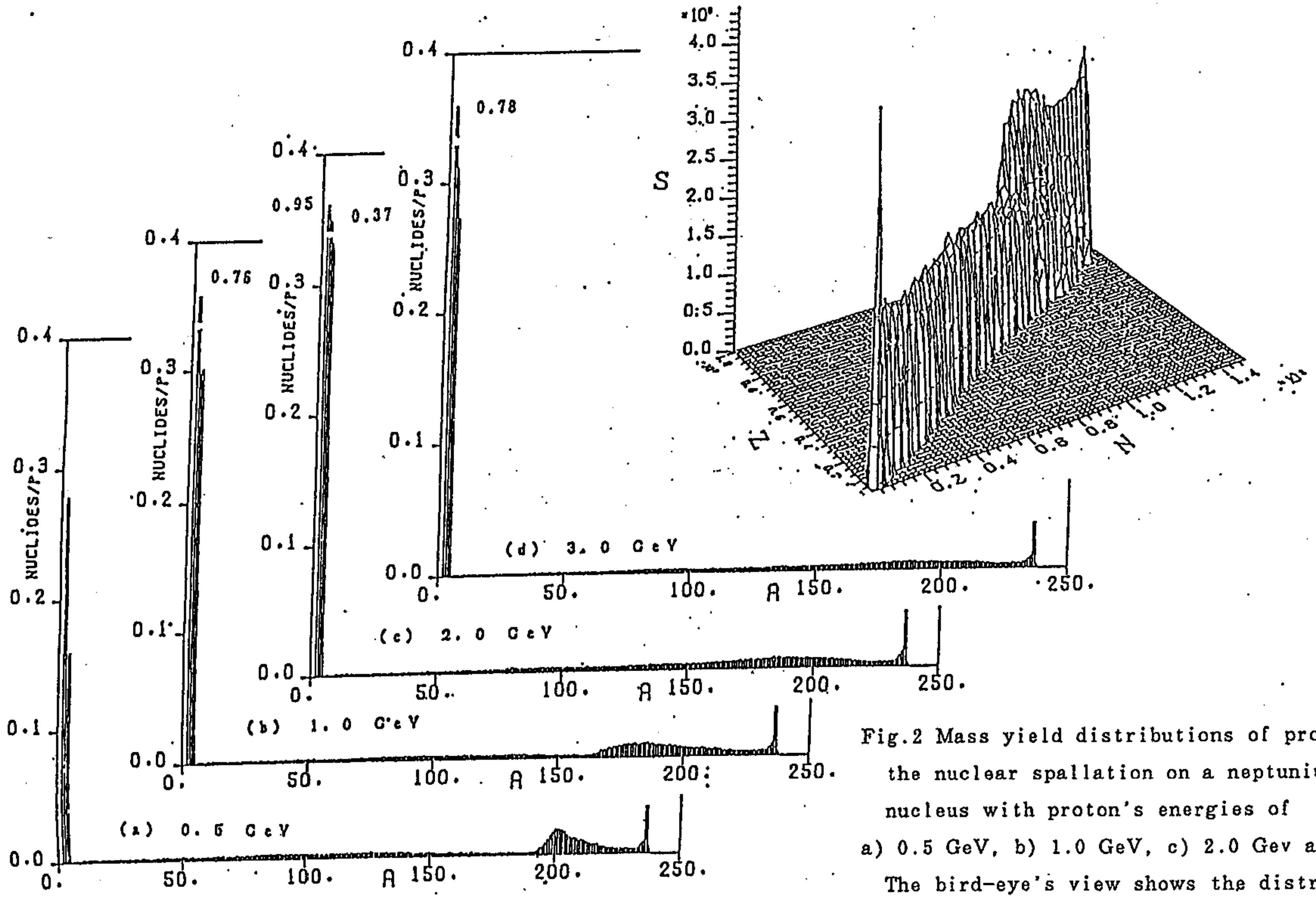


Fig.2 Mass yield distributions of products in the nuclear spallation on a neptunium-237 nucleus with proton's energies of a) 0.5 GeV, b) 1.0 GeV, c) 2.0 Gev and d) 3.0 GeV. The bird-eye's view shows the distribution of products from a uranium nucleus bombarded by 1 GeV protons in log-scale on the (N,Z) plane.

Table 1 Number of neutrons emitted from a single nucleus of some actinides, lead and silver nuclei

Energy of protons (MeV)		500	1000	2000	3000
(1)	Ag	5.18	6.15	6.59	5.33
(2)	Pb	10.64	13.09	14.31	12.20
(3)	Nat. U	13.01	16.05	17.32	14.91
(4)	Np 237	12.34	15.30	16.46	14.24
(5)	Am 241	12.41	15.39	16.48	14.28

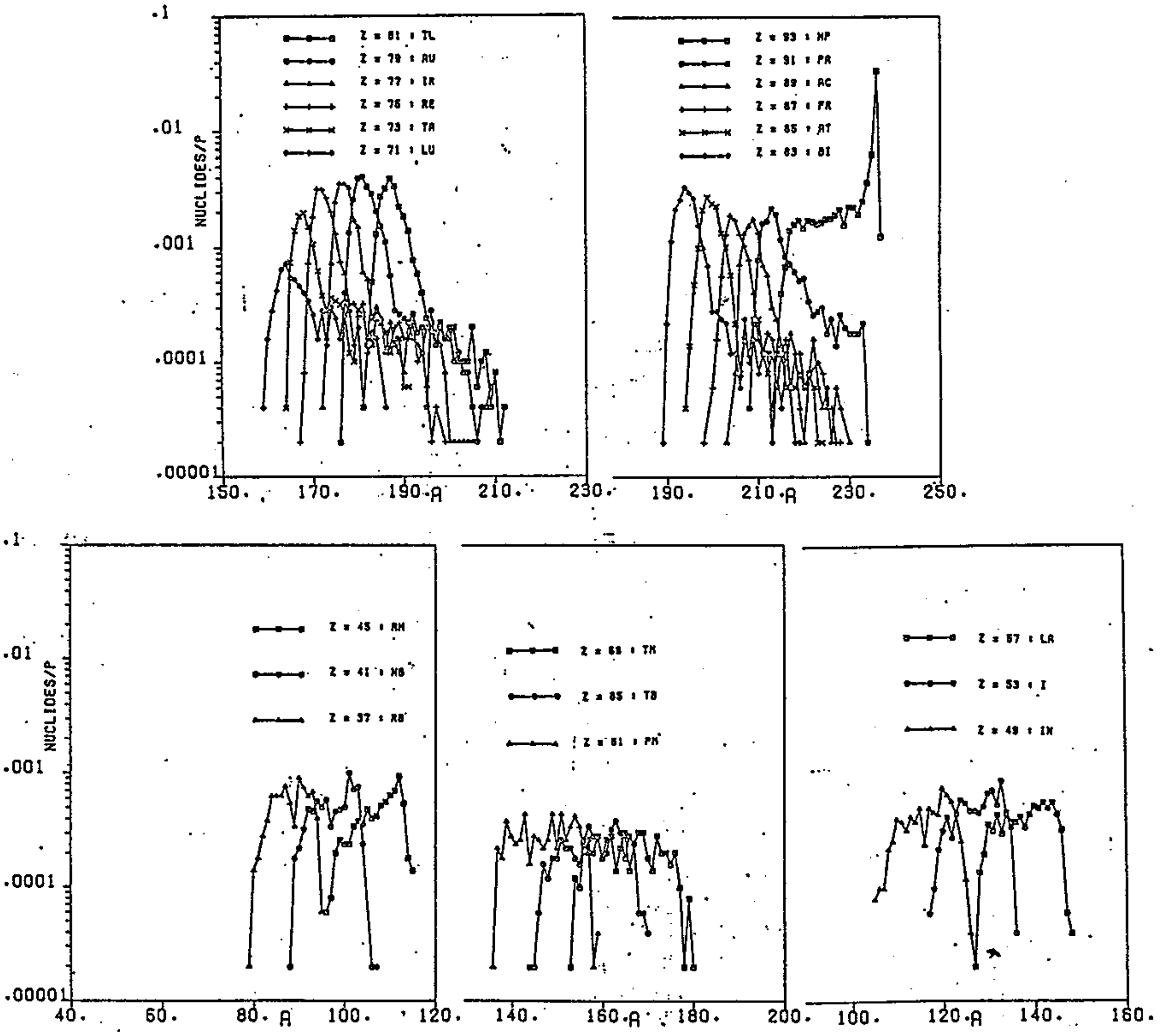


Fig.3 Isotope yield distributions of products with Z = 93-37 in the nuclear spallation reaction of a neptunium-237 nucleus with a 1 Gev proton

bombarding on a ^{237}Np nucleus are shown for atomic numbers Z = 91 − 37. It is aparrent that the maximum in each distribution with Z= 91 − 71 appears on the neutron deficient side on the A-axis except for Np (neutron excess peak :^{236}Np), since the stable nuclide of each element exists in the right tail of each peak. However the isotope distribution changes its form symmetrically with respect to the position of a stable isotope for elements with Z= 69 − 53, and neutron excess isotopes become dominant in the distributions for Z= 49 − 37.

It is also shown how the distribution is affected by the variation of the level density parameter a characterizing the evaporation probability in a highly excited compound nucleus. The residual nuclides from the spallation reaction of a ^{237}Np nucleus bombarded by protons of 500 MeV are examined by evaluating the contribution of level density parameter a to the evaporation calculation in the slow step. The distributions of isotopes of the non-fission products are shown in Fig. 4 with odd atomic numbers Z = 93~83 for a = A/30, A/20, A/10 and A/5 ,where A denotes the mass number and in NUCLEUS the Le Conteur's equation gives A/7.7~A/7.4 to the value of a for the nuclides with the mass number more than 200. When a decreases from A/5 to A/30, the shape of Neptunium distribution (Z=93) in the neutron-deficient side varies from a subsidary peak to a steep slope. The tail of Protactinium(Z=91) peak in the neutron-excess side shrinks and the peak's width becomes wider. The height of the Bismuth peak(Z=83) increases by about one order. Therefore, in order to calculate exactly the product yield of transmuted nuclei, it is neccesary that the value of level density parameter is reasonably fitted to measured data.

Figure 5 illustrates half-life distributions of product yields in the transmutation of three actinide nuclei, natural uranium, ^{237}Np and ^{241}Am, bombarded by a 2 GeV proton, where 9 decay classes represent the time range of half life of a nuclide as described in this figure. There are major products in 1st, 7th(triton) and 9th(heliun and deuteron) classes. It is desired from the transmutation point of view that yields in 7th (1 y − 100 y) and 8th (> 100 y) classes are as possible as small. As seen from the figure, these yields are small except triton. This fact seems quite promising for this transmutation method.

On the other hand, the actual accumulation yield of a residual nuclide due to buildup and decay processes in the spallation reaction may be roughly estimated from product yields on the β decay chain leading to it and thier half-lives. Product yields on β decay chains

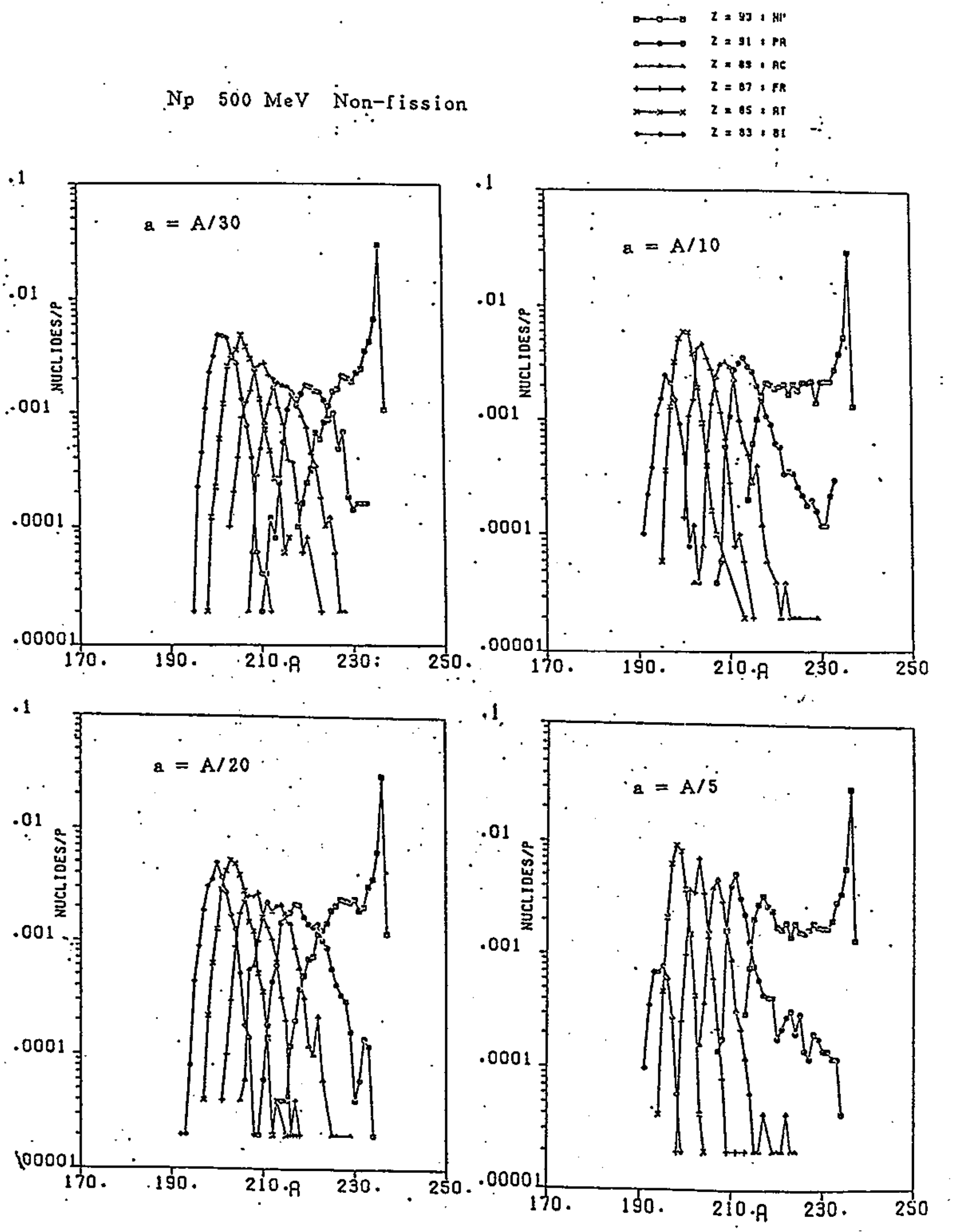


Fig.4 Isotope yield distributions of the non-fission component of products with Z = 93-83 in the nuclear spallation reaction of a Neptunium-237 nucleus bombarded by a 500 MeV proton Level density parameter :

a) A / 30, b) A / 20, c) A / 10 and d) A / 5

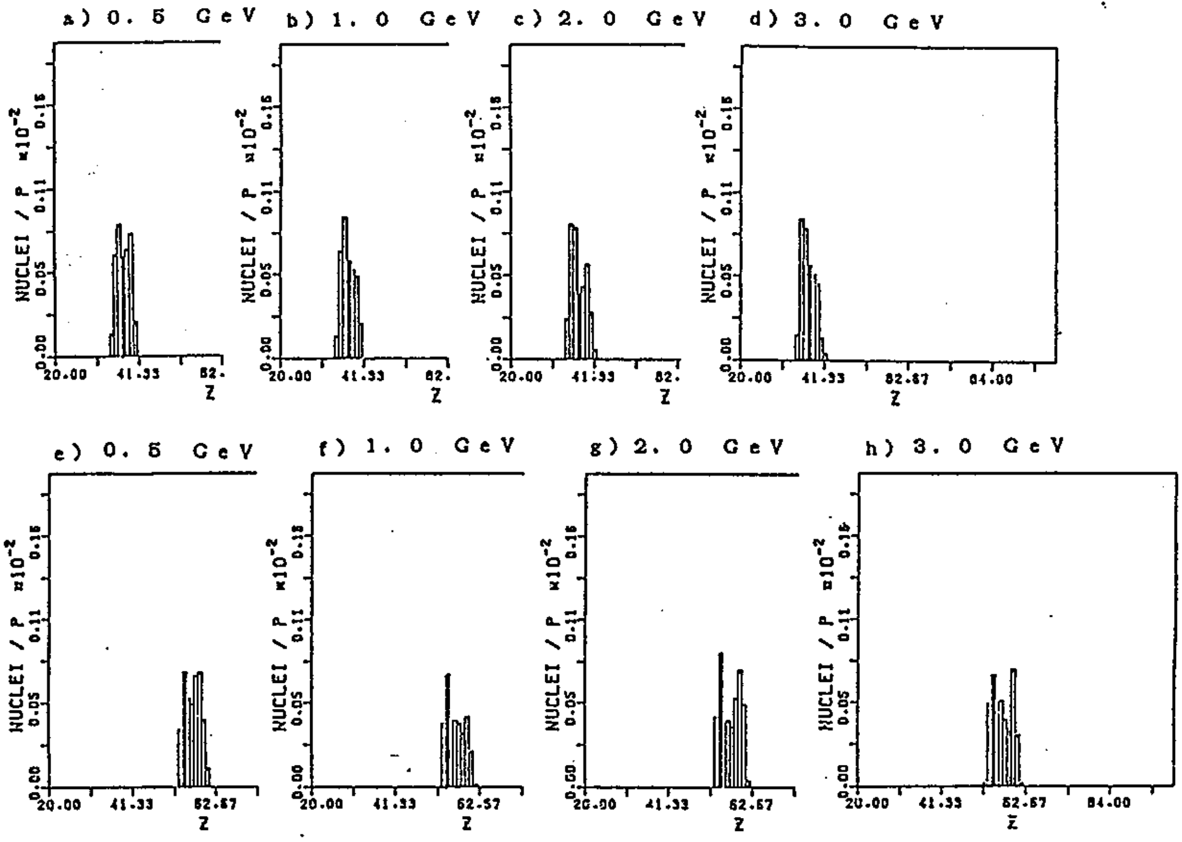


Fig.6 Product yields on the β decay chain leading to ^{90} Sr with incident proton's energies of a) 0.5 GeV, b) 1.0 GeV, c) 2.0 Gev and d) 3.0 GeV and to ^{137} Cs with incident proton's energies of e) 0.5 GeV, f) 1.0 GeV, g) 2.0 Gev and h) 3.0 GeV

DECAY CLASS

1 : OTHERS T(1/2) < 1. E-3 SEC
2 : 1. E-3 SEC < T(1/2) < 1 SEC
3 : 1 SEC < T(1/2) < 1 MIN
4 : 1 MIN < T(1/2) < 1 HOUR
5 : 1 HOUR < T(1/2) < 5 DAYS
6 : 5 DAYS < T(1/2) < 1 YEAR
7 : 1 YEAR < T(1/2) < 100 YEARS
8 : 100 YEARS < T(1/2) < 1. E+8 YEARS
9 : NRTURRL

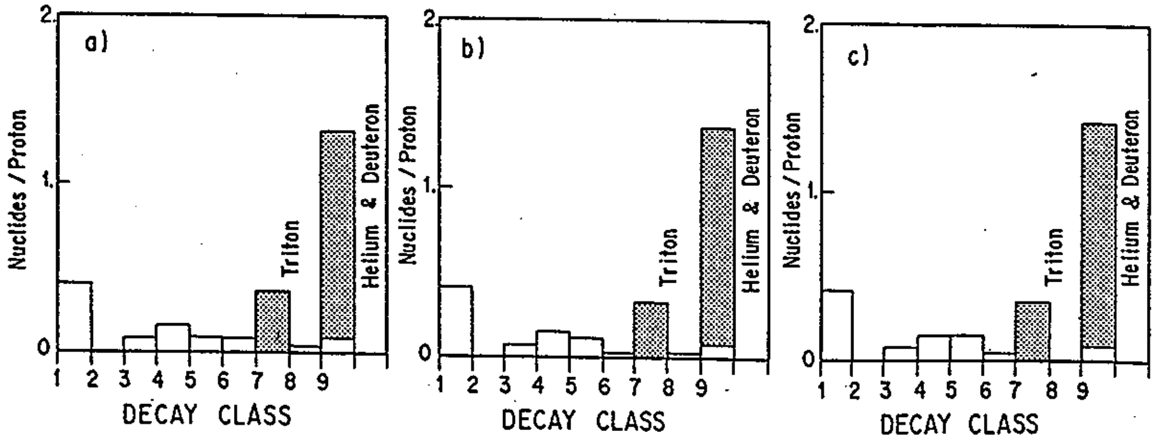


Fig.5 Half-life distribution of product yields in the spallation reaction of a 2 GeV proton with a target nucleus a) natural Uranium, b) Neptunium-237 and c) Americium-241

leading to ^{90}Sr and ^{137}Cs are shown by blacked rectangles in Fig. 6. Most of them are produced mainly from the fission processes after intranuclear cascades. These product yields are smaller by about an order of magnitude than those of non-fission products and the accumulations from the decay chains are seen also to be small. It is advantageous to the transuranic waste transmutation that the production of harmful FP nuclides such as ^{90}Sr is small. Conversely product yields on the Z-axis with mass numbers of 235 and 233 are expected to accumlate in due time to ^{235}U and ^{233}U respectively as shown in Fig.7. It is very interesting that not a small amount of fissile materials ^{235}U and ^{233}U are produced also. The spallation reaction producing a nuclide at the first peak in the non-fission product region is virtually equivalent to " one neutron emission" reaction. There is a flow scheme producing a fissile nuclide from a TRU nucleus by the suitable combination of two "one neutron emission" and one short life β decay. Two production routs of ^{235}U from ^{237}Np and ^{239}Pu from ^{241}Am are shown in Fig.8.

Table 2 summarizes the results of the spallation calculation for a target model (cylinder, length 60 cm, radius 10 cm) of Nat. U. The number of transmted nucleus and emitted neutron per incident proton are listed for the energy range of 1.5 to 3.0 GeV. It is possible that about five nuclei are transmuted per an incident proton of 1.5 GeV in internuclear cascades of high energy nucleons above 15 MeV. The value implies that the annual products of TRU from four units of 1000 MWe LWR can be transmuted by using this method as given in the last line of Table. It should be noted, however, that the contribution of the spallation neutrons with energies below 15 MeV is not taken into consideration in estimating the transmutable amount of TRU. If all those spallation neutrons are used also to transmute TRU, it is estimated that it is possible to transmute the amount of TRU from ten units of 1000 MWe LWR. Figure 9 illustrates an example of a TRU transmter-breeder system. Then a high power proton LINAC(1.5 Gev, 300 mA) has to be developed to drive this system.

Spallation Integral Experiment

For examining the actual efficiency of TRU transmutation, spallation experiments on a target irradiated by high energy protons have to be carried out. Our group are planning a seris of target integral experiments using 500 MeV proton beams in co-operation with KEK

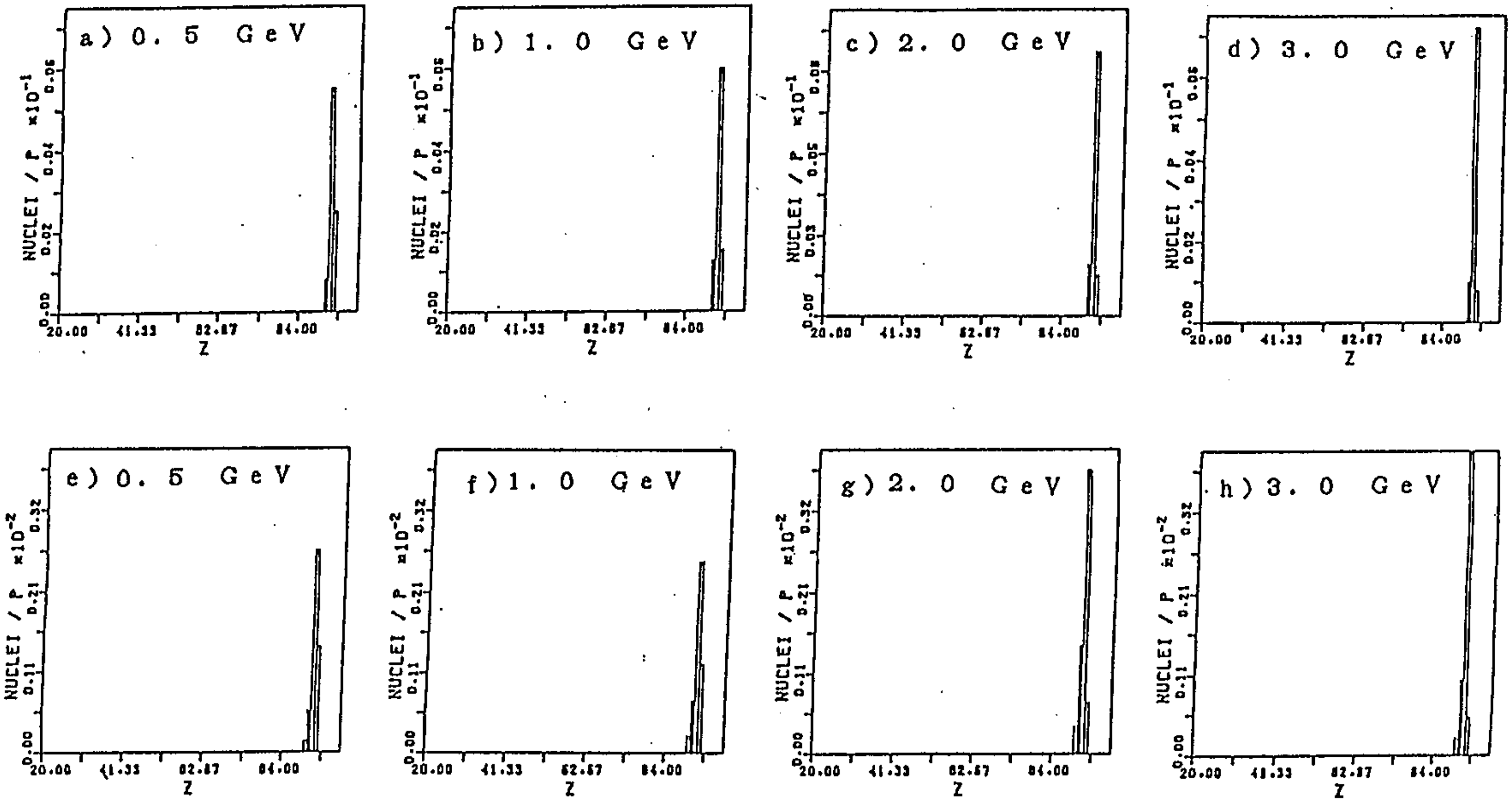
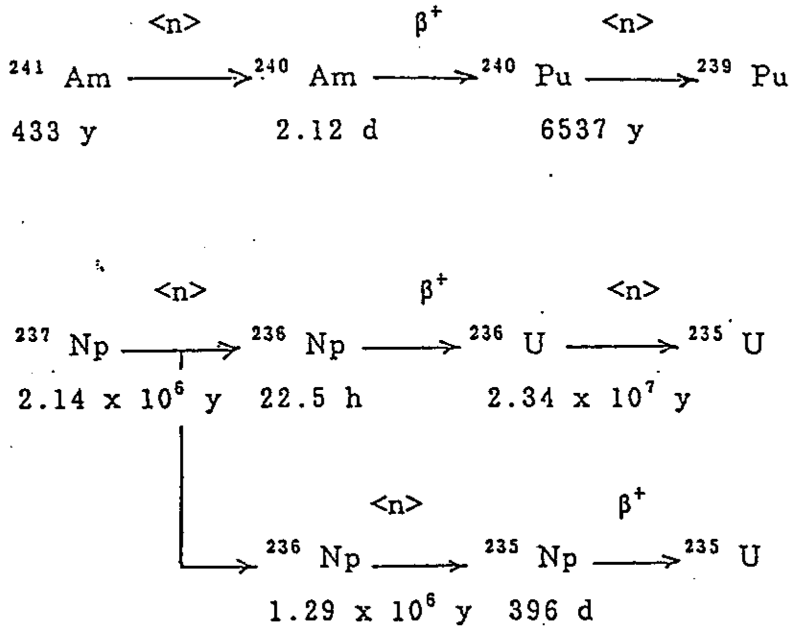


Fig.7 Product yields on the β decay chain leading to ^{235} U and ^{233} U with incident proton's energies of a) 0.5 GeV, b) 1.0 GeV, c) 2.0 Gev and d) 3.0 GeV



<n> : main peak in the spallation reaction

Fig.8 Flow scheme of fissile production from TRU due to the suitable combination of double transmutation and short life decay reactions

Table 2 Numbers of transnuted nuclei and emitted neutrons per incident proton in a uranium target

Incident Proton Energy (GeV)	1.5	2.0	2.5	3.0
Transmuted Nuclei / Incident proton *1	5.3	6.8	8.3	9.6
Emitted Neutrons / Incident proton *2	57.9	73.8	88.6	102.8
Number of LWR units *3 (Their Annual Products of TRU transmuted in the spallation)	4.0	5.1	6.2	7.2

*1 Target size : Radius 10 cm, Length 60 cm
*2 Most of them with the energy lower than 15 MeV
*3 1 GWe, TRU annual products 23.1 kg (33,000 MWd/t)
Proton beam 0.3A, Plant factor 0.75

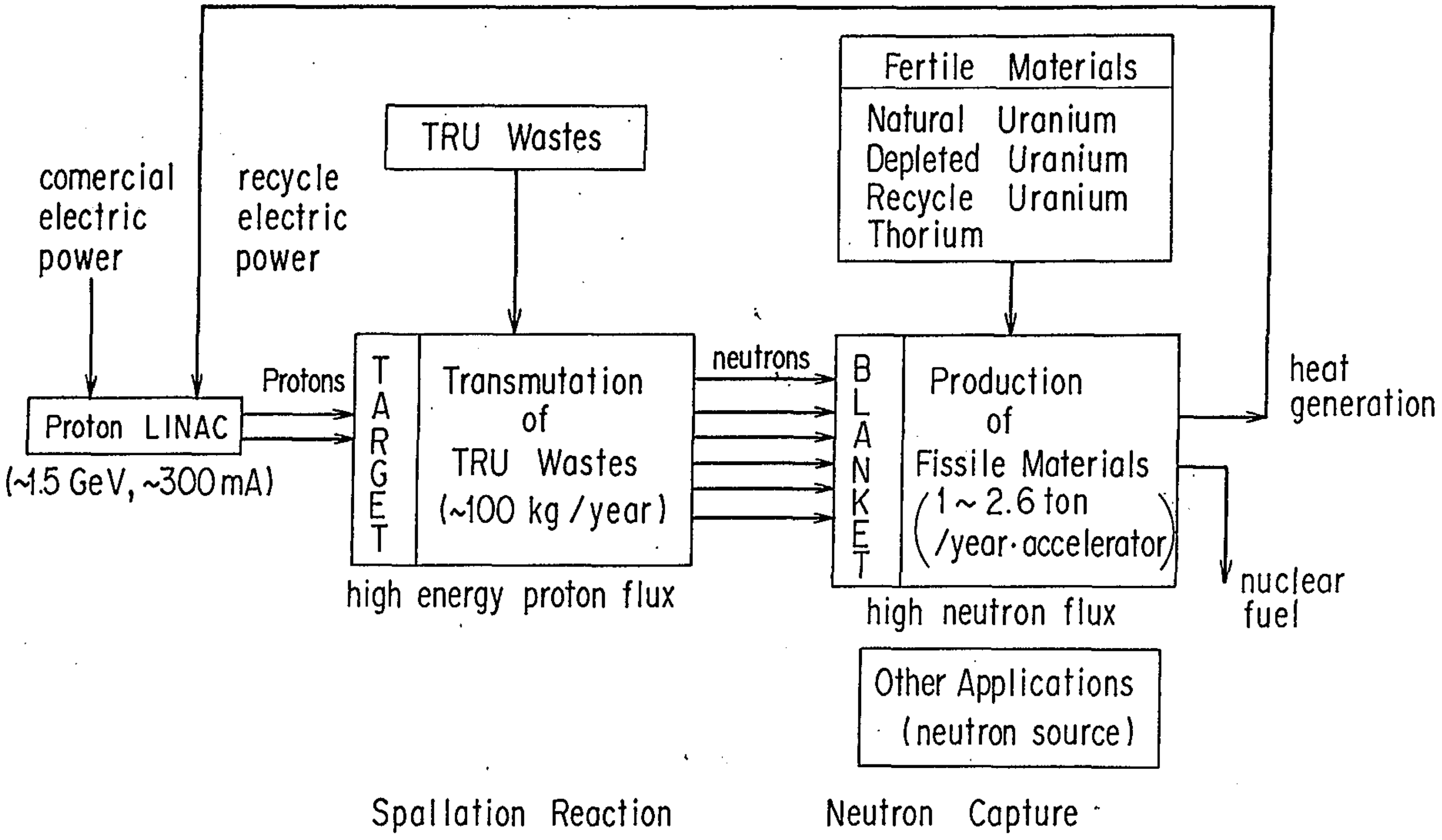


Fig.9 An example of the TRU transmutation target-blanket system driven by an high power proton LINAC

groups.(High Energy Physics Labolatory at Thukuba) At first the spallation products and emitted particles in the lead target are measured as the preliminary experiment to ascertian the spallation transmutation of TRU and to validate and upgrade the calculational model and the computer codes. For the next step, the depleted uranium target is used to simulate the TRU spallation experiment before the direct irradiation of TRU nuclides.

Summary and Conclusion

In order to make evaluations of TRU transmutation in proton-induced spallation reactions, simulation codes has been developed. Many Monte Carlo computations have been done to calculate the nuclear spallation reaction of TRU nuclides. From these results, we summarize as follows.

(1) Calculated results of all the produced nuclei and number of emitted neutrons show that an energy brought into a nucleus by an incident proton for the TRU transmutation saturates around 2 GeV, independent of the mass of a target nucleus.

(2) Many high energy neutrons are generated in the spallation reaction and can be used for the breeding of fissile nuclides, the creation of intense neutron source and the transmutaion of TRU in the lower energy reaction.

(3) In the nuclear spallation reaction, more neutron deficient nuclides are produced than in the fission and mostof them have half-lives shorter than 100 y, some of which can be used as the source of unstable nuclide beam. Few harmful nuclides such as ^{90}Sr are produced. There is the feasibility of producing fissile materials by the multi irradiation of protons on a TRU target also.

(4) From these results it seems that the transmutation of TRU in the spallation reaction is more promising.

The main study program in the near future will include the following :

(a) the upgrade of precision of computational results of nuclear data in the high and intermediate energy range due to the sufficient compilation of nuclear data,

(b) calculations of the decay and buildup processes for spallation and fission products by using the extended DCHAIN,

(c) design analysis of a transmuter model,

(d) measurement of reaction products in the proton-induced spallation integral experiments on a uranium target.

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