Transmutation of Fission Products through Accelerator

H. Nakamura , S. Tani, T. Takahashi and O. Yamamura
Power Reactor & Nuclear Fuel Development Corporation
1-9-13, Akasaka, Minato-ku, Tokyo, 107, Japan

Abstract

The transmutation of fission products through particle accelerators has been studied under the OMEGA program. The photonuclear reaction has also been investigated to be applied to transmuting long-lived fission products, such as Cesium and Strontium, which have difficulties on reaction with neutrons due to its so small cross section. It is applicable for the transmutation if the energy balance can be improved with a monochromatic gamma rays in the range of the Giant Dipole Resonance generated through an excellent high current electron linear accelerator. The feasibility studies are being conducted on the transmutation system using it through an electron accelerator.

1. Introduction

The high level radioactive waste (HLW) generated from the reprocessing of spent fuel is one of issues in the backend of nuclear fuel cycle. In Japan, a present national policy for the HLW is to vitrify and dispose it in a deep geological repository after 30-to-50-year storage for cooling down the decay heat. The researches and developments have been devoted to the technologies for waste disposal and the methodologies for safety assessment. In parallel, the long-term R&D program, "OMEGA," on nuclides for partitioning and transmutation was initiated in 1988 by the Japanese Atomic Energy Commission in order to explore the new way utilizing the HLW as useful resources and reduce the burden to the geological disposal. The program led by the Science and Technology Agency has been conducted with the collaboration of three major research organizations; the Japan Atomic Energy Research Institute, the Power Reactor and Nuclear Fuel Development Corporation(PNC) and the Central Research Institute of Electric Power Industry.

The program is composed of major R&D, as shown in Fig.1. The nuclides are partitioned from HLW according to half-life and potential value for utilization. The minor actinides (MA; ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm, etc.) and long-lived fission products (FP; ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs, etc.) are transmuted into short-lived or stable nuclides.

2. Transmutation of Fission Products

The methods of transmutation of the long-lived nuclides in the HLW are roughly classified into those using neutron and those using protons or photons. The neutron methods are available to transmute actinides having comparatively large fission cross sections for fast and thermal neutrons. The fission reactors are expected as neutron sources for the transmutation of long-lived minor actinides. The studies in OMEGA program have indicated that the fast reactors have an advantage over the thermal reactors in transmuting actinides efficiently and in generating less secondary waste. Thus, the concept of utilization of the electric power plants of fast reactors has been developed into the acitinide recycle as an advanced nuclear fuel cycle.

2.1 Methodologies of Transmutation of Fission Products

For the transmutation of fission products, Tc and I, having comparatively large cross sections for thermal neutron, the neutron methods could be conceptually applied to transmutation by the thermal or fast reactor if the core was modified to have a specified region. However, Cs and Sr as typical fission products having so small cross sections for thermal and fast neutrons, demands extremely high neutron fluxes for transmutation up to 10^{17} to 10^{18} n/cm^{2}s in fission reactors. It may be hard to obtain this high neutron fluxes at the present days. The fission reactors are limited to use in the transmutation of fission products. Thus, as another alternative of incineration, the utilization of high energy particles through accelerators has been considered, referring the recent technological advances and backgrounds of accelerators.

2. 2 Alternatives for Transmutation of Fission Products

The feasibility of the transmutation by the particle accelerators using spallation and spallation-neutron and photonuclear reaction has been evaluated, focusing on the energy balance in the transmutation.

Transmutation of Tc and Cs as target nuclides are referenced in this study. Tc as representative of nuclides has large cross sections for thermal neutron. Therefore the spallation-neutron method and the muon catalyzed fusion method are relatively effective for the transmutation, as shown in Table 1.

As for Cs, the spallation-neutron method is relatively potential to the transmutation with high flux neutrons supplied through the spallation reaction. In the method, the high energy protons impinge on heavy metal such as lead placed in the center of the target. The neutrons generated by spallation reaction are thermalized and supplied to the transmuting reaction (n,γ) of fission products. The muon-catalyzed fusion method is effective for transmutation of Cs. In this method ^{137}Cs is transmuted into ^{136}Ba through the (n,2n) reaction of neutrons generated by the muon and D-T fusion in the target. These methods are relatively more advantageous than the proton spallation method and photonuclear reaction method, as shown in Table 1.

The evaluation of these methods indicates that the spallation-neutron method is relatively feasible. Therefore a hybrid system as accelerator-driven reactor can be a candidate of the transmutation of fission products. However more technology developments are required for the system to be used for the transmutation of fission products, from the point of energy balance. The muon-catalyzed fusion is still future technology. Therefore it is needed to research other alternatives.

In this context, the photonuclear reaction method would be one of alternatives, with the advantage of its simpleness of transmutation, less secondary-waste production, and the experienced accelerator technology.

But it still has the energy balance problem. In order to improve the energy balance, the transmutation by the photonuclear reaction is re-evaluated through monochromatic γ rays generated from a high power electron linear accelerator.

3. Photonuclear Reaction for Transmutation

The photonuclear reaction is caused by γ rays: a nuclide is radiated and excited resonantly with γ rays of 10 - 20 MeV in the range of the Giant Dipole Resonance, and then a neutron is released from a nuclei and the nuclei is transmuted. The photonuclear reaction has uniform cross sections for the actinides and fission products. The cross sections of Sr and Cs have the threshold at around 8 MeV and the maximum peak at 300 or 200 mb as shown in Fig. 2. There exist competitive reactions as well as photonuclear reaction; the pair electron creation, photoelectric effect and the Compton effect. Among them, the largest is the pair electron creation that has cross section of around 8000 mb. With competitive reactions, the photonuclear reaction is expected to be caused by 3 to 5% of monochromatic γ rays in the target. In the reaction, a nuclei of ^{137}Cs is transmuted by the monochromatic γ rays around 15 MeV and the transmutation requires 400 MeV energy, as shown in Fig. 3. On the contrary, with the Bremsstrahlung γ ray it requires up to 4700 MeV energy. Therefore the photonuclear reaction method can be improved, if monochromatic γ ray is provided to the target. It is assumed that the criteria of the energy balance for transmutation is 10% of the usable energy that is 1100MeV with 200 MeV released energy through fission reaction, with 6% of fission yield of ^{137}Cs, and with 33% of the conversion of thermal power to electricity.

3.1 Accelerator for Photonuclear Reaction

In the large amount of transmutation by the photonuclear reaction, well-qualified and high-current electron beam is required. For example, with the transmutation of 40 kg of ^{137}Cs produced in a 1000 MWe LWR in a year, a few amperes of beam current and beam energy around some hundreds of MeV will be needed, being taken into the efficiency of conversion from electron beam to γ rays in the range of 10 to 20 MeV. This requires to develop an accelerator for the high quality electron beam with the energy range of 100 - 1GeV and high current. However, as shown in Fig. 4, the electron linacs in the world have been so far developed in order to elevate the beam energy for the high energy particle physics, and even in the medium beam energy, there is little experience for developing high current accelerator. As for the high current beam, it has to be well controlled to avoid the beam-break-up phenomenon which causes damage to accelerator guides. The heat removal from the tubes and other components has to be deliberately designed to avoid the thermal structural deformation disturbing beam stability. To get monochromatic γ rays with high intensity, the conversion of the electron beam into monochromatic γ rays, such as the laser Compton scattering et. al., is the key technology. Therefore it is needed to research these technical feasibilities by using an experimental accelerator.

3.2 The Research on High Current Electron Accelerator

It is technically and also financially quite difficult to achieve some hundreds MeV energy and the current of a few amperes at one step. Thus, the experimental high power electron linear accelerator, with 10 MeV beam energy and the maximum / average current of 100mA / 20mA is under development. The main specification and the basic structure of the accelerator are shown in Table 2 and Fig. 5, respectively.

The experimental accelerator basically consists of electron gun, chopper, pre-buncher,buncher,accelerator guides with traveling wave resonant rings and beam dump. It is designed to facilitate the study of key technologies and high intensity beam. The beam is delivered with a current of 100 mA, 4 mA pulse and 50 Hz pulse repetition. The accelerator will be completed in March 1997 but an injector experiment is planned to be conducted in 1995.

The components of the accelerator are deliberately designed through experiments and analyses. The injector system is designed to consist of a 200kv DC gun, magnetic lens, a RF chopper, chopper slits, a prebuncher,and a buncher. The electric gun is designed to be available on a pulse-to-pulse repetition rates from single pulse to 50 pulses/sec. In the chopper consisted of a RF chopping cavity and a slit, three magnetic fields are mixed together for adjusting RF field amplitude and phase, and the chopped beam can be led to the beam center line. Bunching in the injector is conducted by the prebuncher. The traveling-wave resonant ring accelerates or decelerates electrons depending on their phase with respect to the buncher RF field.

The accelerator tubes are characterized by the traveling-wave-ring and are excited with microwave power at a frequency of 1.25 GHz. The accelerating tube has a cylindrical, disk-loaded shape made by oxygen free high-purity copper. The structures maintain a constant axial electric field over its length. The length of accelerating section is 1.2 m with 13 of cavities and two coupling cavities. In the high power linac operation, considerable amount of heat is expected to be generated in accelerator structure. The dimension of the accelerating section is determined by the analyses of heat transfer and thermal stress.

A beam dump of the high power and low energy beam(200kw of 10 MeV electron beam) is a challenging technology, in which electrons with 10MeV energy, concentrates within a range of only a few centimeters. The energy deposition causes high thermal power densities in a beam dump. The klystron used in the accelerator were developed specially to operate in continuous waves and pulse with good efficiency(over 60%). The output windows were designed and tested with pill-box type windows. Based on the test with collaboration of KEK, it has been verified that the klystron can generate more than 1.2MW RF.

3.3 The Studies on the Transmutation System

The studies have been conducted to conceptualize a system for transmuting waste targets with design principles of simpleness, reliability and safety. The targets, solid or liquid, and its effective cooling are being investigated. The handling of the target with high radioactivity has to be easy and safe in the operation with the remote system. The estimation of transmutation shows, as shown in Fig. 6, that Sr or Cs targets can be incinerated during 300 days and the created nuclides can be decayed by 400 days' cooling. In this estimation, the long-lived nuclides would be transmuted in short time with less residual waste produced in the target. This means that the photonuclear method is simple in the transmutation. The system should be co-located with the reprocessing plant in order to reduce the risk with transmutation. For continuous and long duration of operation in transmutation, the accelerator as gamma ray source has to be reliable. Thus, the electron accelerator may be suitable for transmutation of fission products due to its simpleness in operation and maintenance.

4. Conclusion

The R&D activities for the transmutation of fission products are in progress in PNC under the OMEGA program. The transmutation methodology is evaluated on the energy balance through the comparison of neutron, proton and photon methods. It is indicated that neutron methods including spallation-neutron method would be available for the transmutation of fission products having large cross sections for neutron such as Tc, and that the photonuclear method would be advantageous for the transmutation of Cs and Sr having so small cross sections for neutron. More studies and experiments are required to evaluate the feasibilities on the transmutation system.

It should be reminded that the OMEGA program is not only intended to seek short-term alternatives for present back-end strategies for nuclear energy system, but also to pursue benefits for future generations through long-term basic R&Ds.

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Table 1 The Comparison of the Energy for Transmutation with Methods

(MeV)

Methods	Cs-137	Tc-99
Spallation(Proton)	570	330
Spallation-Neutron (Proton)	510	35
Muon Catalyzed Fusion	195	40
Photonuclear Reaction (Bremsstrahlung)	4700	-
Photonuclear Reaction (Monochromatic)	400	-

Table 2 Main Specification of PNC Electron LINAC

Max. Beam Energy	10MeV
Max. / Ave. Current	100mA / 20mA
Pulse Length	4ms
Beam Repitition	50Hz
Duty	20%
Average Beam Power	200kW
RF Frequency	1.249135GHz
Length of Microwave	24cm
Mode of Accerelation	$2\pi / 3$
Klystron Power	1.2MW
Total Length of Linac	16m

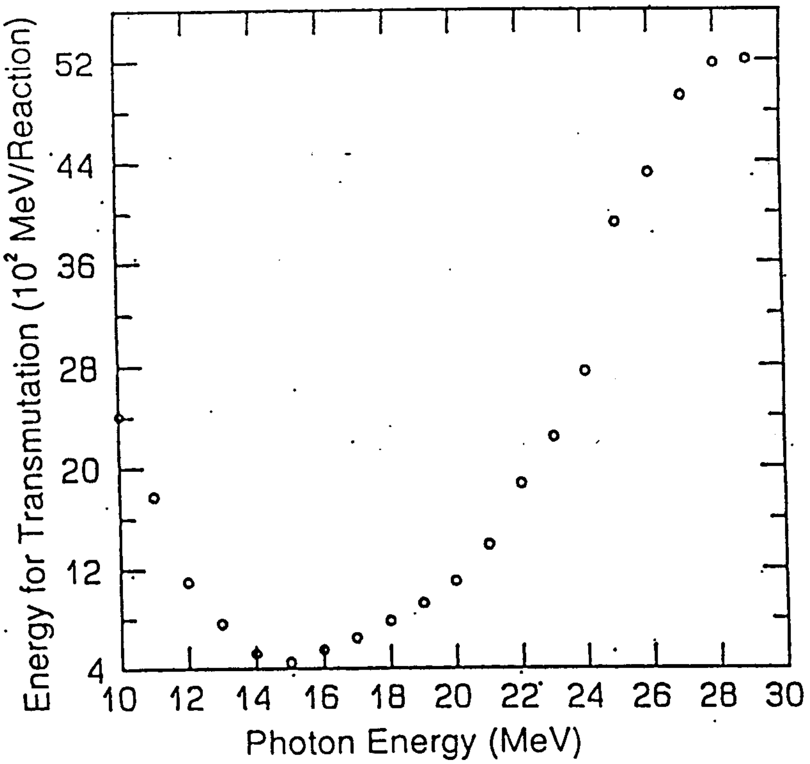


Fig. 3 The Energy for Transmutation of Cs-137 with the Monochromatic γ ray

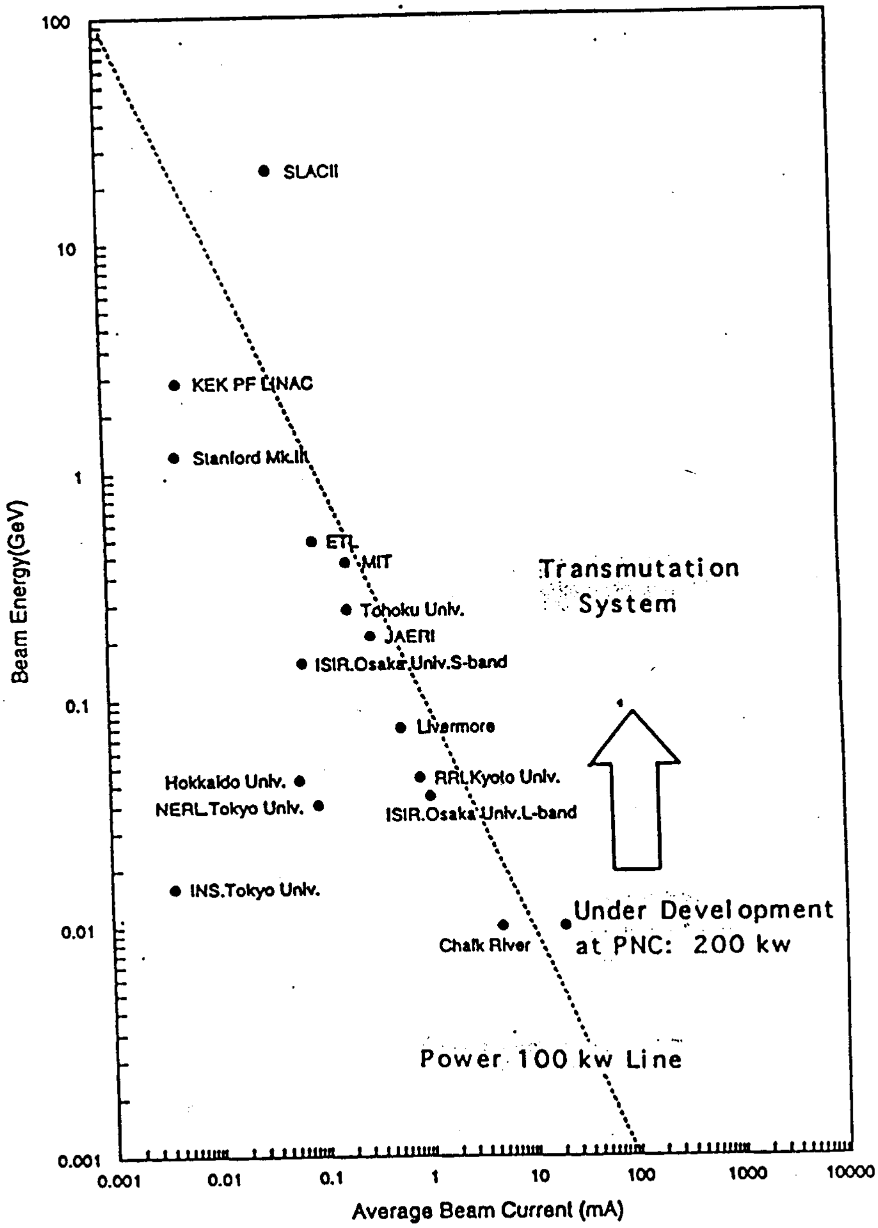


Fig. 4 Electron Linear Accelerator in the World

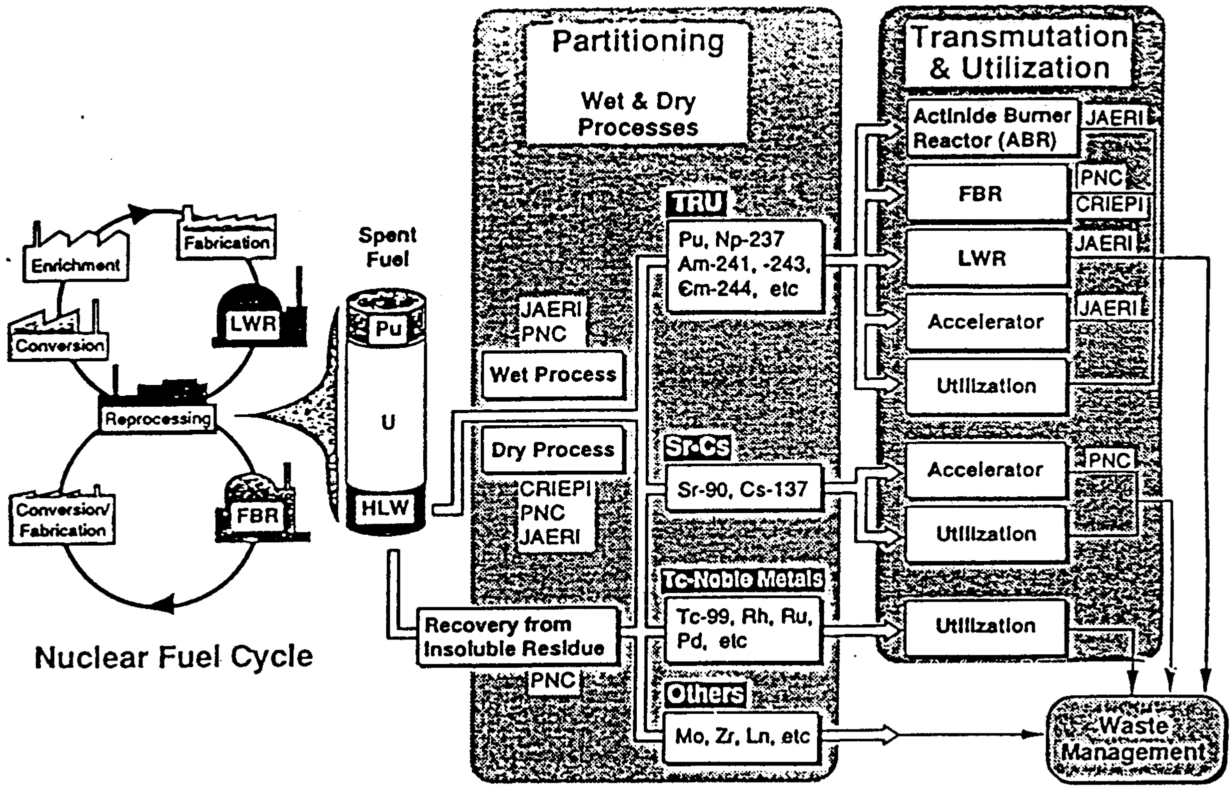


Fig. 1 R&D Activities under OMEGA Program

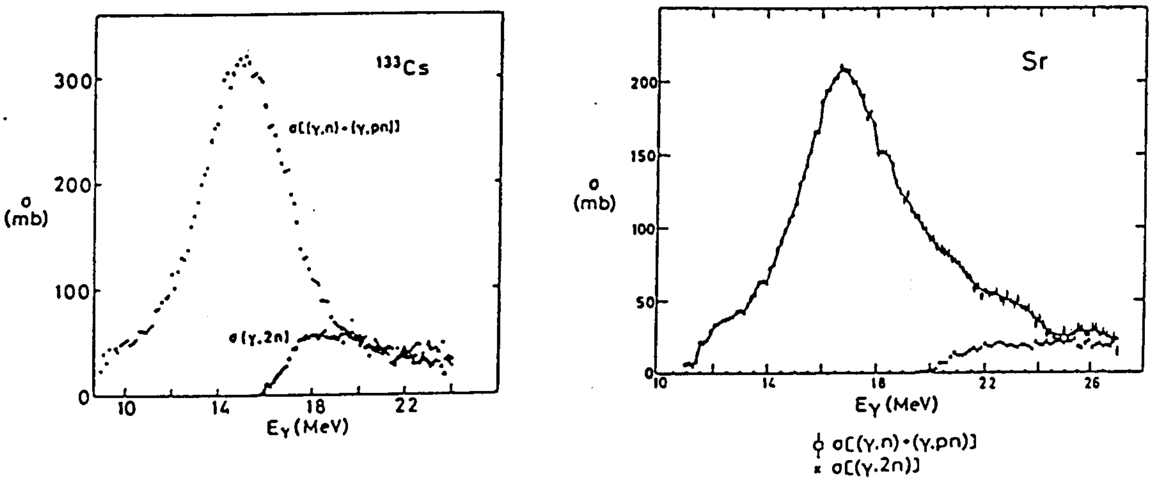
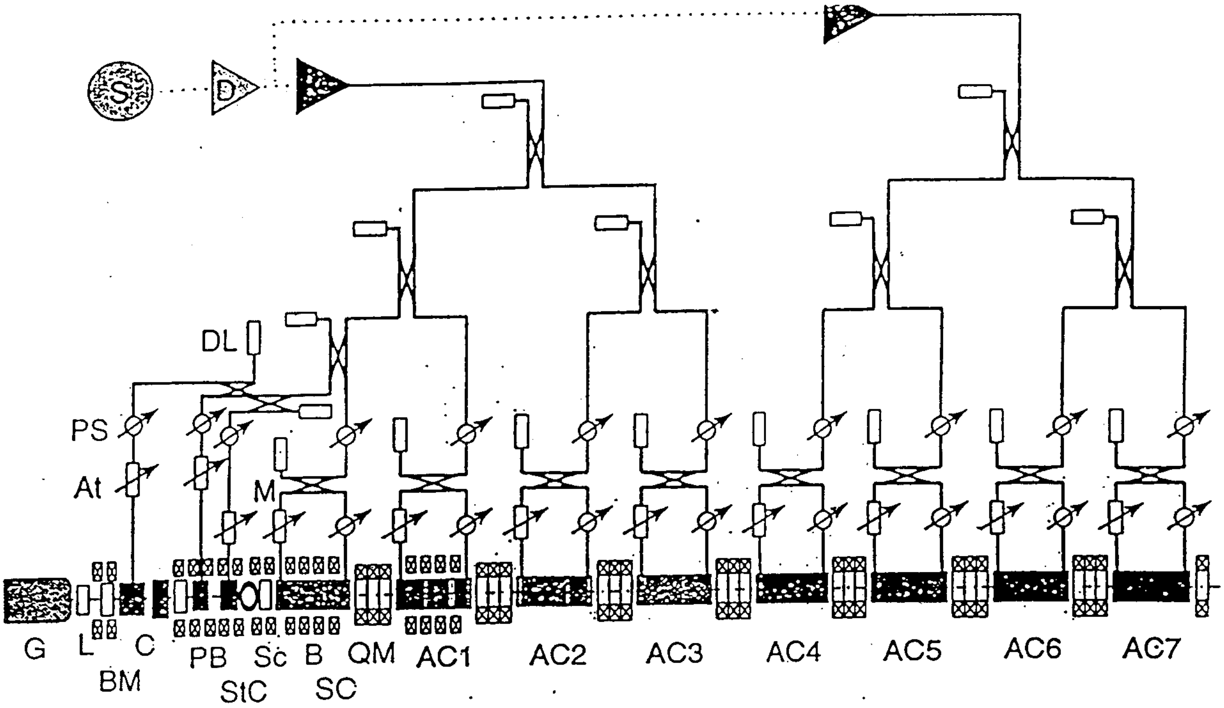
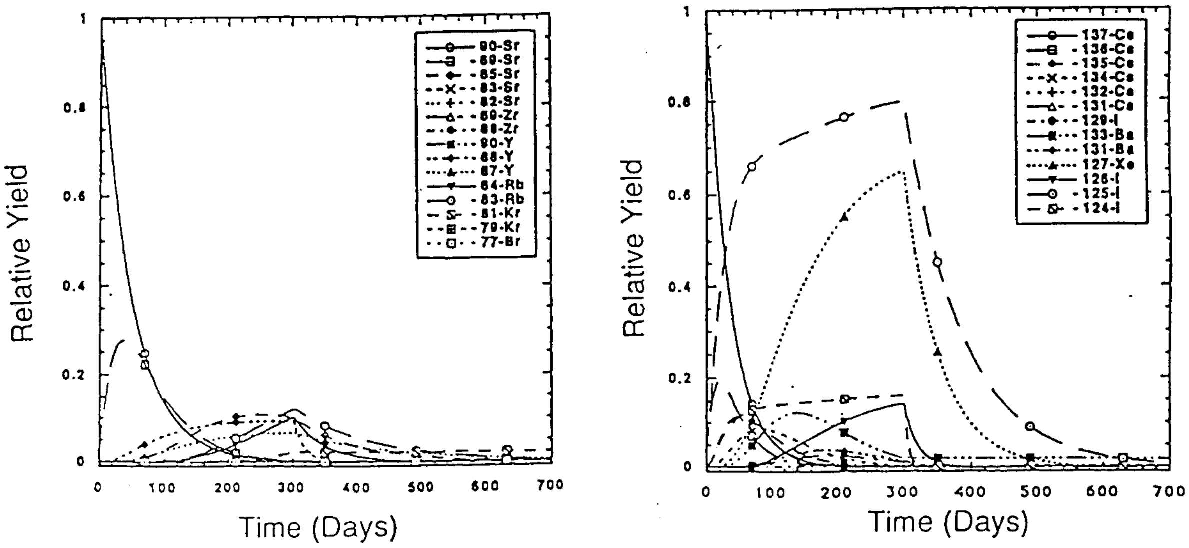


Fig. 2 The Cross Section of Photonuclear Reaction for Cs and Sr (10)



G - Gun L - Lens C - Chopper Cavity and Slit PB - Prebuncher StC - Steeriong Coil Sc - Scraper
B - Buncher Guide QM - Q Magnet SC - Solenold Coil AC - Accelerator Guide BM - Beam Monitor
S - Signal Generator D - Driver K - Klystron PS - phase Shifter At - Attenuator M - Matcher
DL - Dummy Load

Fig. 5 Scheme of CW Electron LINAC of PNC



Target of Nuclides of Sr

Target of Nuclides of Cs

Fig. 6 Transmutation of Cs and Sr Targets by Photonuclear Reaction