Transmutation of Fission Products

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ABSTRACT

Studies of transmutation of long-lived fission products by accelerator and by fission reactor at PNC are described. Four transmutation methods by accelerators are compared in terms of transmutation energy and transmutation rate. A super-powered electron linear accelerator which could transmute a large quantity of fission products has been studied and development of PNC test linac in this line is shown. Fission reactors for transmutation are also explained.

High transmutation rate and small transmutation energy are main requirements for the transmutation of fission products. However, those requirements are difficult to satisfy simultaneously only by accelerator or by fission reactor. Therefore, new transmutation methods which could satisfy both requirements have been also studied at PNC. As the new transmutation methods, moving target method and inertial transmutation method are presented.

1. INTRODUCTION

Long-lived radioactive wastes have been produced and accumulated by the operation of fission reactors. For the time being, one available method for permanent disposal of radioactive wastes is to solidify them into glass and to dispose of them in a stable geologic formation in order to isolate from the human biosphere. Transmutation of radioactive wastes may decrease the radioactivity to be disposed of and hence the risk associated with the disposal of radioactive wastes.

Long-lived nuclides in radioactive wastes are divided into two groups, i. e., fission products and transuranic elements(TRU). Since TRU have a potential of a fission, it has been studied to use the TRU as a fuel in a fission reactor. In this paper, the transmutation of long-lived fission products are discussed.

Accelerator and fission reactor may be applied to the transmutation of fission products, so we have been studying the transmutation of fission products by accelerator and by fission reactor in terms of transmutation rate and energy balance. The transmutation rate of fission products should be several times or more faster than their natural decay rate. The energy balance is not achieved if the transmutation device of fission products consumes more energy than that is originally obtained when the very fission products are produced. Therefore, the energy balance is related to transmutation energy which is the energy required for one transmutation reaction of target nuclide.

Since the fission reactor generates energy itself, the transmutation by fission reactor could meet the energy balance criterion. However, the transmutation rate is low because of its low neutron flux. On the other hand, the transmutation rate by accelerator is higher compared with that by fission reactor because it is possible to create high flux by focusing particle beam. But the energy balance criterion is difficult to be met, since accelerator needs energy from outside to be operated, whereas fission reactor dose not need outside energy to be operated theoretically. Moreover, the quantity of transmuted fission products is small due to the small size of the particle beam and/or the low beam current.

This paper shows comparison of four transmutation methods by accelerators and development of superpowered electron linear accelerator at PNC. Then we explain the fission reactor for transmutation. Recently, we started to study new transmutation methods which could achieve both high transmutation rate and small transmutation energy simultaneously. We will introduce them also in this paper.

2. TRANSMUTATION BY ACCELERATOR

Various methods of transmutation of fission products have been proposed as follows:

(a) transmutation through photonuclear reaction by using bremsstrahlung from accelerated electron (electron method) [l, 2],

(b) transmutation through spallation reaction by high energy proton (proton method) [3, 4],

(c) transmutation through spallation-induced neutron reaction, mostly (n, γ) reaction, by use of secondary neutrons from high energy proton-induced spallation reaction (spallation neutron method) [l, 3] and

(d) transmutation through (n, 2n) reaction by use of high energy neutron generated by muon catalyzed fusion (μCF method) [5, 6].

Above four methods were compared with each other in terms of the transmutation energy and the transmutation rate(effective half-life) by using 3-dimensional Monte Carlo codes. ^{137}Cs was chosen as the target nuclide. The transmutation energies for ^{137}Cs with the effective half-life of 2 years in the four methods are tabulated in Table 1 together with accelerated particles, energies of the particles, particle beam currents and calculation codes which were employed in the calculation.

The transmutation energy was calculated by dividing input energy by the number of transmuted ^{137}Cs. Generally the number of transmuted fission products is proportional to their cross sections for transmutation reactions and the number of beam particles which contribute to the transmutation. Therefore, large cross sections and large number of beam particles which need small energy to be created are favorable for the transmutation energy.

As you can see in Table 1, the μCF method gives the least transmutation energy, and the electron method gives the largest among the four methods. The proton method and the spallation method give almost the same transmutation energy under the condition that the effective half-life of ^{137}Cs is 2 years. Note that the transmutation energy depends on target volume as well as transmutation method employed. We change the target volumes to equalize the effective half-lives in the four methods. Therefore, the target volumes in the four methods in Table 1, which are not mentioned, differ from each other(See Fig. 1).

The electron method uses the giant resonance of the (y, n) reaction to transmute fission products. The y-ray is created as bremsstrahlung from electron. However, since the energy spectrum of the y-rays obtained by bremsstrahlung is continuous, only a small part of the γ-rays contributes to the (y, n) reaction. Furthermore, other reactions such as pair production and Compton scattering are larger than the (y, n) reaction. Thus, most of the γ-rays are lost in these reactions before the transmutation occurs. Those reasons make the energy balance worse in the electron method. If we can make a high intensity y-ray with an exact energy where the resonance (y, n) reaction takes place, the energy balance becomes better[6].

Both the proton method and the spallation neutron method use the spallation reaction. But in the proton method, fission products are hit directly by proton beam, while in the spallation neutron method, they are exposed to neutrons which are produced in Pb target by injecting proton beam. Of course, the secondary neutrons are also utilized for transmutation in the proton method, but most of the fission products are transmuted by protons directly through the spallation reaction. For the transmutation of ^{137}Cs, the proton method is more favorable than the spallation neutron method in terms of the cross section because the cross section for the spallation reaction is larger than that for the (n, γ) reaction. However, about 40 neutrons are produced in Pb target through the spallation reaction by one high energy proton, and those neutrons are used for the transmutation in the spallation neutron method. Therefore, the transmutation energy in the spallation neutron method is smaller than that in the proton method despite the small (n, γ) cross section of ^{137}Cs.

As for the μCF method, muons are created by injecting 25 mA deutron beam with an energy of 4 GeV into a Be target[7]. One muon causes 175 fusions in a deuterium-tritium target with liquid hydrogen density [6, 7], which means that one muon produces 175 neutrons with an energy of 14 MeV. That is the reason for small transmutation energy in the μCF method despite the large input energy to the deutron beam.

We will discuss the energy balance. We estimate the energy which we can use for transmutation of ^{137}Cs to achieve the energy balance. Fission yield of ^{137}Cs is 6% .Released energy during one fission event is about 200 MeV. Suppose thermal to electrical conversion efficiency is 33 %, then the electrical energy produced during one ^{137}Cs generation is 1100 MeV, which is calculated by (200 MeV/0.06)× 0.33. Thus the transmutation energy for ^{137}Cs should not be more than 1100 MeV. If we use more energy than this energy to transmute one ^{137}Cs, we don't actually transmute but produce ^{137}Cs, assuming that transmutation devices are operated by using the energy generated by fission reactors. Actually we should use much less than this energy in order to gain public acceptance for the transmutation. So if it is assumed, for example, that the only 10 % of 1100 MeV energy can be used for transmutation of one ^{137}Cs, the upper limit of electrical energy available for the transmutation is 110 MeV. All the transmutation energies in Table 1 are beyond this energy even in the μCF method. Hence it seems that the transmutation by accelerator is difficult to meet the energy balance criterion.

In Fig 1, effective half-lives of ^{137}Cs in the four methods are shown as a function of the target volume. This figure indicates that the effective half-life increases with increasing target volume. This is because the increase of the target volume is greater than that of the number of transmutation reactions. Effective half-life depends on the conditions such as beam energy and beam current. However, according to various papers on the transmutation by accelerator [1-7] and also this result, we might say that the order of the effective half-life of ^{137}Cs is in several years.

3. DEVELOPMENT OF SUPER-POWERED ACCELERATOR

To transmute a large quantity of radioactive wastes by accelerator, high intensity beam is needed. PNC started development of a super-powered electron linear accelerator. Our goal of the development is to construct a CW(continuous wave) accelerator whose maximum beam energy and time-averaged beam current is 100 MeV and 100 mA, respectively. To approach this goal, we are now constructing a test electron linac which accelerates 10MeV-100mA electrons with duty factor of 20 %. Fig. 2 presents the mapping of electron power of accelerators in the world. The main specifications of our test linac are summarized in Table 2[8]. Fig. 3 and Fig. 4 show the block diagram and the bird's-eye view of the test linac, respectively. At present, our study of the linac is focused on the development of klystron and accelerating tube for high power RF.

The klystron to be developed has output power of 1.2 MW and RF frequency of 1.25 GHz. In order to obtain this high power, developments of electron gun, collector and output window are needed for stable operation of the accelerator. High conversion efficiency from electricity to RF is also important.

Between 1989 and 1990, the first trial klystron was manufactured, which was subjected to its performance test. The test revealed the total efficiency of 62 %[9]. In 1990, the second trial klystron was manufactured based on the data obtained by the test of the first trial klystron. The first CW mode operation was carried out with this second trial klystron. The maximum RF output power of 330 kW was obtained[9]. This value is about a quarter of the target value. The lower output is due to the higher temperature of RF output window than predicted in the design. Therefore, improvement of the window will be carried out.

Good energy transfer efficiency from RF to beam is also of vital importance for our test linac. For this purpose, accelerating tube under development is one of traveling wave type which has a resonant ring, since this type of accelerating tube has a potential for higher energy transfer efficiency. Thermal stress by microwave whose power amounts to 300 kW per accelerating tube causes a deformation of cavity structure. To avoid the deformation, an effective cooling system has been studied by some calculations and experiments.

Beam break-up(BBU) problem is expected in the operation of a high current accelerator. So it is necessary to study the problem of induced higher electric modes caused by beam-RF interaction. Some calculations are carried out for the BBU problem.

4. TRANSMUTATION BY FISSION REACTOR

Fission reactors for transmutation are also studied at PNC. We have studied the transmutation of ^{137}Cs in fission reactors which we call transmutation reactors[10]. Fig. 5 shows the concept. The transmutation reactor is a high-flux fast reactor which has a Cs region inside the core. The Cs region is surrounded by the fuel and the reflector. In the Cs region, ^{137}Cs is loaded in the chemical form of deuteroxide $^{137}CsOD$, because the density of this form is higher than that of metallic form, and furthermore the deuterium(D) plays a role as moderator. In the fuel region, solid fuel is loaded. Taube studied a transmutation reactor for the transmutation of ^{90}Sr and ^{137}Cs[11] For that reactor he proposed a liquid fuel to obtain a high power density which leads to a high neutron flux. However, we use a solid fuel which is more realistic than the liquid fuel.

Fast neutrons generated in the fuel region are thermalized due to mainly deuterium in the Cs region, and the thermalized neutrons are used to transmute ^{137}Cs, since the neutron capture cross section of ^{137}Cs is approximately one order of magnitude larger for thermal neutron than for fast neutron. Nevertheless, the thermal neutron capture cross section of ^{137}Cs is very small (- 0.25b)[12], and therefore high thermal neutron flux is necessary to transmute it effectively. It is desirable that the Cs region has the thermal neutron flux level of 10^{16} n/cm2s, but it is very difficult to attain this flux level within conditions of an ordinary fast reactor such as power density, burnup reactivity and power peaking[10].

Since the transmutation reactor is a fission reactor, it produces ^{137}Cs itself along with transmuting ^{137}Cs. If the transmutation reactor produces more amount of ^{137}Cs than that is transmuted, it is nonsense. Therefore, the transmutation reactor is required to transmute a large quantity of fission products, at least, more than the quantity produced in it. Changing parameters such as power density and volume of Cs region, we calculate the quantity of transmuted ^{137}Cs, the transmutation rate, and so on. As for the transmutation rate in which the produced ^{137}Cs in the transmutation reactor is taken into account, the transmutation reactor with power density of 500 W/cc, burnup reactivity of 3 $\%\Delta k$ and power peaking of 1.7 can transmute only a few percent of loaded ^{137}Cs per a year[10]. The result indicates the difficulty of effective transmutation of ^{137}Cs only by fission reactor with a solid fuel. If a special fuel such as particle fuel and liquid fuel which could achieve high power density is used, the transmutation may become more efficient[1 1, 13].

5. NEW TRANSMUTATION METHODS

It seems that the transmutation methods by accelerator and by fission reactor which have been studied so far have difficulty in satisfying the requirements for the transmutation rate and the transmutation energy simultaneously. So we have been also studying new transmutation methods which might achieve both high transmutation rate and small transmutation energy at the same time. We introduce moving target method first and then inertial transmutation method.

5.1 Moving target method

Moving target method is based on the resonant neutron capture of fission products. Neutron capture cross sections of fission products generally have resonance regions at certain energies and some of them present large values at some resonance energies. Fig. 6 shows the neutron capture cross section of ^{99}Tc[14]. It can be seen that there is a very sharp resonance peak at neutron energy of about 5.6 eV. The maximum peak is about 7000 b. If strong beam of neutrons with this energy can be produced, high transmutation rate might be expected. But it is difficult to produce monochromatic or highly controlled neutron beam.

Instead of accelerating neutrons, we accelerate the target nuclei to have the resonance reaction with thermal neutrons in the moving target method[15]. Since the total momentum is conserved in any coordinate system without external forces, the neutron energy can be controlled in a relative manner to match the resonance peak in the laboratory frame where target nucleus stands still. The concept is illustrated in Fig. 7. Physical phenomena do not depend on inertial frames. If the resonance reaction of the target with neutron is observed in the target rest frame, it can be also observed in the neutron rest frame. The only difference is the resonance energy of neutron capture observed in the two frames. The resonance energy, E_{res}, in the target rest frame is observed as

$$E_{res} = \frac{M}{m} E_{res} \quad \cdots\cdots\cdots\cdots\cdots\cdots \quad (1)$$

in the neutron rest frame, where m is neutron mass and M is the mass of the target nucleus. In the case of ^{99}Tc ,the resonance neutron capture takes place at neutron energy of about 5.6 eV in the target(^{99}Tc) rest frame, which is converted to target energy of about 550 keV in the neutron rest frame. In other words, ^{99}Tc must be accelerated to the energy of 550 keV to have the resonance reaction with thermal neutron.

Transmutation device in the moving target method is shown in Fig. 8. Target FP atoms are ionized and accelerated to the resonance energy, and then they are circulated in a magnetic field. However, if each FP atom is ionized, only a small number of FP atoms can be circulated in a realistic magnetic field due to the repulsive electric force between the FP atoms. But it is necessary to circulate a large number of FP atoms in order to transmute fission products in large quantities. Therefore, microparticles made of FP atoms, which we call matrons, are circulated instead of individual FP atoms. Matrons are produced and charged in a matron source[16, 17]. After the distribution of particle size is controlled, the matrons are accelerated and circulated in the magnetic field. During the circulation, the matrons are exposed to the thermal neutrons which are produced by fission reactor or accelerator, and the resonance reactions are brought about.

Parameters of the device for the transmutation of ^{99}Tc are summarized in Table 3. The matron radius is 0.01 μm. The each matron contains 2.93X 105 atoms, and its charge number is 293, which means that one atom per 1000 atoms is charged. One matron can be circulated with the radius of 3.4 m under the magnetic field of 10 T. However, the target of 0.5 mol(-1018 matrons) must be circulated for about 14 hours to achieve an effective transmutation[15]. This poses a rather tough confinement problem. Although confining the matrons with 0.5 mol atoms is easier compared to confining the same number of ionized atoms, the confinement force has to be still very strong. Hence, neutralization of matron charge by electrons is needed to reduce the confinement force.

The device can transmute an amount of ^{99}Tc produced by a fission reactor of 1 GW_e with an effective half-life of 14 hours. Note that the natural half-life of ^{99}Tc is 2.1X 105 years. This moving target method provides high transmutation rate. But further studies such as effective confinement of matrons are still necessary to optimize the device.

5.2 Inertial transmutation method

Another new approach we are studying is to utilize inertial confinement fusion. In this method, fission products are placed outside the deuterium(D)-tritium(T) core, forming FP shell as can be seen in Fig. 9. They are compressed together with the DT core by laser or particle beam, and irradiated by 14 MeV neutrons produced by DT fusion inside the pellet.

Fission products are transmuted mainly through (n, 2n) reaction. For ^{90}Sr and ^{137}Cs, (n, 2n) cross sections at neutron energy of 14 MeV are 100 and 10 times, respectively, larger than (n, y) cross sections at thermal neutron energy. Therefore, the (n, 2n) reaction is more favorable than the (n, y) reaction for transmutation of such nuclides which have small neutron capture cross sections. Furthermore, compressing the FP shell leads to the enhancement of (n, 2n) reaction due to high density. As a result, high transmutation rate can be expected in this method.

A simple calculation indicates the enhancement of the reaction. Suppose that a neutron with energy of 14 MeV is produced in the center of the DT core, the probability P_0 that the neutron reacts with FP in the FP shell is given as

$$P_0 = 1 - \exp(-n\sigma d). \cdots\cdots\cdots\cdots \tag{2}$$

Here n, σ and d are defined as number density of the FP, (n, 2n) cross section of the FP, and thickness of the FP shell, respectively. Since the (n, 2n) reaction is dominant in this case and the thickness of the FP shell is very thin (the order of 100 pm), multiple scattering of neutron and (n, y) reaction are omitted in eq. (2). If the pellet is compressed with a ratio of 1000, the number density n becomes 1000 times larger than that before compression and the thickness d becomes d/1 O, which leads eq. (2) to

$$P = 1- \exp(-100n\sigma d). \cdots\cdots\cdots\cdots\cdots\cdots \quad (3)$$

For instance, we assume that the FP is ^{90}Sr and the thickness of FP shell is 500 μm, then, using $n=1.7\times10^{22}cm^{-3}$ and σ=1 .7b, we can calculate the ratio of the reaction probability after compression to that before as

$$\frac{P}{P_0} \cong 93. \cdots\cdots\cdots\cdots\cdots\cdots \quad (4)$$

Thus we might obtain high transmutation rate in this inertial transmutation method.

Fig. 10 shows effective half-life of ^{90}Sr as a function of the thickness of Sr region(FP shell) before compression. It is assumed that the compression ratio is 1000 and the radius of DT core before compression is 500 μm. From this figure, the effective half-life can be seen in the order of minute, which means that very high transmutation rate can be obtained in this method.

Transmutation energy for ^{90}Sr is shown in Fig. 11 as a function of the thickness of Sr region. This calculation is based on a simple statistical model[18]. Compression ratio of 1000 and DT core radius of 500 μm are assumed again. It is also assumed that the temperature of DT core after compressions is 10 keV which is demanded in order to get the energy gain by the inertial confinement fusion[19]. The temperature of Sr region is determined as 1.7 keV from the assumption of pressure balance between the DT core and the Sr region after compression. In this calculation, we neglect the conversion efficiencies from input energy to laser or particle beam energy and from the laser or the particle beam energy to the internal energy of pellet. We also neglect the effect of a-heating. If those efficiencies and α-heating are not taken into consideration, small transmutation energy in the order of 10 MeV can be obtained in this method. At this stage, the inertial transmutation method meets the criterion on the energy balance as well as on the transmutation rate.

The inertial transmutation method is very attractive because of not only its uniqueness of approach (for example, it incorporates the effect of compression) but also the high transmutation rate and the possibility of small transmutation energy. But the following problems are yet to be investigated;

* Possibility of compression of pellet which contains fission products,

* Interaction of laser or particle beam with fission products,

* Manufacturing process of the pellet, and so on.

6. CONCLUSION

Studies of transmutation of fission products at PNC have been discussed. First, four transmutation methods by accelerator were compared in terms of transmutation energy and effective half-life for ^{137}Cs. It was found from this comparison that the transmutation energy was the least in the μCF method and the largest in the electron method among the four methods discussed here. But the difficulty in constructing the transmutation devices for these methods may be the opposite, namely, the device in the μCF method may be the most difficult to be constructed. As for effective half-lives of ^{137}Cs in the four methods, all of them were calculated to be in the order of several years.

A problem with transmutation by accelerator is that it is difficult to transmute a large quantity of fission products by the present accelerators because of their small beam currents. To transmute a great deal of fission products, high power accelerator is needed. As one of such accelerators, we have been developing the super-powered electron linear accelerator at PNC. As a first step, we are developing a 10 MeV-100mA test electron linac, especially, high power klystron and accelerating tube for high beam current. The klystron achieved its maximum output power of 330 kW. The accelerating tube with resonant ring is under construction.

Concept of the transmutation reactor was described. The transmutation reactor has ability to transmute a large quantity of fission products. However, the transmutation rate is low for fission products whose thermal neutron capture cross sections are small, since a very high flux of thermal neutrons is difficult to obtain. Increasing power density of the transmutation reactor is the most efficient way to increase the neutron flux, but high power density brings a problem of removal of heat. Present technology for the heat removal restricts the power density, which leads to the difficulty in producing high neutron flux. Therefore, it may be impossible to obtain a high transmutation rate only by using a fission reactor.

Finally, two new transmutation methods were discussed. One was the moving target method, and the other was the inertial transmutation method. The moving target method uses the resonance reaction between fission product and neutron. In this method, matrons made of FP atoms, instead of the neutrons, are accelerated to have the resonance reaction with thermal neutrons. A device of this method was described and parameters of the matron were also given. Confinement of the matrons is a serious problem. In general, the resonance reaction has a large cross section. Hence the moving target method gives a very high transmutation rate. But this concept is quite new and much investigation is yet to be done.

On the other hand, the inertial transmutation method utilizes the inertial confinement fusion. In this method, pellet which contains fission products is compressed, and the fission products are transmuted by high energy neutrons produced by the DT fusion inside the pellet. It was found that this method as well as the moving target method gave a high transmutation rate owing to the effect of the compression. Small transmutation energy was also obtained. But in the calculation of the transmutation energy, a simple statistical model was used and the conversion efficiencies were not taken into account. More exact calculation should be done for further study.

Concerning the moving target method and the inertial transmutation method, they have a potential to satisfy both high transmutation rate and small transmutation energy at the same time. But they are in a preliminary study phase, and much studies are necessary to establish both methods.

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Table 1. Transmutation energy for ^{137}Cs

Effective half-life : 2 years
Target : cylinder

Transmutation method	Accelerated particle	Energy [MeV]	Current [mA]	Calculation code	Transmutation energy [MeV]
Electron method	e-	100	2000	EGS4	4700
Proton method	p	500	900	NMTC	570
Spallation neutron method	p	1500	300	NMTC+MCNP	510
μCF method	d	4000	25	MCNP	200

Table 2. Main Specifications of PNC Test Linac

Beam Energy	10 MeV
Maximum Beam Current	100 mA
Average Beam Current	20 mA
Pulse Length	4 ms
Pulse Repetition	50 Hz
Duty Factor	20 %
Output Beam Power	200 kW
RF Frequency	1249 MHz
Total Length of Accelerator	16 m
Klystron Output Power	1.0 MW

Table 3. Parameters of Device for Transmutation of ^{99}Tc in Moving Target Method

Matron Source	
Matron Radius	0.01 μm
Number of Atoms in Matron	2.93×10^{5}
Charge Number	293
Electric Field on Surface	4.2×10^{9} V/m
Acceleration Voltage	550 kV
Total Current	0.682 A
Storage	
Total Amount of Target	0.5 mol
Confinement Time	14 hr
Thickness of Target	20 cm
Matron Density	3.4×10^{11} cm^{-3}
Radius of Gyration	3.4 m (for 10T)
Length of Target Region	70.5 cm (for 10T)
Neutron Field	
Flux	10^{15} cm^{-2}sec^{-1}
Energy	Thermal

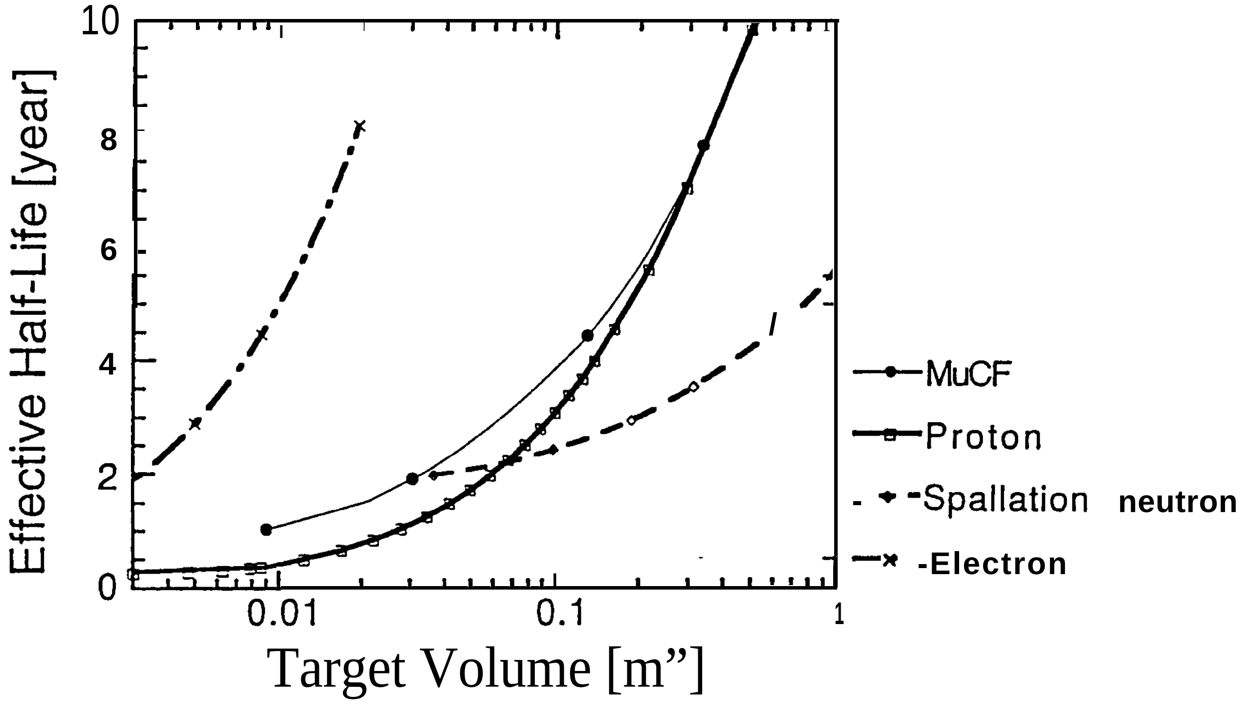


Fig. 1 Effective Half-Life of ^{137}Cs.

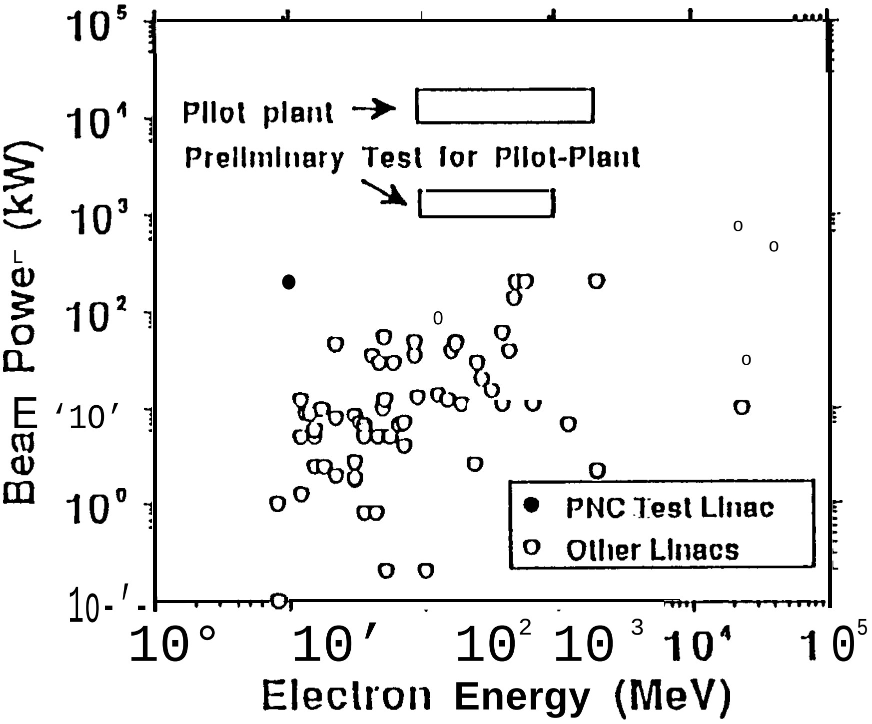


Fig. 2 Map of Electron Linacs.

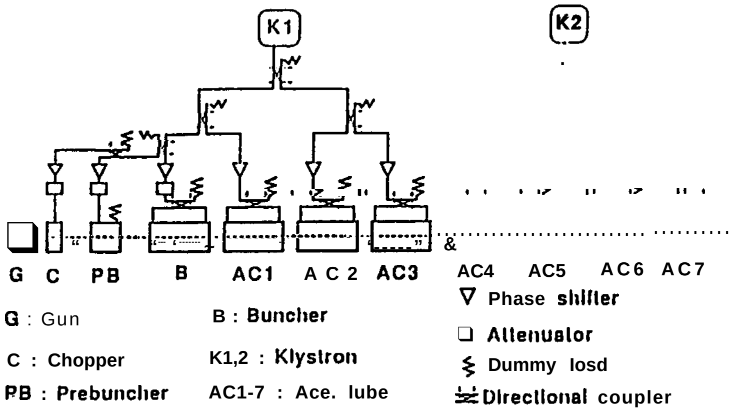


Fig. 3 Block Diagram of PNC Test Linac.

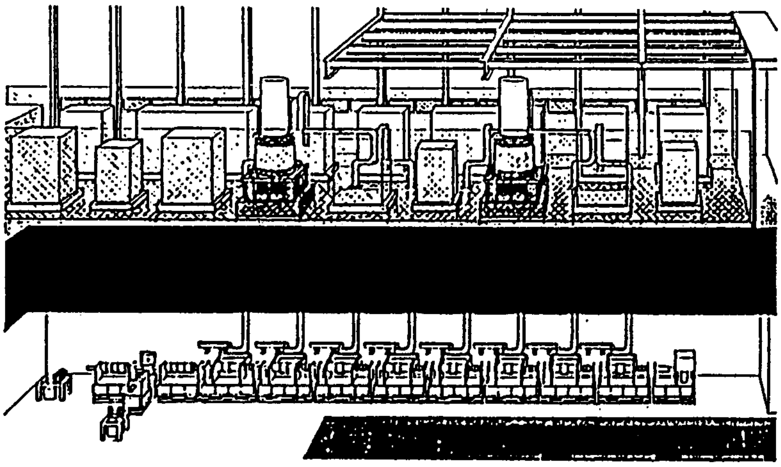


Fig. 4 Bird's-eye View of PNC Test Linac.

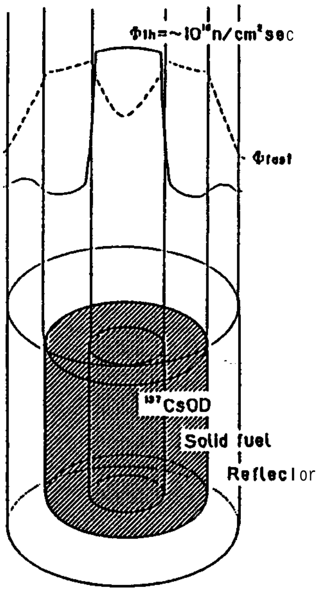


Fig. 5 Transmutation Reactor.

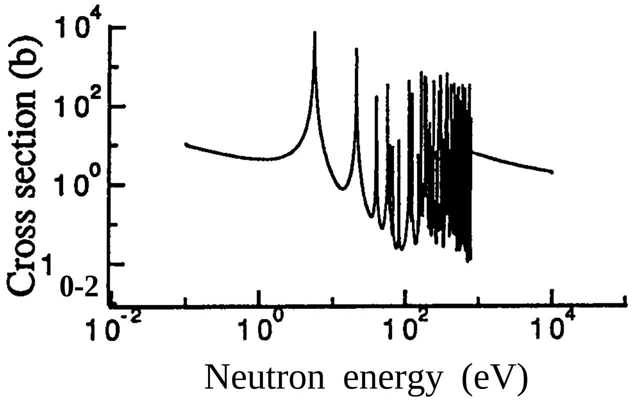
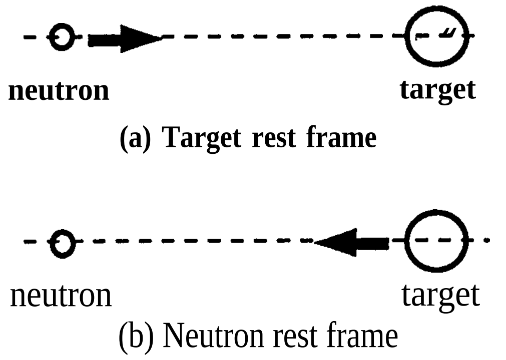


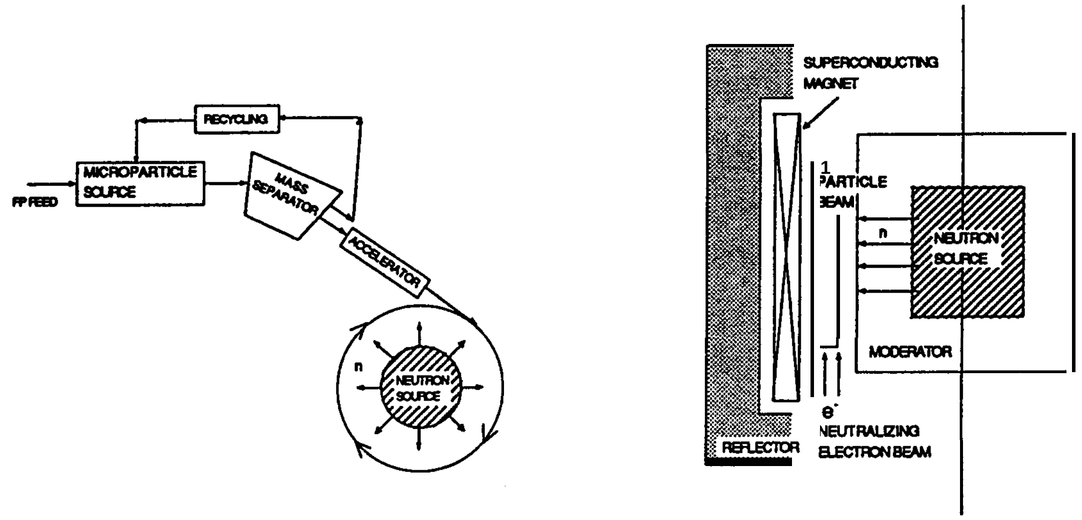
Fig. 6 Neutron Capture Cross Section of ^{99}Tc.



(a) Target rest frame

(b) Neutron rest frame

Fig. 7 Concept of Moving Target Method.



(a) total system

(b) reaction vessel

Fig. *8* Transmutation Device in **Moving** Target Method.

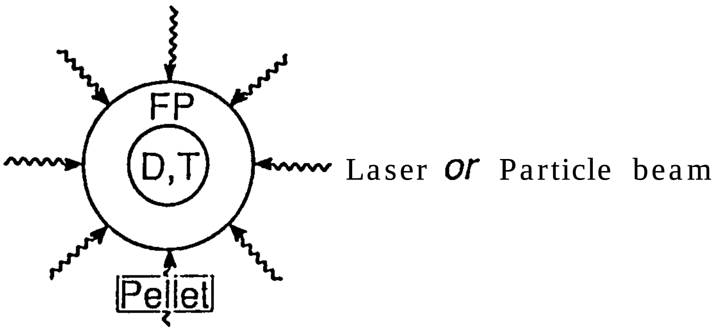


Fig. 9 Concept of Inertial Transmutation Method.

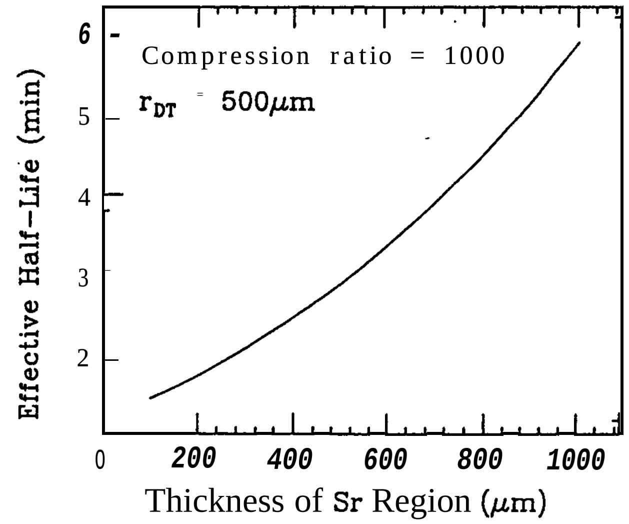


Fig. 10 Effective Half-Life of ^{90}Sr.
(Compression ratio=1 000, DT core radius=500μm)

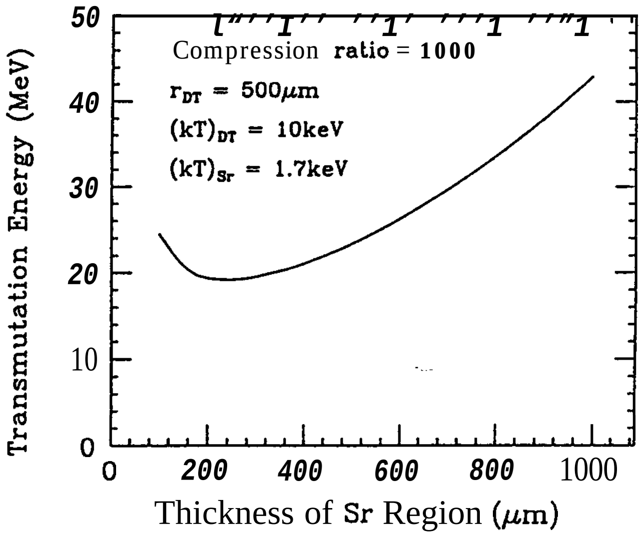


Fig. 11 Transmutation Energy for ^{90}Sr.
(Compression ratio=1000, DT core radius=500μm,
DT temperature=10keV, Sr temperature=1.7keV)