Transmutation of Fission Products and Transuranium by High Energy Neutron

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Abstract

We discuss the feasibility of the system that incinerate radioactive fission products (FP) and transuranium (TRU) by using high energy neutrons. As high energy neutron sources, μCF reaction, fusion reaction, and spallation reaction were investigated.

In the system that utilizes μCF reaction, a subcritical core made of FP and TRU is bombarded by 14 MeV neutron generated via μCF reaction. Monte Carlo neutron transportation code (MCNP) and evaluated nuclear data (ENDF/B-VI) were used to simulate neutron

transportation in the core. To generate μ- mesons, a 4 GeV-25 mA deutron accelerator is used. The results of the simulation show that the driver energy can be completely supplied by transferring fission energy to electric energy; to operate this system, no energy is required.

As the system that utilizes an inertial fusion reaction, two types of target were investigated. The first type uses a spherical wall, thickness about 20 cm and radius 50 cm, composed of ^{90}Sr, ^{90}SrO, ^{137}Cs, or $^{137}Cs_2O_2$. The target is bombarded by 14 MeV neutrons generated by inertial confinement fusion at the center of the wall. Neutronics in the wall were simulated to estimate the probability of neutron utilization per a generated neutron. The second type uses a micro target that is composed of DT fuel and ^{90}Sr. An analytic model of the implosion of the target was formulated to evaluate the energy that is required to transmute radioactive waste (incineration energy).

In the system that utilizes a spallation reaction, damages of structural material is a severe problem. To estimate the damage of structural material, atomic displacement, H production, He production, and energy deposition were calculated using computer codes, LAHET and HTAPE.

Introduction

Methods of incinerating radioactive wastes, fission product (FP) and transuraniums (TRU), have been vigorously investigated in the last decade. [(1),(2),(3)] The use of the neutron capture reaction and the fission reaction have received the greatest attention, as ways to incinerate FP and TRU, respectively.

Recent measurements showed the thermal neutron-capture cross-sections of ^{90}Sr and ^{137}Cs were 15.3(4) mb and 0.25 b[5], respectively. If a fission reactor, similar to that proposed by Taube[1], was used to transmute these nuclei, with the requirement of an incineration half-life of 2 years, the thermal neutron-flux in the device would have to be as high as 6.5 x 10^{17} and 4.0 x 10^{16} n/cm^2/sec for ^{90}Sr and ^{137}Cs, respectively. Such a high flux is beyond the technology of present reactors.

To overcome this difficulty, we have investigate the incineration systems utilizing high energy neutron, especially 14 MeV neutrons. High incineration rate can be expected because the (n,2n) cross-section is typically 2 barn for fission products at 14 MeV neutrons.

As the high energy neutron sources, μCF reaction, fusion reaction, and spallation reaction were investigated. To evaluate the feasibility of these systems as an incinerator, we calculated the neutronics of the systems. Incineration half-life and incineration energy were quantitatively calculated.

Section A Incineration of ^{90}Sr and ^{137}Cs by Inertial Fusion

This section discusses a system of nuclear transmutation in which ^{90}Sr or ^{137}Cs is incinerated using 14 MeV neutrons produced by inertial fusion. The dimensions of the incineration system using inertial fusion reactor could be smaller than that using tokamak fusion reactor. This makes it possible to achieve high incineration rate.

Fig. A-1 shows the cross section of the target. The 14 MeV neutrons produced by DT fusion reaction at the center are used to trigger incineration reactions. The radius of the central void was chosen as 50 cm to achieve high incineration rate. The cell of ^{137}Cs is made of Cs metal with the density of 1.87 g/cm^3 or Cs_2O_2 with the density of 4.25 g/cm^3. The inventory of ^{137}Cs is 4.18 x 10^3 kg in both cases. The cell of ^{90}Sr is made of Sr metal with the density of 2.6 g/cm^3 or SrO with the density of 4.7 g/cm^3. The inventory of ^{90}Sr is 2.75x 10^3 kg; in this case the number of ^{90}Sr is the same as the number of ^{137}Cs. A carbon wall with the thickness of 2 cm or 20 cm was used as a refractor. The neutronics in the target was simulated by the Monte Carlo neutron-transportation code (MCNP).[6]

Column 3 in Table A-I shows the probabilities of the (n,2n) and (n,γ) reactions for ^{90}Sr per an input of a 14 MeV neutron. About 50% of an incident neutron is used in incineration reactions. In contrast with ^{137}Cs, the probability of neutron utilization do not increase so much. The contrast could be explained with the difference of neutron-capture cross-section between ^{90}Sr and ^{137}Cs; the neutron-capture cross-section of ^{90}Sr for thermal neutron is 0.015 barn[4] and that of ^{137}Cs is 0.25 barn.[5]

If 1 GW_{th} fusion reactor was used, the generation rate of 14 MeV neutrons would be 2.5 x 10^{20} n/see. Column 3 in Table A-III shows the calculations of the incineration half-life, where 1 GW_{th} fusion reactor was used as a generator of 14 MeV neutrons. In this case, the incineration half-life of 2 - 3 years is achieved.

Column 4 in Table A-III shows the calculations of the incineration energy, where a total gain factor of 10 was assumed. In this case, a 14 MeV neutron can be created with electric energy of 1.76 MeV. Electric energy of 2- 4 MeV is required to incinerate one ^{90}Sr or ^{137}Cs.

Column 5 in Table A-III shows reduction amounts by incineration per year for ^{90}Sr and

^{137}Cs, where 1 GW_{th} fusion reactor was used as an incinerator as well as Column 3. About 500 - 1,300 kg of ^{90}Sr or ^{137}Cs could be reduced by the system.

Section B Incineration of ^{90}Sr by an Inertial Fusion Target

This section discusses the inertial confinement fusion system as a transmutator of ^{90}Sr. The target of this system is composed of DT fuel, with fission product around the fuel. After compression of the target, a 14 MeV neutron produced by DT reaction can be used to transmute the highly compressed fission product via (n,2n) reaction. The neutron reaction rate could be very high because the target is highly compressed. Furthermore, the inventory of fission product could be very small. An analytic model of the implosion of the target was formulated to evaluate its internal energy and the probability of neutron utilization. From the results of this calculation, we could evaluate the energy that is required to transmute ^{90}Sr.

Fig. B-1 depicts a target design used in our analytic model where a spherically shaped DT fuel (radius r_1) is surrounded by a spherical shell (outer radius r_2) made of ^{90}Sr. In the formula of the analytic model, ionized electronics in the compressed target were treated as a Fermi-Dirac gas because a high degeneracy effect was expected, especially for Sr that could be highly ionized. The detail of our model will be published elsewhere.

In Fig. B-2, incineration energy, E, was plotted as a function of internal energy of the compressed target, U. In this calculation, the temperature of DT fuel and that of the ^{90}Sr after compression is 10 keV and 1 keV, respectively, and the compression ration, K, of DT fuel is varied as $10^3, 10^4, 10^5$. The numbers in the two-line parentheses show the gain factor G (upper) and the annual transmutation value for ^{90}Sr. The gain factor, G, is defined as the output energy of DT reaction divided by the internal energy of a compressed target. The annual transmutation value was deduced by assuming 1 GW_{th} fusion reactor.

The ratio of U^{FP} to U^{DT} is illustrated in Fig. B-3 to show how the compression of the DT-FP combined target is difficult compared to that of a simple DT target. In this calculation, the number of ^{90}Sr was set as equal to the number of triton and deutron pairs. Three curves show the ratio of the internal energy of ^{90}Sr to that of DT fuel, where the corresponding temperature of the ^{90}Sr are written alongside the curves, 10 keV, 3 keV, and 1 keV, respectively. The temperature of DT fuel was set as 10 keV.

Section C Incineration of Fission Product and Transuranium by μCF

This section discusses a system of nuclear transmutation in which fission products (FP) and transuranium (TRU) are incinerated using 14 MeV neutrons produced by muon-catalyzed fusion (μCF) and a subcritical core composed of FP and TRU. High generation rate of 14 MeV neutrons can be obtained because the repetition number of DT fusion reaction per one μ- is large. Fig. C-1 is a conceptual picture of the system and Fig. C-2 shows the r-z cross-section of the subcritical core. This hybrid core, composed of FP and TRU, has the advantage that the output of thermal energy by the fission reaction could be used as the input of electric energy to the driver that produces μ- mesons. We adapted the He-cooled particle fuel concept designed by M. Todosow et al.[7], which is shown in Figs. C-3a, b, and c; this fuel has a high efficiency of heat transfer because of its large surface-to-volume ratio.

The 14 MeV neutrons produced by μCF are used to transmute ^{90}Sr by the (n,2n) reaction. The outcoming neutrons from the ^{90}Sr cell transmute TRU through fission reactions, and ^{99}Tc through (n,γ) reactions. This fission energy is converted into electric energy to supply 4 GeV - 25 mA Deutron beam power, which is used to produce μ- mesons. We also evaluated the production of tritium that is consumed as a fuel for μCF. The neutronics in the system was analyzed by MCNP.

Table C-I shows the probability of the (n,2n), (n,γ), and (n,f) reactions for each nuclide in the core per an input of a 14 MeV neutron. The incineration half-life for each nuclide was calculated from these quantities, where a generation rate of a 14 MeV neutron was 3.1 x 10^{19} n/see.') The calculations of the incineration half-lives for each nuclide are shown in Column 7 of Table C-I. The incineration half-lives of ^{90}Sr, ^{99}Tc, and TRU are 1.6 years, 1.6 years, and 0.6 years, respectively.

In the core shown in Fig. C-2, the number of fission reactions is 1.08 per an input of 14 MeV neutrons, which corresponds to thermal energy of 210 MeV. This energy can be converted into electric energy of 70 MeV when the efficiency of thermal-to-electric conversion is 1/3. Because the electric energy required to generate one 14 MeV neutron is 64 MeV, this system is energetically complete and closed within itself.

Table C-II shows the amount transmuted in this system each year for each nuclide. The incineration amounts of ^{90}Sr, ^{99}Tc, and TRU are 40 kg, 96 kg, and 245 kg per year, respectively.

For the core design shown in Fig. C-2, the probability of T production was 0.98 per an input of 14 MeV neutron; therefore, almost all the T consumed by μCF reaction can be supplied within the system.

Fig. C-4 shows the multiplication factor of the core, k_{eff}, as a function of the inventory of TRU. The core dimensions in Fig. C-2 have a k_{eff} value of 0.68, which is much smaller than a critical k_{eff} value of 1.0.

Section D Damage of Structural Material by Spallation Neutron

This section discusses a darnage of structural material of an incinerator that utilizes a spallation reaction. Fig. D-1 shows the cross section of the target, which is composed of a lead target (cell #3-6), a TRU cell (cell #l), and structural walls (Cell #7-13). The structural walls is stainless-steel with thickness of 0.5 cm or 2 cm. By bombarding a lead target with a 1 GeV - 10 mA proton beam, high energy neutrons and charged-particles are produced.

Computer codes [9] of LAHET and HTAPE were used to estimate the damages of structural material. Table D-I shows the calculations: neutron flux, atomic displacement, Hydrogen production, Helium production, and energy disposition. The rate of the atomic displacement in the side wall (cell #10) is as large as 50% of that in the beam window (cell #7).

Acknowledgement

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References

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Figure Captions

Fig. A-I The cross section of the target composed of a fission product material and a refrector wall. The 14 MeV neutron produced by DT fusion reaction at the center are used to trigger incineration reactions.

Fig. B-1 A cross section of a micro-target used in our model, where spherically shaped DT fuel (radius r_1) is surrounded by a spherical shell (outer radius r_2) made of ^{90}Sr.

Fig. B-2 Incineration energy, E, is shown as a function of the target's internal energy, U. The corresponding compression ratio, K, of DT fuel are written alongside the curves in the figures. In this example, the temperature of inner DT fuel and outer ^{90}Sr were chosen as 10 keV and 1 keV, respectively. The numbers in the two-line parentheses show the gain factor G (upper) and the annual transmutation amount of ^{90}Sr.

Fig. B-3 Internal energy ratio of ^{90}Sr to DT fuel are plotted as a function of the DT fuel compression ratio. The corresponding temperature of the ^{90}Sr (T^{FP}) are written alongside the curves. The temperature of DT fuel is chosen as 10 keV.

Fig. C-1 A conceptual picture of the incineration system, composed of a 4 GeV -25 mA Deutron accelerator and a FP-TRU hybrid core.

Fig. C-2 The r-z cross section of the subcritical core composed of FP (^{90}Sr and ^{99}Tc) and TRU. ^{6}Li is used to produce T fuel for the μCF reaction through the $^{6}Li(n,\alpha)$ reaction.

Fig. C-3a The cross-section of a particle fuel. TRU carbide is covered with carbon of 30 μm thickness and ZrC of 30 μm thickness. The radius of each material is shown in the figure.

Fig. C-3b The z cross-section of a fuel rod. The particle fuel in Fig. C-3a is set in a fuel cell; the fuel is cooled by He gas, and the wall material is Zr. The radius of each cell is shown in the figure.

Fig. C-3C Fuel rods in the core.

Fig. C-4 The k_{eff} of the core as a function of the inventory of TRU.

Fig. D-1 The r-z cross-section of the subcritical core composed of a TRU core, a refractor, and a lead target. Numbers in the figure show ID number of each cell.

Table A-I

The Probabilities of the (n,2n) and (n,γ) Reactions for ^{137}Cs per an Input of a 14 MeV Neutron

rge Material	Refractor Thickness	Reaction Probabilities		
		(n,2n)	(n,γ)	sum
^{137}Cs	2 cm	46.6%	0.4%	47.0%
^{137}Cs	20 cm	46.7%	28.3%	75.0%
$^{137}Cs_2O_2$	2 cm	43.8%	4.1%	47.9%
$^{137}Cs_2O_2$	20 cm	43.9%	40. 1%	84.0%

Table A-II

The Probabilities of the (n,2n) and (n,γ) Reactions for ^{90}Sr per an Input ofa14MeV Neutron

Target Material	Refractor Thickness	Reaction Probabilities		
		(n,2n)	(n,γ)	sum
^{90}Sr	2 cm	49.6%	2.6%	52.2%
^{90}Sr	20 cm	49.6%	9.3%	58.9%
^{90}SrO	2 cm	41.6%	4.9%	46.5%
^{90}SrO	20 cm	41.7%	11.9%	53.6%

Table A-III

The Incineration Half-Life, the Incineration Energy, and the Annual Reduction Amounts

Target Material	Refractor Thickness [cm]	Half-Life [Year]	E^{inc} [MeV]	Reduction [10^3 kg]
^{137}Cs	2	3.4	3.7	0.77
^{137}Cs	20	2.2	2.3	1.13
$^{137}Cs_2O_2$	2	3.4	3.7	0.77
$^{137}Cs_2O_2$	20	1.9	2.1	1.28
^{90}Sr	2	3.1	3.4	0.55
^{90}Sr	20	2.7	3.0	0.82
^{90}SrO	2	3.5	3.8	0.49
^{90}SrO	20	3.0	3.3	0.57

Table C-I

Reaction Probability and Incineration Half-Life for Each Nuclide

Nuclide	Inventory	Reaction Probability per an Input of 14 MeV Neutron				Half-Life	
	[kg]	(n,2n)[%]	(n,γ)[%]	(n, f)[%]	total [%]	[year]	
^{237}Nu	72.2	0.00	28.73	9.62	38.35	0.34	(1.35)’
^{241}Am	83.9	0.00	40.07	15.54	55.61	0.27	(0.96)’
^{243}Am	15.2	0.00	4.66	2.27	6.93	0.39	(1.18)[1]
^{244}Cm	3.0	0.00	0.53	0.51	1.04	0.51	(1.03)’
^{238}Pu	2.7	0.00	0.68	0.90	1.58	0.31	(0.54)’
^{239}Pu	100.4	0.00	16.80	59.61	76.41	0.23	(0.30)’
^{240}Pu	46.5	0.00	0.90	8.31	9.21	0.90	(1.00)[a]
^{241}Pu	15.5	0.00	0.27	10.95	11.22	0.24	(0.25)’
^{242}Pu	8.1	0.00	0.16	1.02	1.18	1.21	(1.40)’
TRU(sum)	347.5	0.00	92.80	108.73	201.53	0.31	(0.57)’
cc	72.7	0.00	0.54	0.00	0.54	47,000	
Zr^{c}	831.9	5.04	10.23	0.00	15.27	25.52	
^{90}Sr	113.9	31.69	1.78	0.00	33.47	1.62	
^{99}Tc	277.9	0.34	72.60	0.00	72.94	1.64	
T	0.1	0.20	0.00	0.00	0.20	6.41	
D	446.0	3.14	0.62	0.00	3.76	25,000	
’^{6}0	1783.8	0.00	1.07	0.00	1.07	45,000	
^{6}Li	6.7	0.00	97.85’	0.00	97.85	0.49	
^{27}Al	272.0	0.00	1.13	0.00	1.13	380	
Cr^{c}	175.3	0.90	10.10	0.00	11.00	13.10	
Fe^{c}	1259.3	2.99	54.94	0.00	57.93	16.65	

a: incineration half-life determined by (n, f) reaction rate.

b: (n,α) reaction probability.

c: average of natural abundance was adapted for nuclear data.

Table C-II

Reduction Amounts by Incineration Per a Year for each FP and TRU

	Waste Reduction by Incineration [kg/year][a]
^{237}Np	29.0
^{241}Am	43.1
^{243}Am	6.8
^{244}Cm	1.5
^{238}Pu	2.0
^{239}Pu	90.4
^{240}Pu	23.3
^{241}Pu	14.5
^{242}Pu	3.2
TRU(sum)	244.5
^{99}Sr	39.6
^{99}Tc	95.8

a: To deduce this quantity for elements of TRU, incineration half-life including only contribution of fission reaction was used.

Table D-I

Neutron Flux, Atomic Displacement, Hydrogen Production, Helium Production, and Energy Disposition in the Target

Cell #	Neutron Flux [10^{-3} cm^{-2}]	dpa Per Year	H. Prod. per P	He Prod. per P	Energy Dep. [kW]
1	0.0389	0.0153	0.945	0.295	797
2	0.0054	0.021	----	----	0.004
3	2.61	10.28	3.35	0.58	4926
4	0.652	2.57	0.59	*0.035*	853
5	0.0602	0.237	0.015	----	5.95
6	0.0187	0.074	----	----	0.31
7	1.54	6.07	0.150	0.015	86.0
8	----	----	----	----	----
9	0.095	0.374	0.015		1.70
10	0.751	2.96	*0.14*	*0.03*	69.75
11	0.295	1.16	0.055	----	47.85
12	0.076	0.299	0.055	0.005	19.73
13	0.0087	0.034	----	----	----

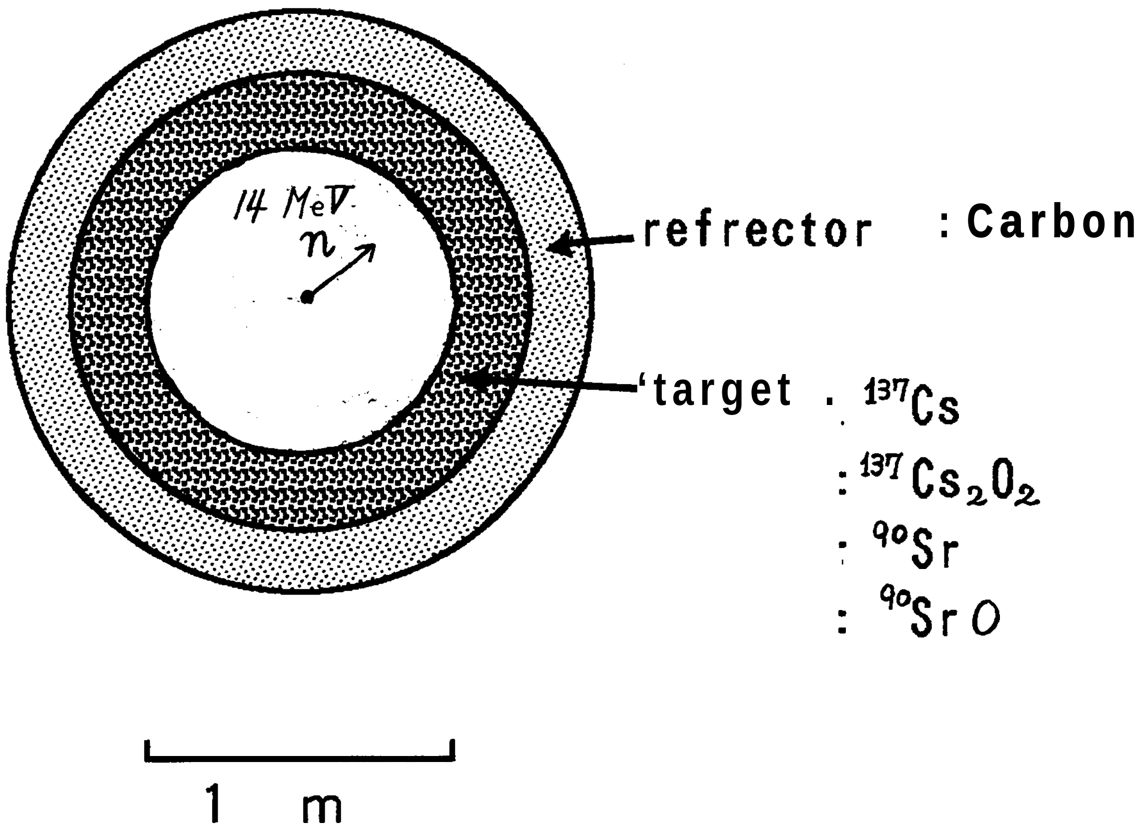

14 MeV
n
refrector : Carbon
target . ^{137}Cs
: $^{137}Cs_2O_2$
: ^{90}Sr
: ^{90}SrO
1 m

Fig.A- 1

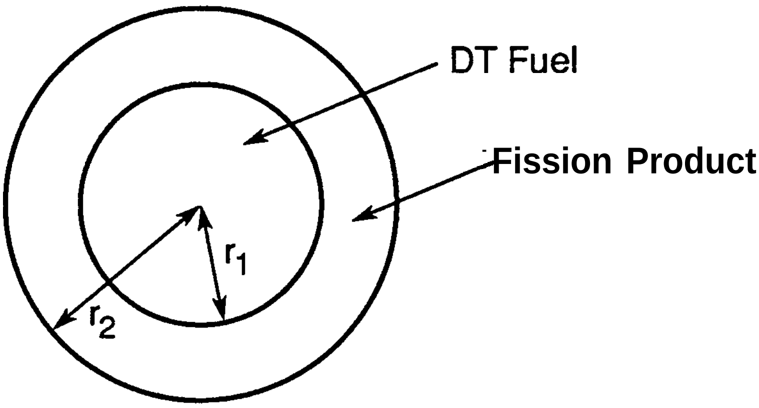
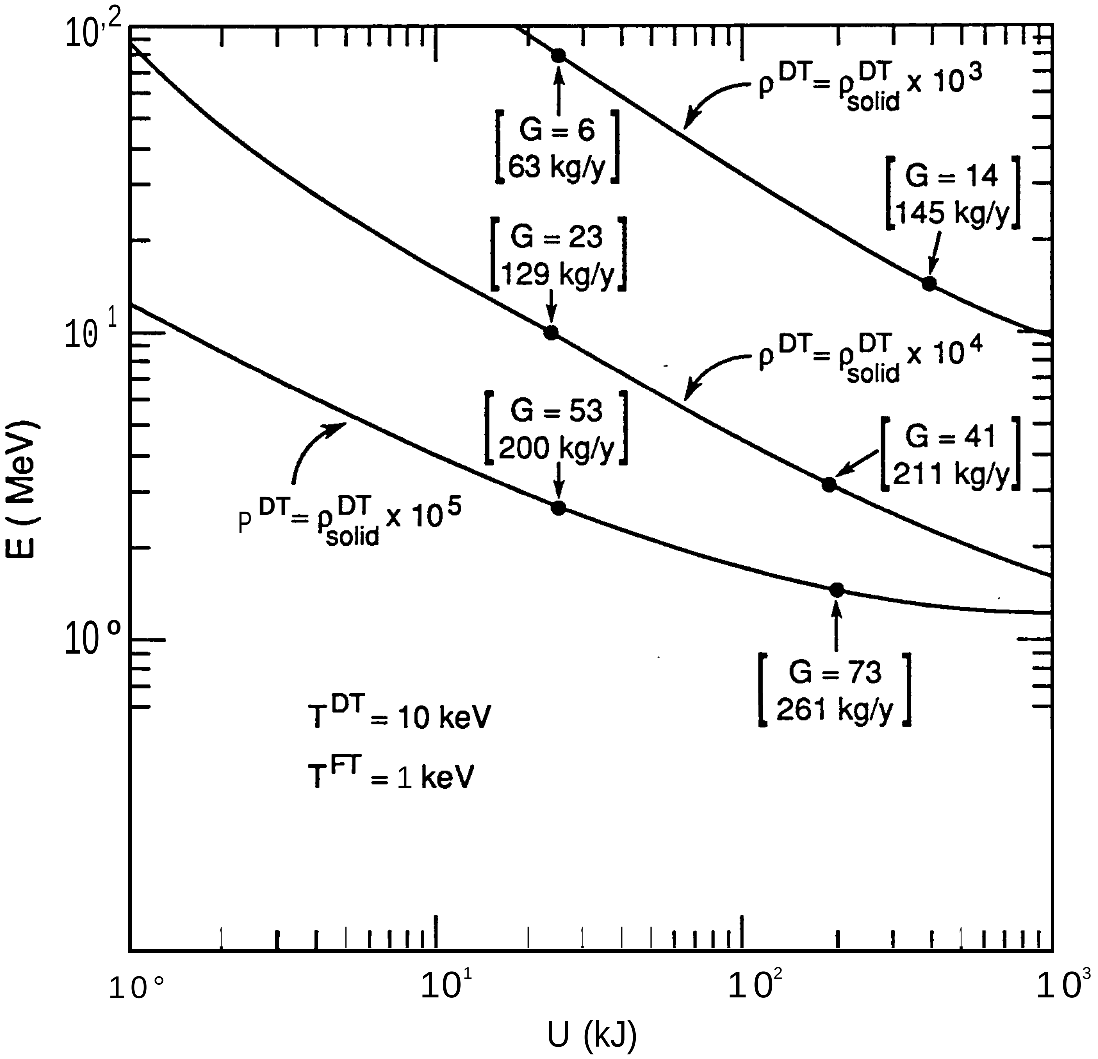
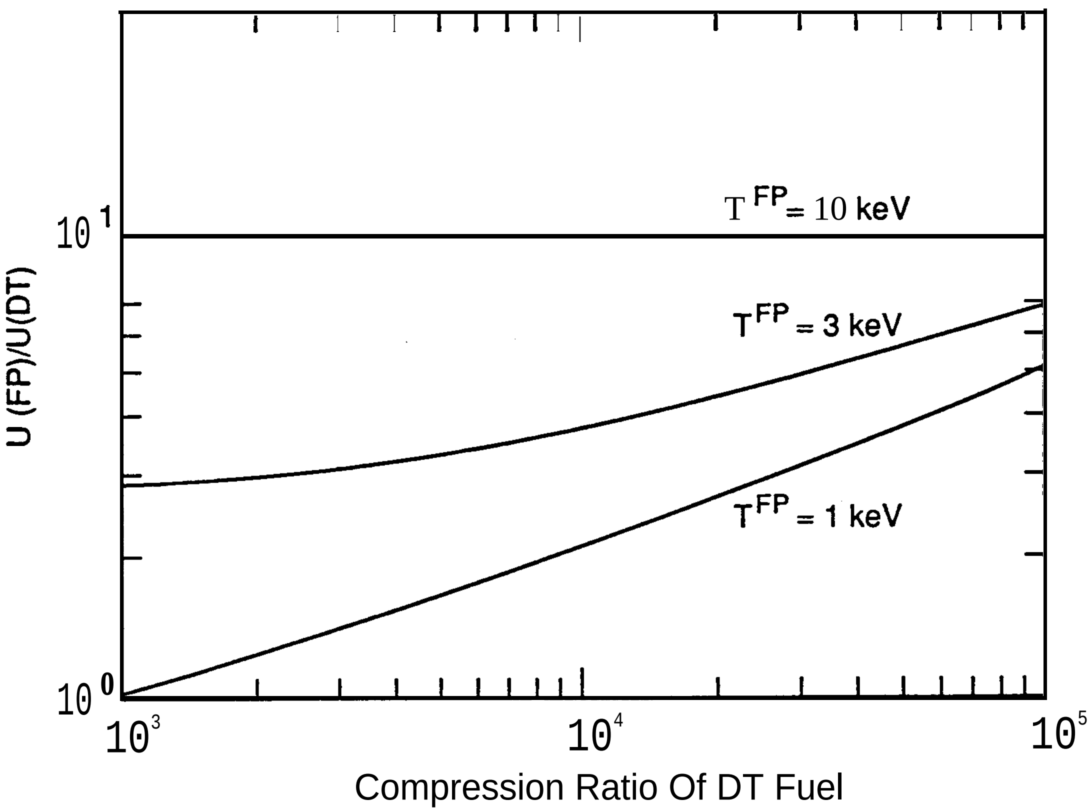


Fig. B-1



$\rho^{DT} = \rho^{DT}_{solid} \times 10^3$
$\rho^{DT} = \rho^{DT}_{solid} \times 10^4$
$\rho^{DT} = \rho^{DT}_{solid} \times 10^5$
[G = 6 63 kg/y]
[G = 14 145 kg/y]
[G = 23 129 kg/y]
[G = 53 200 kg/y]
[G = 41 211 kg/y]
[G = 73 261 kg/y]
T^{DT} = 10 keV
T^{FT} = 1 keV
E (MeV)
U (kJ)
10^2
10^1
10^0
10^0
10^1
10^2
10^3

Fig. B-2



T^{FP} = 10 keV
T^{FP} = 3 keV
T^{FP} = 1 keV
10^1
10^0
U (FP)/U(DT)
10^3
10^4
10^5
Compression Ratio Of DT Fuel

Fig. B-3

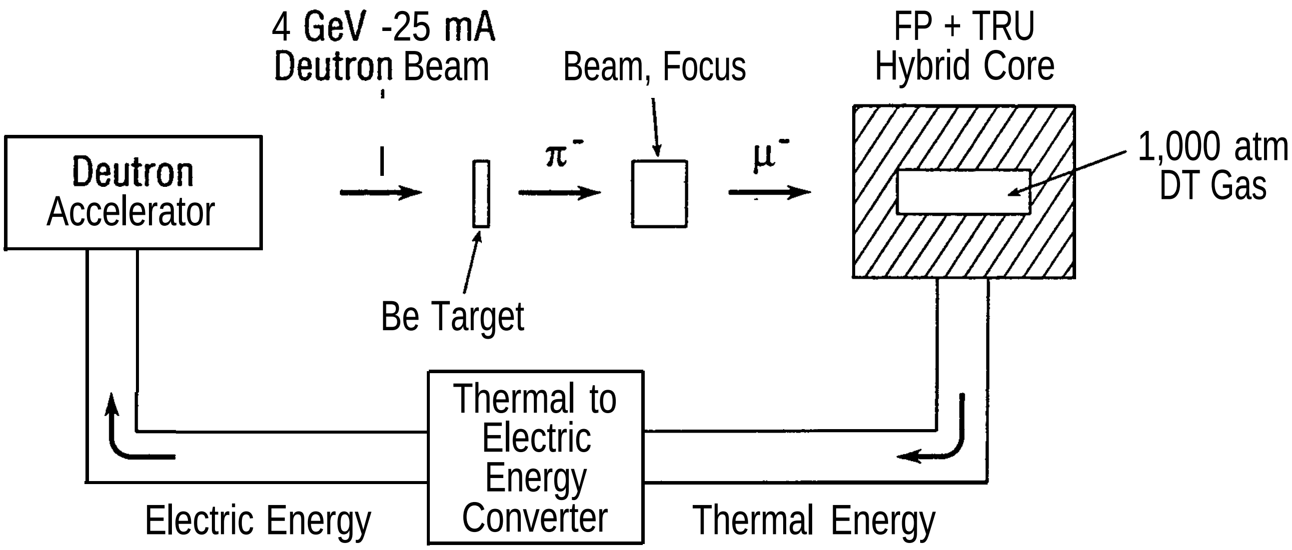

4 GeV -25 mA
Deutron Beam
Beam, Focus
FP + TRU
Hybrid Core
Deutron
Accelerator
I
π⁻
μ⁻
1,000 atm
DT Gas
Be Target
Thermal to
Electric
Energy
Converter
Electric Energy
Thermal Energy

Fig. C-1

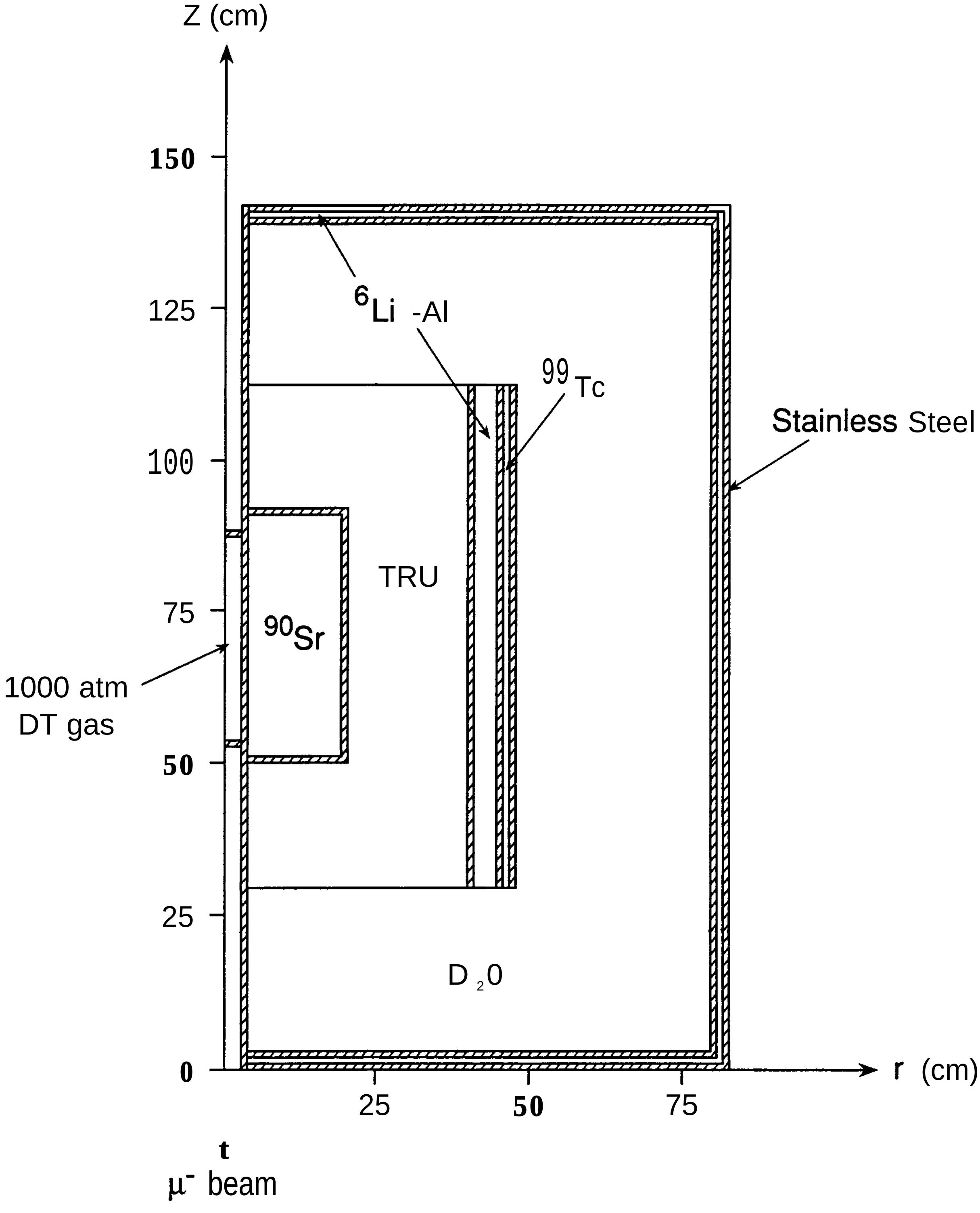
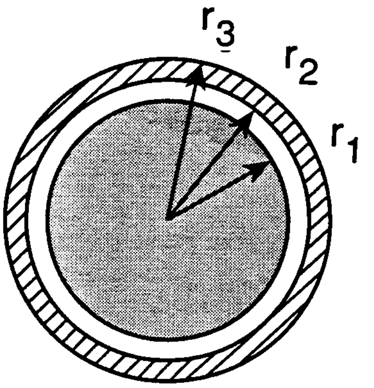

Z (cm)
150
125
100
75
50
25
0
25
50
75
r (cm)
6Li -Al
99 Tc
Stainless Steel
TRU
90Sr
1000 atm
DT gas
D 2 0
t
μ- beam

Fig. C-2



r_1 = 190 µm (TRU)C_2 11.4 glee

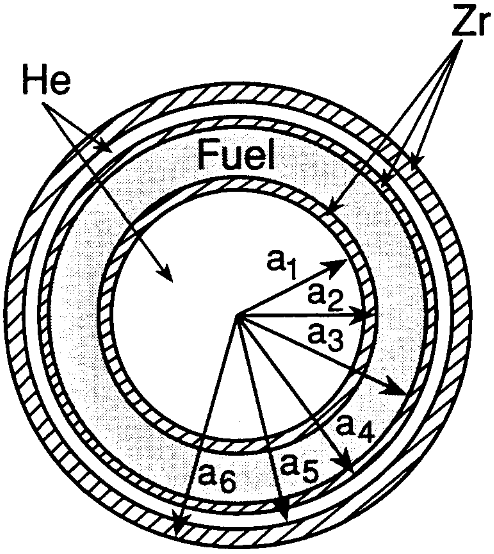
r_2 = 220pm C 1.0 glee

r_3 = 250pm ZrC 6.7 glee

solid fraction = 0.64

void fraction = 0.36

Fig.c3a



a_1 = 2.5 cm

a_2 = 2.7 cm

a_3 = 3.7 cm

a_4 = 3.9 cm

a_5 = 4.2 cm

a_6 = 4.5 cm

Fig.c3b

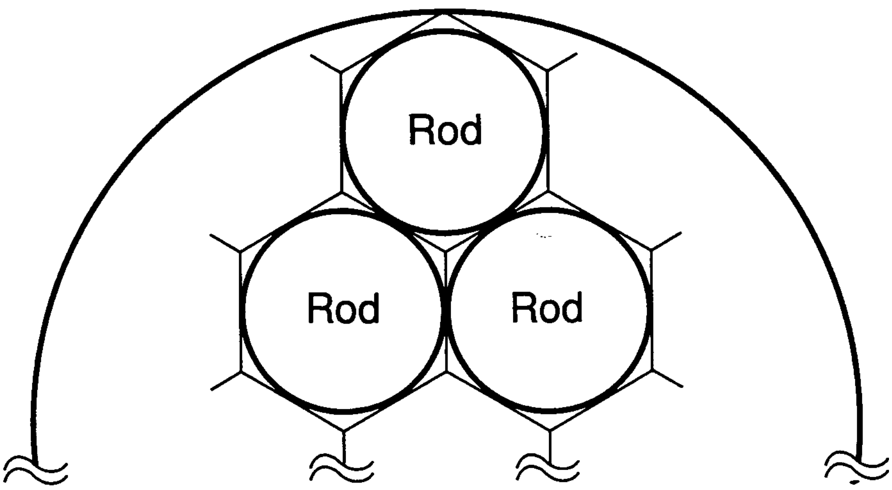


Fig.c3c

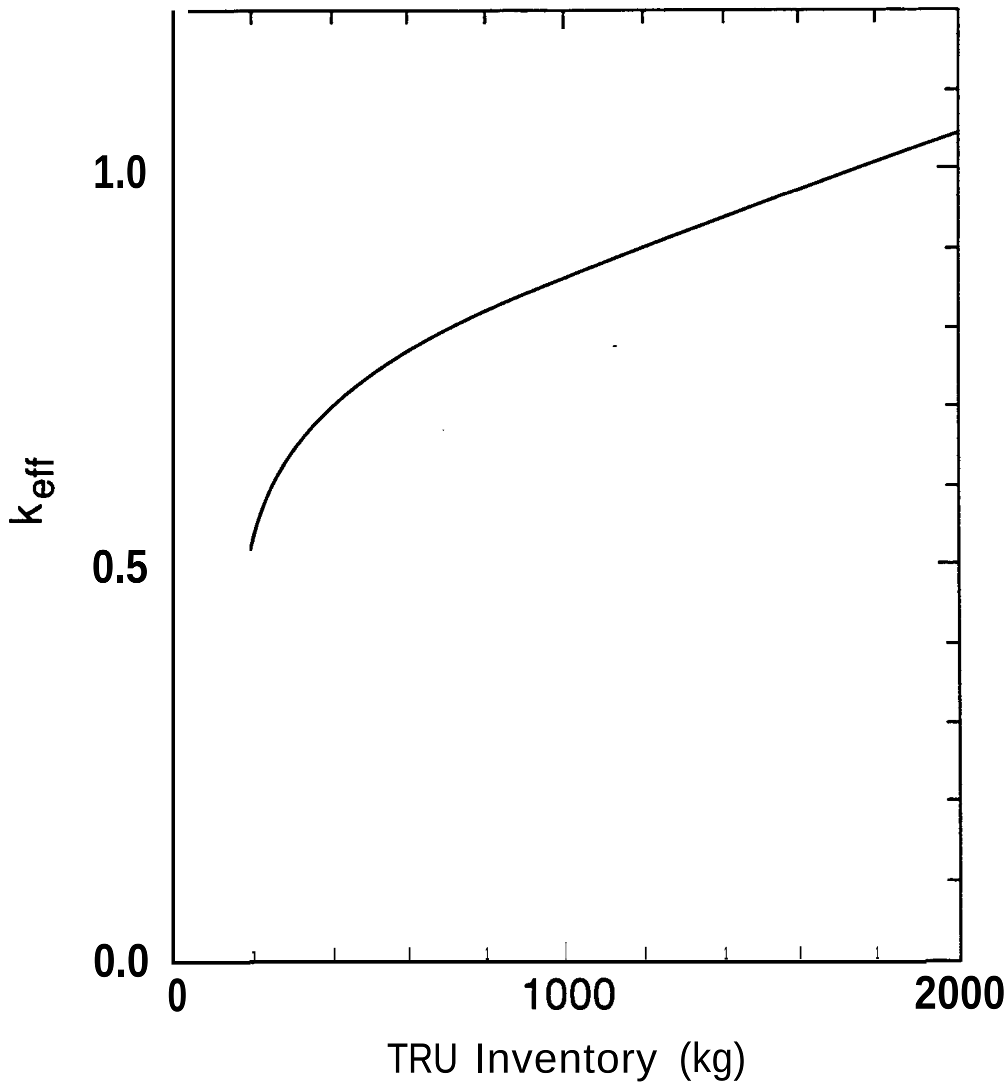


Fig. C 4

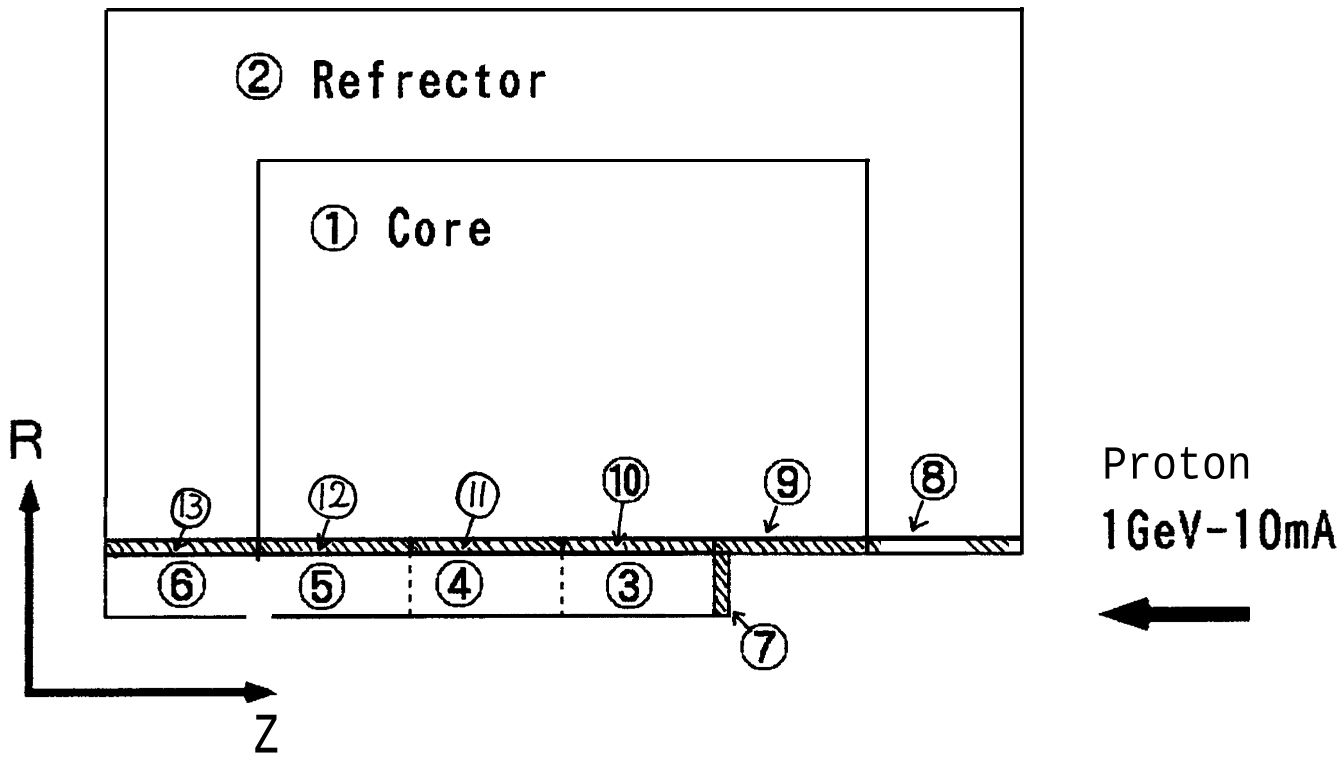


Fig. D-1