Challenge of Transmutation of Long-Lived Nuclides

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   objectives and goals of transmutation;
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2. **Incentives to apply accelerator&fusion technology for waste transmutation**

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   application of hard spectra (transmutation through (n,2n) reaction).

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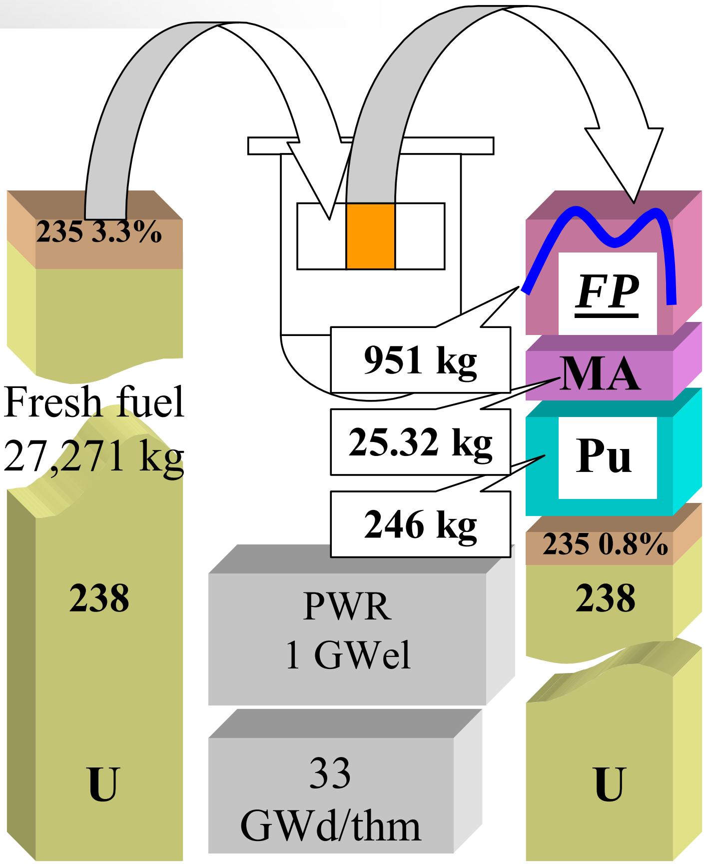
1. Challenge of Waste Transmutation

- waste characterization;
- objectives and goals of transmutation;
- current status of waste transmutation in fission reactors.

Composition of Spent Nuclear Fuel



What is Nuclear Waste?

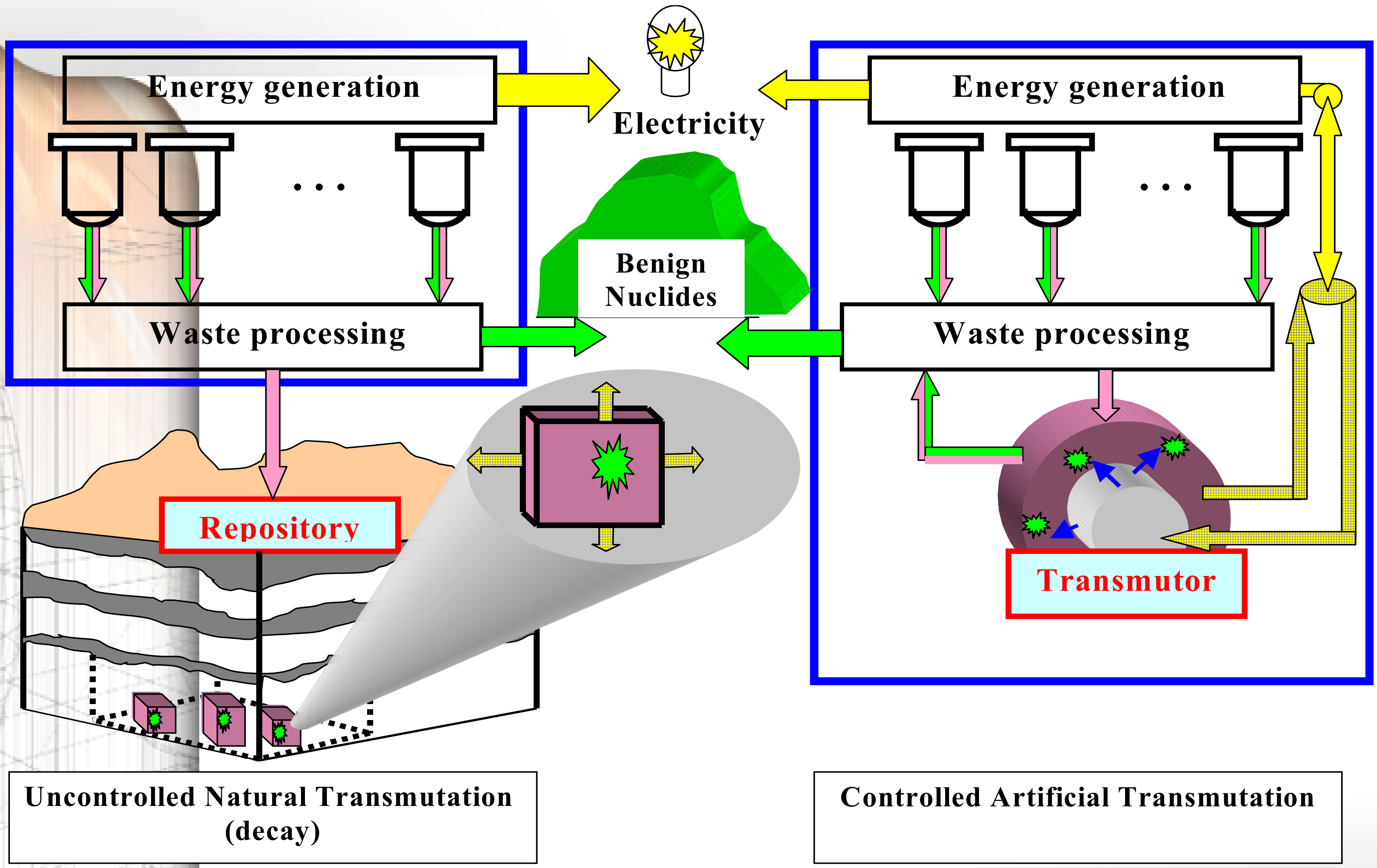
Fission Products ?

Minor Actinides?

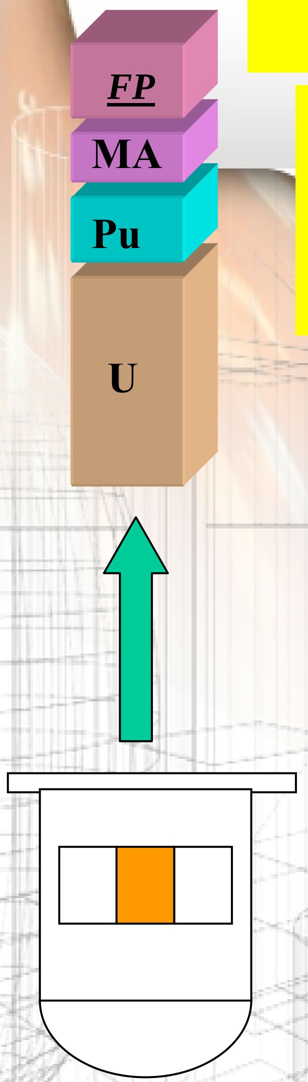
Plutonium ?

Uranium ?

Waste Management Options



Terminology of hazard from radioactive nuclides



Mass (g)

m$_i$ **Source term**

Activity (Bq)
Nuclear properties added

$$A_i = m_i \times \frac{\lambda_i N_{Av}}{Mol_i} = m_i \times \gamma_i$$

Potential Hazard

Toxicity (ALI)*
Properties of dose absorption in the human body added

$$T_i = A_i \times \frac{1}{ALI_i} = m_i \times \frac{\lambda_i N_{Av}}{Mol_i} \times \frac{1}{ALI_i} = m_i \times$$

Radiological Risk

Radiological risk

(rem/m^3)
Properties of retention in geological strata added

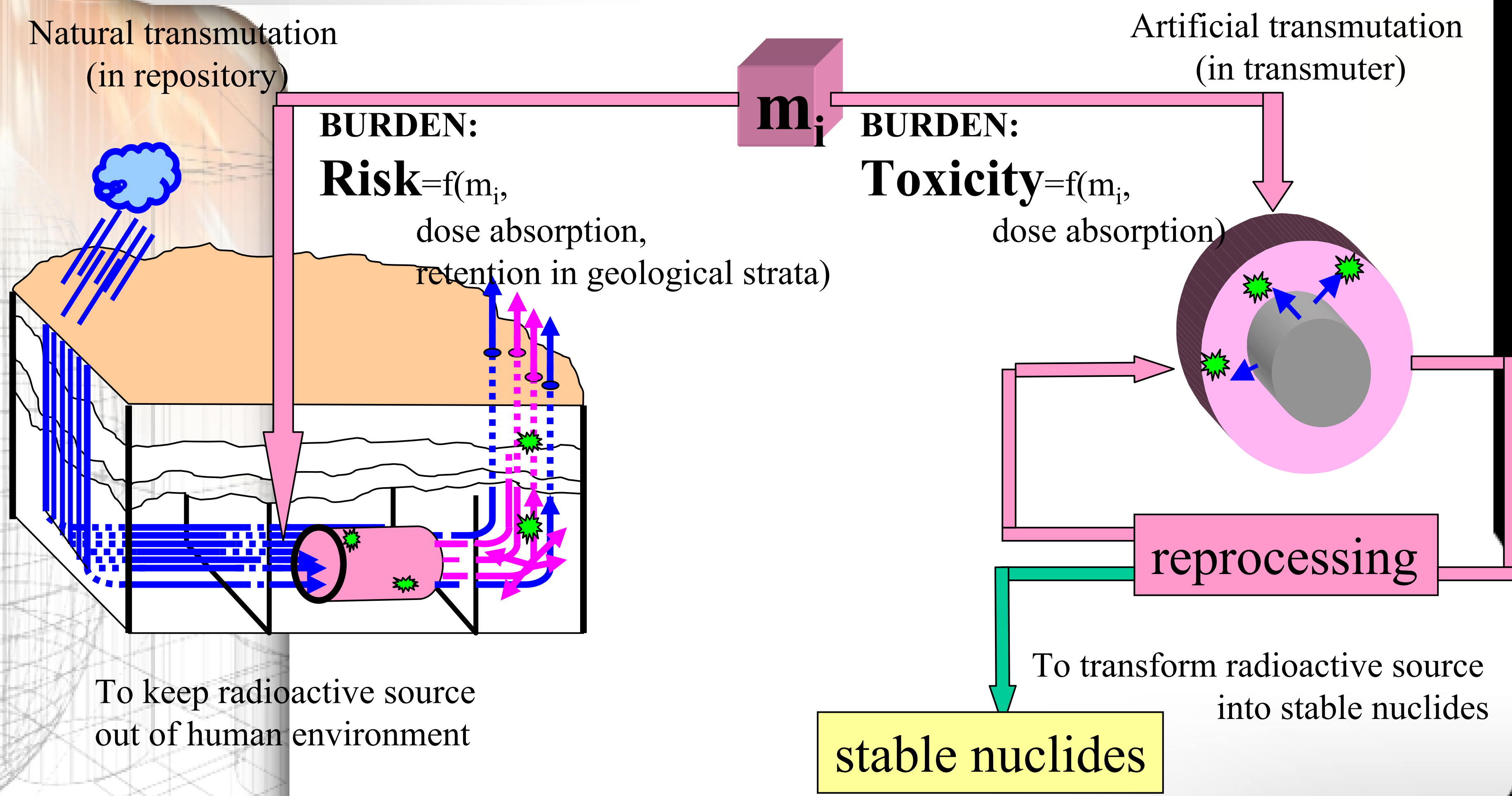
$$R_i = T_i \times f\left(\lambda_i, F_i, K_i, DD_i, t_i\right)$$

*Annual Limit on Intake (ALI) - is the annual intake of a given radionuclide by "Reference Man" which would result in ...ther a committed effective dose equivalent of 5 rems (stochastic ALI) or a committed dose equivalent of 50 rems to an ... or tissue (non-stochastic ALI)

Objectives of FP Transmutation

To eliminate the radiological risk

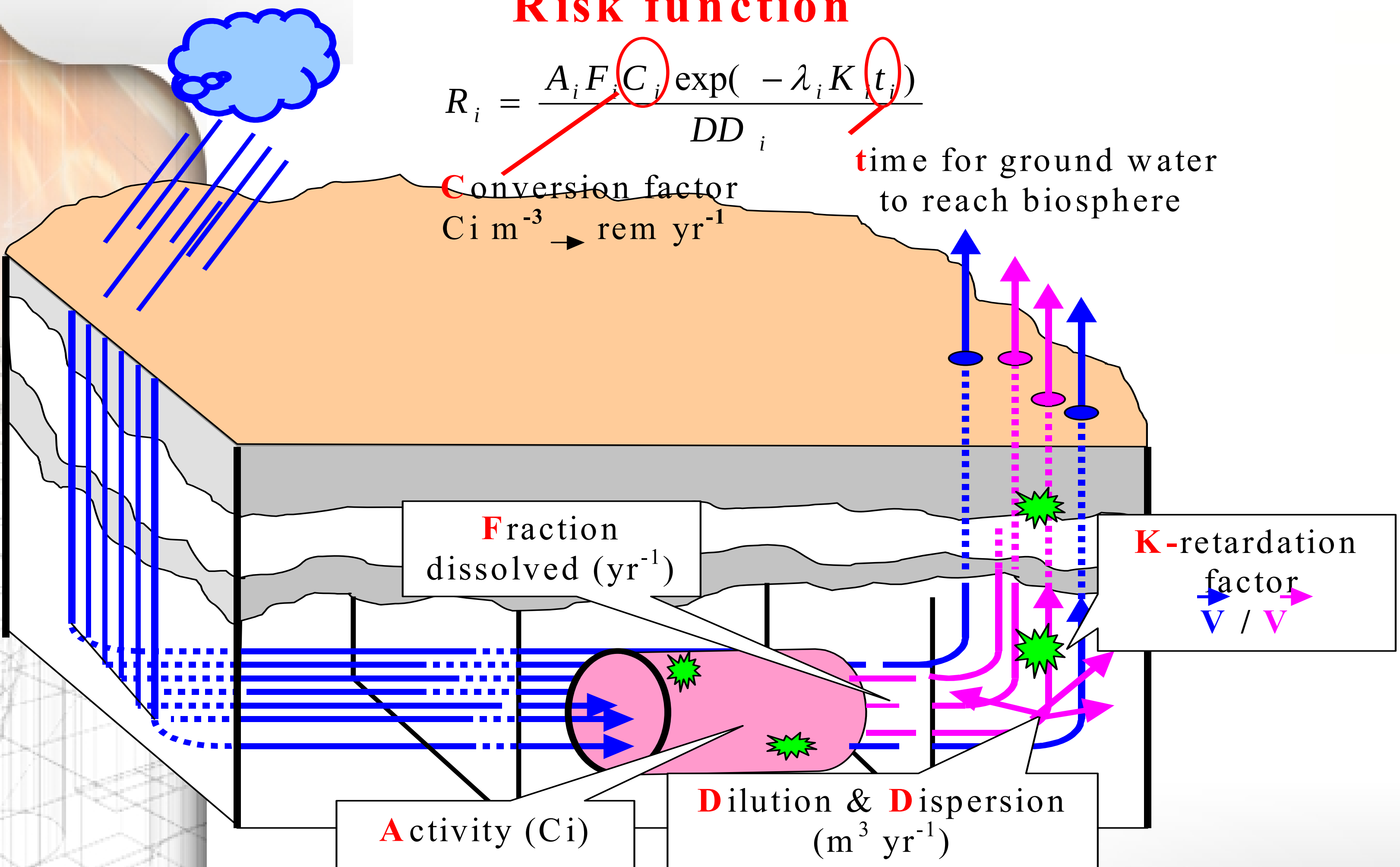
To eliminate the source of radiological risk



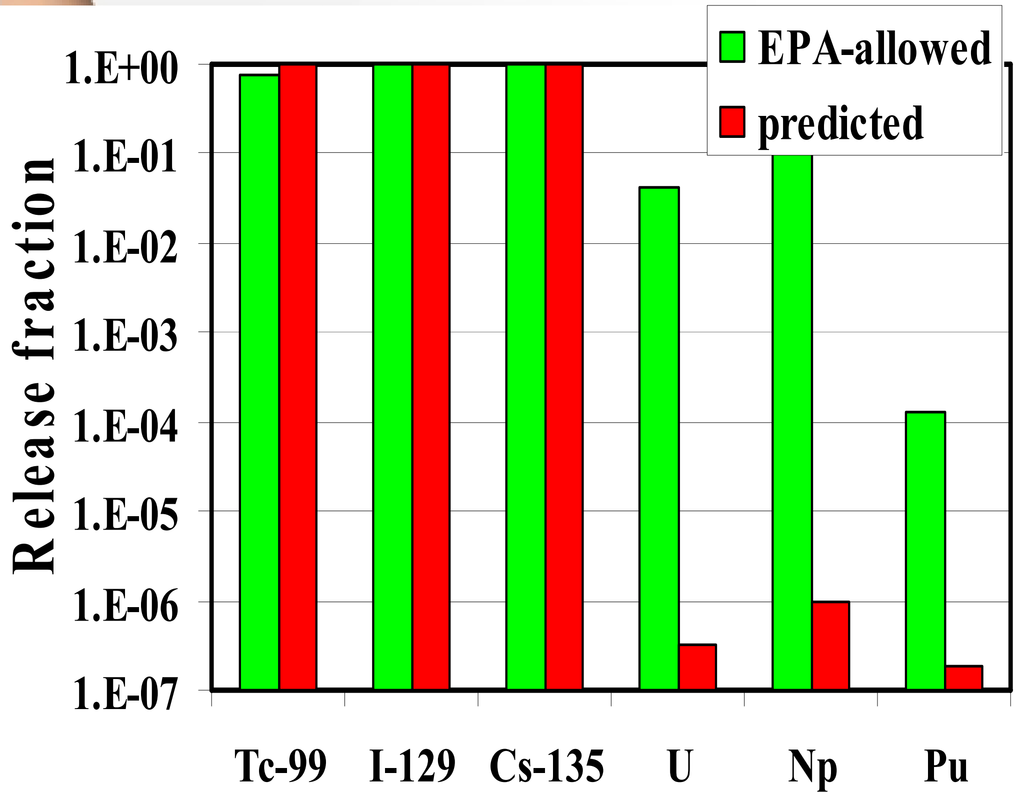
Characterization of Waste Storage

Risk function

$$R_i = \frac{A_i F_i C_i \exp(-\lambda_i K_i t_i)}{DD_i}$$



Cumulative releases from spent fuel in 10,000 years (Yucca Mountain project)*



EPA - Environmental Protection Agency

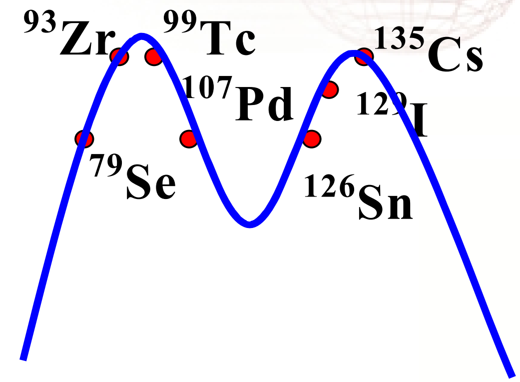
Standards on cumulative yields are taken from:

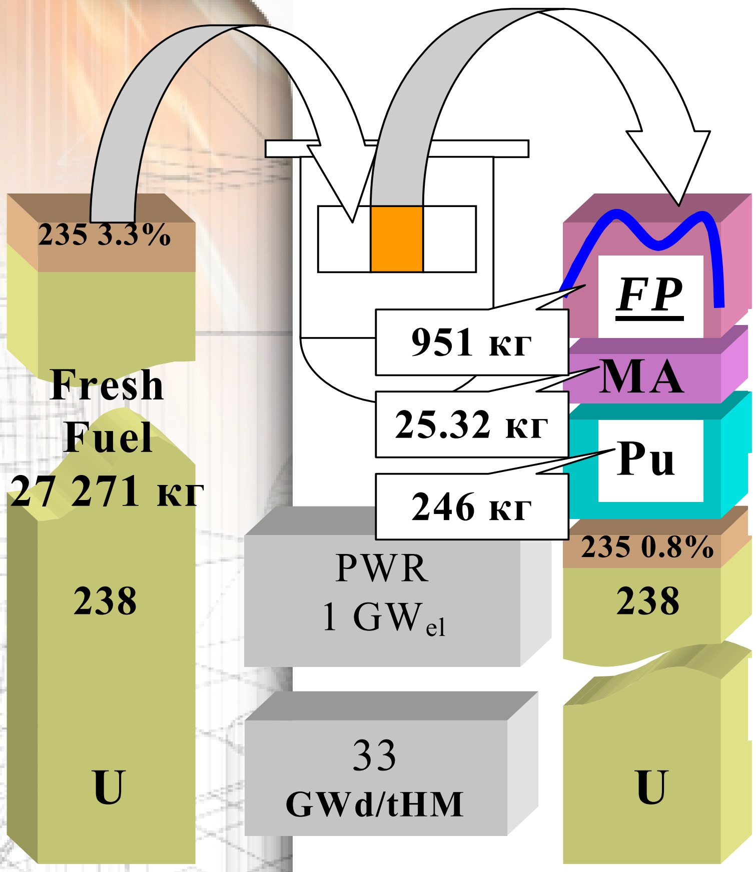
The Code of Federal Regulations, Title 40, Part 191- Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Wastes

* T.PIGFORD, Actinide Burning and Waste Disposal, *Proc. Int. Conf. Next Generation of Nuclear Power Technology*, Berkley, University of California, 5 October, UCB-NE-4176 (1990)

Incentives for Recycle of Heavy Metals

Fission Products $T_{1/2}$ >10^4лет :





MA-recycle → **Energy production (&protection of recycled Pu)**

^{237}Np: (n,γ)^{238}Np→ ^{238}Pu

^{241}Am: (n,γ)^{242}Am→^{242}Cm→ ^{238}Pu

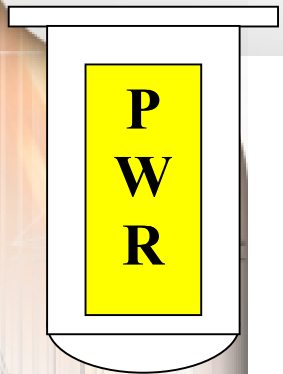
Pu - recycle → **Energy production (&complete use of uranium)**

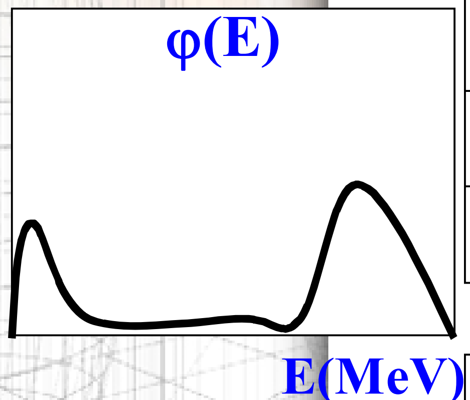
U - recyle → **Energy production (&uranium saving)**

Neutron balance in fast and thermal reactors*

(normalized to one fission)

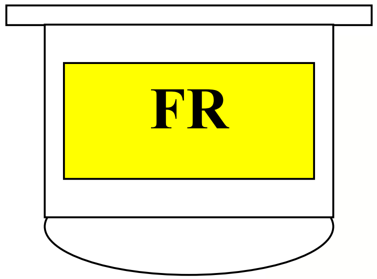
$\varphi = 10^{14}$ n/cm^2 s

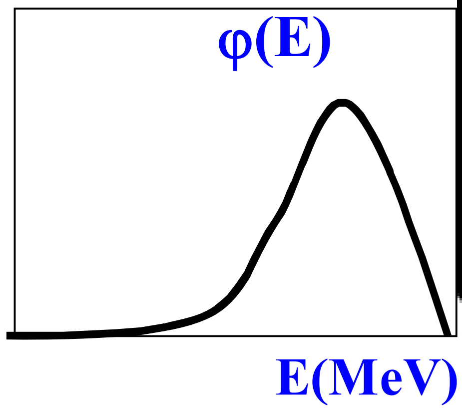




PWR		FR
2.9	Neutron Generation	2.9
	Neutron Captures	
1	Energy generation	1
0.4	Capture in fissiles	0.13
1.3	Fuel breeding	0.85
0.25	Other absorptions	0.2
2.95	Total	2.18
<0	Neutron Excess	>0

$\varphi = 10^{15}$ n/cm^2 s





* Shimizu, A., Fujii-e,Y.: *Prog. Nuclear Energy*, 29,25 (1995)

Concepts of Nuclear Energy Systems with FP Transmutation

Equilibrium Nuclear Society	Self-Consistent Nuclear Energy System	Concept Of Radiation Equivalency	Radiologically Clean Nuclear Power
H.Sekimoto, et al, Preliminary study of future society in nuclear quasi-equilibrium,	Y.Fujii-e, et al, An approach to Self-Consistent Nuclear Energy System,	E.Adamov, et al, Attainment of radiation equivalency in nuclear power radioactive product management,	M.Salvatores, et al, A global physics approach to transmutation of radioactive nuclides,
Journ. Nucl. Sci.Techn. v.28, p.941-946, **1991**	Trans. Amer. Nucl. Soc., v.66, p.342-344 **1992**	Nucl.Techn., v.104, p.233-240 **1993**	Nucl.Sci and Engineering, v.116, p.1-18 **1994**

Fast Spectrum

Fission Products – candidate for transmutation

7 nuclides: ^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, ^{135}Cs	**29** nuclides: ^{79}Se, ^{85}Kr, ^{90}Sr, ^{93}Zr, ^{93m}Nb, ^{94}Nb, ^{99}Tc, ^{106}Ru, ^{102}Rh, ^{107}Pd, ^{108m}Ag, ^{109}Cd, ^{113m}Cd, ^{121m}Sn, ^{126}Sn, ^{125}Sb, ^{129}I, ^{134}Cs, ^{135}Cs, ^{137}Cs, ^{146}Pm, ^{147}Pm, ^{151}Sm, ^{152}Eu, ^{154}Eu, ^{155}Eu, ^{158}Tb, ^{166m}Hm, ^{171}Tb	**2** nuclides: ^{99}Tc, ^{129}I	**19** nuclides: ^{79,82}Se, ^{81,85}Kr, ^{87}Rb, ^{90}Sr, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{113}Cd, ^{115}In, ^{126}Sn, ^{129}I, ^{135,137}Cs, ^{142}Ce, ^{144}Nd, ^{151}Sm

Isotope Separation

Present status of FP transmutation problem

L.H. Baetsle, et al, «Status and assessment report on actinide and fission product partitioning and transmutation, an OECD Nuclear Energy Agency Review» **Int. Conf., GLOBAL'99, Wyoming, USA, 1999**	"As a general conclusion it can be postulated that transmutation of ^{99}Tc and ^{129}I in present reactors is not encouraging because of the long transmutation half-lives and the huge inventories of FP required. Special-purpose high flux reactors or ADS systems may perhaps improve the prospects". **Main conclusion: "transmutation of ^{99}Tc and ^{129}I in present reactors is not encouraging…"**

Intern.Sem. on Advanced Nuclear Energy Systems Toward Zero Release of Radioactive Wastes, Susono, Japan, 2000

M.Salvatores, **Transmutation: Issues and Perspectives after a Decade of Revival**	"…transmutation, even if limited to ^{99}Tc and ^{129}I (^{135}Cs being out of question) is a formidable one, implies the presence in the fuel cycle at equilibrium of very large amount of LLFP. The use of ADS can slightly help in terms of neutron availability, but the decrease of the support ratio… makes all the scenarios of LLFP transmutation very unlikely". **«... all the scenarios of LLFP transmutation very unlikely»**
Y.Fujii-e, **Nuclear Energy System for a Sustainable Development Perspective – Self-Consistent Nuclear Energy System**	"…It should be noted that SCNES and geological disposal can complement each other". **«... SCNES and geological disposal can complement each other»**

Goal of waste transmutation: To Stop Waste Accumulation in Nuclear Energy System

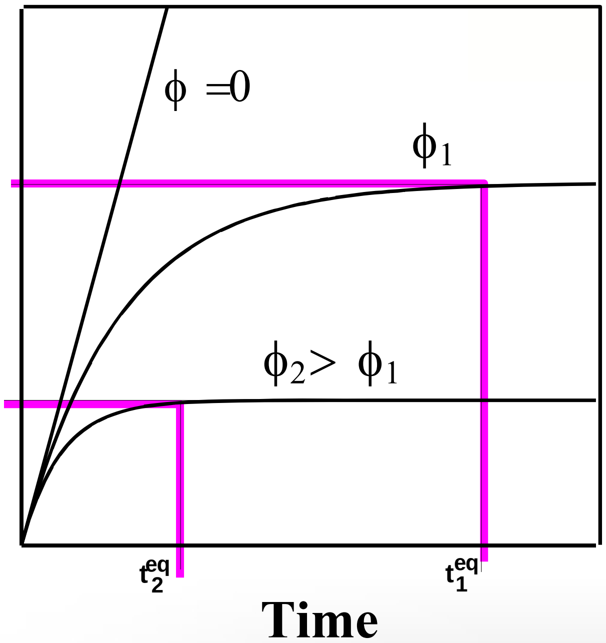
$$\frac{dN_i}{dt} = Y_i - (\lambda + \sigma\phi)_i N_i \Rightarrow N_i^{eq} = \frac{Y_i}{(\lambda + \sigma\phi)}$$

Characteristics of Transmutation Efficiency

- **Equilibrium Mass**
- **Time to approach equilibrium**

N_1^{eq}

N_2^{eq}



Time to Approach Equilibrium for Key Fission Products

Nuclide	Half-life (yr)	Effective half-life (yr)		Time to approach equilibrium (yr)**	
		Fast Spectrum*	Thermal Spectrum*	Fast Spectrum	Thermal Spectrum
^{79}Se	6.5×10^4	7.3×10^2	2.1×10^3	2.41×10^3	7.07×10^3
^{90}Sr	**29**	**29**	**29**	**≈100**	**≈100**
^{93}Zr	1.5×10^6	730	790	2.44×10^3	2.61×10^3
^{99}Tc	2.1×10^5	110	51	365	170
^{107}Pd	6.5×10^6	44	733	146	2.44×10^3
^{126}Sn	1.0×10^5	4.2×10^3	4.2×10^3	1.4×10^4	1.4×10^4
^{129}I	1.6×10^7	157	51.2	522	170
^{135}Cs	2.3×10^6	310	170	1.04×10^3	562
^{137}Cs	**30**	**30**	**30**	**≈100**	**≈100**

*) thermal spectrum: average energy -1eV, neutron flux- 10^{14} n/(cm^2· s);
fast spectrum: average energy -200 KeV, neutron flux - 10^{15} n/(cm^2· s).

**) 90% of assymptotic level.

Current Status of Waste Transmutation in Fission Reactors

Large-scale transmutation of Fission Products requires Nuclear Power System based exclusively on Fast Fission Reactors

Neutron Excess available in Fast Reactors is generally limited to isotope transmutation option, thus it requires introduction of large-scale isotope-separation technology

Time to approach equilibrium for long-lived fission product lasts for several centuries.

Neutron excess is absolutely necessary but not sufficient enough condition to perform effective transmutation of fission products

2. Incentives to apply accelerator&fusion technology for waste transmutation

- to improve the environment for transmutation of Fission Products

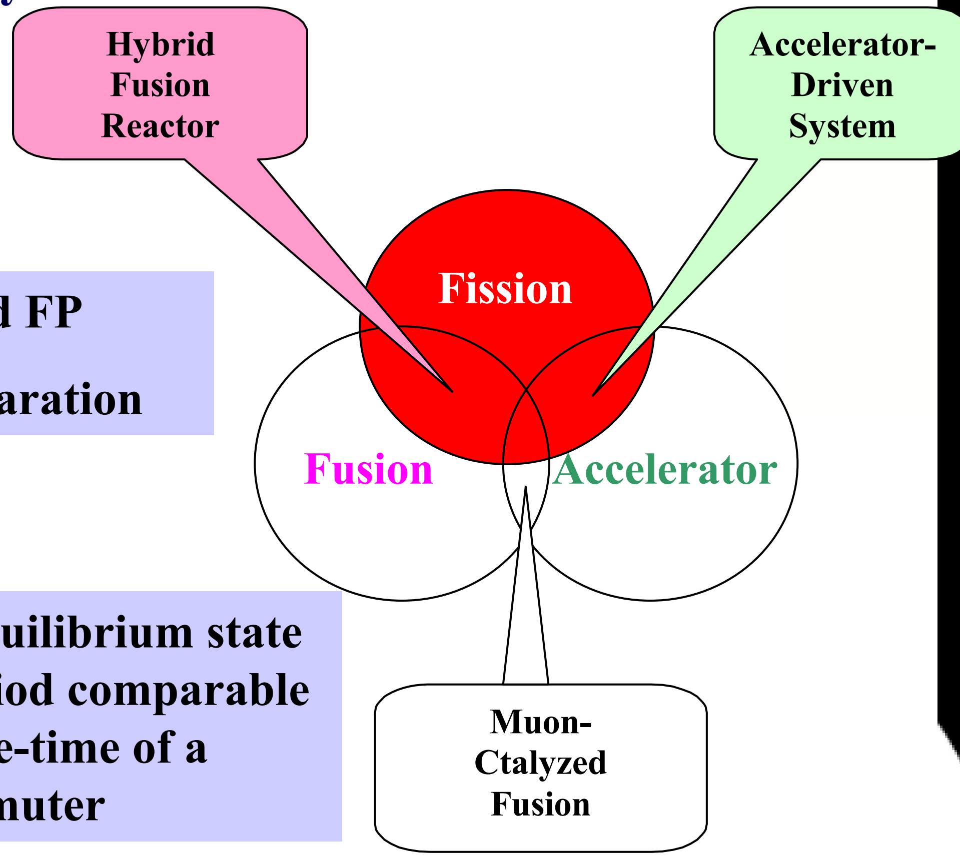
External Neutron Sources:
To improve quality of the neutron field in Nuclear Power System

Quality:

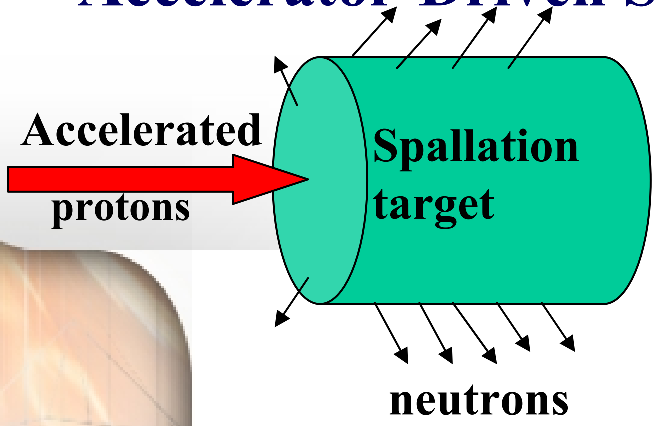
- **Neutron excess**
- **Neutron spectrum** → $\sigma\varphi$ ← **Neutron flux**

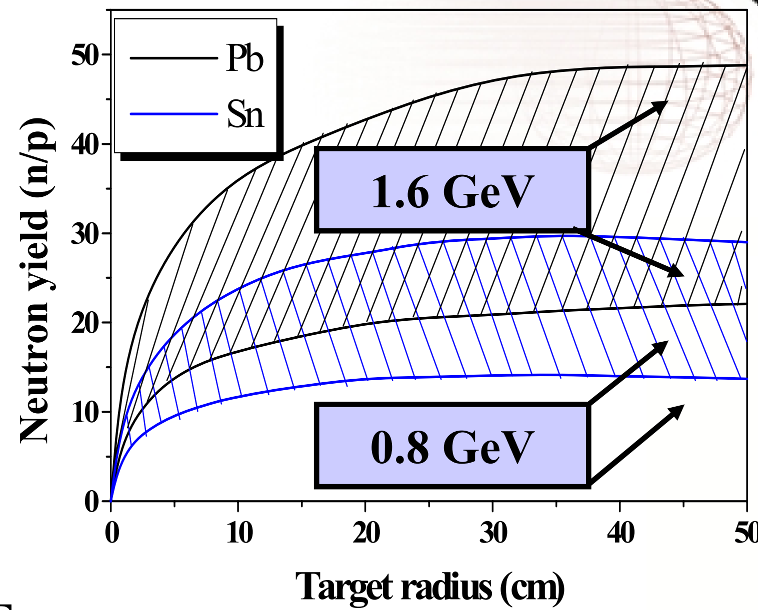
Neutron excess → **To avoid FP isotope separation**

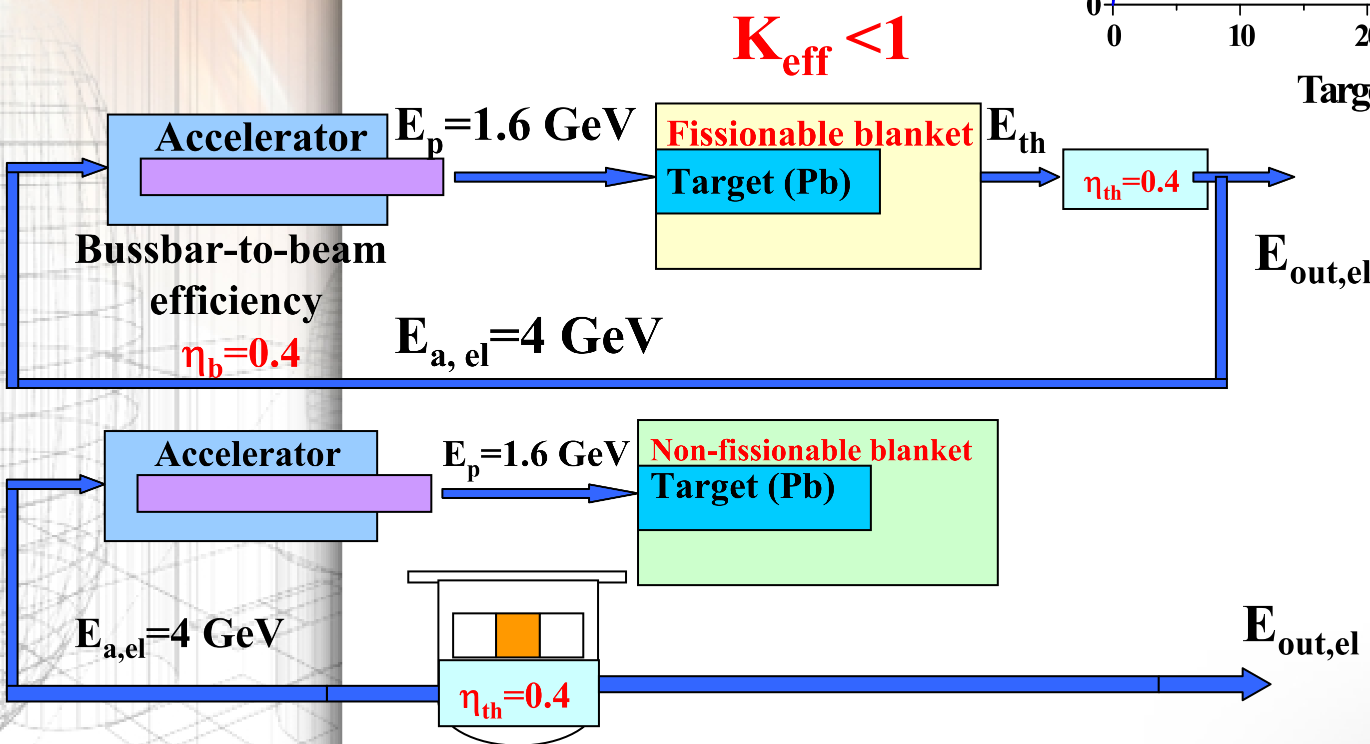
$\sigma\varphi$ → **To approach equilibrium state for the time period comparable with the life-time of a transmuter**



Accelerator-Driven System

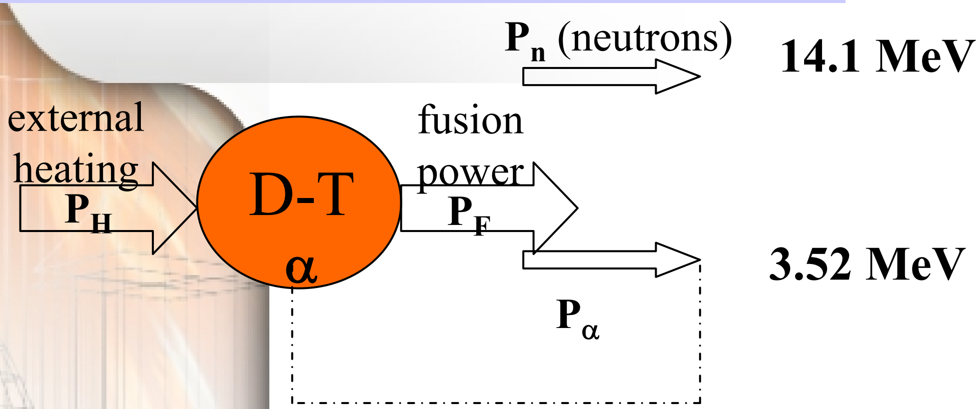






Fusion Neutron Source

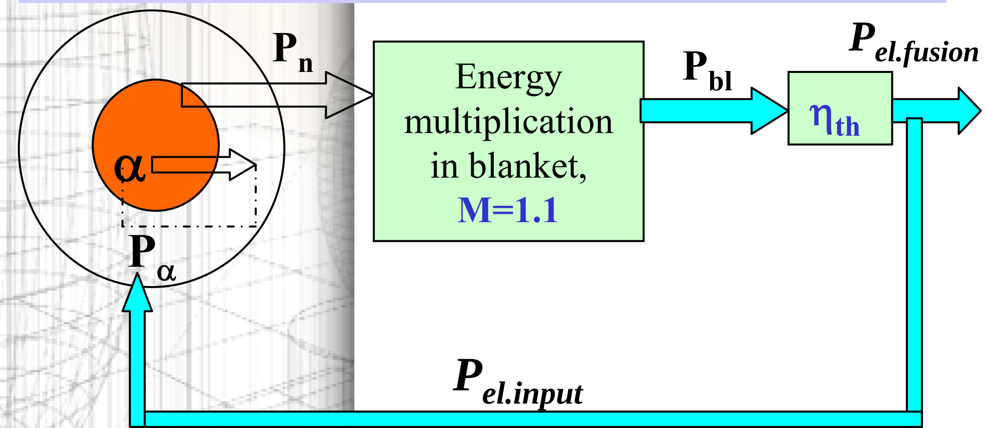
Essence of D-T fusion process



Thermal utilization efficiency:

$$Q_{th} = \frac{P_{fusion}}{P_{thermal\ \ input}}$$

Energy flows in Fusion Neutron Source

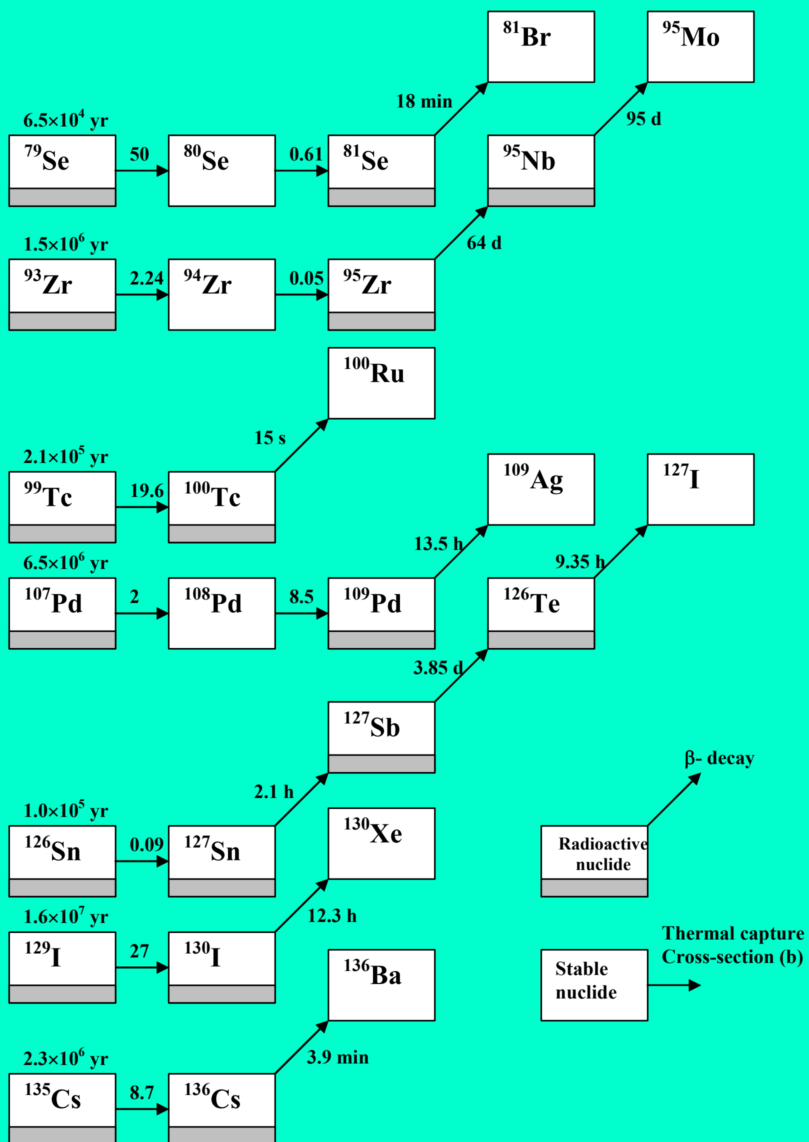


Break-even energy conditions (energy self-sustaining in FNS):

$$Q_{th}^{be} = \frac{1}{\eta_{th}\eta_{heating}} = 5$$

Neutron requirements to perform isotope transmutation

Transmutation chains



Neutron requirements

(per one fission)

$$n_{req,i} = Y_i \times \sum_{j=1}^{m} (m - j)$$

m **–is the maximum number of isotopes in the transition chain;**

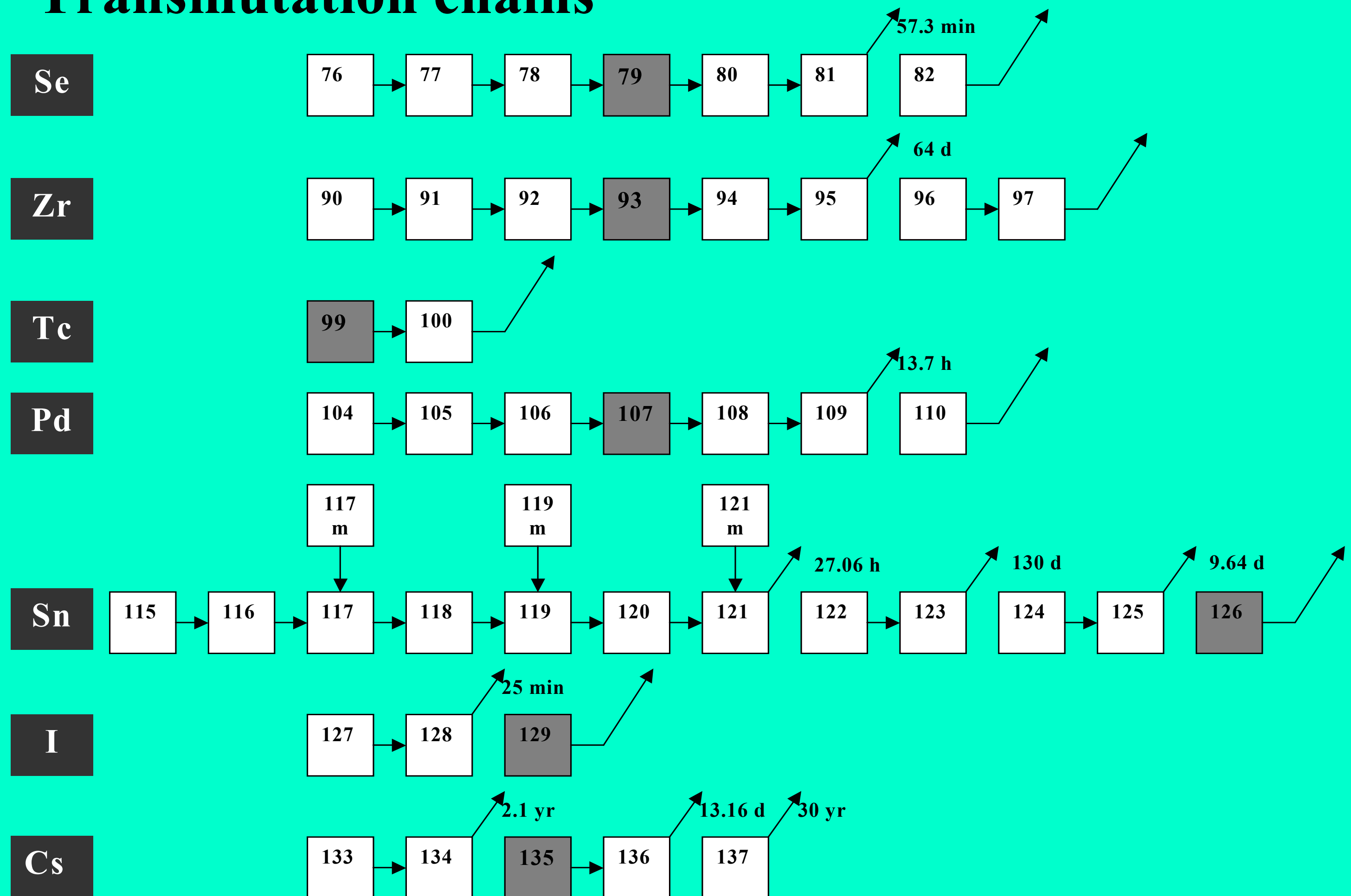
j **– is the target isotope number in this chain (for isotope transmutation** ***j*****=1);**

Y_i –fission yield of ***i*****–th isotope (atoms per fission)**

Thermal spectrum (fission U-235)	Fast spectrum (fission Pu-239)
2.66E-01	**2.94E-01**

Neutron requirements to perform elemental transmutation

Transmutation chains

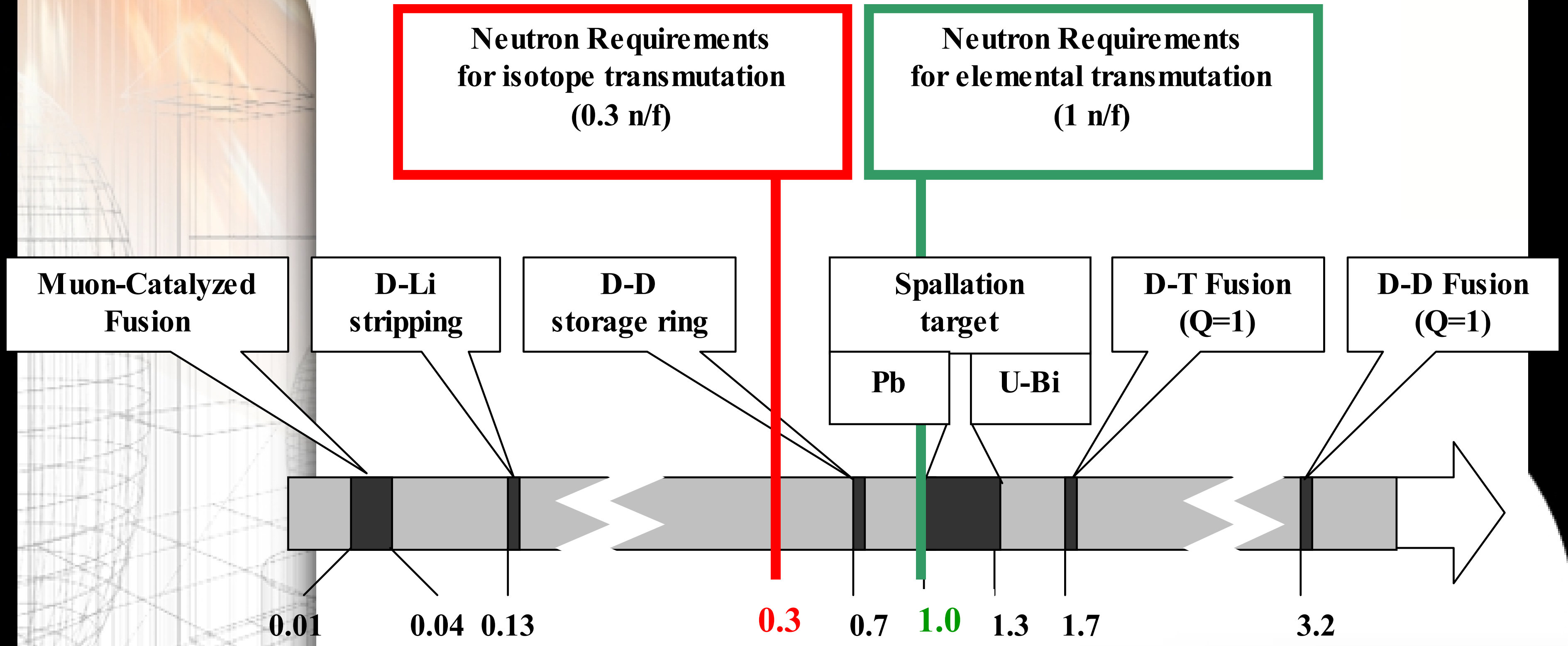


Neutron requirements (per one fission)

Thermal spectrum (fission U-235)	Fast spectrum (fission Pu-239)
0.93	**1.02**

Break-even transmutation requirements

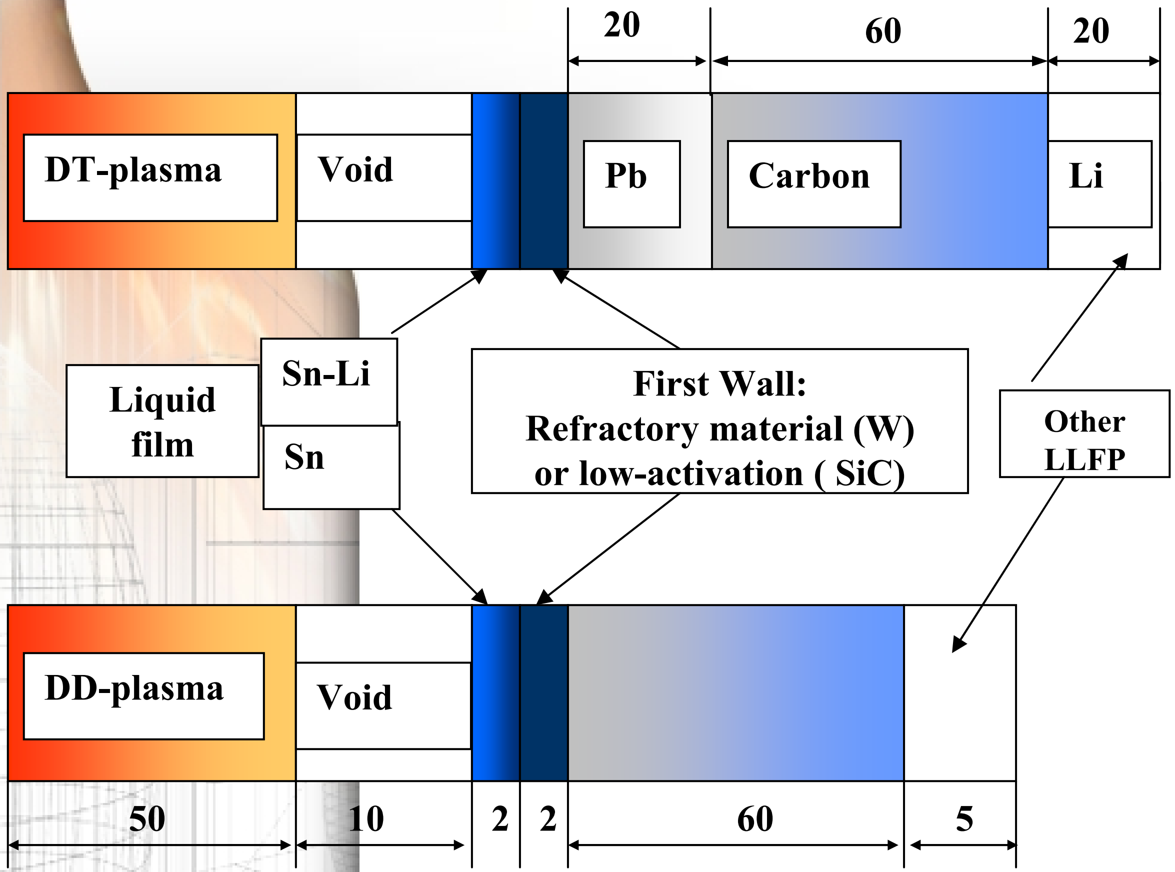
(maximum neutron production per one fission dedicated to neutron source)

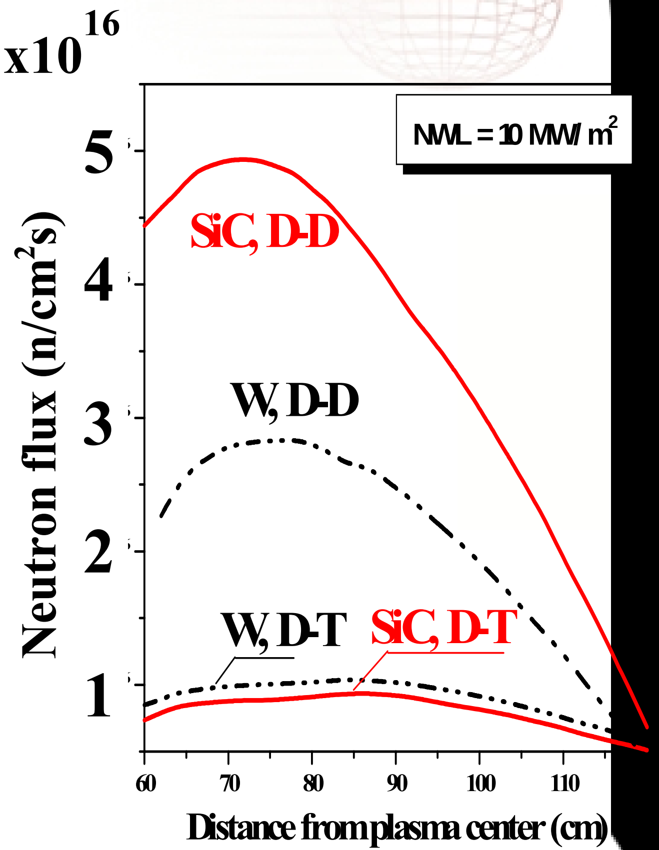


3. Transmutation in Fusion Neutron Source

- application of moderated spectra (transmutation through neutron capture)
- application of hard spectra (transmutation through (n,2n) reaction)

Principal layout of blanket concept for LLFP transmutation





* Targeted NWL in Advanced Power Extraction (APEX) project
M.ABDOU, Exploring Novel High Power Density Concepts for Attractive Fusion Systems, *Fusion Eng.Des*. **45**, 145 (1999))

Fusion-Driven Transmutation of Sn-126 (Fusion Neutron Source: NWL=10 МВт/м^2

Nuclide	$T_{1/2}$ (yr)	Effective half-life T_{eff} (yr)			
		Fast spectrum**	Thermal spectrum**	D-T	D-D
^{126}Sn	10^5	4.2×10^3	4.2×10^3	30.2	10.6
^{99}Tc	2.11×10^5	110	51	12.6	9.5
^{129}I	1.57×10^7	157	51.2	10.3	8.1

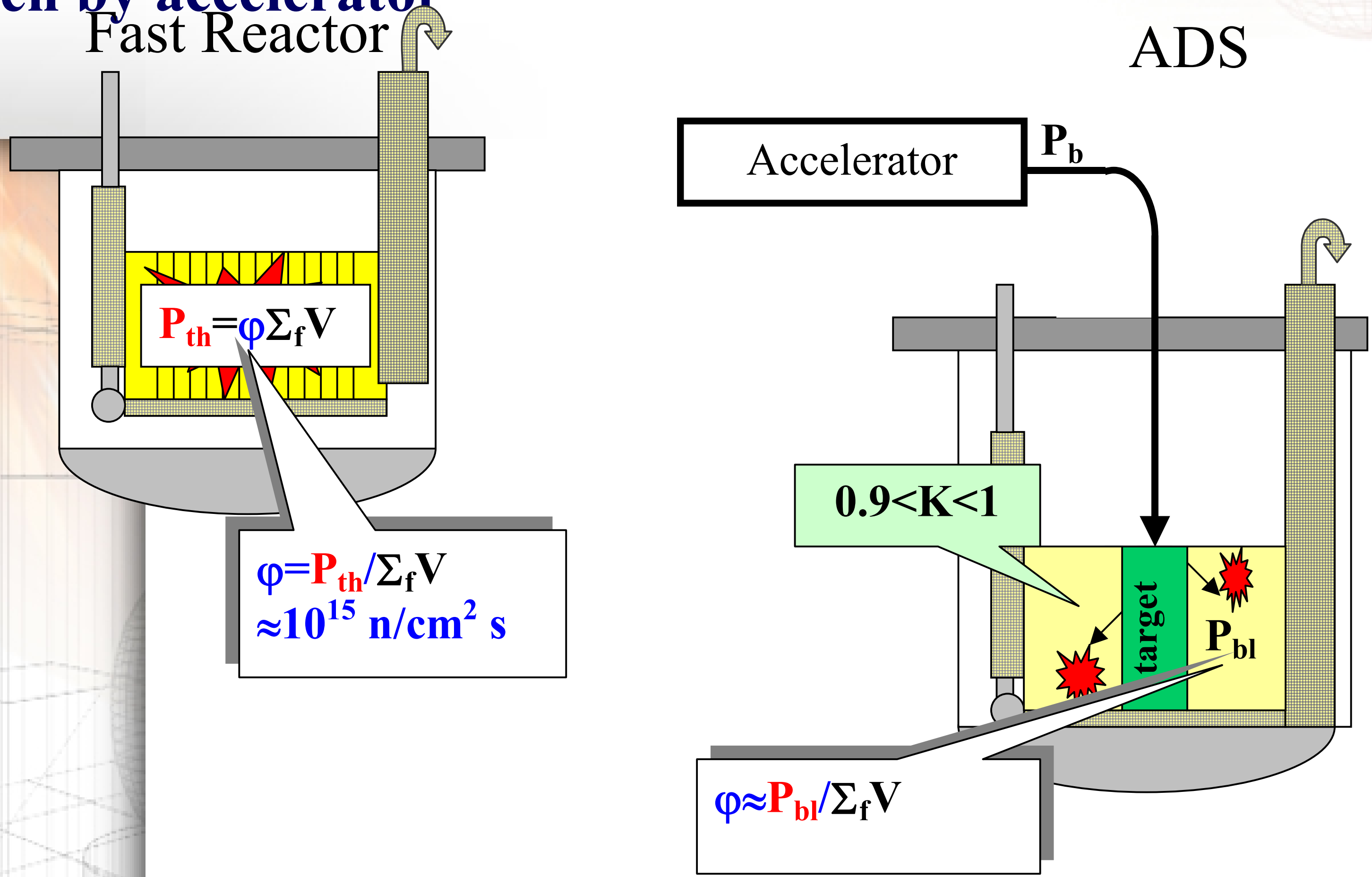
*) Elemental transmutation

**) thermal spectrum: average energy - 1 eV, neutron flux – 10^{14} n/(см^2·s)
fast spectrum,average energy– 200 кэВ, neutron flux - 10^{15} н/(см^2·с)

4. Transmutation in Accelerator Driven System

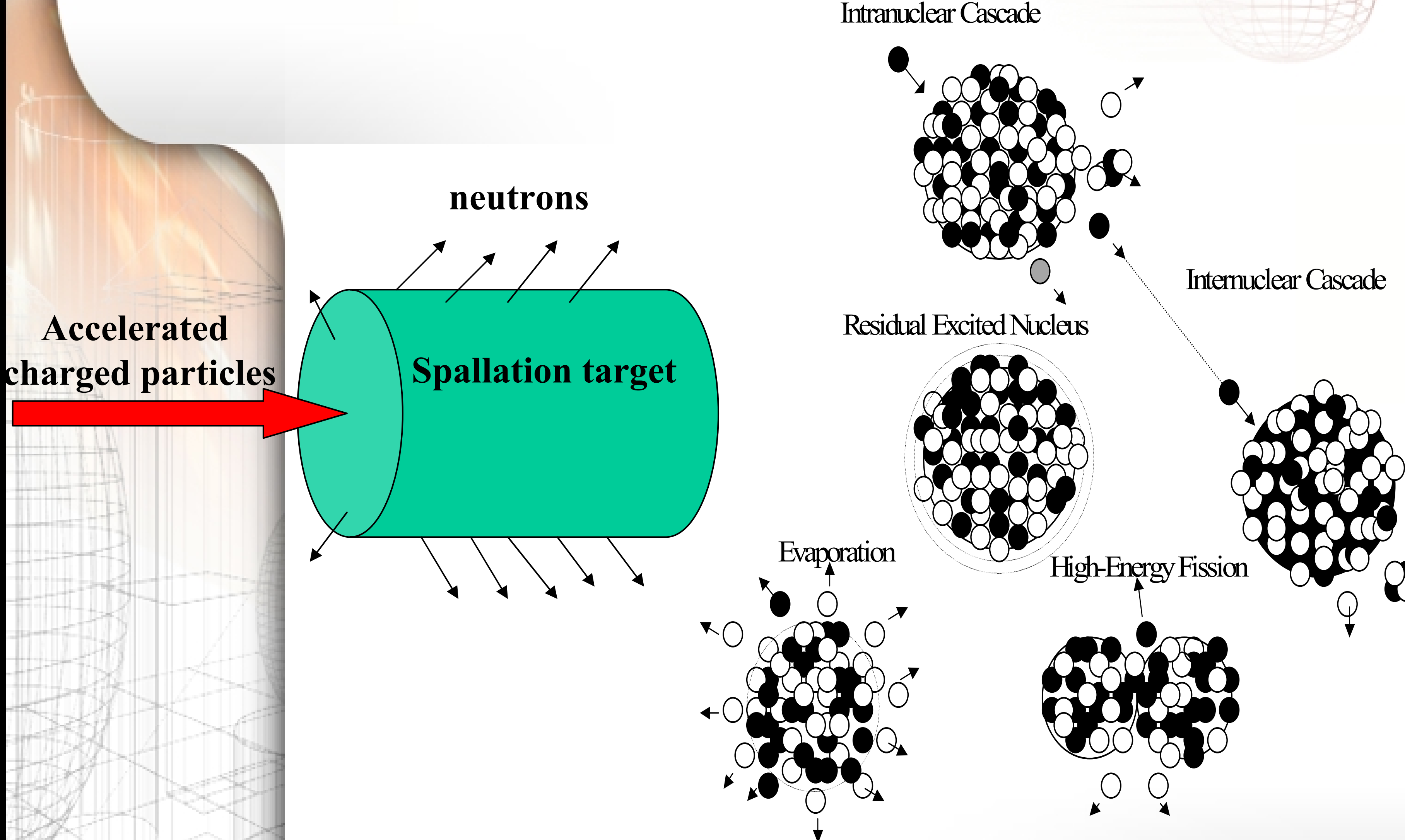
- Neutronics of the ADS components;
- Proton-induced transmutation in ADS target:
- stable nuclides to radioactive
- radioactive to stable

Transmutation environment in slightly subcritical blanket driven by accelerator



Transmutation environment in the ADS blanket similar to that of reactor

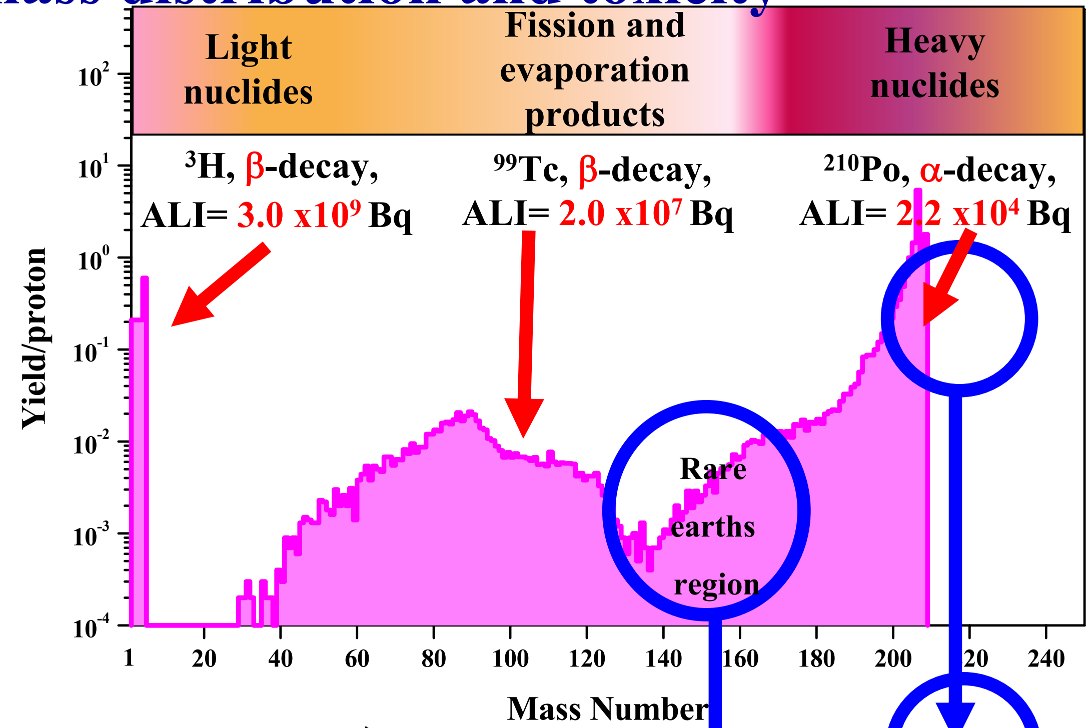
Neutron production in ADS

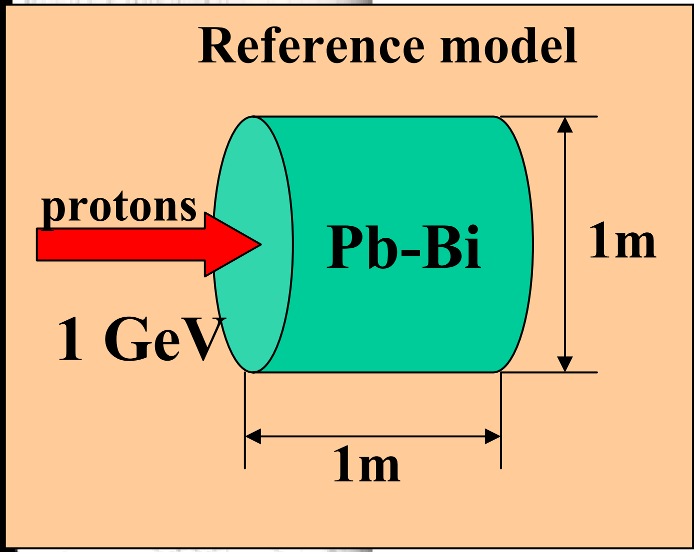


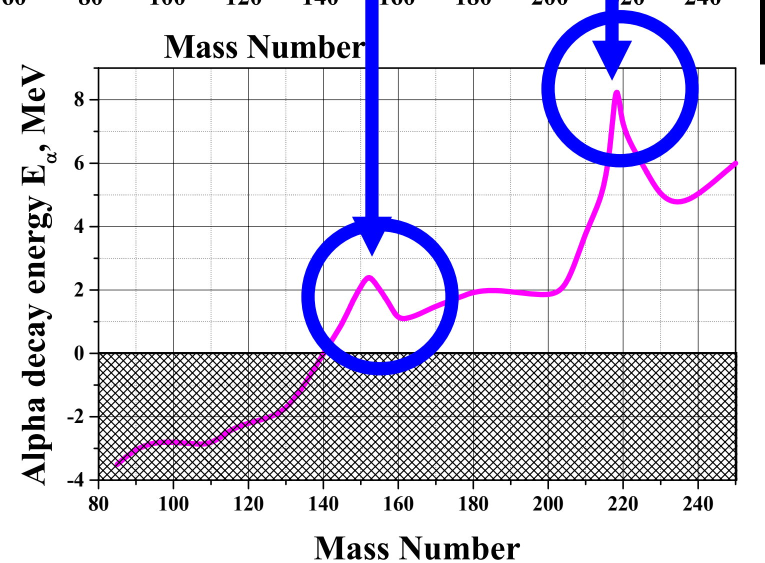
Spallation products: mass distribution and toxicity*

*Annual Limit on Intake (ALI) - is the annual intake of a given radionuclide by "Reference Man" which would result in either a committed effective dose equivalent of **5 rems** (stochastic ALI) or a committed dose equivalent of **50 rems** to an organ or tissue (non-stochastic ALI)**

** CFR.20.B - The Code of Federal Regulations, Part 20: Standards for Protection Against Radiation, Appendix B

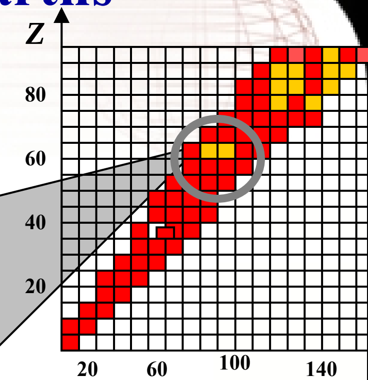






Fragment of Chart of the Nuclides Containing Rare Earths

^{149}Dy 4.20 m 0.49 s	^{150}Dy 7.17 m	^{152}Dy 2.38 h	^{153}Dy 6.4 h	^{154}Dy 3x10^{6}y	^{155}Dy 9.9 h
^{148}Tb 1.08 h 2.30 m	^{149}Tb 4.118 h 4.16 m	^{150}Tb 3.48 h 5.8 m	^{151}Tb 17.609 h 25 s	^{152}Tb 17.5 h 4.2 m	^{153}Tb 2.34 d
^{147}Gd 38.06 h	^{148}Gd 74.6 y	^{149}Gd 9.28 d	^{150}Gd 1.79x10^{6}y	^{151}Gd 124 d	^{152}Gd 1.1x10^{14}y
^{146}Eu 4.59 d	^{147}Eu 24.1 d	^{148}Eu 54.5 d	^{149}Eu 93.1 d	^{150}Eu 36.9 y 12.8 h	^{151}Eu 47.8
^{145}Sm 340 d	^{146}Sm 1.03x10^{8}y	^{147}Sm 1.1x10^{11}y	^{148}Sm 7.x10^{15}y	^{149}Sm 13.8	^{150}Sm 7.4
^{144}Pm 363 d	^{145}Pm 17.7 y	^{146}Pm 5.53 y	^{147}Pm 2.62 y	^{148}Pm 41.29d 5.370 d	^{149}Pm 2.212 d
^{143}Nd 12.18	^{144}Nd 2.3x10^{15}y	^{145}Nd 8.30	^{146}Nd 17.19	^{147}Nd 10.98 d	^{148}Nd 5.76



stable

$30d < T_{1/2} < 5x10^{8}y$

$10m < T_{1/2} < 30d$

$T_{1/2} < 10m$

$99.9\% < \varepsilon + \beta$

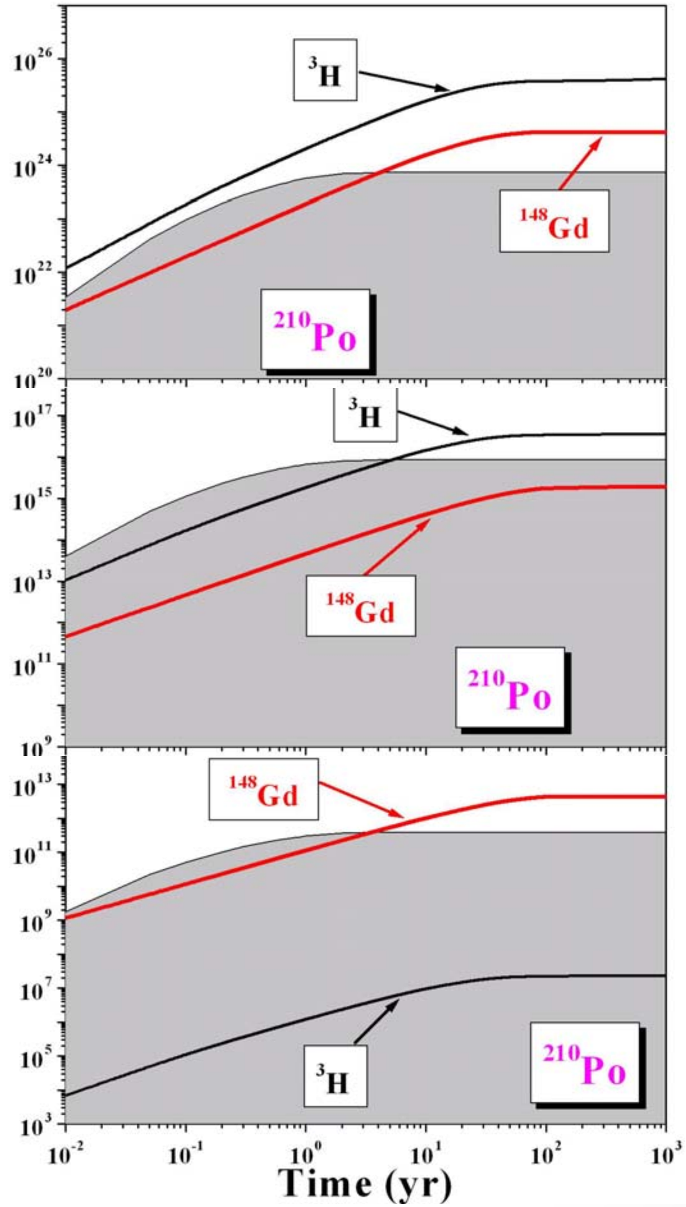
$99.9\% < \alpha$

Time-dependent accumulation of key nuclides in bare Pb-Bi target*

Inventory (at.)

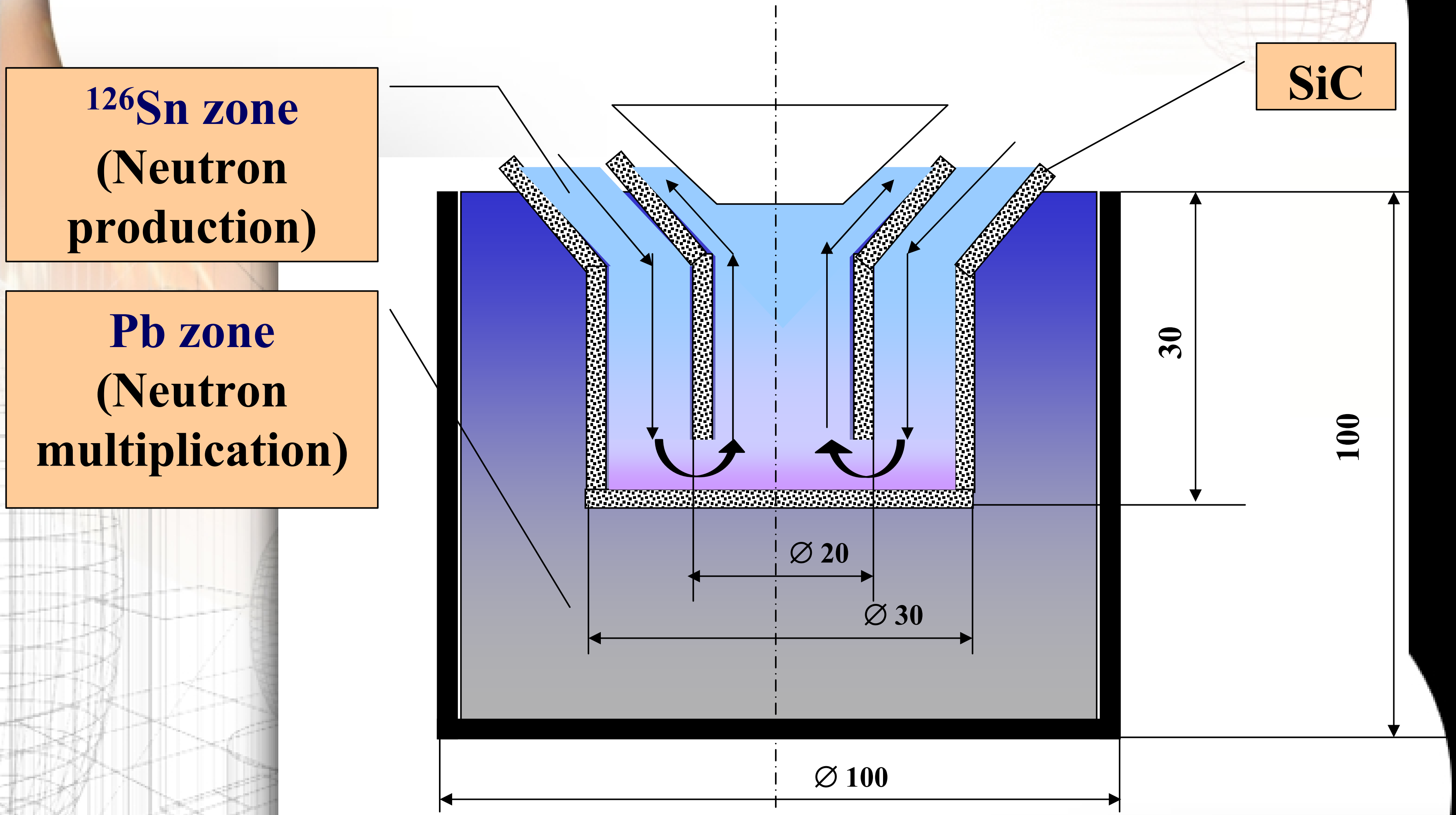
Activity (Bq)

Toxicity (ALI)

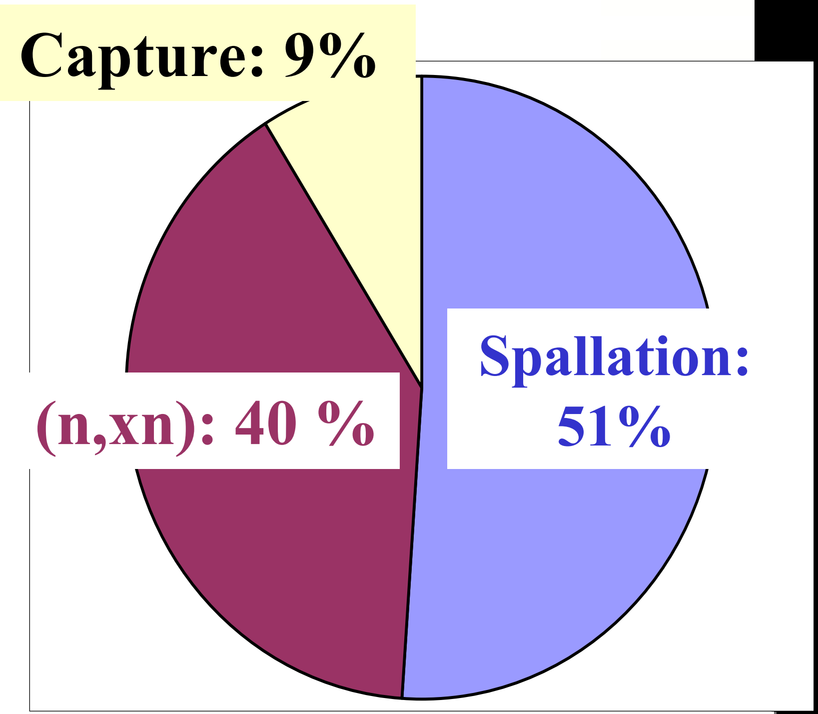


* diameter 1m, length 1 m.
Proton beam:
1 GeV, 50 mA

Conceptual target design module



Transmutation Characteristics of ^{126}Sn



<u>**Transmutation rate**</u>

1.8 ат.^{126}Sn per one initial proton

or

74.3 g/yr per 1 mA proton beam

Effective life-time of ^{126}Sn
(beam: 1 GeV, 10 mA)

60 yr

5.Summary

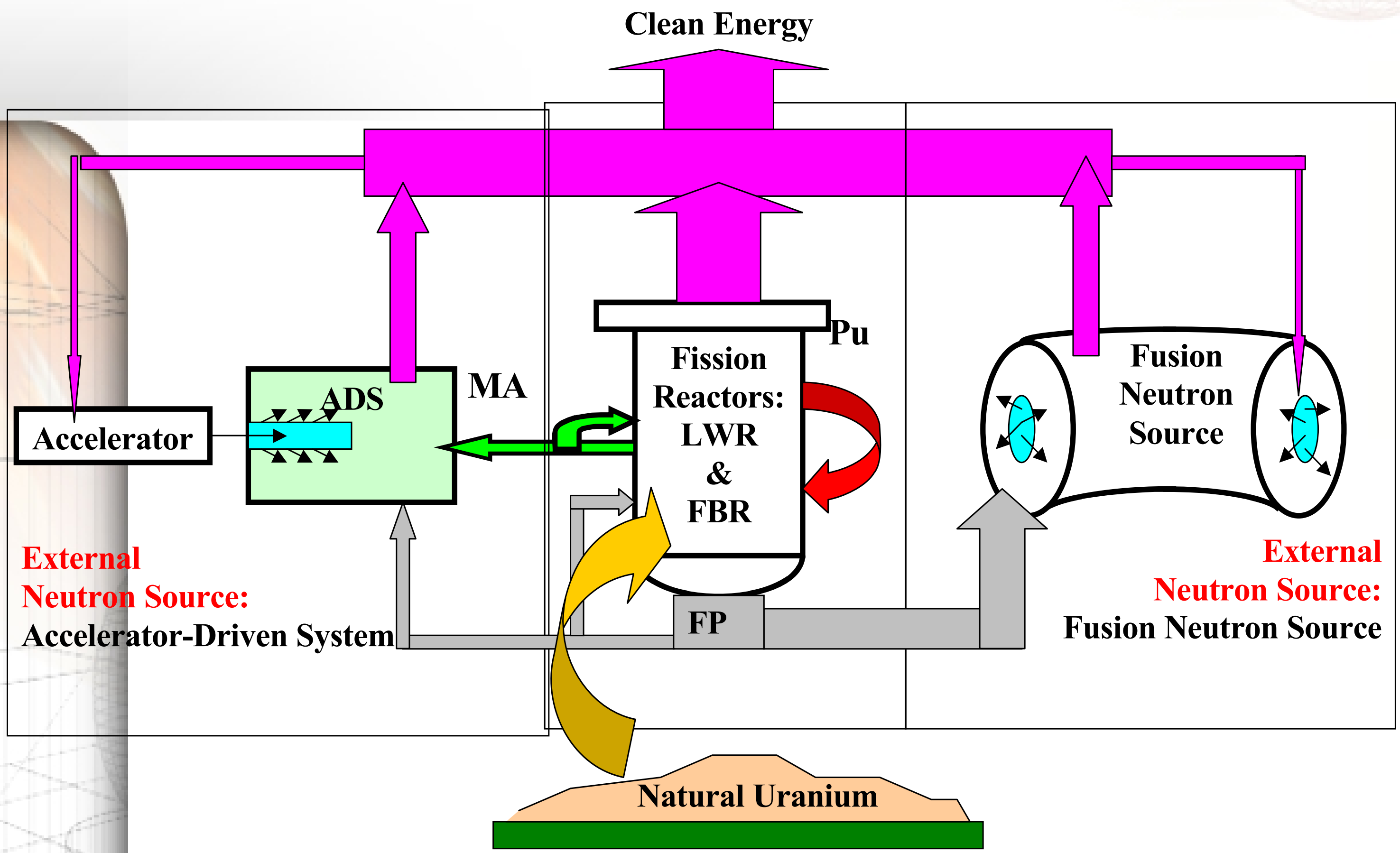
Introduction of advanced Accelerator and Fusion Driven waste transmuters might make the way to

establishment

of

wasteless nuclear energy system:

Image of Wasteless Nuclear Energy System



Freeman DYSON

”The fundamental problem of the nuclear industry is not reactor safety, not waste disposal, not the dangers of nuclear proliferation, real though all these problems are.

The fundamental problem of the industry is that nobody any longer has any fun building reactors.

… The spirit of the schoolhouse did not prevail….

And so the really good reactors were never invented”.

Disturbing the universe. (p.104-106)
Harper and row publication (1979)

Thank You

for Your attention…