Outline: Transmutation for Nuclear Waste Yacine Kadi

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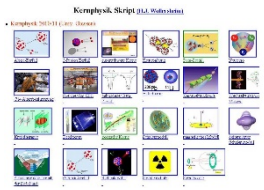
web-page: https://web-docs.gsi.de/~wolle/ and click on



1. The problem of nuclear waste
2. Nuclear reactor
3. Transmutation and incineration
4. Accelerator Driven System ADS

The basic process is nuclear transmutation

1919 Rutherford ($^{14}_{7}N + {}^{4}_{2}He \rightarrow {}^{17}_{8}O + p$) ^{210}Po accelerator!

1940 E.O. Lawrence/USA and W.N. Semenov/USSR proposed to use a particle accelerator as a neutron source

1941 G. Seaborg produced the first µg of ^{239}Pu with the Berkeley 60 inch cyclotron

1950 E.O. Lawrence proposed the Materials Testing Accelerator (MTA) at the Lawrence Livermore Radiation Lab, to produce 239Pu from Oak Ridge depleted Uranium

1952 W.B. Lewis in Canada proposed to use an accelerator to produce ^{233}U from Thorium for CANDU reactors (electro-breeder concept)

- MTA and Lewis‘ projects dropped or slowed down when
  a) rich Uranium deposits were discovered in USA, and
  b) it was realized that it required several hundred *mA* of beam intensity, hundred of *MW* to produce the beam

What is the nuclear waste transmutation

To transmute means to convert one element to another and by extension one isotope to another

The main physical process to perform useful transmutation is nuclear fission. One example of transmutation by fission can be:

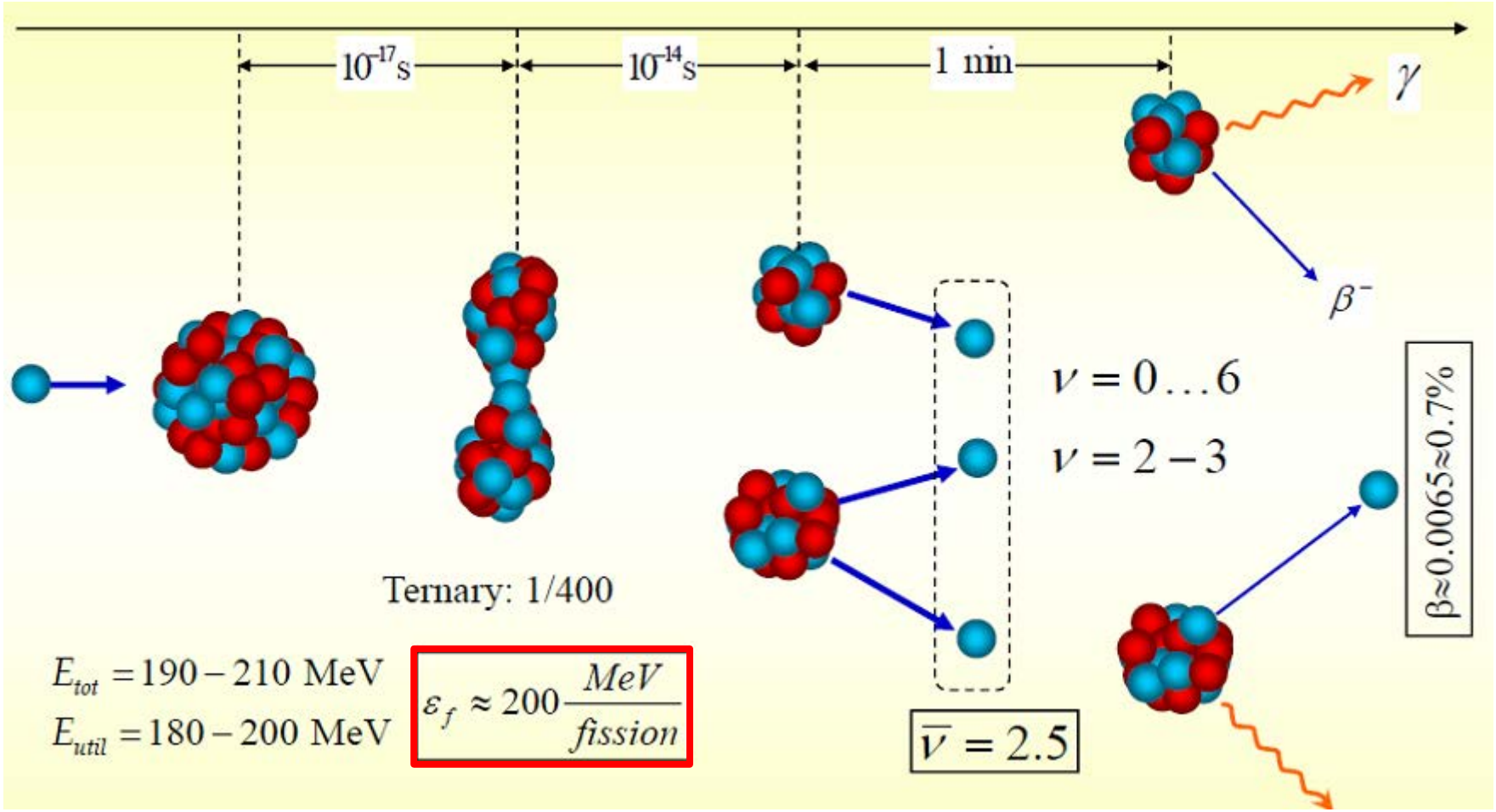
$$n + {}^{239}Pu\ (24000\ years) \rightarrow {}^{134}Cs(2\ years) + {}^{104}Ru(stable) + 2n + 200\ MeV$$

Sometimes one or several captures and decays must take place before fission can perform the useful transmutation:

$$n + {}^{240}Pu(6600\ years) \rightarrow {}^{241}Pu(14\ years)$$
$$n + {}^{241}Pu(14\ years) \rightarrow {}^{134}Xe(stable) + {}^{105}Rh(35\ hours) + 3n + 200\ MeV$$

$$n + {}^{241}Am(432\ years) \rightarrow {}^{242}Am(16\ hours)\ [capture]$$
$${}^{242}Am(16\ hours) \rightarrow {}^{242}Cm(163\ days)\ [\beta^- - decay]$$
$${}^{242}Cm(163\ days) \rightarrow {}^{238}Pu(88\ years)\ [\alpha - decay]$$
$$n + {}^{238}Pu(88\ years) \rightarrow {}^{142}Ce(stable) + {}^{95}Zr(64\ days) + 2n + 200MeV$$

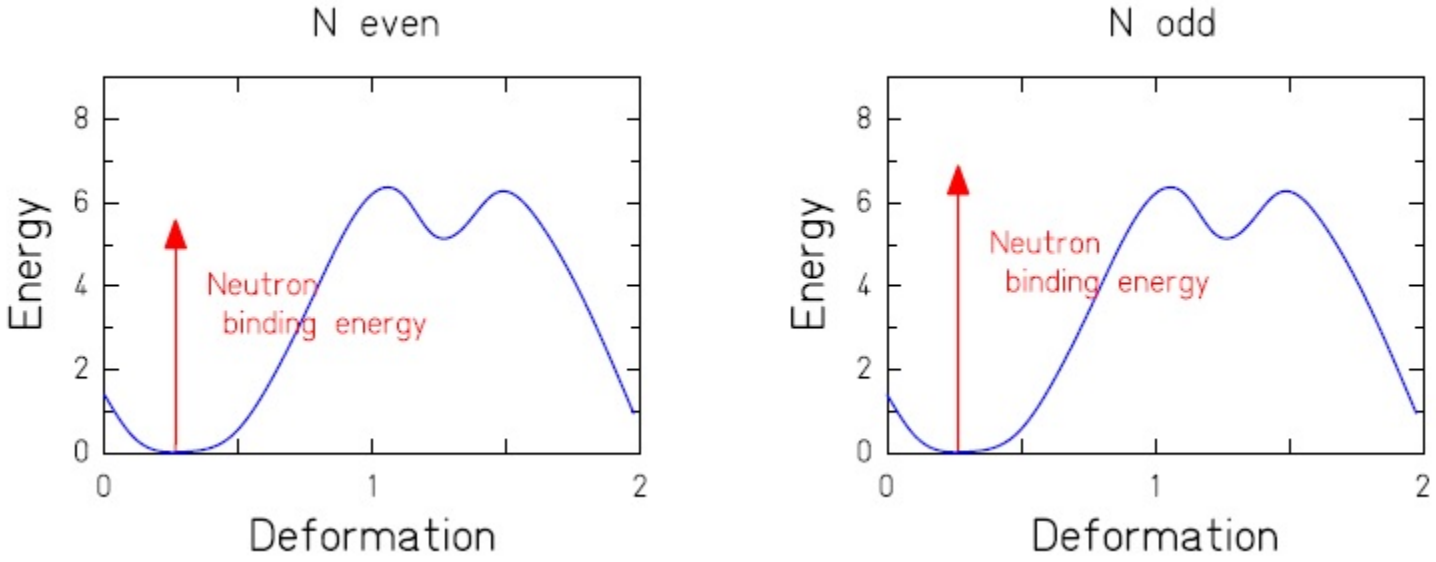
Characteristic properties of nuclear fission



Primordial heavy nuclei

^{232}Th: Z=90, N=142
^{234}U: Z=92, N=142 (0.0055%)
^{235}U: Z=92, N=143 (0.72%)
^{238}U: Z=92, N=146 (99.2745%)

excitation by thermal neutrons



Only nuclei with odd neutron number fission after capture of thermal neutrons.

Nuclei with even neutron number are fertile.
(By capture of thermal neutrons, a nucleus with odd neutron number is formed.)

The only natural nuclear fuel for conventional fission reactors is ^{235}U.

Absorption- and fission cross sections for neutrons

We start with thermalized neutrons,

η is the average number of fission neutrons per thermalized neutron.

$$\eta = \frac{\sigma_{fission}}{\sigma_{fission} + \sigma_{abs}} \cdot \langle \nu \rangle$$

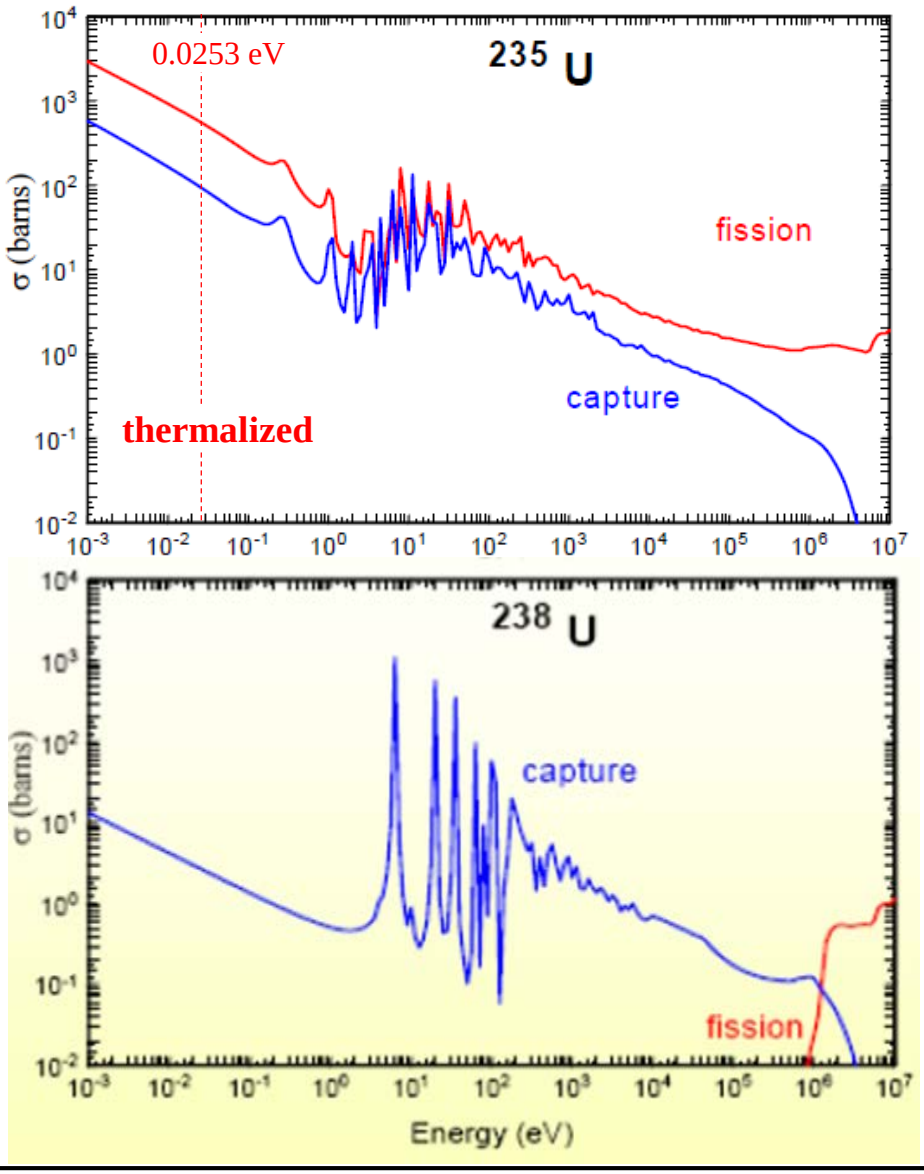
$$\sigma_{fission} = \frac{0.72}{100} \cdot \sigma_f\left(^{235}U\right) + \frac{99.28}{100} \cdot \sigma_f\left(^{238}U\right) = 4.20\,b$$

$$\sigma_{capture} = \frac{0.72}{100} \cdot \sigma_a\left(^{235}U\right) + \frac{99.28}{100} \cdot \sigma_a\left(^{238}U\right) = 3.43\,b$$

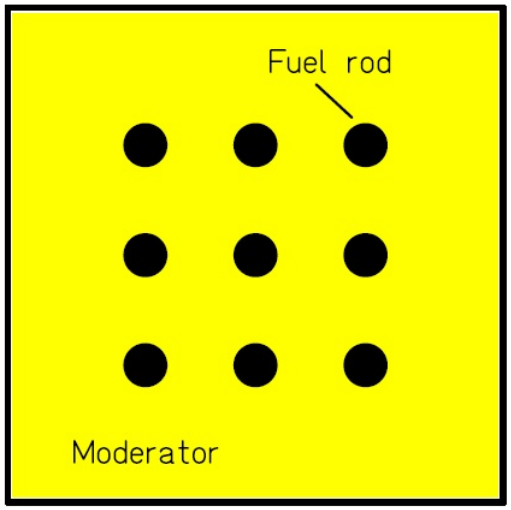
For **^{235}U**: σ_f = 584 b and σ_a = 97 b, <ν>=2.4
For **^{238}U**: σ_f = 0 b and σ_a = 2.1 b

Effective value of **η = 1.3** for natural Uranium is too small for chain reaction.

⟶ ^{235}U has to be enriched to 3% (η=1.8)



Nuclear reactor



Controlled chain reaction

"Fuel" ^{235}U enriched from 0,72% to 3,5%.

Fission → 2.2 to 3 neutrons,
exactly one of those induces another fission (criticality = 1)

Control of reactor possible due to delayed neutrons.

Variation of criticality by rods, absorbing neutrons.

Problem of nuclear waste

Which kind of waste?

Fission products

Plutonium —
Dangerous waste or fuel?

Minor actinides

The time scale

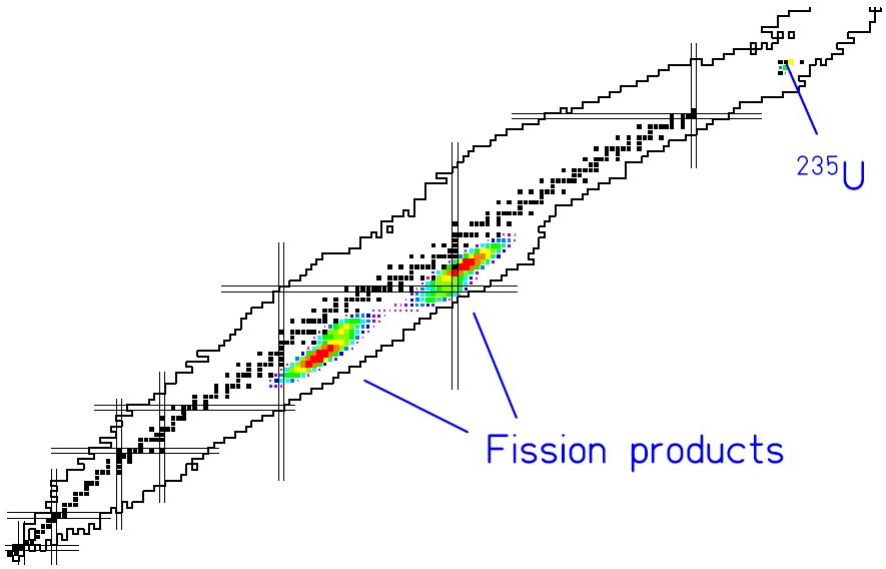
Possible solutions

Deposition — safe?, accepted?

Transmutation and incineration by nuclear reactions —
"Cleaning up" in reasonable time?

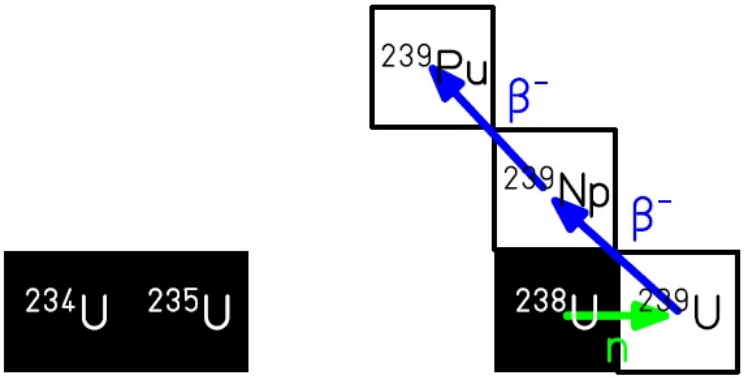
Nuclear reactions in the reactor

Fission:



^{129}I (15.7·10^{6} y)
^{107}Pd (6.5·10^{6} y)
^{135}Cs (2·10^{6} y)
^{93}Zr (1.5·10^{6} y) …

neutron-breeding:



Pu, Am, Cm, Np isotopes

List of the reaction products

- ^{235}U: Fuel (total reserve for ≈ 200 years)
- Fission products:

Isotope	lifetime	900 MW, 1 year
^{79}Se	70000 years	0.1 kg
^{93}Zr	1.5 million years	15.5 kg
^{99}Tc	210000 years	17.7 kg
^{107}Pd	6.5 million years	4.4 kg
^{126}Sn	10000 years	0.44 kg
^{129}I	15.7 million years	3.9 kg
^{135}Cs	2 million years	7.7 kg

- ^{239}Pu: Fuel or dangerous waste?

Isotope	lifetime	900 MW, 1 year
^{239}Pu	24119 years	123.1 kg

- Other actinides produced by breeding: many not fissile by neutrons

Balance: After a few years, the fuel is consumed.
Problem: Increased neutron capture → poisoning

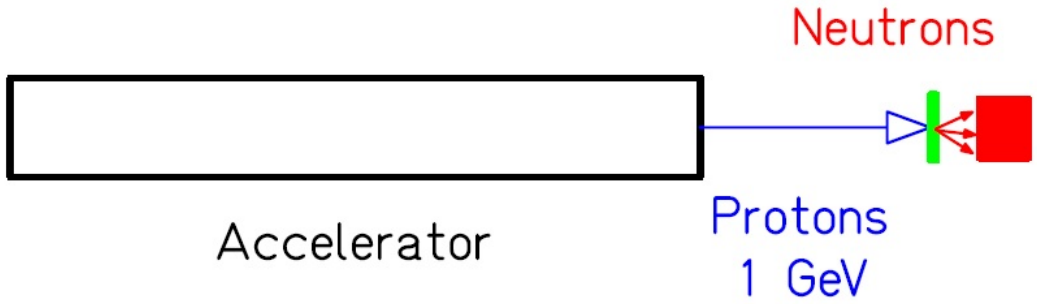
The spallation process

Several nuclear reactions are capable of producing neutrons

Nuclear Reactions	Incident Particle & Typical Energies	Beam Currents (part./s)	Neutron Yields (n/inc.part.)	Target Power (MW)	Deposited Energy Per Neutron (MeV)	Neutrons Emmitted (n/s)
(e,γ) & (γ,n)	e^- (60 MeV)	5×10^{15}	0.04	0.045	1500	2×10^{14}
$H^2(tn)He^4$	H^3 (0.3 MeV)	6×10^{19}	10^{-4} — 10^{-5}	0.3	10^4	10^{15}
Fission			≈ 1	57	200	2×10^{18}
Spallation (non-fissile target)	p (800 MeV)	10^{15}	14	0.09	30	2×10^{16}
Spallation (fissionable target)			30	0.4	55	4×10^{16}

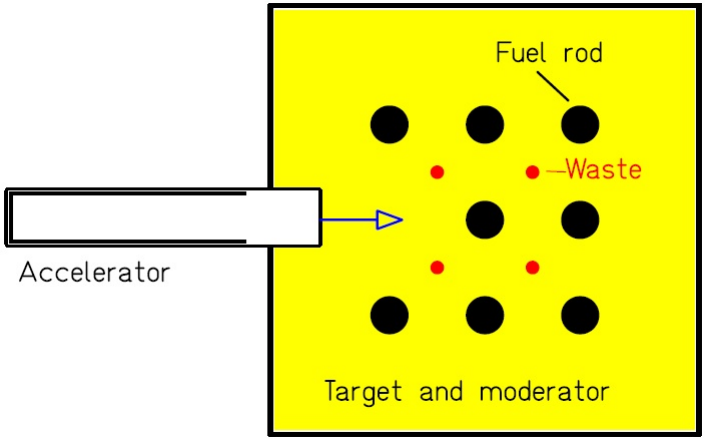
Transmutation of radioactive waste

- By spallation neutrons



high energy consumption

- By an hybrid reactor (accelerator driven system ADS)?

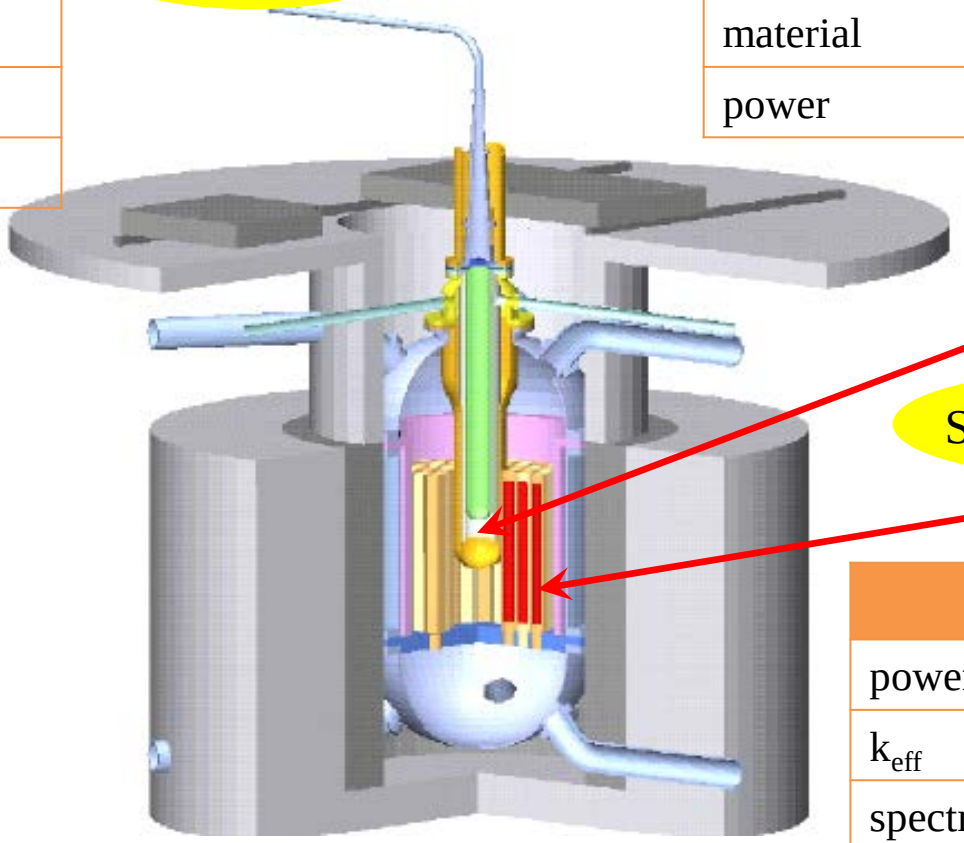


A subcritical reactor with additional neutrons produced by 1 GeV protons.

Transmutation with an accelerator driven system (ADS)

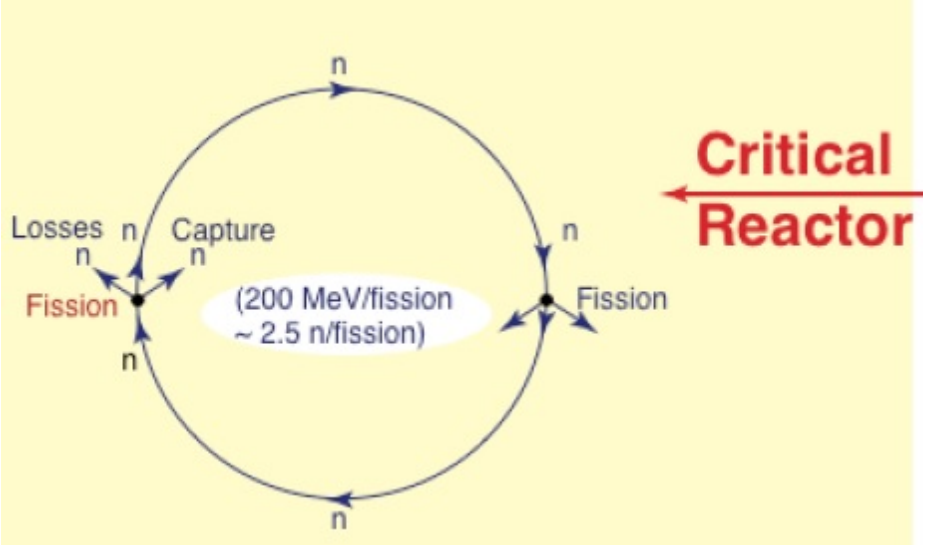
accelerator	
particles	protons
energy	600 MeV
current	2.4-4 mA
mode	CW
MTBF	> 250 h

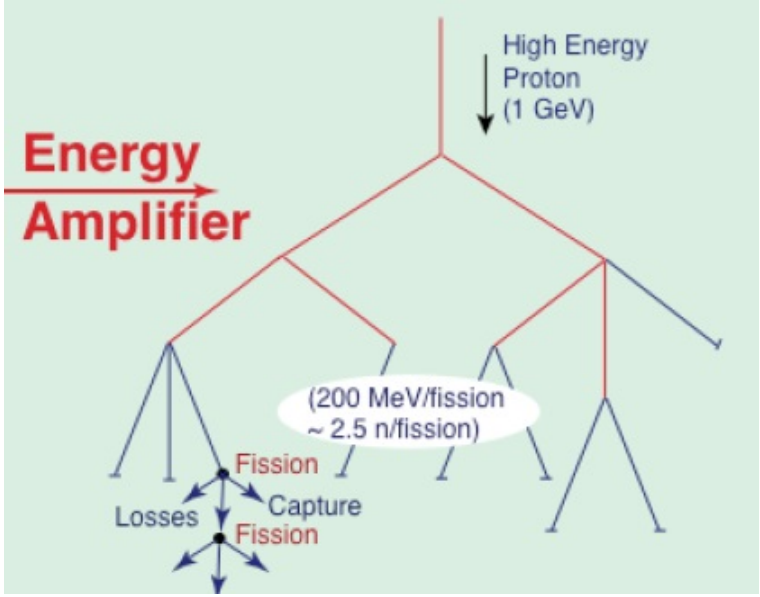
target	
main reaction	spallation
output	$2 \cdot 10^{17}$ n/s
material	LBE (**Pb**Bi coolant)
power	2.4 MW



reactor	
power	~85 MW_{th}
k_{eff}	0.95
spectrum	fast (flexible)
fuel	30-35% Pu MOX
coolant	LBE (PbBi)

Critical versus Sub-critical Systems





effective neutron multiplication factor

$$k = \frac{production}{absorption + losses}$$

$$energy\ gain(G) = \frac{energy\ produced\ by\ EA}{energy\ provided\ by\ beam} = \frac{G_0}{(1-k)}$$

self-sustained process: k=1

(if k<1 the reactor stops)
(if k>1 the reactor is super-critical

externally driven process: k<1 (k=0.98)
G_0=2.7 for Pb

$$E_{tot} = G \cdot E_p$$

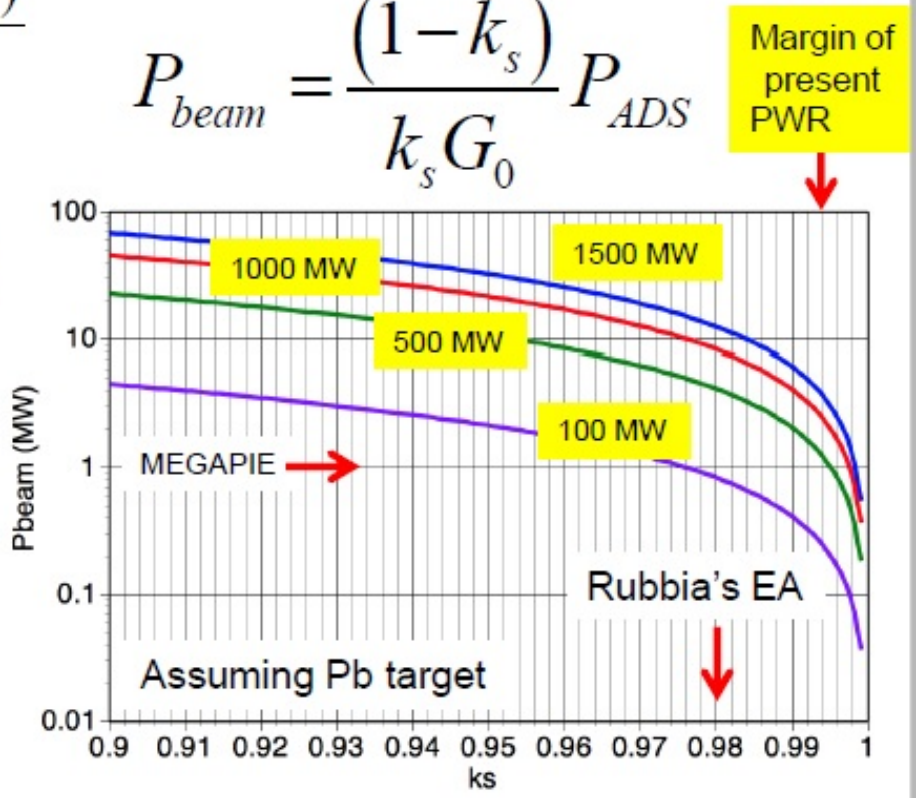
EA ≡ energy amplifier

Energy gain in ADS systems

$$G = \frac{G_0\left(E_b, Material, Geometry\right)}{1 - k_s}$$

- For a given power output, the energy gain (choice of k_s and G_0) determines the accelerator power. **Trade-off between accelerator power and criticality margin**
- Modulating the beam intensity allows variations in the power output (complementary with a fluctuating renewable energy source)
- **Neutronics with thorium very favourable compared to uranium** $t_{1/2}$ (^{233}Pa) ~ 27d; $t_{1/2}$ (^{239}Np) ~ 2.3d! What was a problem in the use of thorium in critical reactors becomes an advantage in the case of ADS

$$P_{beam} = \frac{\left(1 - k_s\right)}{k_s G_0} P_{ADS}$$

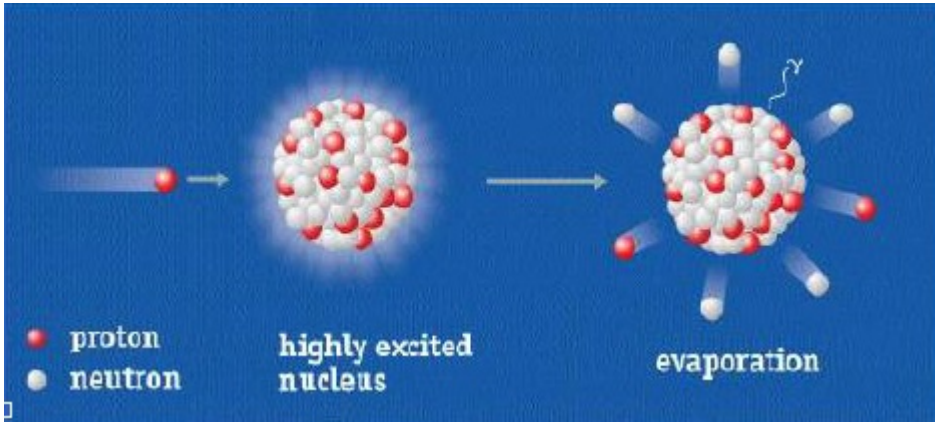


With PSI separate turns cyclotron
(3 mA and 1.8 MW, with 0.59 GeV protons).
P_{ADS} = 243 MW_{th} with k = 0.98
P_{ADS} = 486 MW_{th} with k = 0.99.

Neutron production

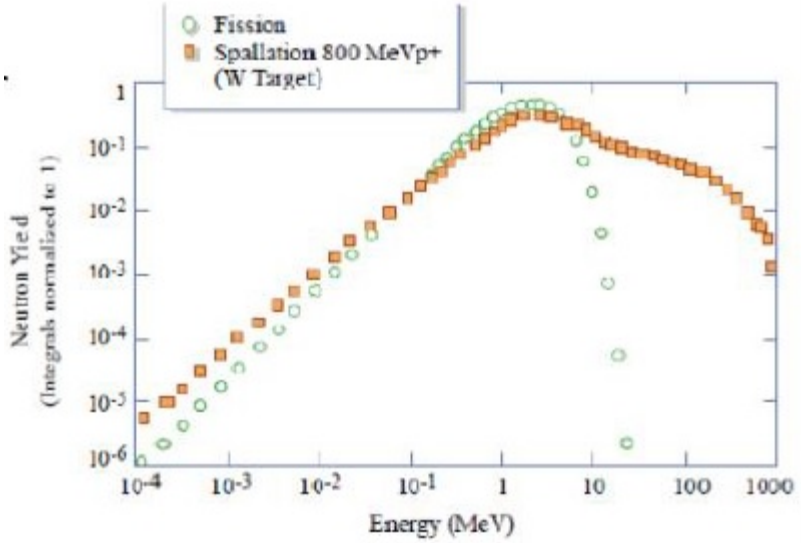
Neutrons are produced through the spallation process on heavy nuclei

- at 1 GeV, each proton produce 32 neutrons by spallation



Spallation: A nuclear process in which a high energy proton excites a neutron rich nucleus which decays sending out neutrons (and other particles)

The average energy deposited on the target, about 50 MeV/A, is lower than for deuteron induced nuclear processes.
Neutrons with a broad energy spectrum peaked at 1 MeV

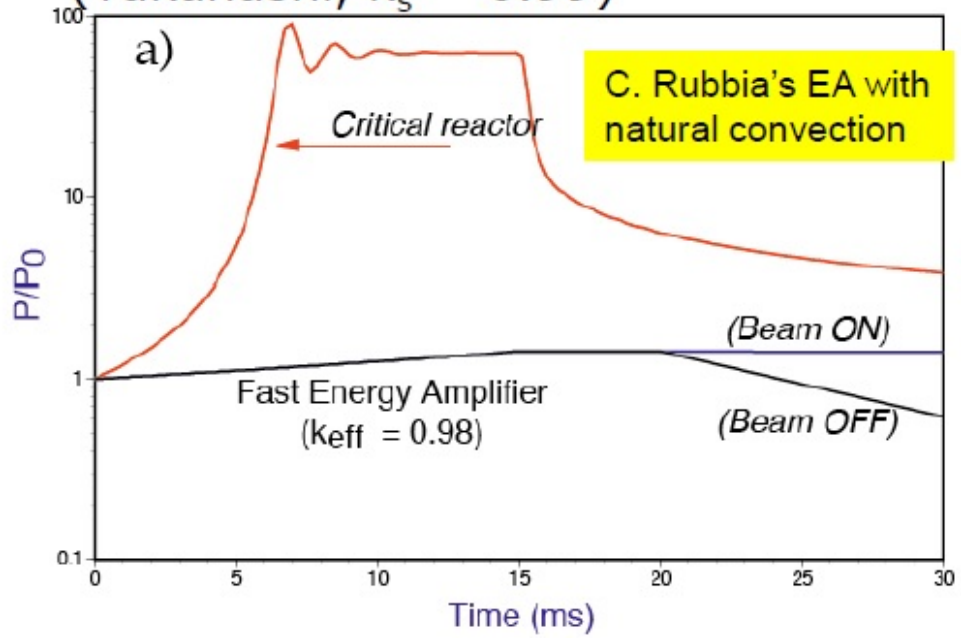


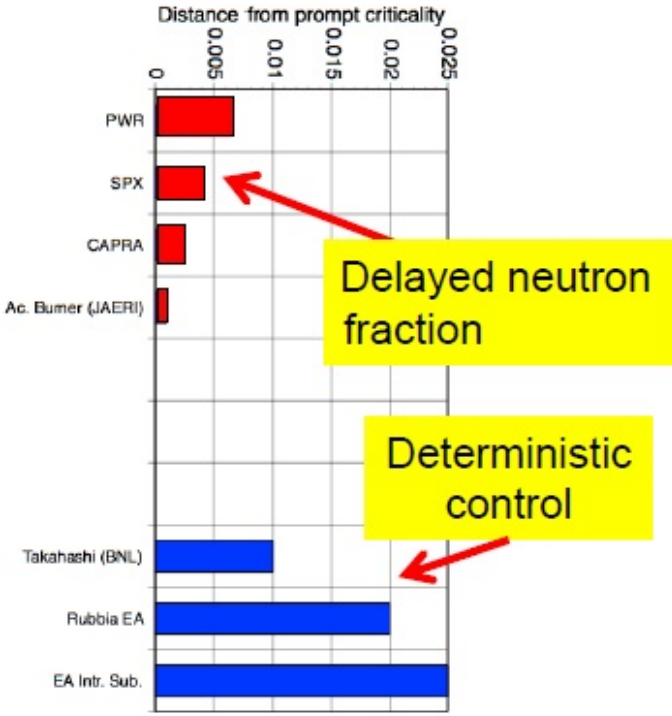
Guidelines for ADS parameters choice

- **Safety**:
  - Eliminate criticality accidents by making the system subcritical (void coef., T coef., β_{eff} no longer "critical" parameters)
    This requires an external proton source!
  - Operate system with passive safety elements to avoid core melting or limit its consequences, borrowing features from US advanced fast critical reactor designs;
  - Avoid dangerous coolants such as liquid sodium (use lead) Generation IV?
- **Waste management**:
  - Use (1) fast neutrons, (2) thorium fuel, and (3) recycle long-lived transuranic actinides (TRU) to minimize waste.
- **Military proliferation**:
  - Use thorium fuel (small Pu prod., ^{233}U very difficult mixture)
  - Avoid Pu separation (Purex), use pyroelectro reprocessing instead (developed for uranium at Argone N.L.)

Physics of subcritical systems

- Subcritical systems are insensitive to delayed neutron fraction (β); **safety margin** (distance from prompt criticality) **is a design choice**, it is not imposed by Nature!
- k_s = 0.975 makes the system subcritical under all conditions (after ^{233}Pa decay)
- The reactivity changes only very slowly; the beam can be switched off very quickly, reducing k_s to k_{eff}. It is possible to choose a higher k_s in order to reduce the load on the accelerator (Takahashi, k_s = 0.99)





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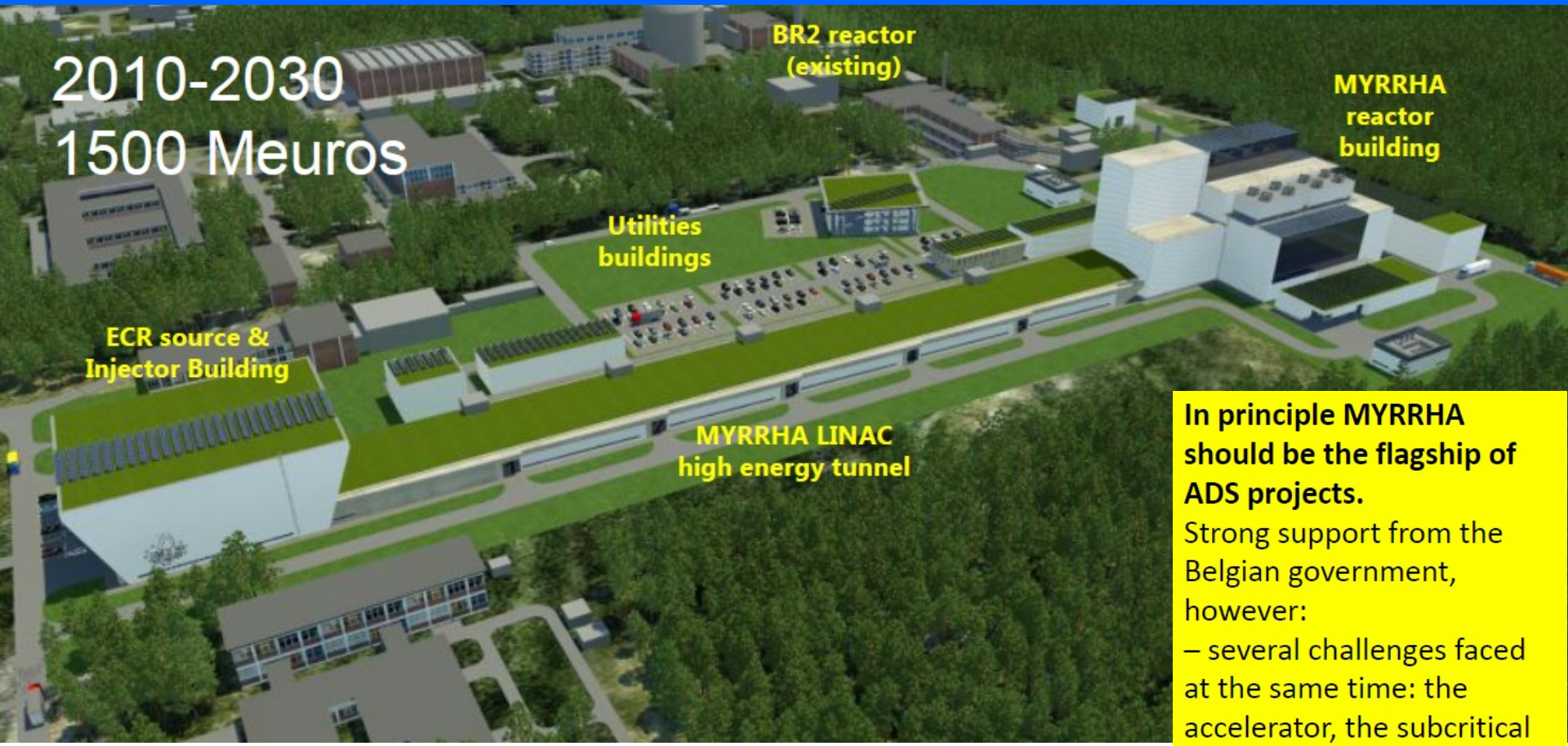
Myrrha project



In principle MYRRHA should be the flagship of ADS projects.

Strong support from the Belgian government, however:

– several challenges faced at the same time: the accelerator, the subcritical core and their coupling

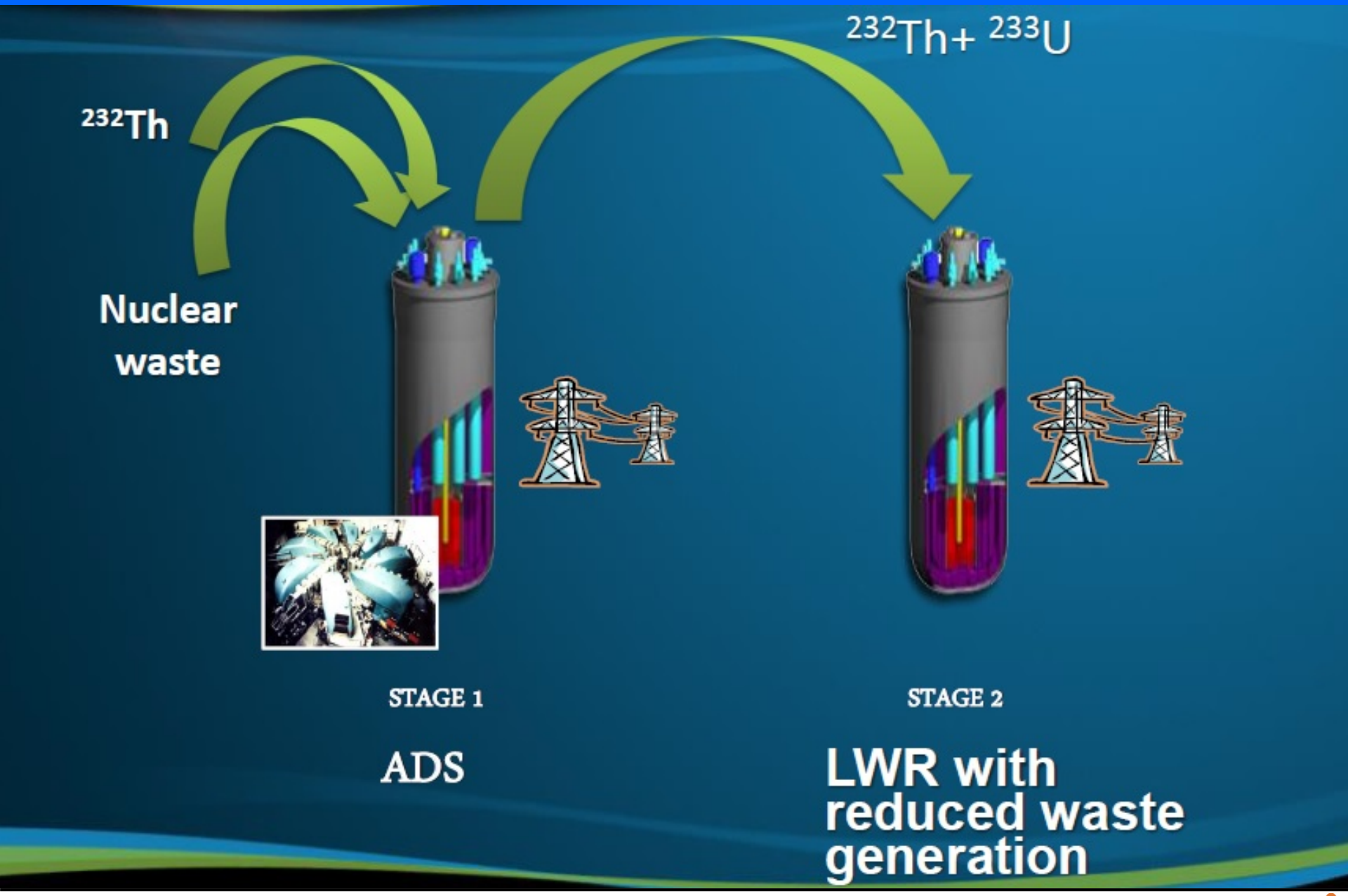
– no thorium in the plans

– only partially funded

– not before 2025?

– will not remain an ADS, and will be turneds into a critical reactor

Myrrha project



The Hybrid System (ADS)

Principle:

- **Slightly under-critical**
- **Controllable by spallation neutrons, produced by 1 GeV protons**

Advantages:

Can be operated with $^{232}Th \rightarrow {}^{233}U$

- **^{232}Th reserves sufficient for 21000 years**

Supports some ´poisoning´

- **Can transmute or incinerate nuclear waste**
- **Is insensitive to some variation of the criticality during long operation**
- **Long operation of fuel rods**

Problems:

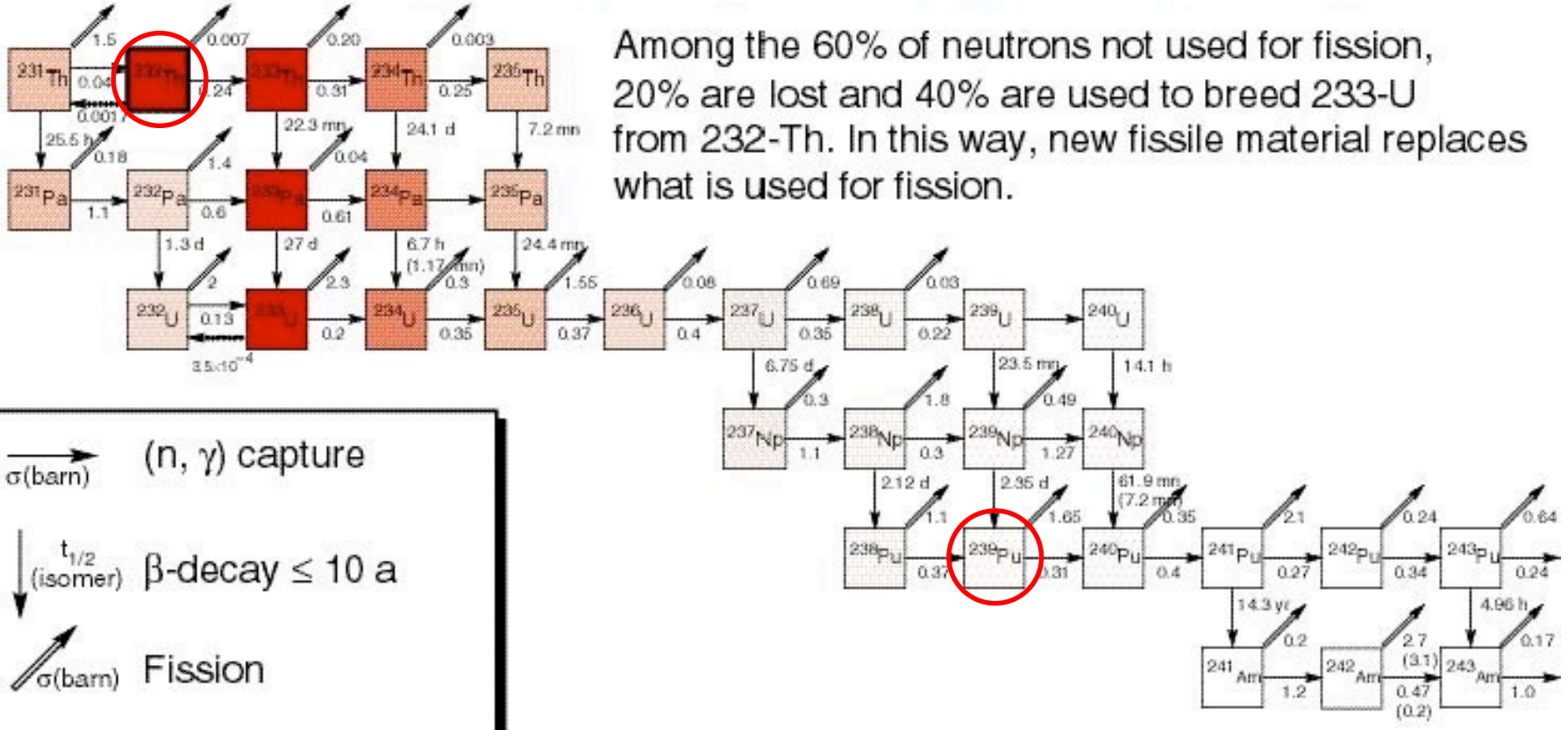
Nuclear reactions up to 1 GeV must be known

- **Yield of spallation neutrons**
- **Production of radioactive nuclei by spallation**
- **Material damages due to irradiation**

The Hybrid System (ADS)

CHOICE OF FUEL: Thorium [$^{232}ThO_2$ (+ $^{233}UO_2$)]

$$n + {}^{232}Th(1.4\times10^{10}\ a) \rightarrow {}^{233}Th(22.3\ m) \rightarrow {}^{233}Pa(27\ j) \rightarrow {}^{233}U(1.6\times10^{5}\ a)$$



Among the 60% of neutrons not used for fission, 20% are lost and 40% are used to breed 233-U from 232-Th. In this way, new fissile material replaces what is used for fission.

σ(barn) (n, γ) capture

$t_{1/2}$ (isomer) β-decay ≤ 10 a

σ(barn) Fission

(n, 2n) (E_n: ≥ 6 MeV)

Thorium is a major asset to minimize waste production

It takes 7 neutron captures to go from ^{232}Th to ^{239}Pu

Pu equilibrium concentration ~ 10^{-4} as opposed to 15% in U-Pu system

Precision Measurements of isotopic yields

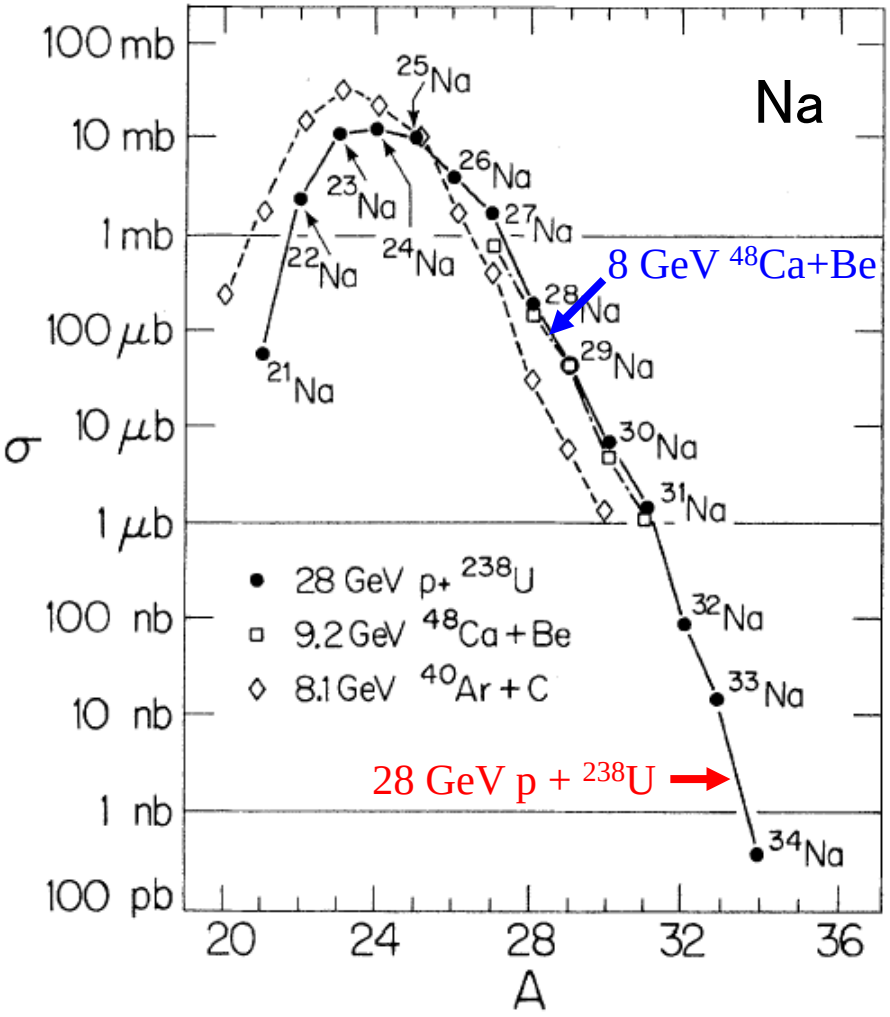
- ^{197}Au (800 A MeV) + ^{1}H
- ^{208}Pb (1 A GeV) + ^{1}H
- ^{208}Pb (1 A GeV) + ^{2}H
- ^{208}Pb (500 A MeV) + ^{1}H
- ^{238}U (1 A GeV) + ^{1}H
- ^{238}U (1 A GeV) + ^{2}H
- ^{238}U (1 A GeV) + ^{208}Pb

Development of nuclear-reaction models

Proton- versus heavy-ion induced reactions

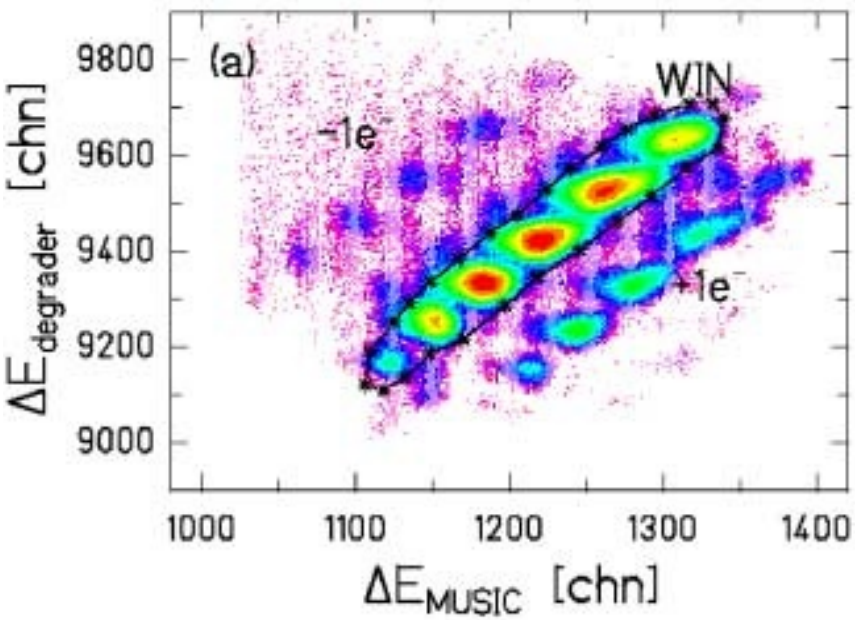
Proton- and heavy-ion induced reactions give very similar isotope distribution:

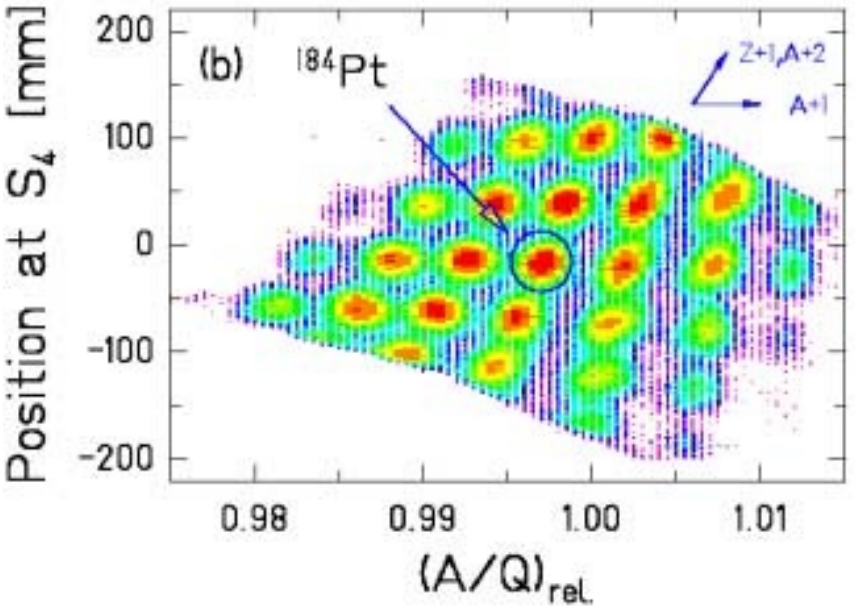
Target fragmentation:	GeV p + A_{target} → A
Projectile fragmentation:	GeV/u A_{proj} + p → A
are equivalent	



Identification Pattern

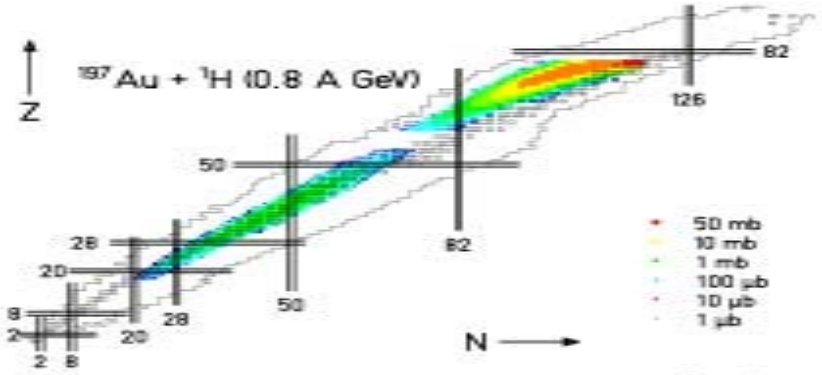
$^{208}Pb + {}^{1}H$, 1GeV/A

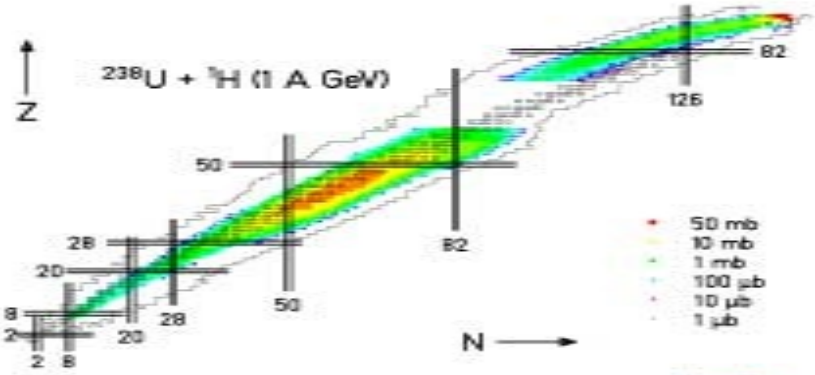


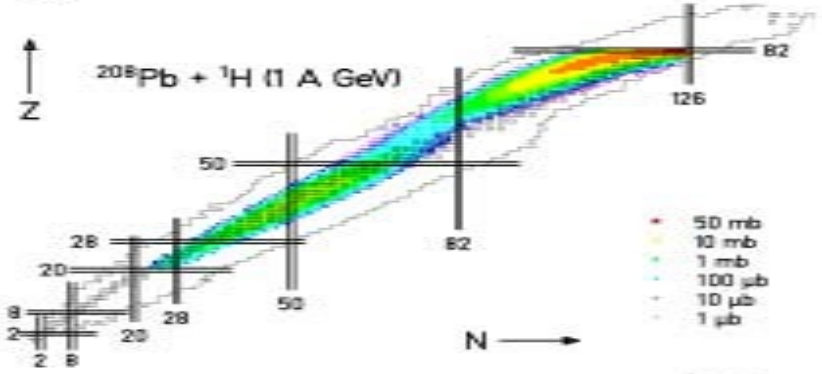


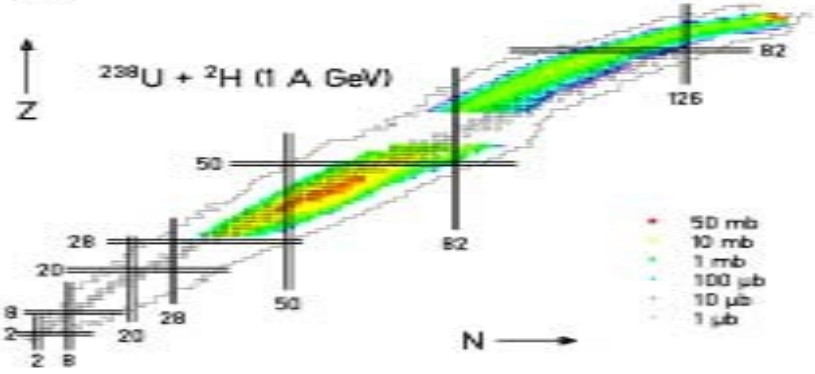
T. Enqvist et al., Nucl. Phys. A686 (2001), 481

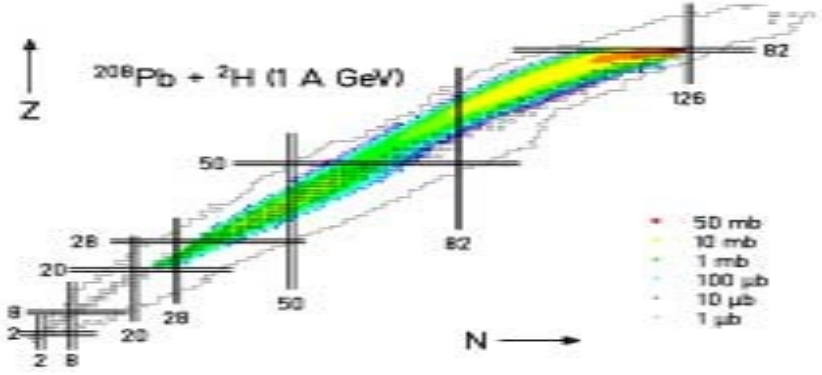
Production cross sections











F. Rejmund et al. NPA 683 (2001) 540
J. Benlliure et al. NPA 683 (2001) 513
T. Enqvist et al. NPA 686 (2001) 481
T. Enqvist et al. NPA 703 (2002) 435
M. Bernas et al. to be published
PhD: B. Mustapha, E. Casarejos, J. Taieb, M.V. Ricciardi, J. Pereira