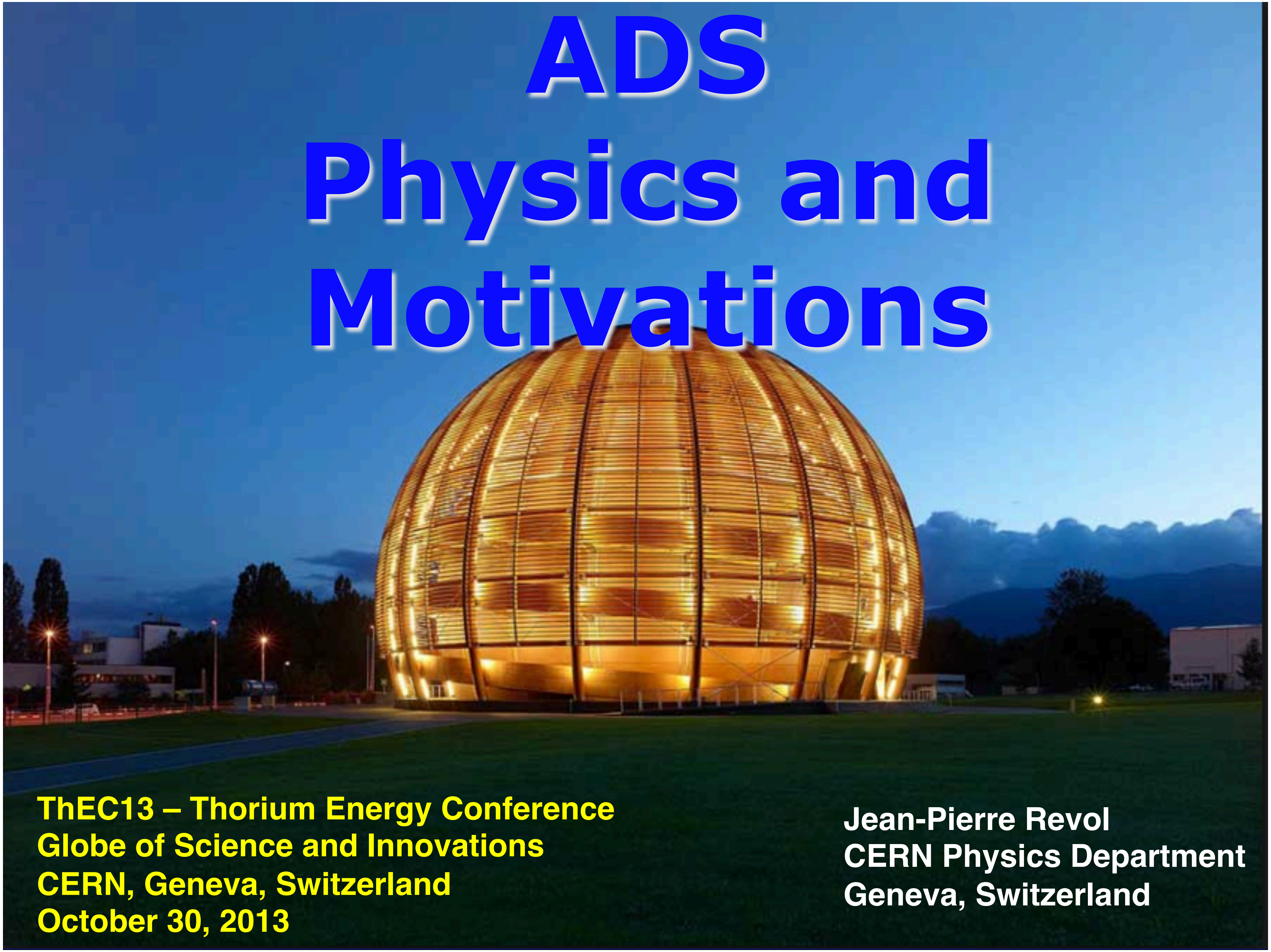
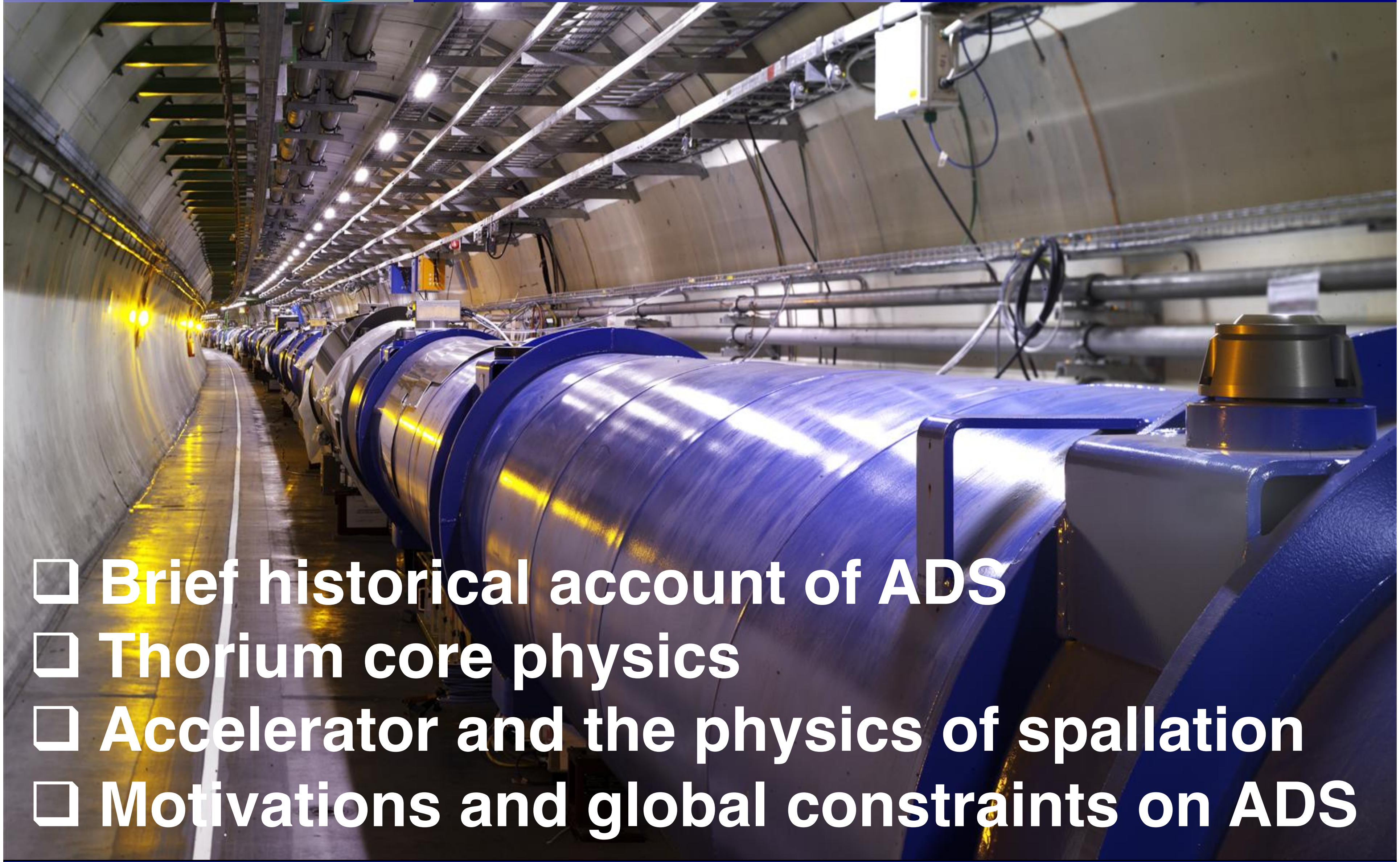


# ADS Physics and Motivations

**ThEC13 – Thorium Energy Conference**
**Globe of Science and Innovations**
**CERN, Geneva, Switzerland**
**October 30, 2013**

**Jean-Pierre Revol**
**CERN Physics Department**
**Geneva, Switzerland**

# Outline



- ❑ Brief historical account of ADS
- ❑ Thorium core physics
- ❑ Accelerator and the physics of spallation
- ❑ Motivations and global constraints on ADS

# Accelerator Driven Systems

## ☐ The basic process in ADS is nuclear transmutation

- 1919 Rutherford ($^{14}N_7 + {}^4He_2 \rightarrow {}^{17}O_8 + {}^1p_1$) **$^{210}$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 $^{239}$Pu 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)


Keystone

# Accelerator Driven Systems

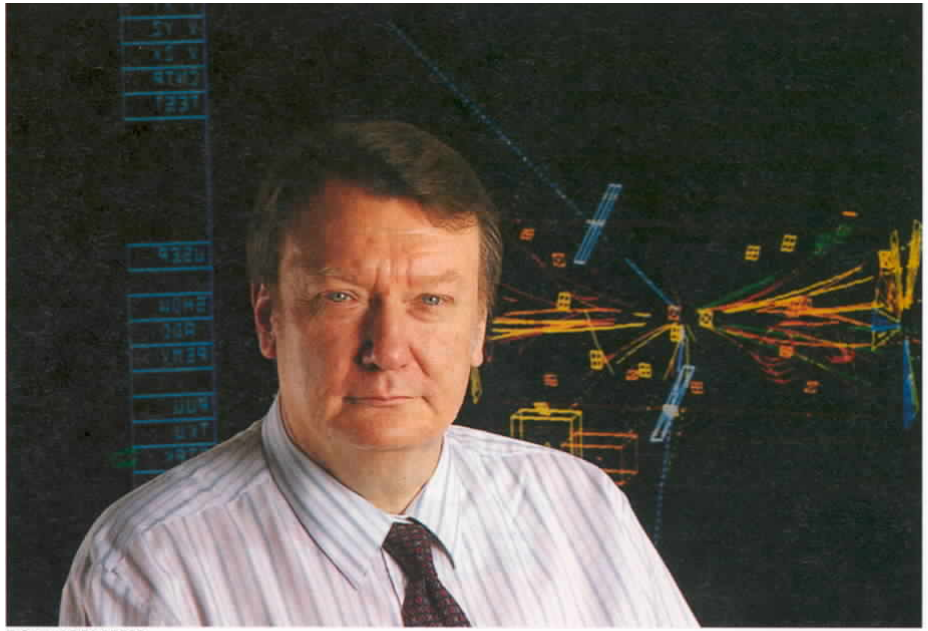
- MTA and Lewis' projects dropped or slowed down when (a) rich uranium deposits were discovered in the USA, and (b) it was realized that it required several hundred mA of beam intensity, hundreds of MW to produce the beam! [*Pu, no amplification*]

  **today ≤ 10 MW beams seem sufficient**

- Renewed interest in ADS in the 1980's, when the USA decided to slow the development of fast critical reactors (Fast Flux Test Facility @ Argone National Lab.):
  - H. Takahashi at Brookhaven National Lab: several proposals of ADS systems (PHOENIX), including the **idea of burning minor actinides** (Fast neutrons – $k_s$~0.99);
  - Ch. D. Bowman at Los Alamos: thermal neutron ADS (**ATW**) with thorium & chemistry on-line for FP and $^{233}$Pa extraction;
  - Japan launches **Options for Making Extra Gains from Actinides** (OMEGA, now JPARC) at JAERI (now JAEA).

# Accelerator Driven Systems

- ☐ In the 1990s, Carlo Rubbia gave a major push to the ADS, by launching a vigorous research programme at CERN based on:
  - ☛ development of **innovative simulation** of nuclear systems
  - ☛ specific **experiments to test basic concepts** (FEAT, TARC)
  - ☛ construction of an **advanced neutron Time of Flight facility** (n_TOF) to acquire neutron cross-section data, crucial to simulate reliably any configuration with new materials (talk by F. Gunsing)



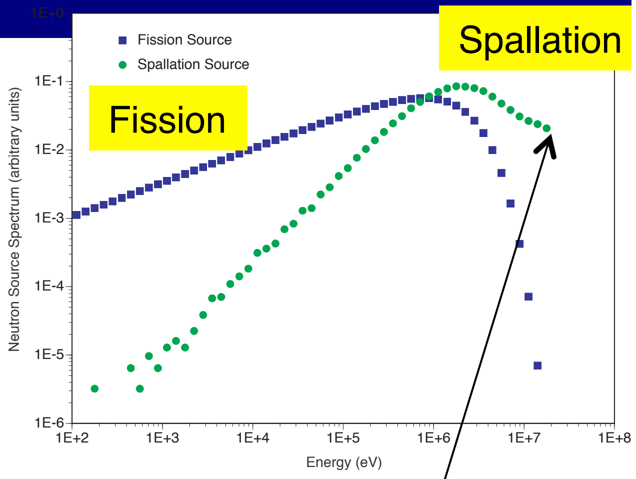
P. Stumpf/SIPA PRESS

Followed by proposals for demonstrators (EU and Italy)

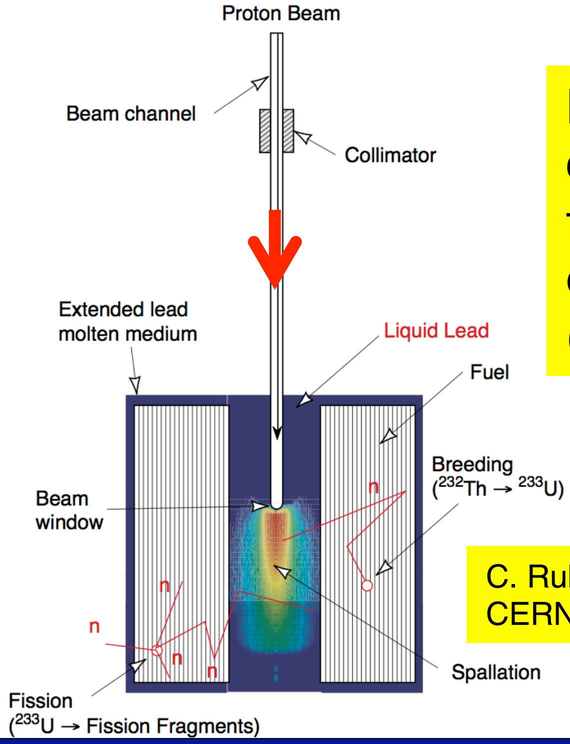
**C. Rubbia triggered a major R&D effort on ADS worldwide**

# Basic elements of an ADS system

- ❑ A particle **accelerator**, to provide a **neutron source**
- ❑ A **core** in which both source neutrons and fission neutrons are at work – restricted here to the case of a **moderator** allowing for a fast neutron spectrum
- ❑ **Two main areas of physics:**
  - ☛ Neutron production by spallation from the beam
  - ☛ Neutron transport and interaction in the core
- ❑ **Physics also drives other ADS elements:**
  - ☛ Cooling (possibility of natural convection)
  - ☛ Electric power production efficiency (go to highest possible temperature)



Non-negligible contribution from the high energy tail (n,xn)

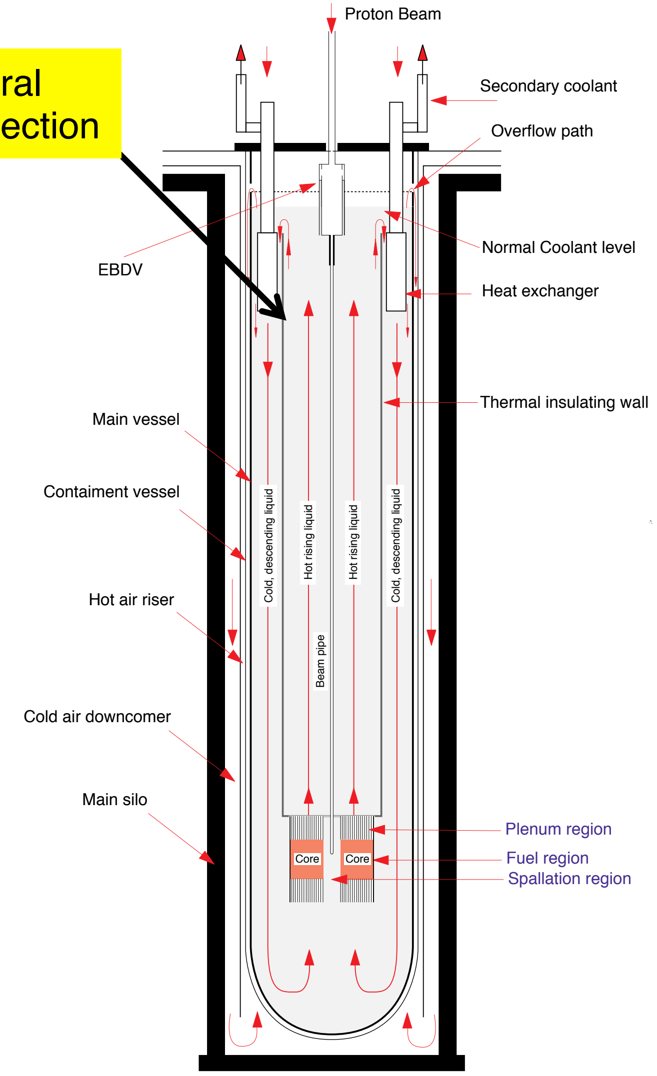


C. Rubbia et al.
CERN/AT/95-44

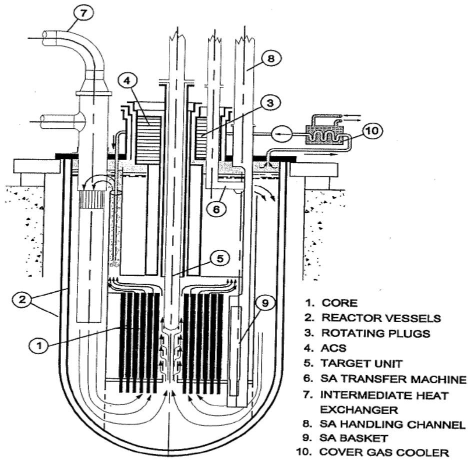
# Realistic ADS system

Energy Amplifier Conceptual Design
C. Rubbia, et al., CERN/AT/95-44 (ET)

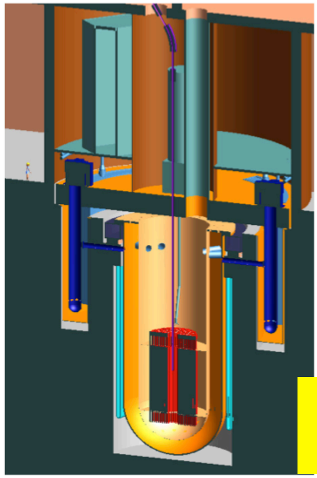
Natural convection



Ansaldo Engineering design
for the Energy Amplifier
Demonstration Facility
EA B0.00 1 200 (Jan. 1999)



AkerSolutions



C. Rubbia
This conf.

- ❑ Today one can simulate in great detail any realistic system of this type (LHC experiments have up to 30 million volume elements in their simulation)
- ❑ The physics is extremely well known! and physics does not fail!
- ❑ Neutron data are improving (n_TOF)

Simulation is never better than the quality of the input data

# Simplified model of subcritical systems

- **Theory of subcritical systems** interesting in itself, to get insights into the physics. Properties are quite different from those of critical systems. (*C. Rubbia, CERN/AT/ET/Internal Note 94-036*)
- **Knowledge of neutron flux geometry important to determine the generated power distribution and the uniformity of fuel burnup**
- Some simplifying assumptions (uniform material and mono-energetic neutrons, small absorption) to get a basic equation similar to that of a critical reactor, but with an **external neutron source term in addition**:

$$\frac{\partial n(\vec{r},t)}{\partial t} = \underbrace{\nu \Sigma_f \Phi(\vec{r},t)}_{\text{Fission}} + \underbrace{\boxed{C(\vec{r},t)}}_{\textbf{Spallation}} - \underbrace{\Sigma_a \Phi(\vec{r},t)}_{\text{Absorption}} + \underbrace{D\nabla^2 \Phi(\vec{r},t)}_{\text{Leakage}}$$

# Simplified model of subcritical systems

- Example of finite system at equilibrium:

Diffusion length

$$\frac{\partial n}{\partial t}=0 \Rightarrow \nabla^2\Phi + \frac{\boxed{\left(k_\infty - 1\right)}}{L_c^2}\Phi = -\frac{C}{D} \qquad \text{with } k_\infty \equiv \frac{\nu\Sigma_f}{\Sigma_a} \quad L_c^2 \equiv \frac{D}{\Sigma_a}$$

- Two regimes corresponding to two classes of solutions:
  - $k_\infty < 1$: the system is intrinsically subcritical (FEAT experiment: $k_\infty \sim 0.93$) – **Solution is an exponential**
  - $k_\infty > 1$: subcriticality comes from the lack of confinement, it is a geometrical issue – **Solution is oscillatory** (C. Rubbia’s EA: $k_\infty \sim 1.2$-$1.3$)

$$C\left(\vec{x}\right) = D\sum_{l,m,n} c_{l,m,n}\psi_{l,m,n}\left(\vec{x}\right) \rightarrow \Phi\left(\vec{x}\right) = L_c^2 \sum_{l,m,n}\frac{C_{l,m,n}}{1-k_{l,m,n}}\Psi_{l,m,n}\left(\vec{x}\right)$$

- All modes are excited

Theorem: $\forall i, k_i < k_1$

$$k_{l,m,n} \equiv k_\infty - L_c^2 B_{l,m,n}^2$$

# Time dependence

- ❑ Diffusion equation (with Φ = βn, where β is the neutron velocity):

$$\frac{\partial n(\vec{x},t)}{\partial t} = \frac{1}{\beta}\frac{\partial \Phi(\vec{x},t)}{\partial t} = D\nabla^2\Phi(\vec{x},t) + (k_\infty - 1)\Sigma_a\Phi(\vec{x},t) + C(\vec{x},t)$$

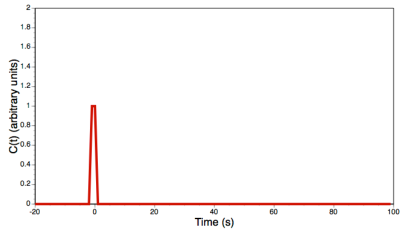
- ❑ Case of a neutron pulse, given by $C_0\delta(t)$, and substituting

$$\Phi(\vec{x},t) = \sum_{l,m,n}\Phi_{l,m,n}\psi_{l,m,n}(\vec{x})f_{l,m,n}(t)$$

provides an equation for the time dependence:

$$\frac{df_{l,m,n}(t)}{f_{l,m,n}(t)} = -\beta\left[DB^2_{l,m,n} + (1-k_\infty)\Sigma_a\right]dt$$, and the general solution

$$\boxed{\Phi(\vec{x},t) = \sum_{l,m,n}\Phi_{l,m,n}\psi_{l,m,n}(\vec{x})e^{-\beta\Sigma_a\left(1-k_{l,m,n}\right)t}}$$



- ❑ Characteristic decay time is shorter as modes become higher. At the criticality limit ($k_{1,1,1}$=1), the mode is infinitely long. Fermi used this to measure the approach of criticality in his Chicago Pile 1, in 1942, and the method is well suited for ADS.

# Neutron multiplication factor

- The neutron multiplication factor depends on the source:

Fission

$$k_{eff} \approx \frac{\nu \Sigma_f \Phi(\vec{r},t)}{\Sigma_a \Phi(\vec{r},t) - D\nabla^2 \Phi(\vec{r},t)}$$

Absorption

Leakage

**Non-fission multiplication**: For fast neutron systems, (n,xn) reactions are not negligible, in particular for the source neutrons

**Source**

$$k_s \approx \frac{\nu \Sigma_f \Phi(\vec{r},t) + C(\vec{r},t)}{\Sigma_a \Phi(\vec{r},t) - D\nabla^2 \Phi(\vec{r},t)} > k_{eff}$$

- Switching off the neutron source not only stops the main power generation, but also moves the system to a smaller k, from $k_s$ to $k_{eff}$.

# ADS energy gain

- A source neutron is multiplied by fissions and (n,xn) reactions. Since $k_s<1$, neutron production stops after a limited number of generations:

$$N_0\left(1+k_s+k_s^2+k_s^3+k_s^4+\ldots+k_s^n\right)=N_0 M=N_0\frac{k_s^{n+1}-1}{k_s-1}\approx\frac{N_0}{\left(1-k_s\right)}$$

- The energy gain G is a characteristic of ADS:

Energy/fission

n/p

$$G\equiv\frac{\textit{Energy produced in EA}}{\textit{Energy injected by the beam}}=\frac{0.18k_sN_0}{\nu\left(1-k_s\right)E_b}=\frac{G_0k_s}{\left(1-k_s\right)}\approx\frac{G_0}{\left(1-k_s\right)}$$

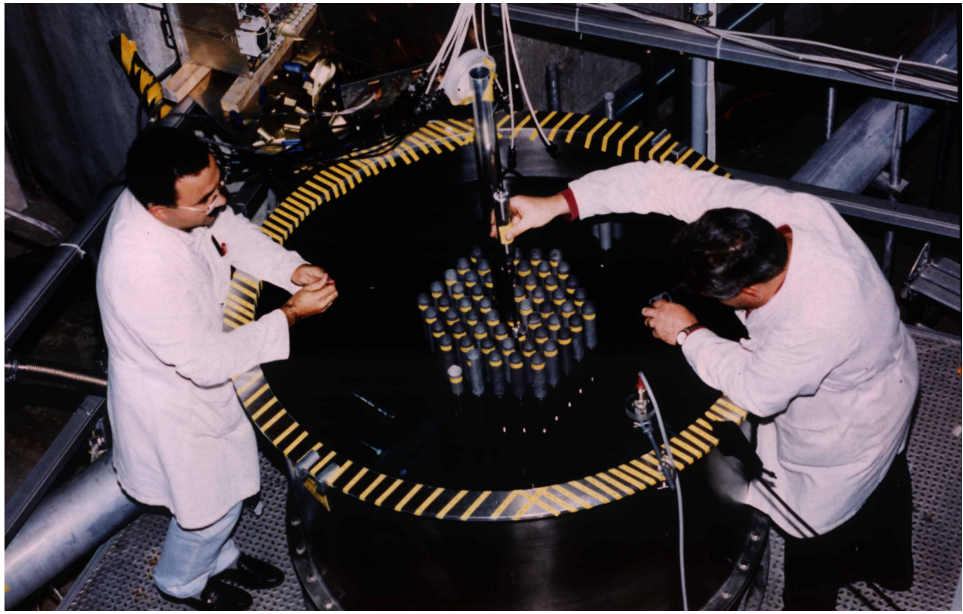
n/fission

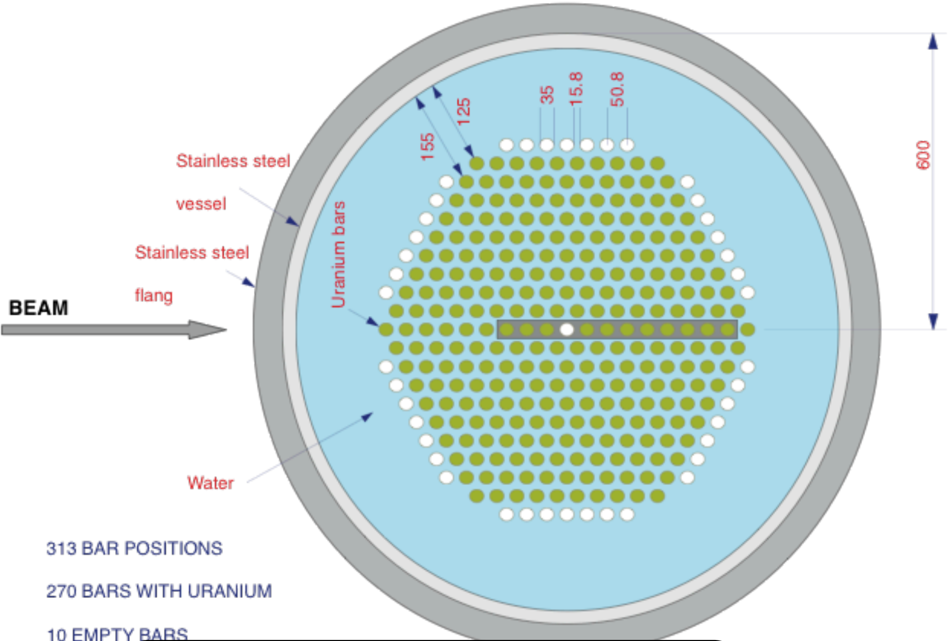
Beam energy

- $G_0$ includes information from the spallation process ($G_0$ ~ 3 for uranium; $G_0$ ~ 2.7 for lead, etc.)

# FEAT at the CERN PS (1994)

- The goal of the **F**irst **E**nergy **A**mplifier **T**est (**FEAT**) at the CERN PS was to check the **basic concept of energy gain**, and **validate the innovative simulation** developed by C. Rubbia and his group.



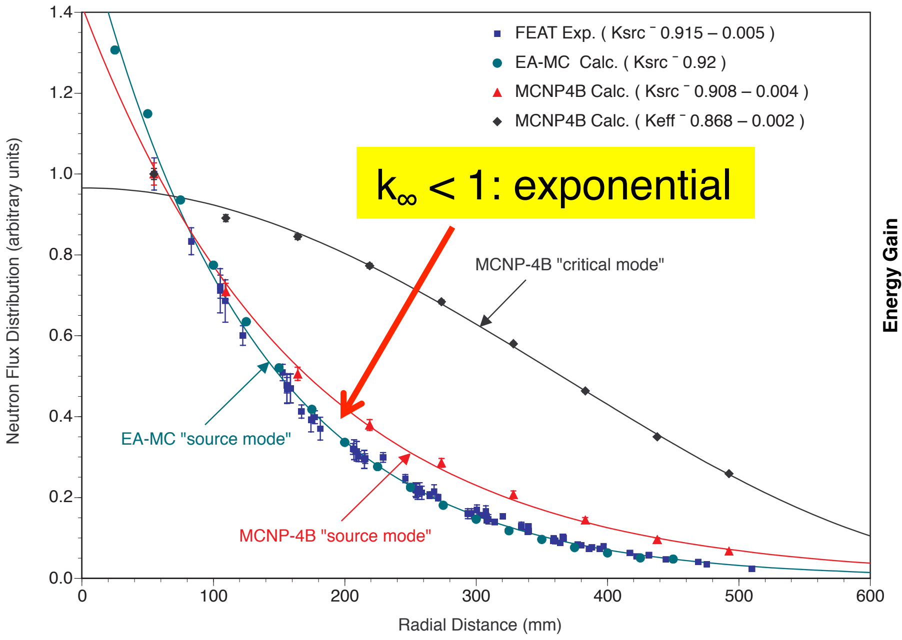


3.62 t of natural uranium; $k_{eff} \sim 0.9$
(Spanish nuclear engineering school)

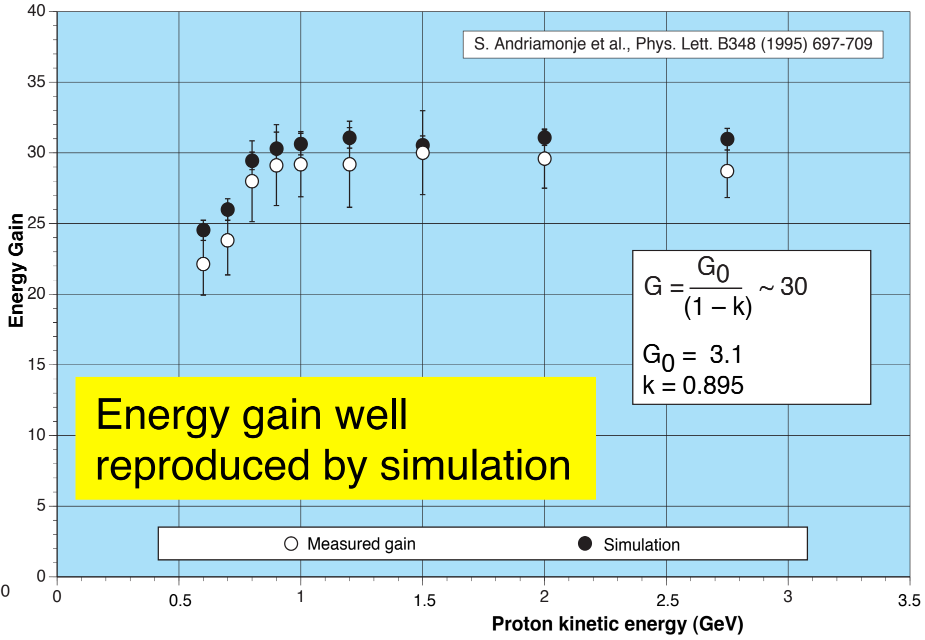
Juan Antonio Rubio

- Count fissions
- Measure temperature

# Main results from FEAT



$k_\infty < 1$: exponential



S. Andriamonje et al., Phys. Lett. B348 (1995) 697-709

$G = \frac{G_0}{(1-k)} \sim 30$

$G_0 = 3.1$

$k = 0.895$

Energy gain well reproduced by simulation

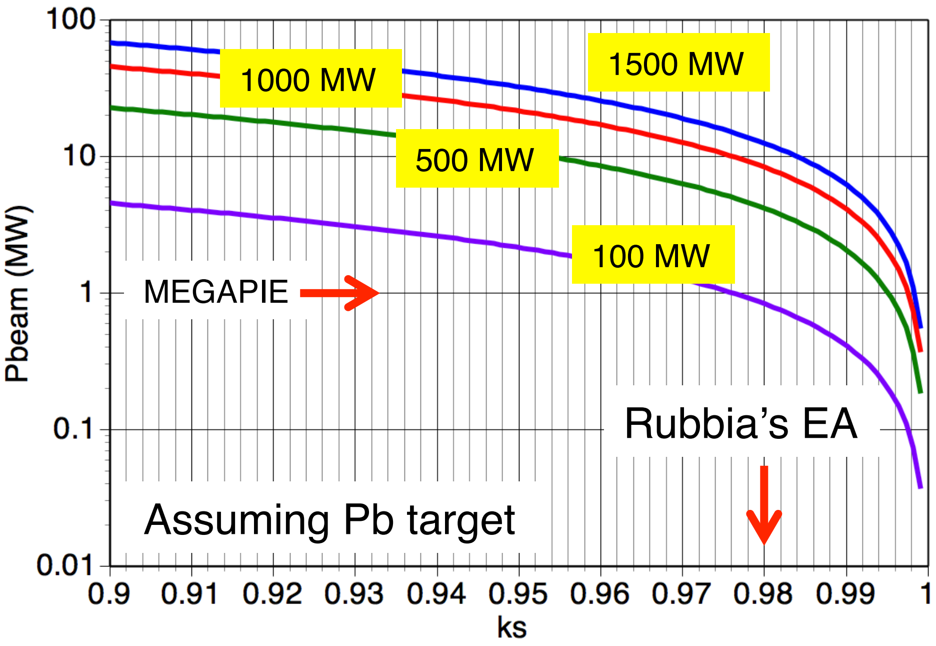
- Two important results from FEAT:
  - ☛ **Optimum beam energy reached at about 800 MeV**, with slow decrease at higher energies (ionization vs nuclear cascade production). Above 900 MeV, the neutron yield scales with proton energy. At much higher energies the gain drops because of pion production.
  - ☛ **Simulation validated** from spallation to energy production

# Energy gain in ADS systems

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

- 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).

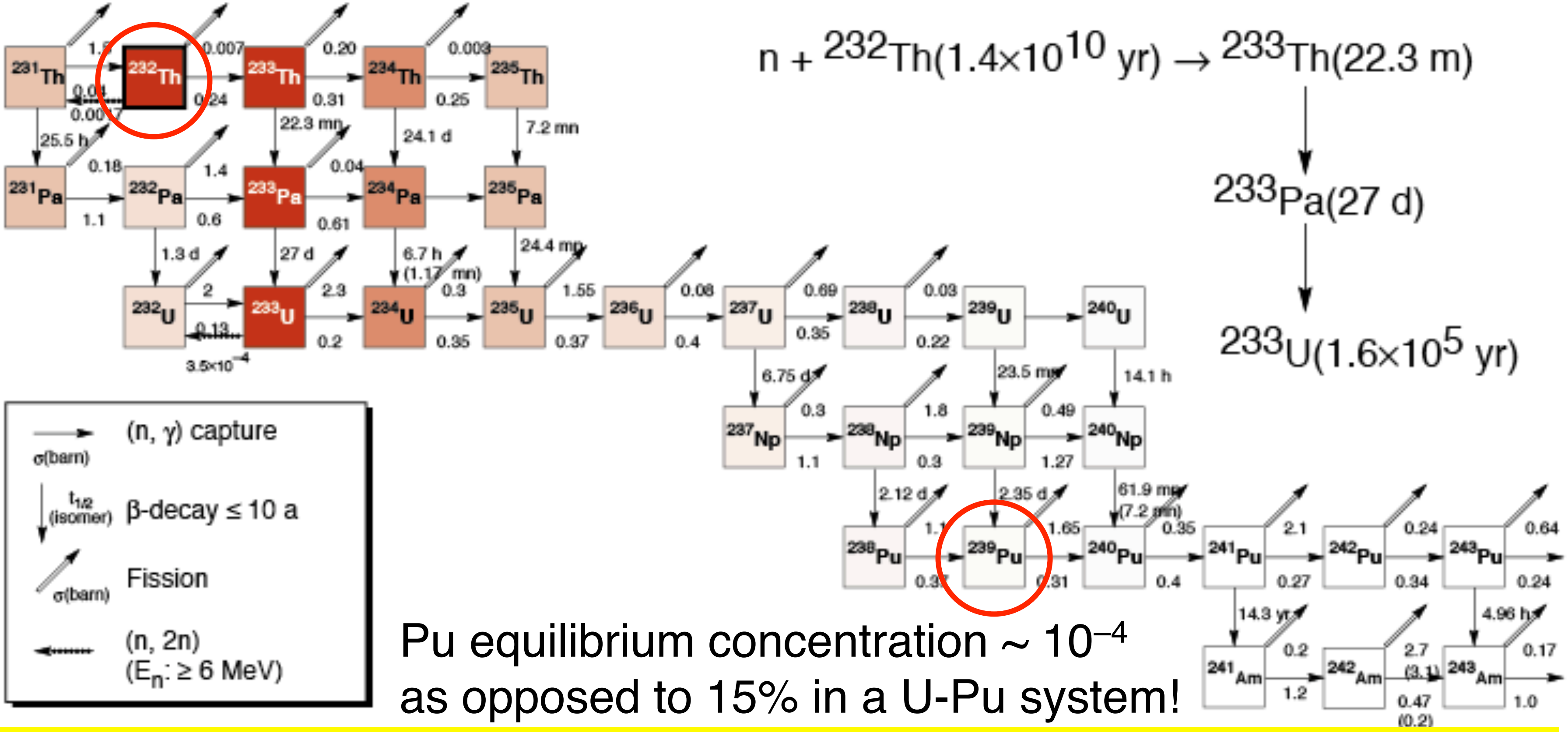
$P_{ADS}$ = 243 $MW_{th}$ with k = 0.98

$P_{ADS}$ = 486 $MW_{th}$ with k = 0.99.

# Physics of a thorium core

## ❑ Thorium in a fast neutron flux:

- ☛ It takes 7 neutron captures to go from $^{232}$Th to $^{239}$Pu!
- ☛ $^{233}$Pa decay 10 times slower than $^{239}$Np



$$n + {}^{232}\mathrm{Th}(1.4\times10^{10}\ \mathrm{yr}) \rightarrow {}^{233}\mathrm{Th}(22.3\ \mathrm{m})$$

$$\downarrow$$

$$^{233}\mathrm{Pa}(27\ \mathrm{d})$$

$$\downarrow$$

$$^{233}\mathrm{U}(1.6\times10^{5}\ \mathrm{yr})$$

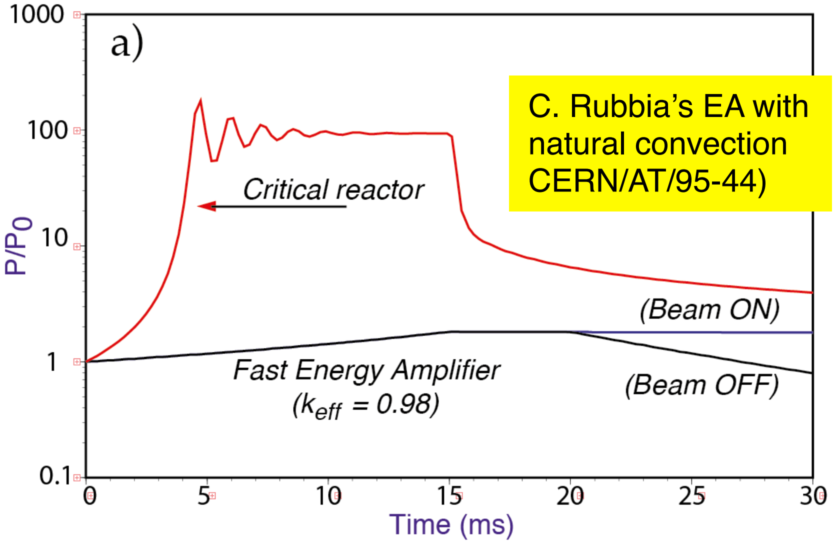
Pu equilibrium concentration ~ $10^{-4}$
as opposed to 15% in a U-Pu system!

Thorium is a major asset to minimize waste production or to destroy waste

# Physics of subcritical systems

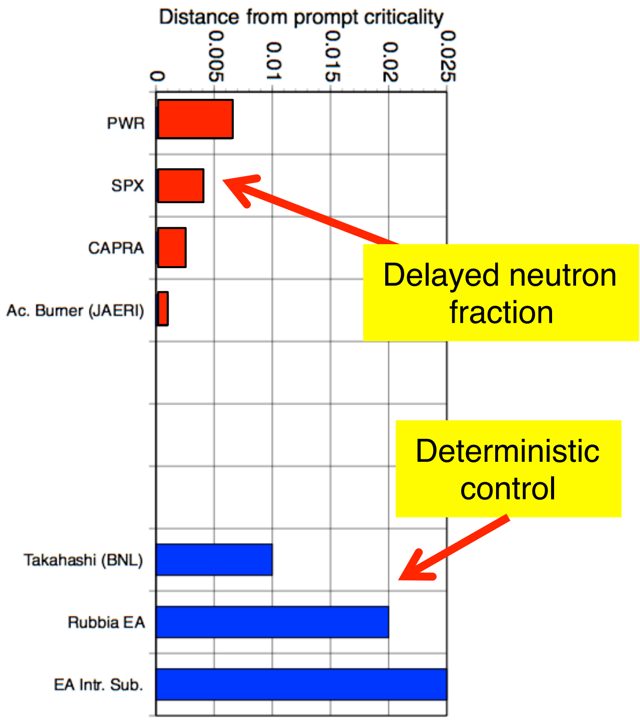
- Subcritical systems are insensitive to delayed neutron fraction (β); **safety margin** (distance from prompt criticality) **is a design choice**, it is not imposed by Nature!
- The reactivity changes only very slowly in a EA with $k_s$ = 0.98; but other choices are possible (Takahashi, $k_s$ = 0.99 or Rubbia with AkerSolutions, $k_s$ = 0.997)
- The beam can be switched off very quickly, reducing $k_s$ to $k_{eff}$.

Accidental reactivity insertion of 255$/s for 15ms



C. Rubbia's EA with natural convection CERN/AT/95-44)

1) Nothing much happens in a subcritical system
2) An accelerator can be switched off in a time which is negligible compared to the typically response time of a critical system to reactivity insertion of the order of 5 ms.

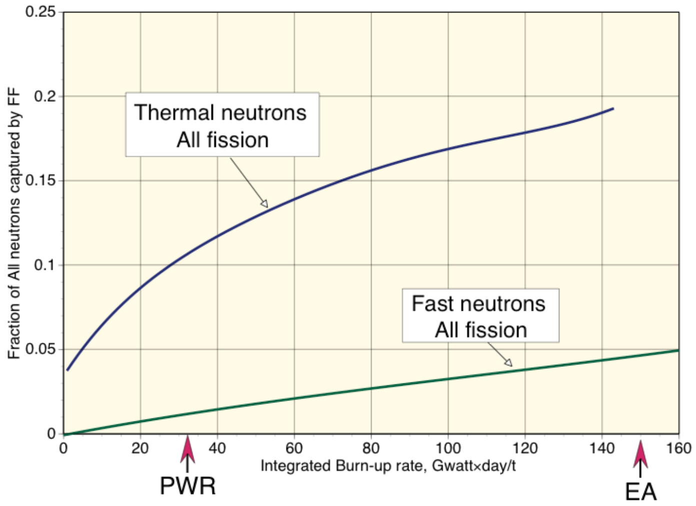


Delayed neutron fraction

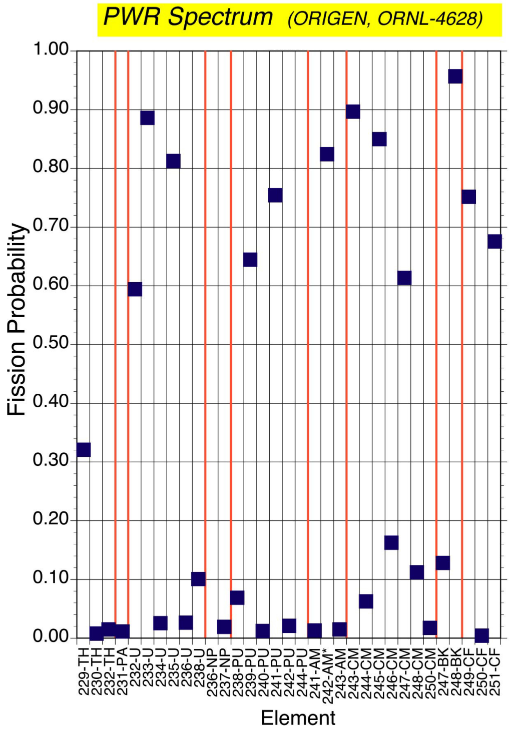
Deterministic control

# Why fast neutrons

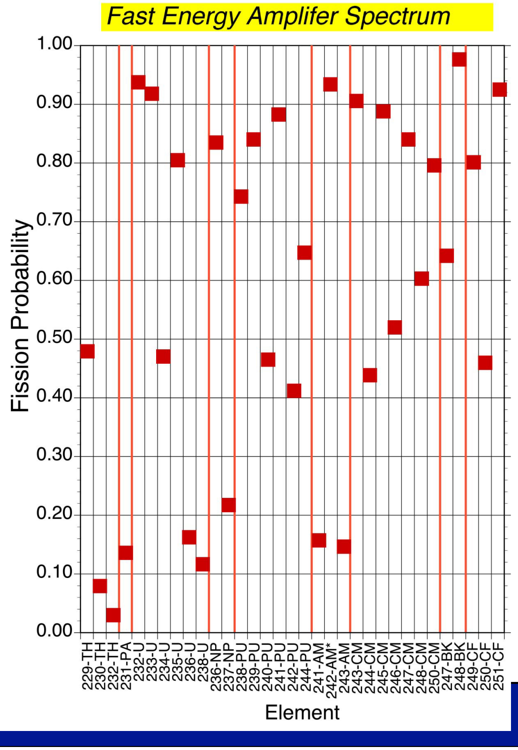
- **Use fast neutrons**:
  - Enhances TRU fission probability
  - No need to separate out Pu! (**Pyro-Electro reprocessing**)
  - Reduces captures on FF, extends burnup (120 GW.day/t achieved in fast electro-breeder at Argonne N.L., in EA simulation)



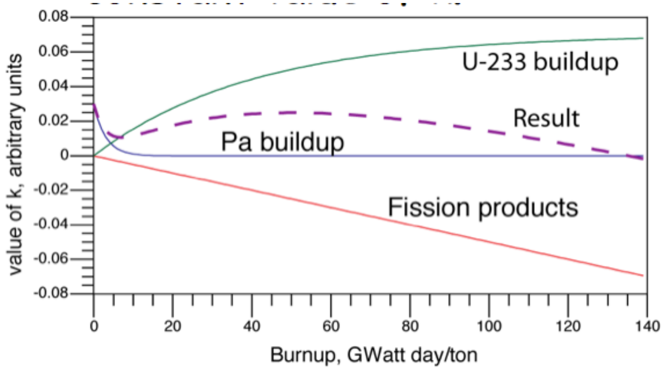
Thermal Neutrons



Fast Neutrons

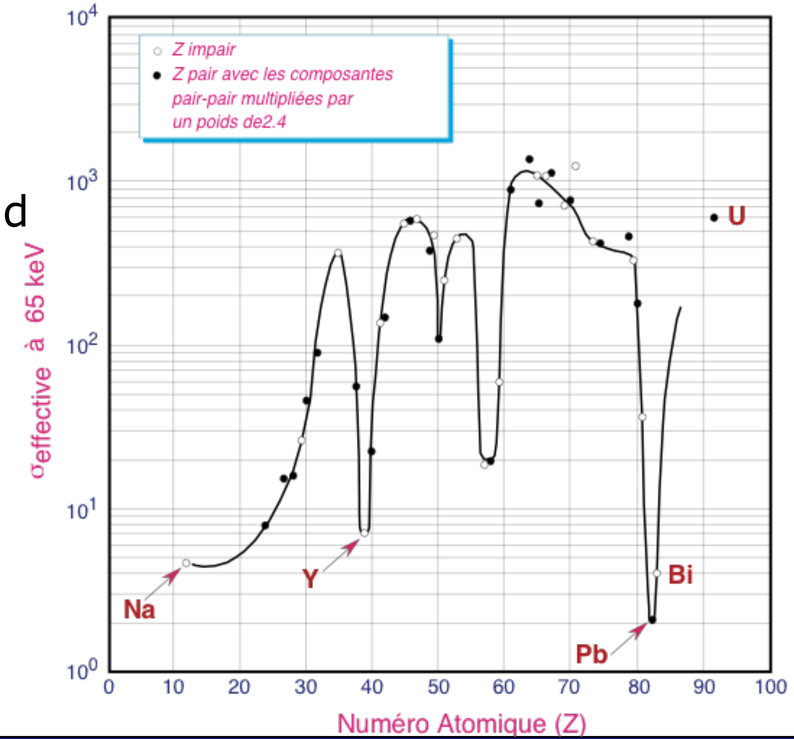


Beautiful demonstration by C. Rubbia of extended burnup, using compensation between 233U breeding and captures



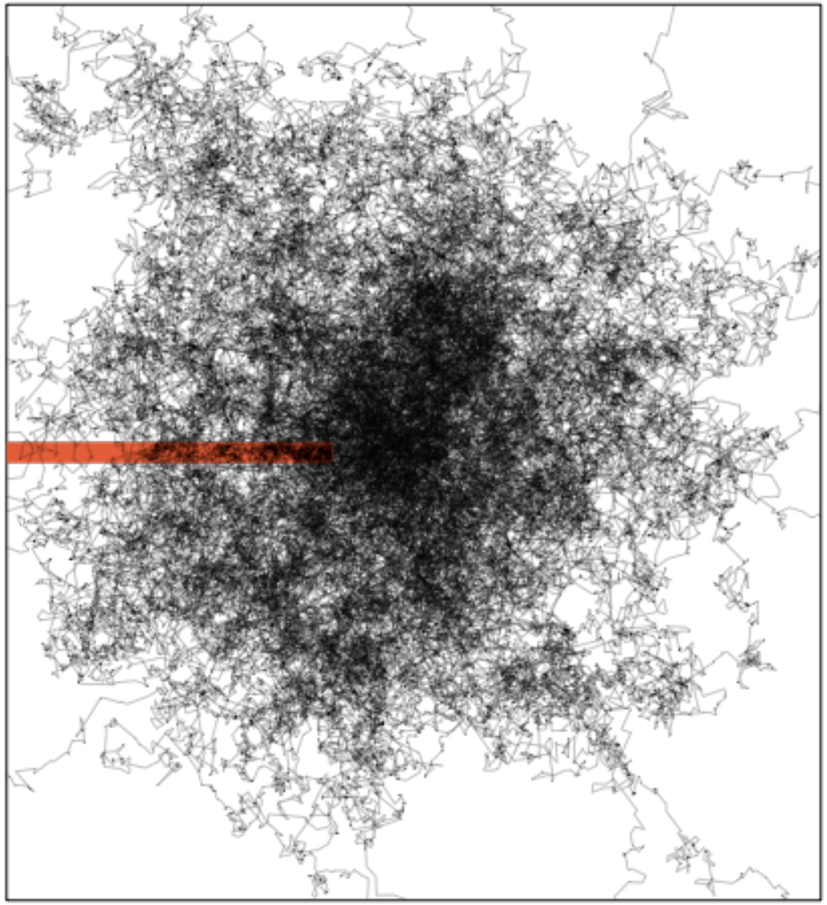
# Example of Carlo Rubbia’s EA

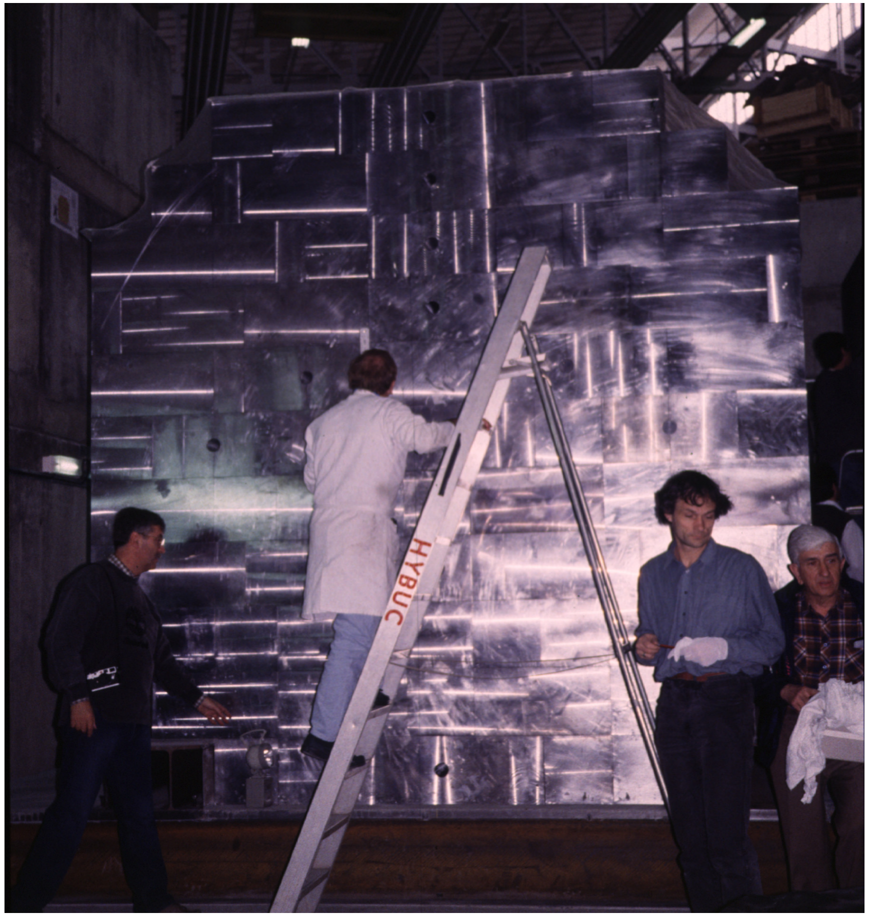
- **Use lead as moderator**: fast neutrons ➔ moderate as little as possible (sodium, gas, lead)
- Lead plays several roles: spallation target, moderator, heat removal agent, containment medium. It is the heavy element most transparent to neutrons.
  - **Excellent spallation target**: neutron yield almost as good as for uranium;
  - Lead **less dangerous than sodium**. Boiling temperature well separated from fusion point (unlike sodium); radiation shield
  - **Excellent coolant**;
  - ***Drawbacks***: **corrosion** at high T but rapid progress in new materials (lead loops, Eurofer), and use of super critical $CO_2$ Brayton power cycle achieving 45% thermal efficiency with inlet temperature of only 550°C (53% at 700°C) MIT V. Dostal, M.J. Driscoll, P. Hejzlar. **Po production** (Pure Pb better than Pb-Bi)



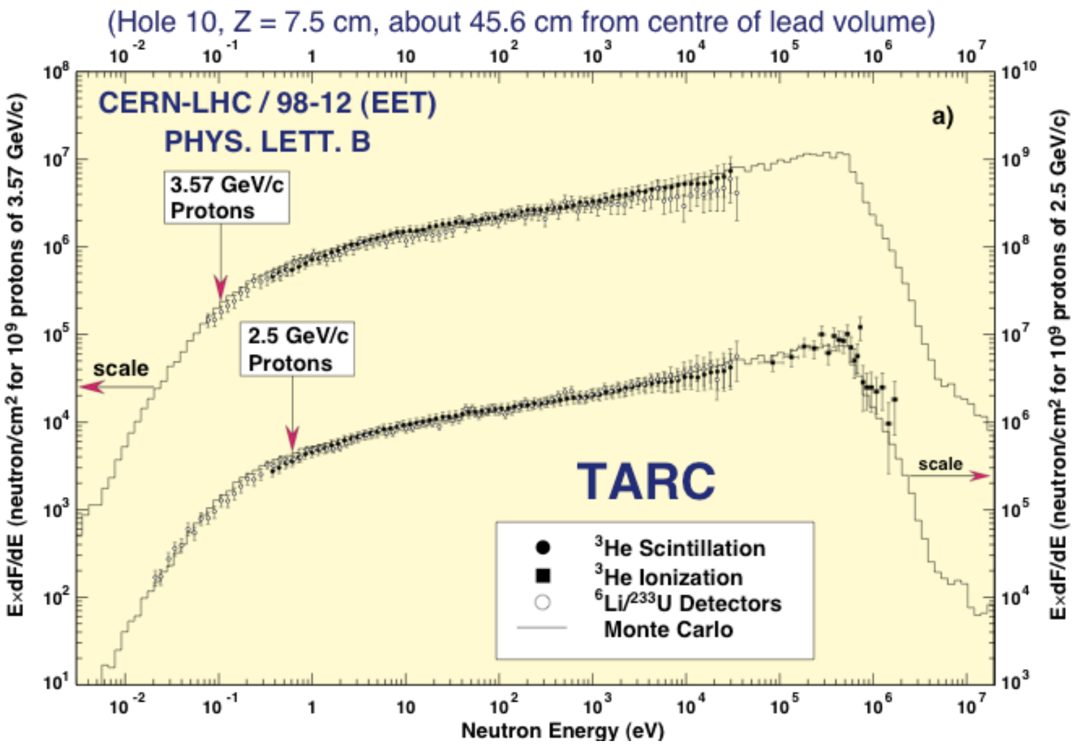
# TARC at the CERN PS

- ❑ Neutron phenomenology studied in great details in the TARC experiment at the CERN PS (1996-1997).
- ❑ Testing both the spallation process and the transport in lead



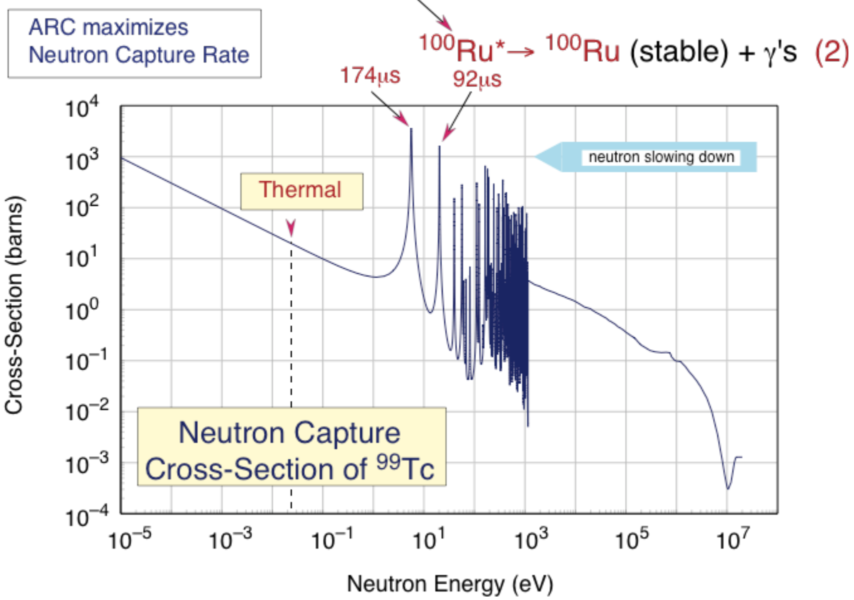


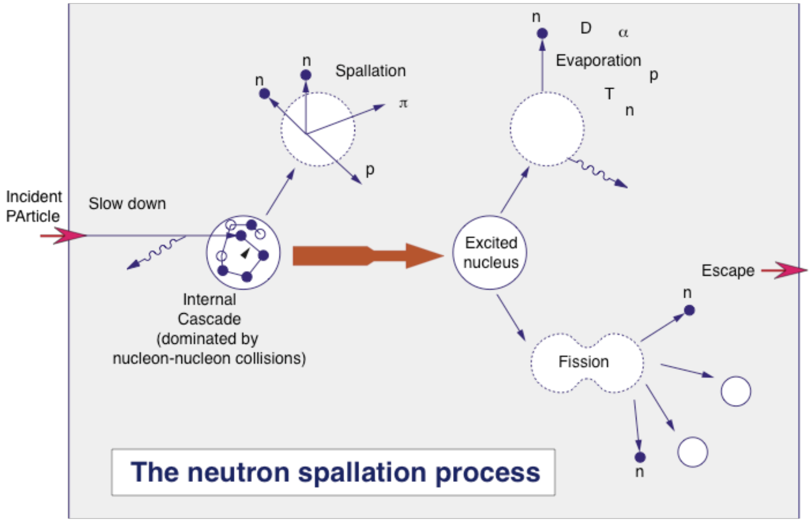
# Results from TARC



$^{99}Tc\ (t_{1/2} = 2.1\times10^5\ \text{ans}) + n \rightarrow {}^{100}Tc\ (t_{1/2} = 15.8\ \text{s}) + \gamma$ prompts (1)

$^{100}Ru^* \rightarrow {}^{100}Ru$ (stable) + $\gamma$'s (2)

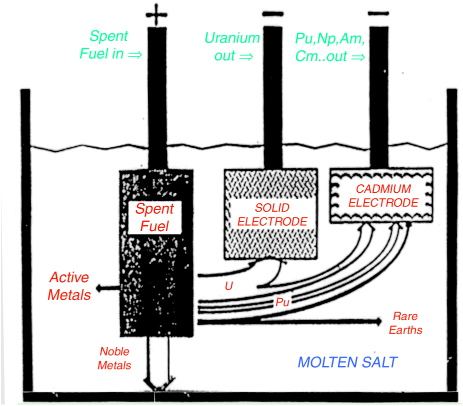


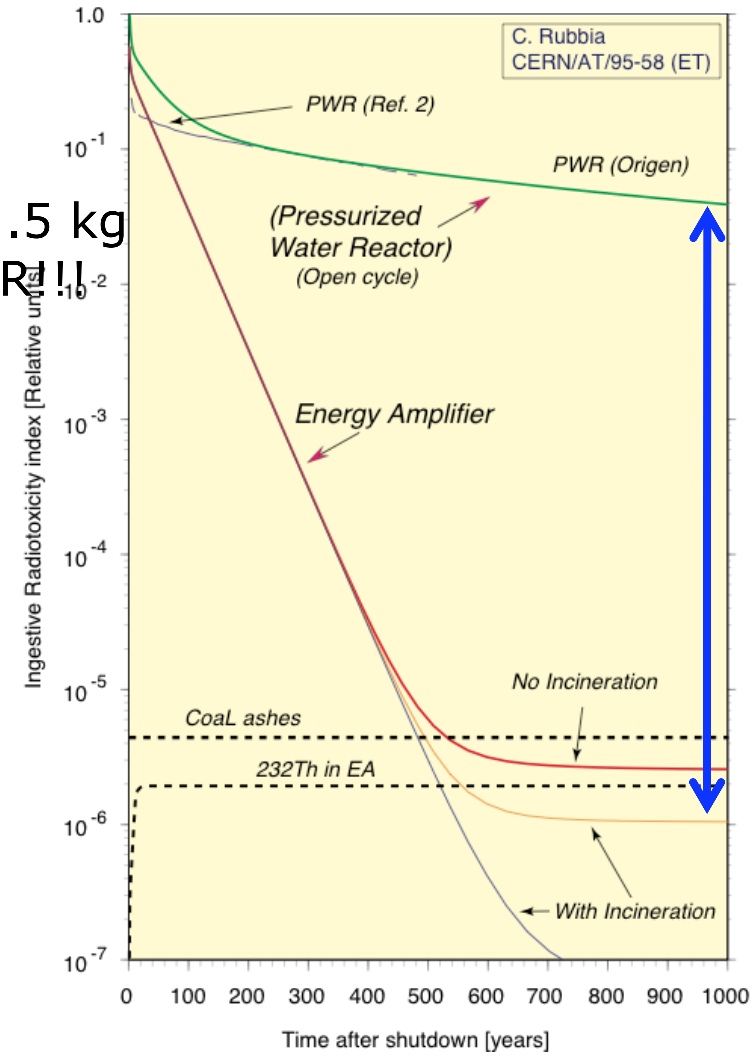


- Demonstrated **Adiabatic Resonance Crossing** for the elimination of long-lived fission fragments, an idea also proposed by C. Rubbia, patent by CERN.

# ADS transmutation performance

- **Destroy** 36 kg of TRU/$TW_{th}$.h (A PWR **produces** 14 kg of TRU/$TW_{th}$.h): based on detailed simulation
- With 100 $MW_{th}$ and k=0.98, one destroys 31.5 kg that is 25% of the waste production of a PWR!!!
- Use pyro-electro reprocessing, adapted to thorium fuel (Argonne National Lab)
  **A major asset for ADS**





- Today the emphasis has clearly shifted toward energy production, because of developing countries.
  - ☛ The two options are not incompatible, since in both cases one wants to minimize TRU production.

# Accelerator requirements

- Choices for an industrial system will rely on specifications in terms of:
  - ☛ **Beam power**: 1-10 MW depending of choice of k value, and desired unit power; $E_{beam} \geq 800$ MeV
  - ☛ **Beam losses**: minimize irradiation of the accelerator components and of the environment; impact on the maintenance (minimize and localize beam losses)
  - ☛ **Reliability**, minimize beam trips (have multiple sources); ease of maintenance
  - ☛ **Beam stability and control**: 1% fluctuation on beam intensity is 1% fluctuation on the thermal power; beam intensity to be varied by a factor ≥ 2
  - ☛ **Energy efficiency**: maximize fraction of electric grid power stored into the beam) Improves with increasing beam power.
  - ☛ **Size, cost**

To be discussed in detailed in this session

# Conclusion

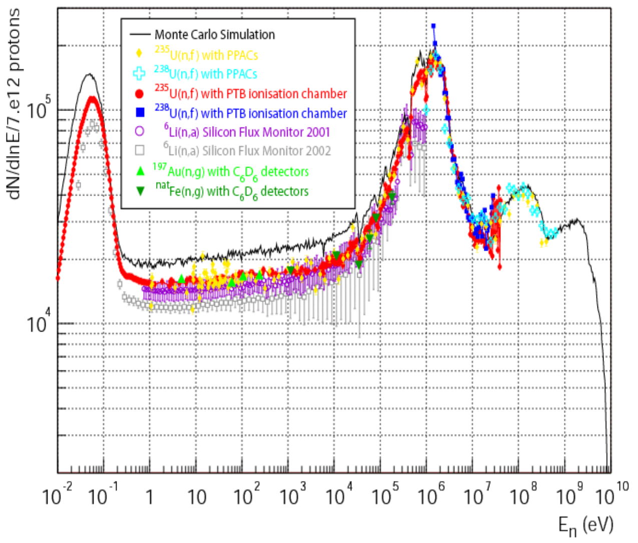
- **The physics of ADS is entirely known, and reliable simulations is available**
- When taking into account the need for safety, waste management and non-proliferation, thorium in a fast neutron ADS is optimum, as shown clearly by Carlo Rubbia
- It is a challenging innovation but there is no show stopper. There is no reason a “demonstrator” of significant power cannot be built now, in order to validate technological solutions, for the various elements of the system.
- This would stimulate targeted R&D, in a coherent way, hopefully in an international collaboration, as global issues should be faced by global responses.

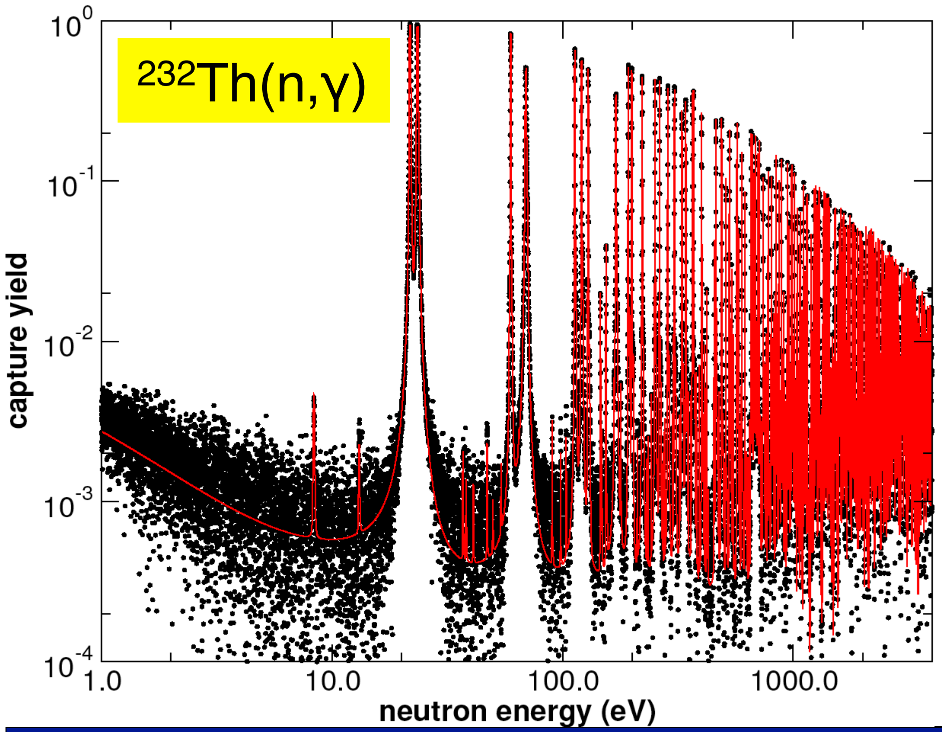
# RESERVE

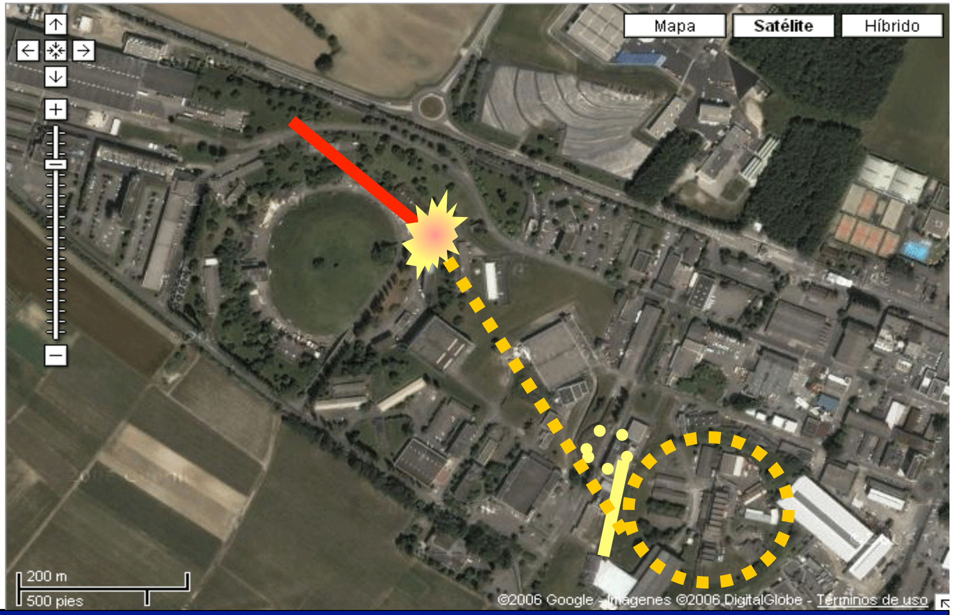
# Neutron cross section data

- **Develop precise and predictive simulation, requires precise input data**:
  - ☛ n_TOF facility at CERN

See talk by Frank Gunsing

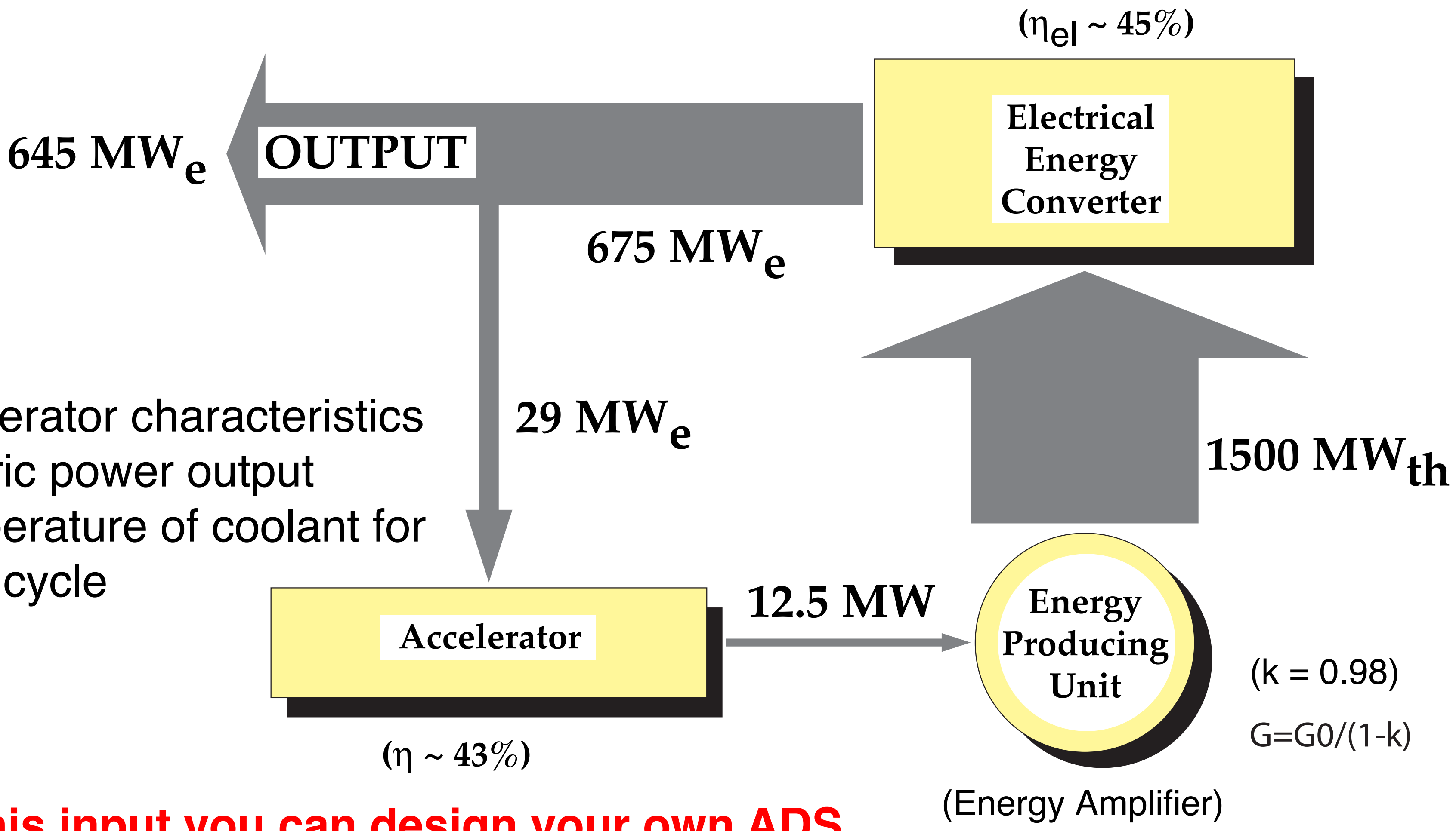






# Architecture of an ADS system

- Example of the generic 1500 $MW_{Th}$ system (Energy Amplifier), designed and simulated by C. Rubbia (CERN/AT/95-44 (ET))



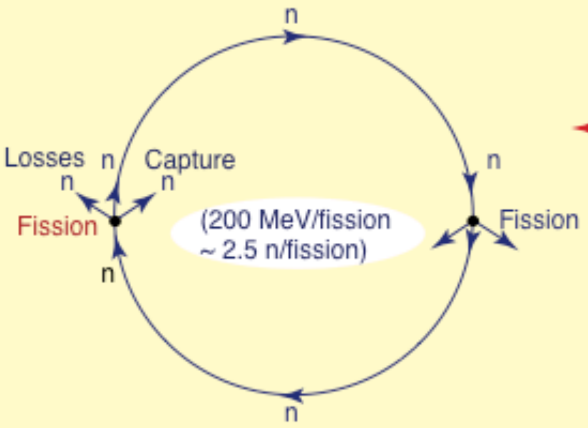
**Input:**

- accelerator characteristics
- electric power output
- Temperature of coolant for Carnot cycle

**With this input you can design your own ADS**

# Critical versus Subcritical Systems

## Chain Reaction



**Critical Reactor** ⟷ **Energy Amplifier**

Effective neutron multiplication factor

$$k = \frac{\text{Production}}{\text{Absorption + Losses}}$$

Self-sustained process:

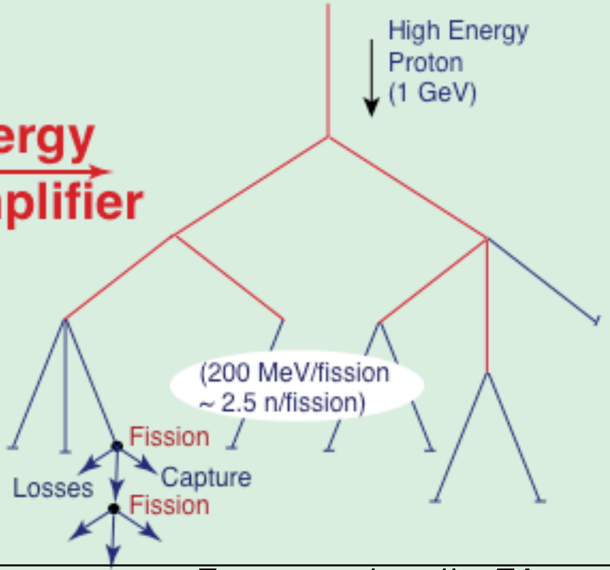
$k = 1$

(if $k < 1$ the Reactor stops

if $k > 1$ the Reactor is supercritical)

⇒ The time derivative of the power kept equal to zero by control

## Nuclear Cascade



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

Externally driven process:

$k < 1$ $(k = 0.98)$

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

Energy Produced

Beam Energy

⇒ Constant Energy Gain

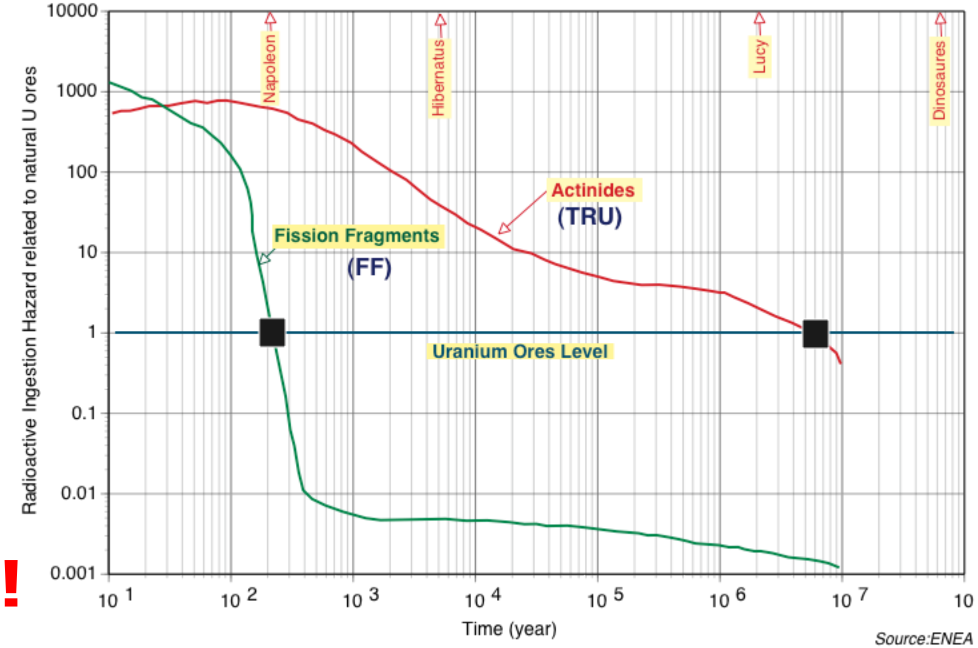
$$N_0\left(1 + k + k^2 + k^3 + k^4 + \ldots + k^n\right) = N_0 \frac{k^{n+1} - 1}{k - 1} \approx \frac{N_0}{1 - k}$$

# Eliminating nuclear waste

- The two components of radioactive waste require different strategies:
  - **TRansUranic elements** (**TRU, 1.1 %**: Np, Pu, Am, Cm, Bk, etc.) result from neutron capture in the fuel and subsequent decays.
    (**~100 t/y of $^{239}$Pu produced in the World + Military Pu**):
    **⇒ eliminated through fission producing energy!**
  - **Fission Fragments**
    (**FF, 4%**: produced in the fission process):
    **⇒ long-lived part eliminated through neutron capture**

**Priority: TRU elimination!**

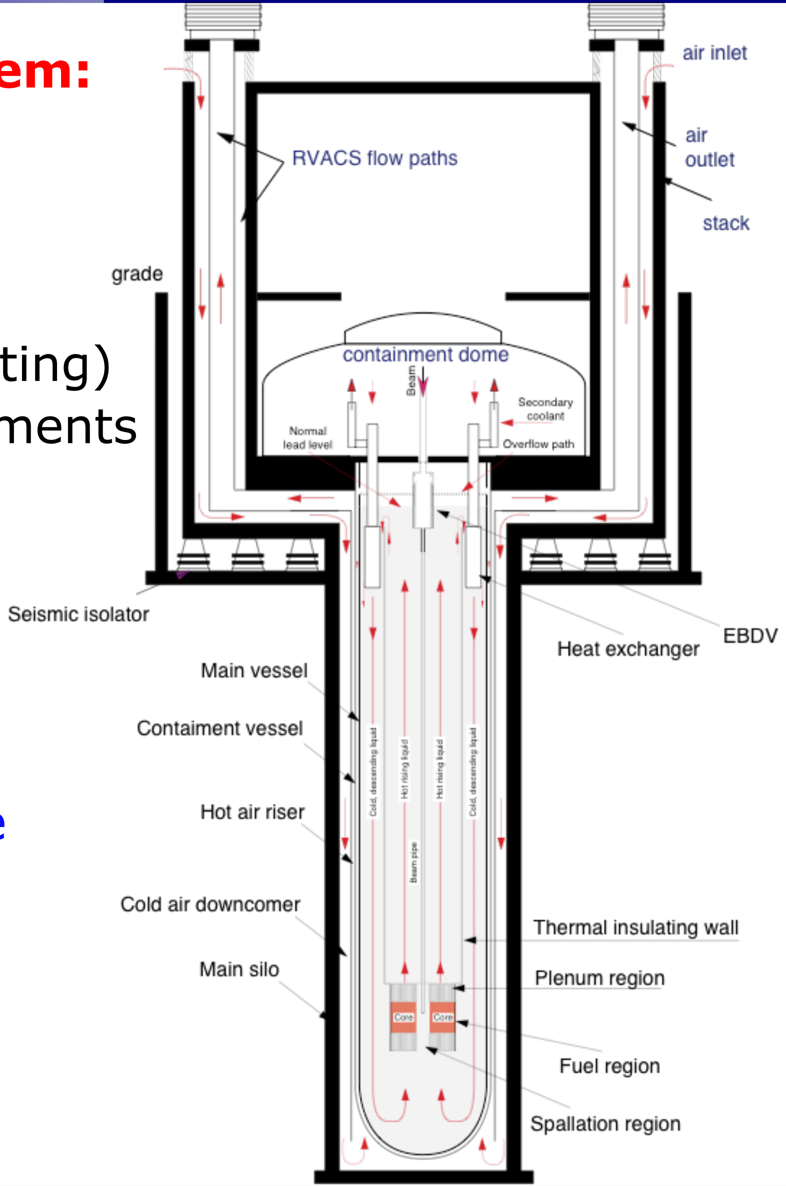
PWR Spent Fuel (ORIGEN2 Updated RadioToxicity)



*Source:ENEA*

# The Energy Amplifier

- ❑ Proton accelerator driven **subcritical system:**
  - ☛ **Fast neutrons** ($10^4$ to $10^6$ eV range)
  - ☛ **Lead** as spallation target, moderator and coolant
  - ☛ Fuel based on **thorium** rather than uranium (minimize waste, less proliferating)
  - ☛ **Deterministic safety** with passive elements



- ❑ C. Rubbia, et al.
  « Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier »,
  CERN/AT/95-44 (ET)

  « A Realistic Plutonium Elimination Scheme with Fast Energy Amplifiers and Thorium-Plutonium Fuel »,
  CERN/AT/95-53 (ET)

# Lead or Lead-Bismuth studies

- ☐ Eurotrans using the many loop facilities existing in Europe
- ☐ Japan, India and China, all have lead or LBE loops, and are making significant progress in corrosion issues (new materials, passivation methods, etc.)

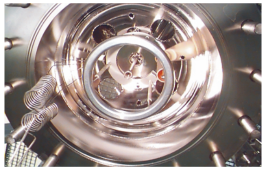
**KALLA Loops**
FZK

**STELLA Loop**
CEA

**CIRCE Loop**
ENEA

**CorrWett Loop**
PSI

**VICE Loop**
SCK-CEN



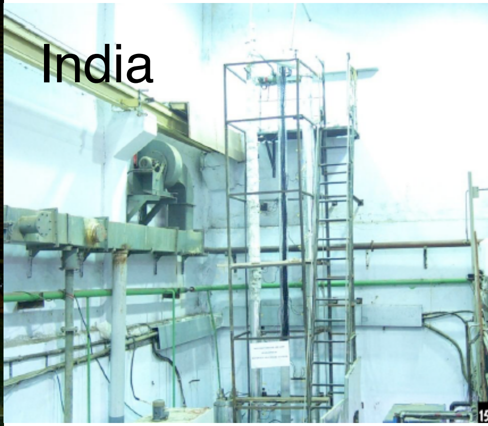
**CHEOPE Loop**
ENEA

**COSTA**
FZK

**TALL Loop**
KTH

**CIRCO Loop**
CIEMAT


J-PARC


India

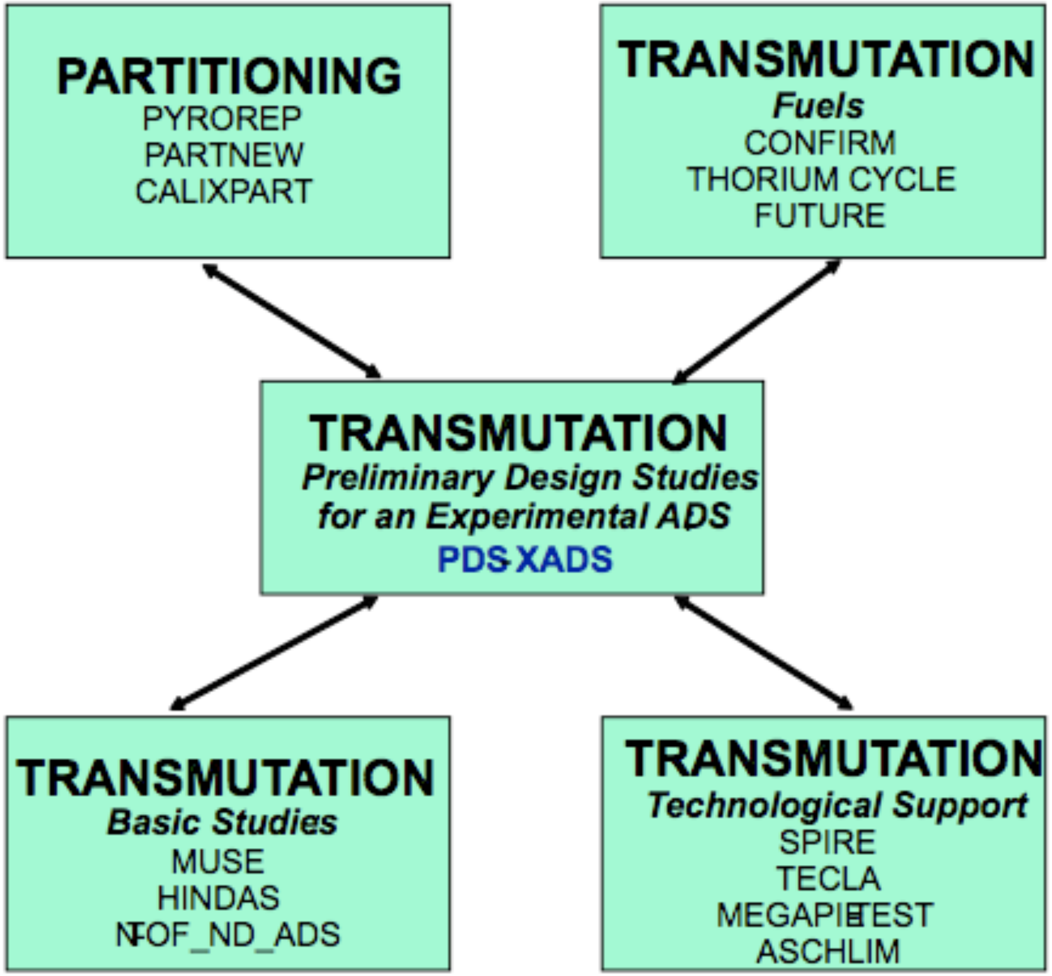
China LBE loop: 550˚C, 6m$^3$/h

中国散裂中子源

*(Shinian Fu, IHEP, Bejing*

# R&D in Europe

Many projects carried out in the EU FP5 and FP6 (Eurotrans) in the field of partitioning and transmutation. All aspects covered.



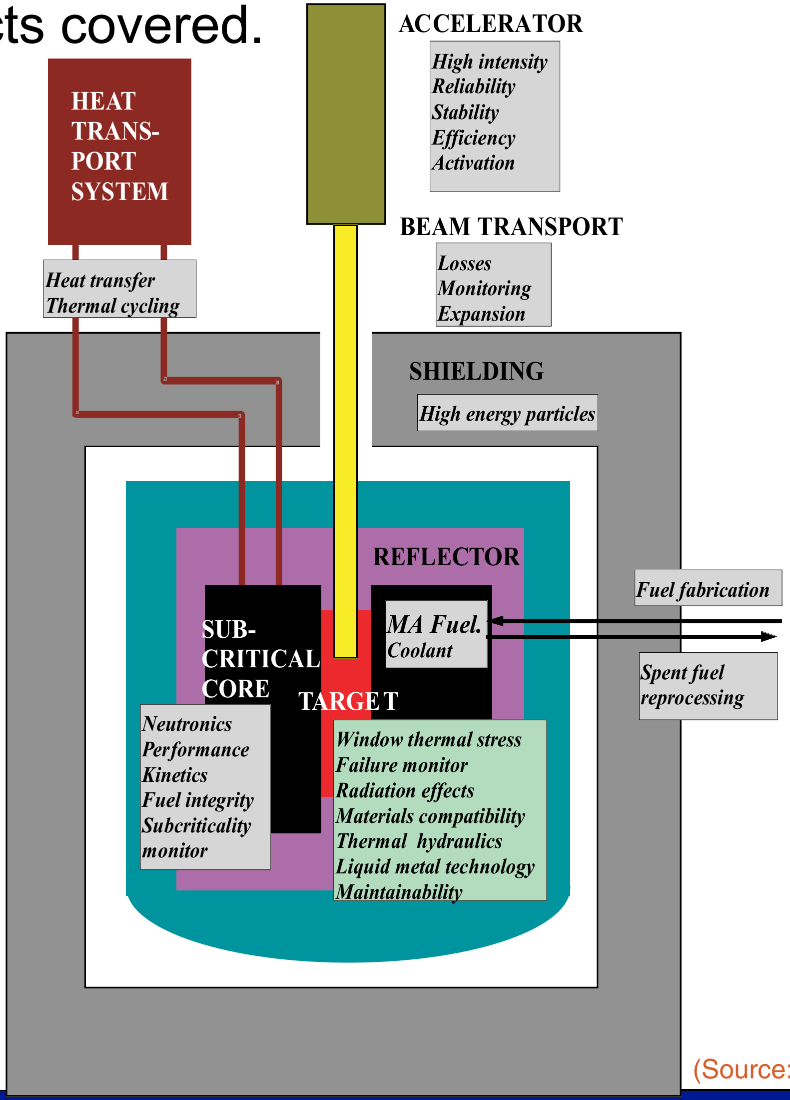
**PARTITIONING**
PYROREP
PARTNEW
CALIXPART

**TRANSMUTATION**
***Fuels***
CONFIRM
THORIUM CYCLE
FUTURE

**TRANSMUTATION**
***Preliminary Design Studies for an Experimental ADS***
PDS-XADS

**TRANSMUTATION**
***Basic Studies***
MUSE
HINDAS
NTOF_ND_ADS

**TRANSMUTATION**
***Technological Support***
SPIRE
TECLA
MEGAPIE TEST
ASCHLIM



(Source:PSI)

# Other developments concerning thorium

- **Example of India**: little uranium resources, but a lot of thorium:
  - Use PWR heavy water reactors (CANDU) and LWR to produce plutonium from their small uranium supply
  - Use sodium cooled U-Pu fast reactor with Th blanket to breed $^{233}U$
  - Reprocess blanket and manufacture $^{233}U$-Th fuel for advanced heavy water reactor

    **Present scheme complicated, not sustainable, not addressing nuclear waste issue. ADS a simplified solution!**

# Pyro-electro reprocessing

- Electrolysis of spent fuel in molten salt. Pu and minor actinides collect on one electrode (electrolyte is LiCl-KCl and actinides as chlorite)
- Pioneered at Argonne National Lab.
- A major asset for ADS