Motto: When you aimed for PERFECTION, you discover it is a moving target.
George Fisher

Acelerator Driven Transmutation Systems - new challenges, new experiments, new instrumentations

Waclaw Gudowski

Nuclear & Reactor Physics

KTH

Program

- Accelerator-driven Transmutation of Nuclear Waste Concept (ATW) – Accelerator Driven Systems (ADS)
- Components of ADS
- Impact of ADS on Nuclear Power
- A short review of some on-going projects and experiments related to ADS
- Experimental & instrumentation challengies
- Conclusions

What is TRANSMUTATION?

Nuclear process where one element transforms into another one via nuclear reactions.

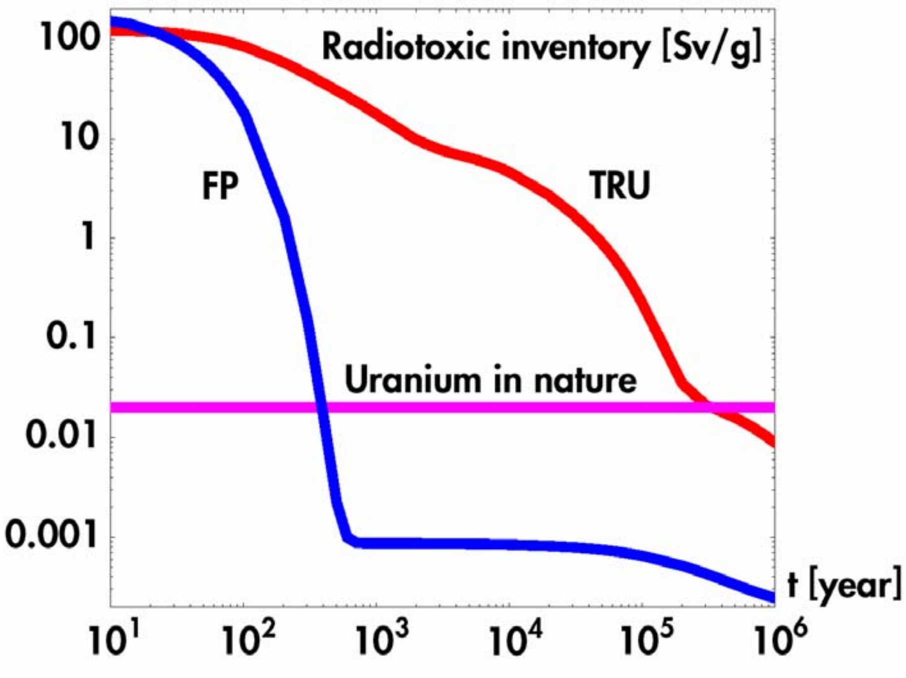
Nuclear transmutation was first demonstrated by Rutherford in 1919, who changed ^{14}N to ^{17}O using energetic α-particles. First accelerator-driven transmutation demonstrated by J.D. Cockroft and E.T. Walton, 1930:

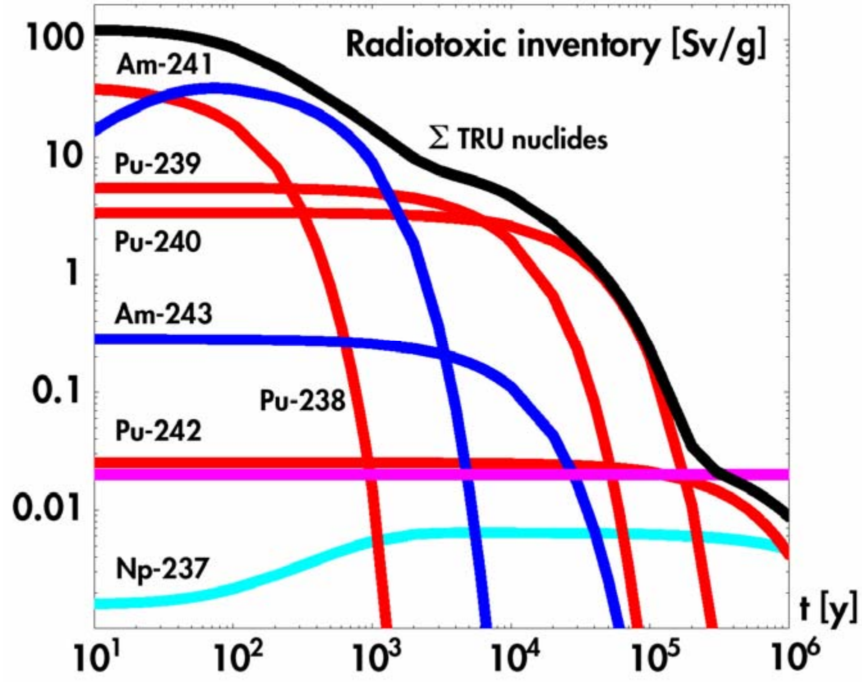
$$^{7}Li + p \rightarrow {}^{4}He + \alpha$$

Then intensively developed by scientists working on first accelerators (G. Seaborg). Plutonium was first produced using just accelerator.

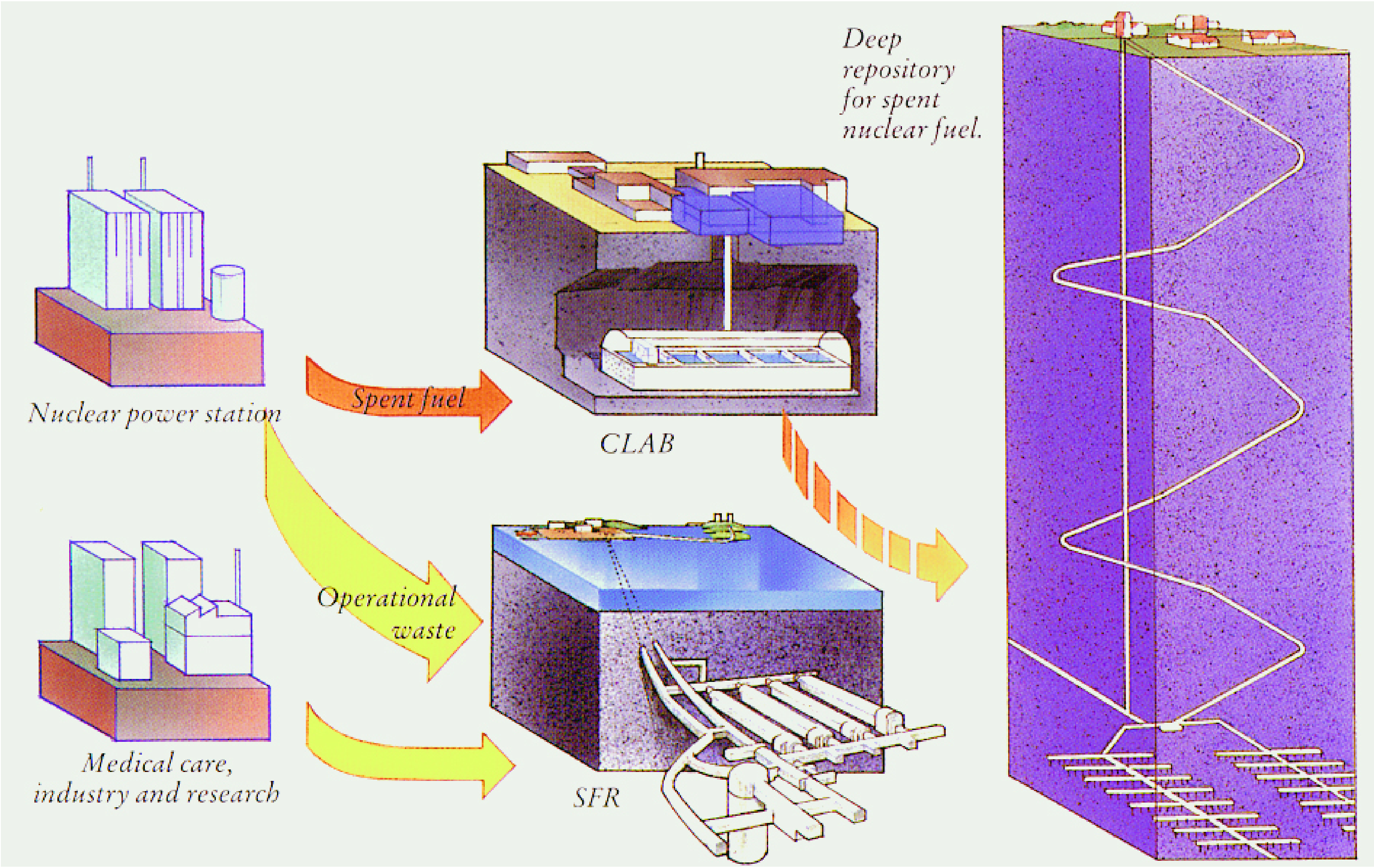
WHY ARE WE INTERESTED IN TRANSMUTATION??

Radiotoxocity of spent fuel reactor fuel (LWR)





”Official” Nuclear Fuel Cycle



Spent Nuclear Fuel containing almost the whole Periodic System of Elements is, however, not a *Mendeleyev Garbage-Can*

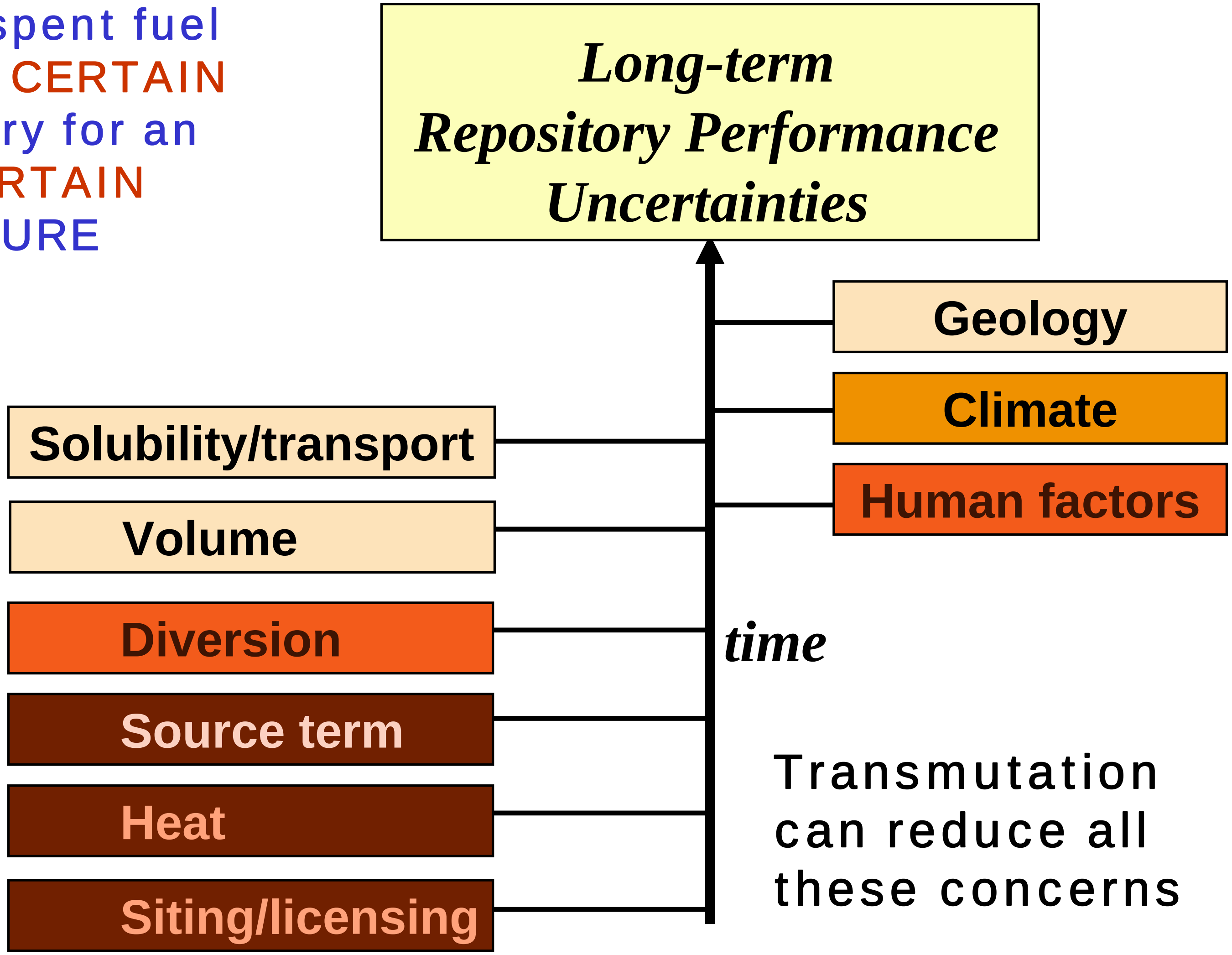
- **Only very few isotopes determine the HAZARDS of the spent fuel repository or/and waste handling:**
  - **In the long term:**
    - **Actinides: Pu isotopes (mainly 239), $^{243/241}Am$ and ^{237}Np**
    - **Fission Products: ^{99}Tc ($T_{1/2}=2.1\times10^5$y) and ^{129}I ($T_{1/2}=1.5\times10^7$y) , (^{135}Cs)**
  - **In the short term:**
    - **^{90}Sr and ^{137}Cs**

Long time uncertainties of geological disposal

DODDERER

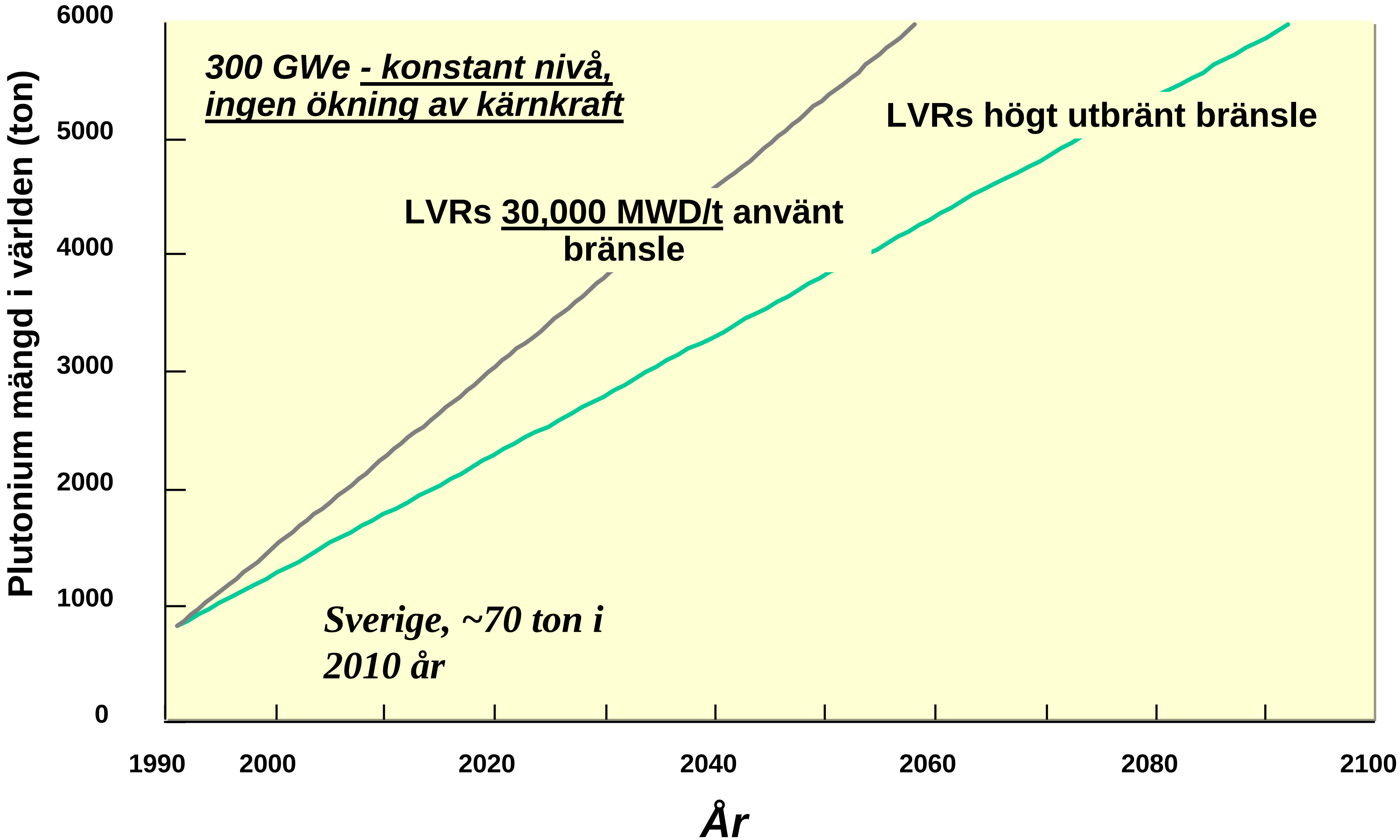
Nuclear spent fuel will get a CERTAIN repository for an UNCERTAIN FUTURE

Dagens Nyheter, 1996



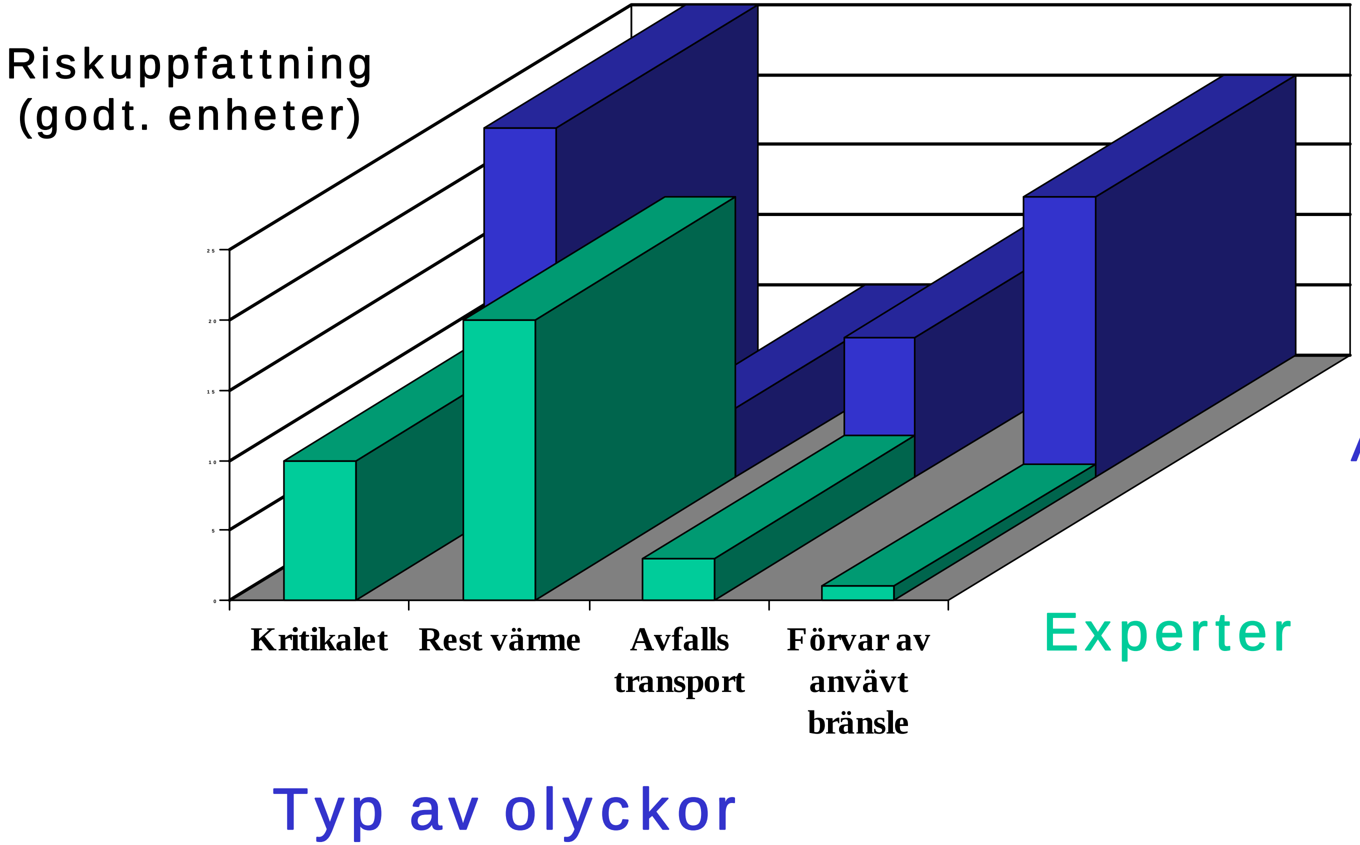
Transmutation can reduce all these concerns

Pu-stock in the world



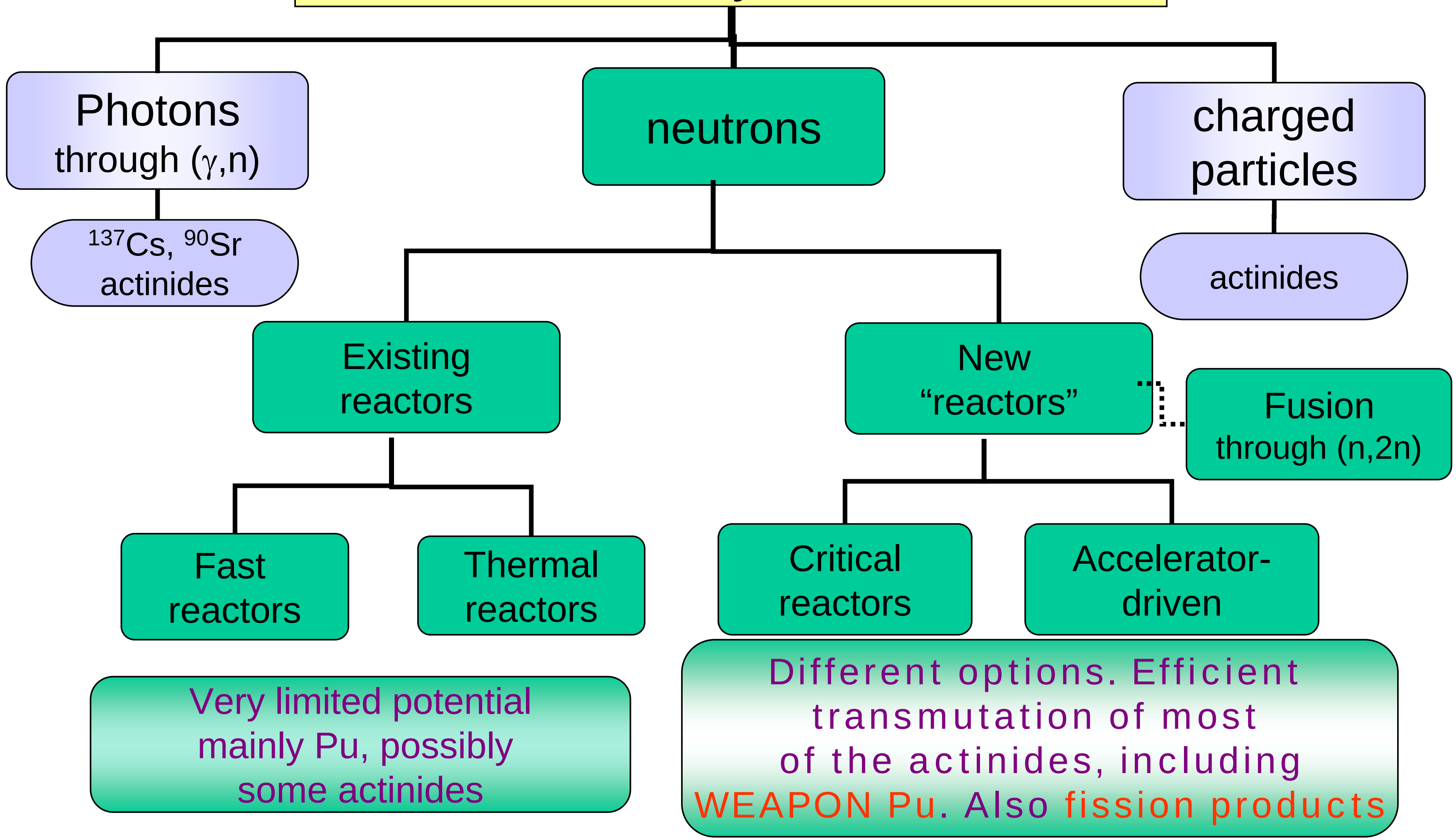
Pu is a problem....

Riskuppfattning för olyckor relaterade till kärnkraften



Transmutation - kan lösa de flesta problem som allmänheten oroar sig för....

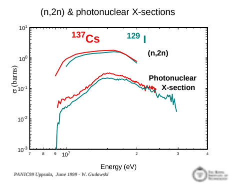
Transmutation Processes by

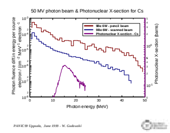


Which method should be chosen?

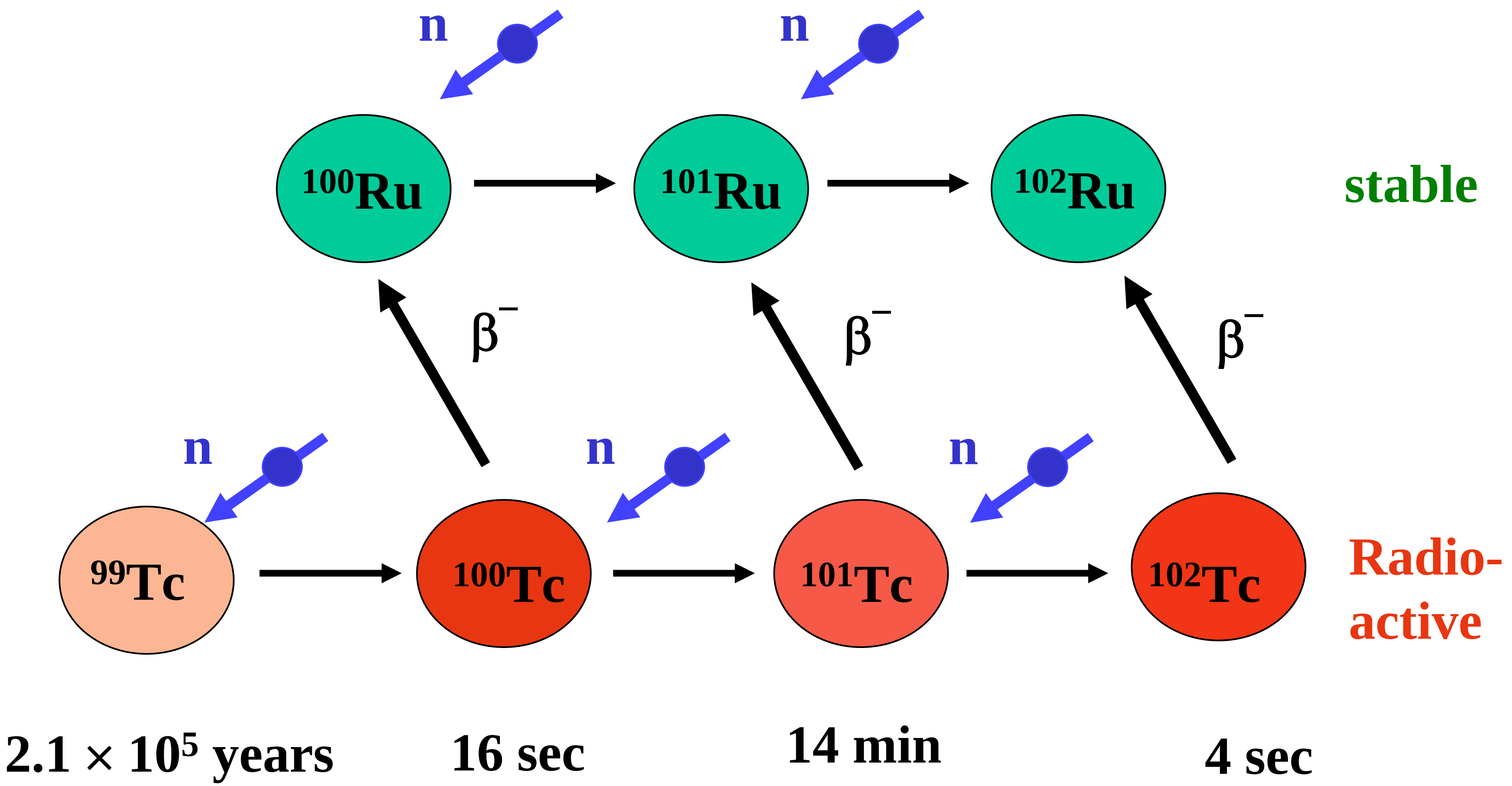
Only fission-enhanced neutron transmutation is economical energetically (exothermic)

- in spite of a fact that (n,2n) reaction X-sections “fit” fusion neutrons, energetic cost is about 200-300 MeV/nuclei for FP transmutation
- direct spallation-transmutation costs ab. 300-600 MeV/nuclei
- (γ,n) even worse, about 4000-5000 MeV/nuclei (assuming electricity production with nuclear power one would produce more e.g. ^{137}Cs than transmute)





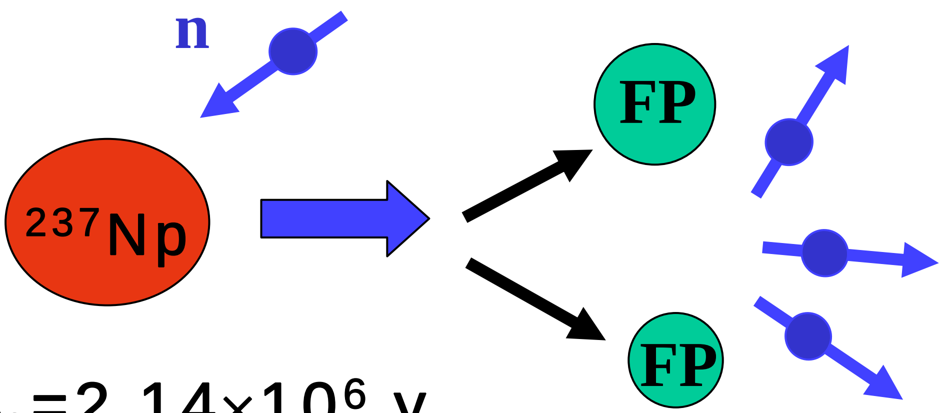
Transmutation through neutron capture, e.g. transmutation of Technetium



Transmutation through capture and fission.

Transmutation of TRU

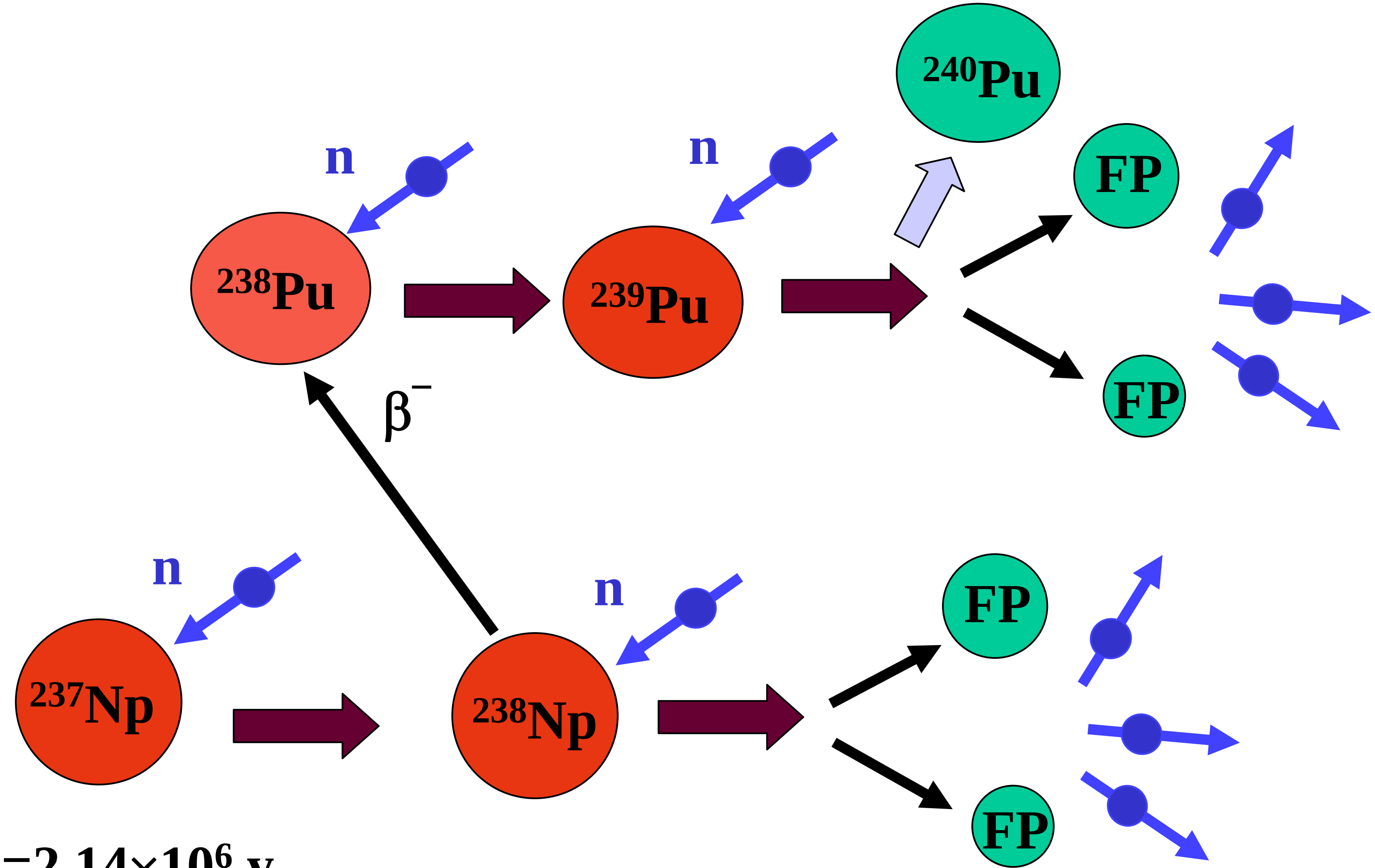
Fast neutrons



Fission (energy)
+ neutrons

$T_{1/2} = 2.14 \times 10^6$ y

Slow neutrons



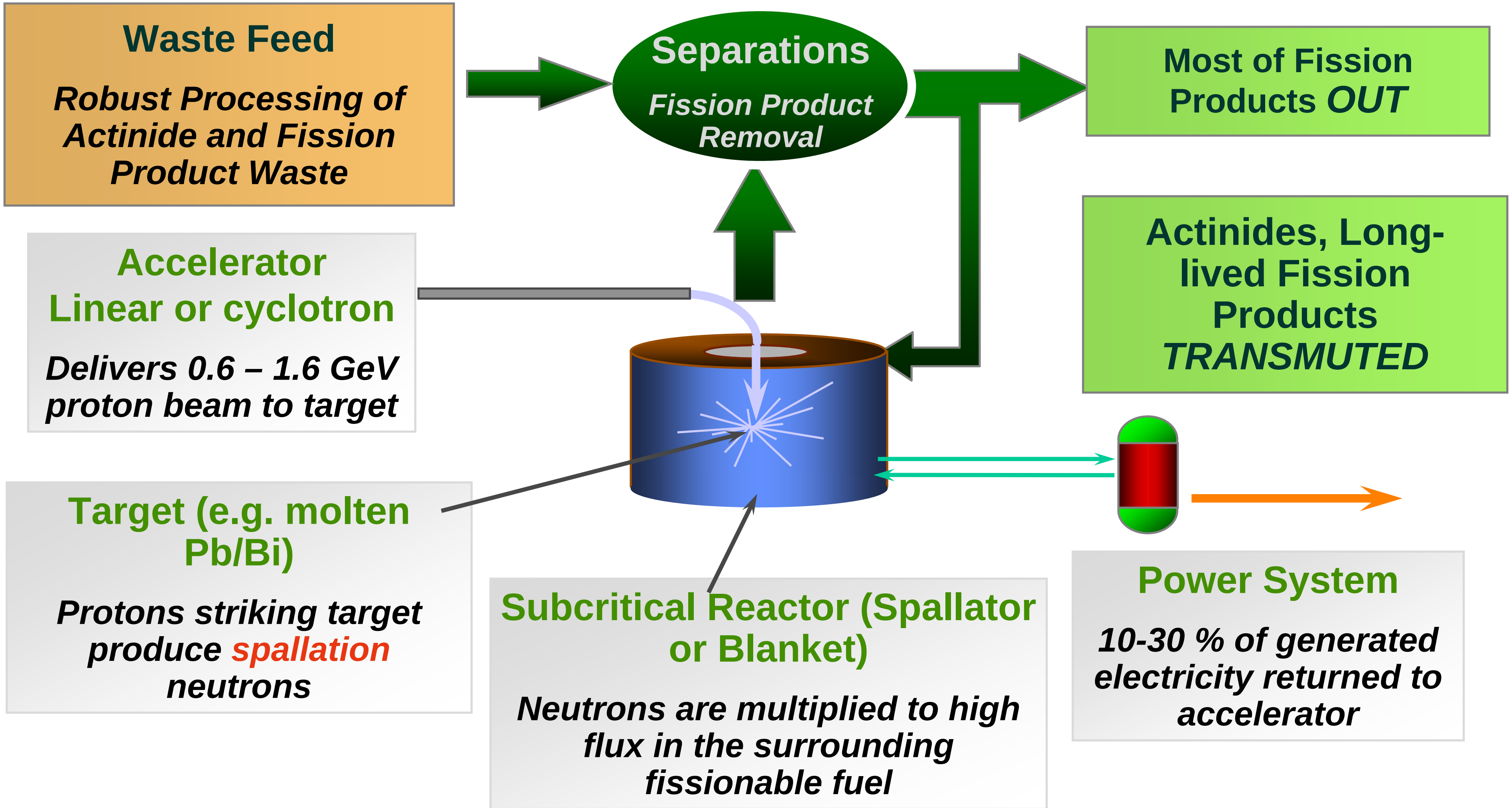
$T_{1/2}$=2.14×10^6 y

$T_{1/2}$=2.1 days

So why Accelerator-Driven Transmutation with neutrons?

- It is exothermic
- It addresses criticality concerns: power control not linked to reactivity feedbacks, delayed neutrons or to control rods, but only to the accelerator drive
- It permits transmutation in dedicated cores, purely transuranic, or other “exotic” fuels
  - subcritical systems work independently of the fuel composition
  - allows constant power during burn-up through variable beam power
  - allows very deep burnup of Pu and other actinides, End of Life inventory not limited by criticality conditions
- Neutronics and thermohydraulics are effectively decoupled

Accelerator driven transmutation Principal Components



What we do at KTH?? A LOT!!

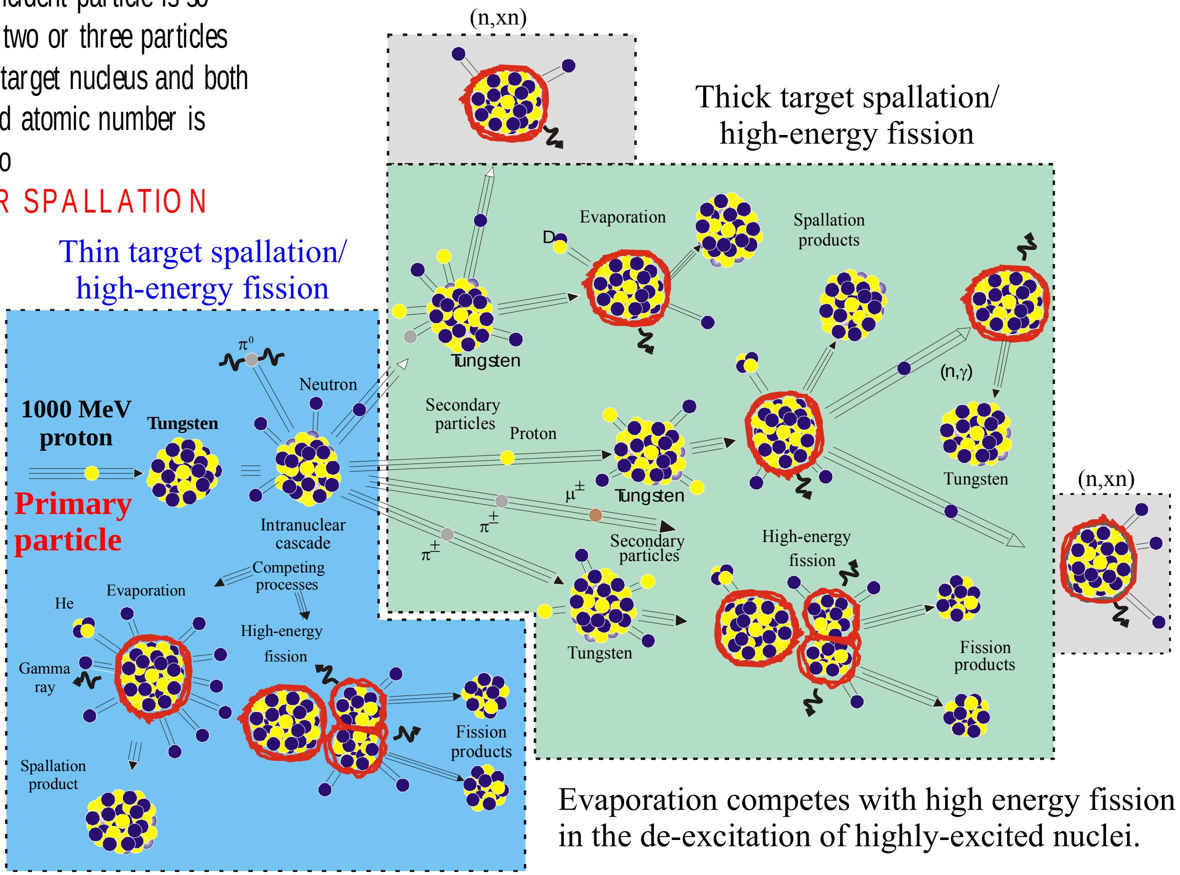
- Think, conceptual designs of ADS, simulate and optimize new ADS, develop new Monte Carlo codes, e.g. burnup Monte Carlo (MCB)
- Transient behaviour and safety aspects of ADS
- From basic physics – spectrum measurements, cross sections to operational measurements
- Radiation damages, simulations, Molecular Dynamics – a path to applications
- Participate in many experiments:
  - 1 MW spallation target, collaboration with Russia, USA (LANL) and France (CEA)
  - MUSE-experiment (a pre-model of ADS) in Cadarache, CEA, in France
  - YALINA experiment in Minsk (pre-model of ADS
  - SAD-experiment in Dubna – the first real ADS

What we do at KTH…

- Many EU-projects in 5th FP(XADS, CONFIRM, MUSE, TECLA, SPIRE, FUTURE, ADOPT, MOST m.m.)
- Important projects in 6th FP – RED-IMPACT, proposal for EUROTRANS, NURESIM + Gen IV
- Technical Working Group – so called Rubbia-group -> European ADS-Roadmapping
- Bilateral collaboration with USA, USA (LANL, ANL), Frace (CEA, CNRS), Germany (FZK), Belgium (SCK-Mol), Korea (KAERI), Japan (JAERI) +more
- Collaboration with IAEA, NEA/OECD

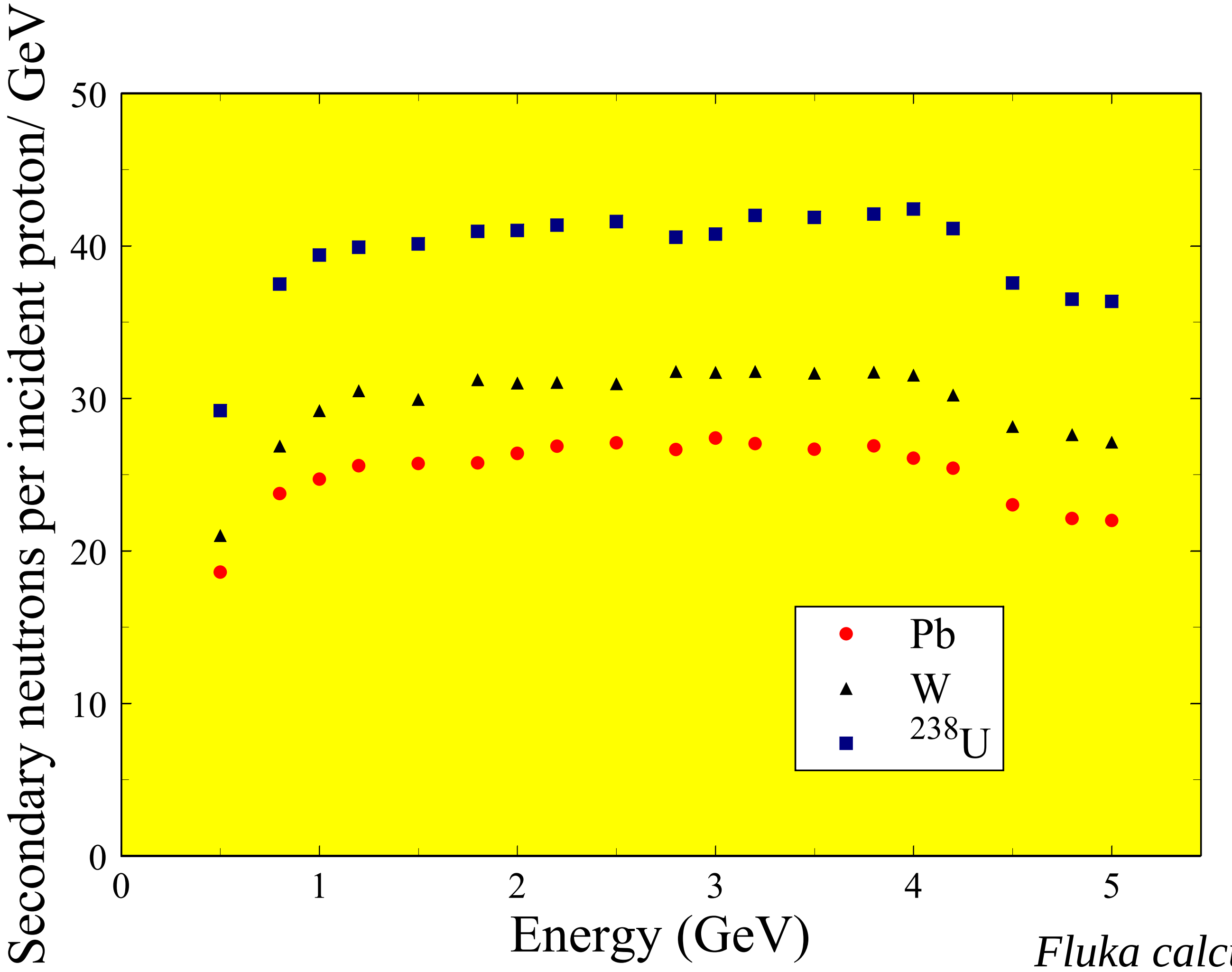
SPALLATION PROCESSES IN THIN/THICK TARGET

SPALLATION: A nuclear reaction in which the energy of each incident particle is so high that more than two or three particles are ejected from the target nucleus and both its mass number and atomic number is changed. Called also NUCLEAR SPALLATION



Evaporation competes with high energy fission in the de-excitation of highly-excited nuclei.

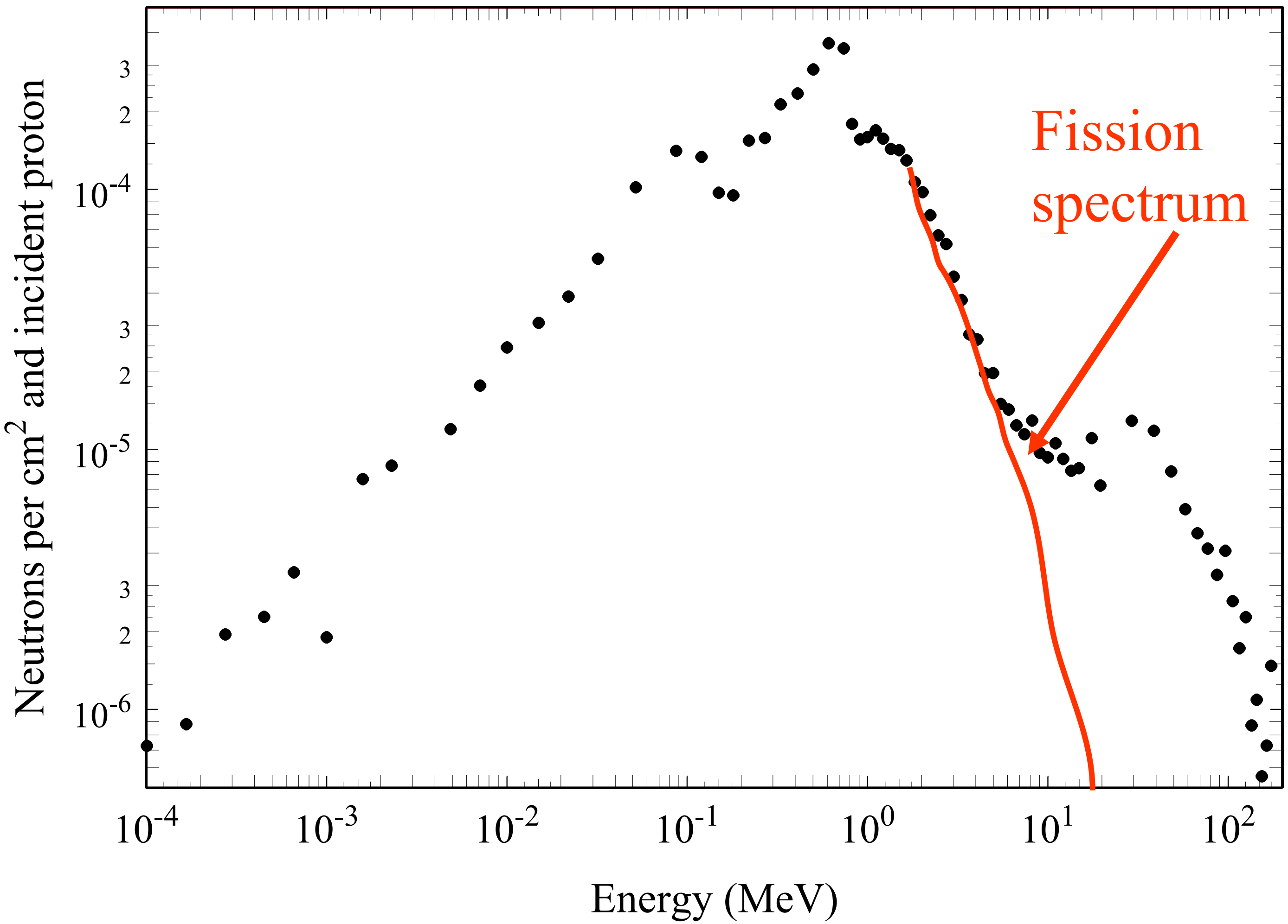
Spallation process is an intense neutron source



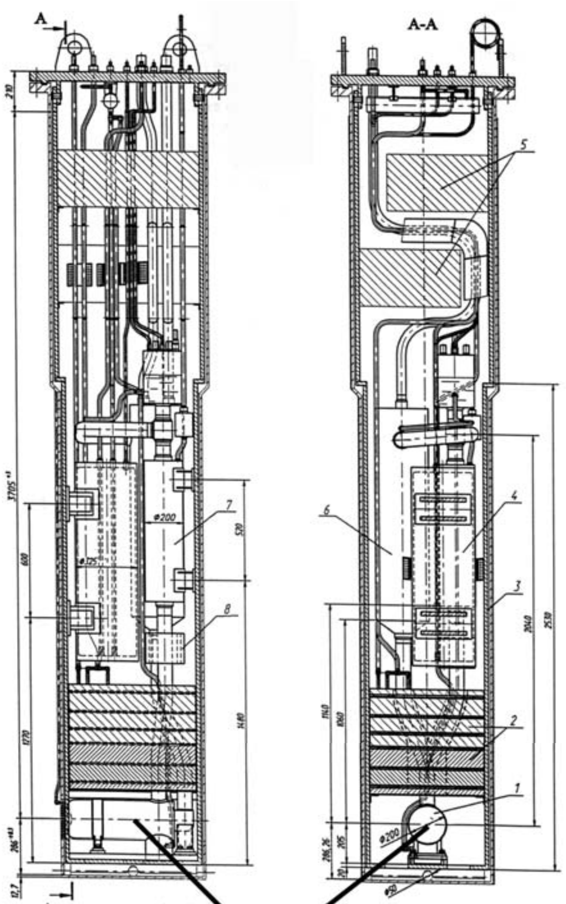
Fluka calculations

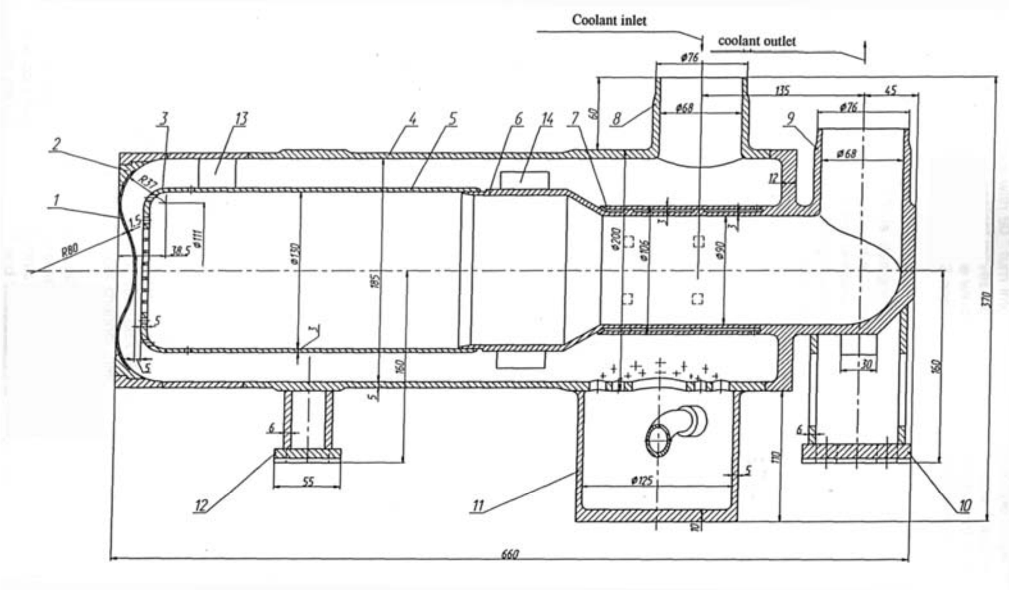
Spectrum of spallation neutrons

Pb/Bi target120x150 cm^2, 1,6 GeV

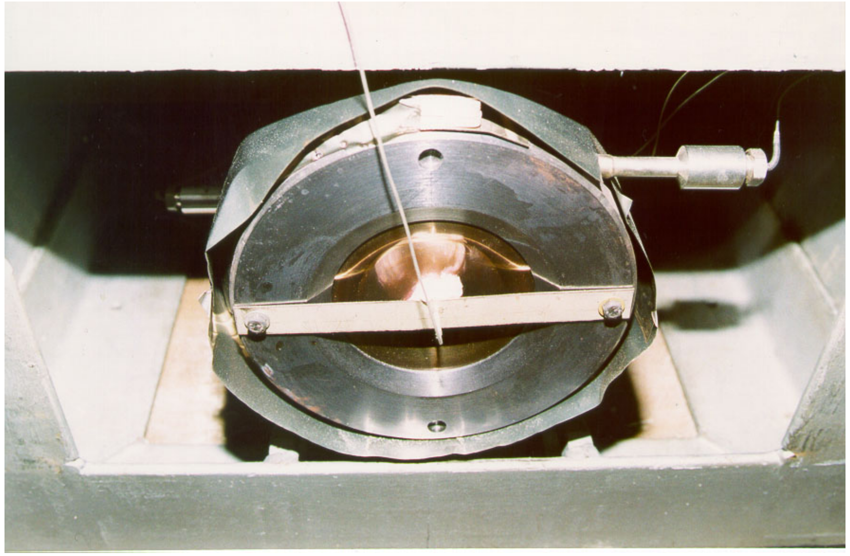


1 MW Liquid Pb-Bi Spallation Target – customizing Pb/Bi Technology

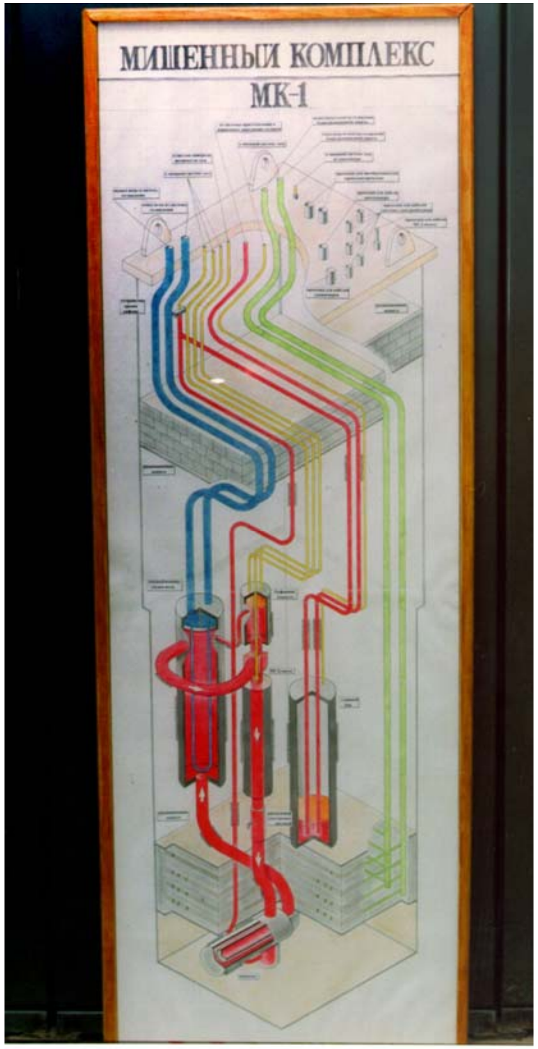


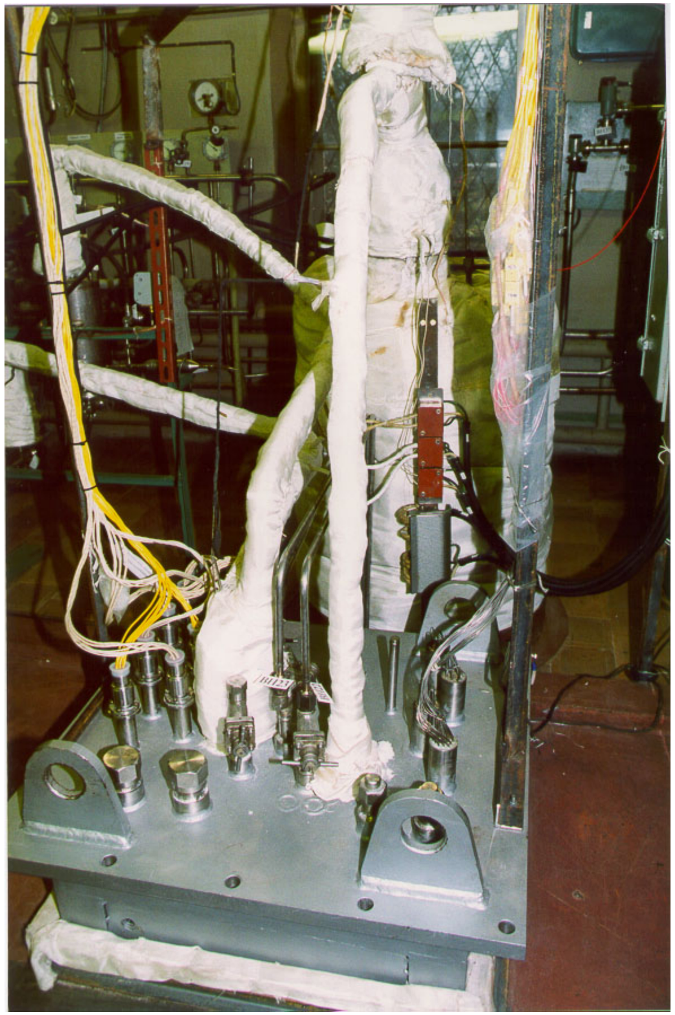


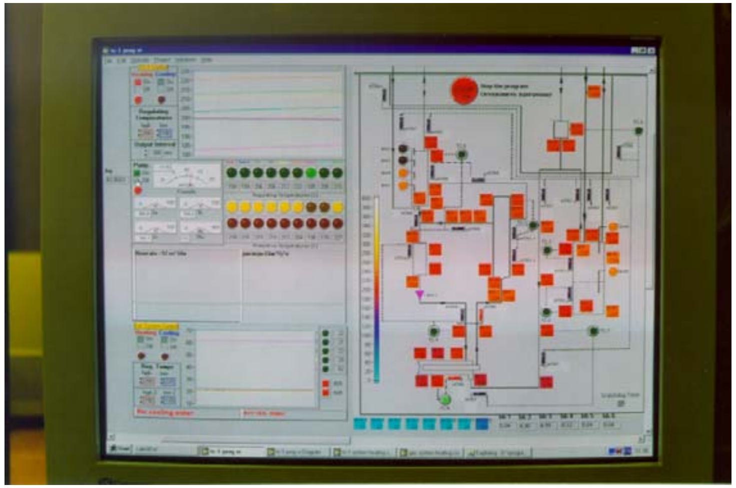
Images of internals of the target



ISTC #559 - 1 MW Pb-Bi spallation target







ISTC #559 - 1 MW Pb-Bi spallation target

What have we learned?

- Customizing Pb-Bi (and Pb) technology – controlling corrosion/erosion processes through:
  - Right choice of the constructional materials (steel)
    - below 400 C – a ”regular” austenitic steel
    - above 450 C – a high Cr-content ferrite-martensitic steel (upper limit ~620 C)
  - Formation of the protecting films on the steel surface
    - protecting films (layers) consist mainly of Fe_3O_4, then FeO, Fe_2O_3 and Cr_2O_3
  - Control of the coolant quality (oxygen and mass transfer control)
    - To keep protective layer of the oxide the concentration of the dissolved oxygen in the alloy should be kept on the level of equilibrium with Fe_3O_4

$$K = \frac{a_{[Fe_3O_4]}}{a^2_{[O_2]} \cdot a^3_{Fe}}$$

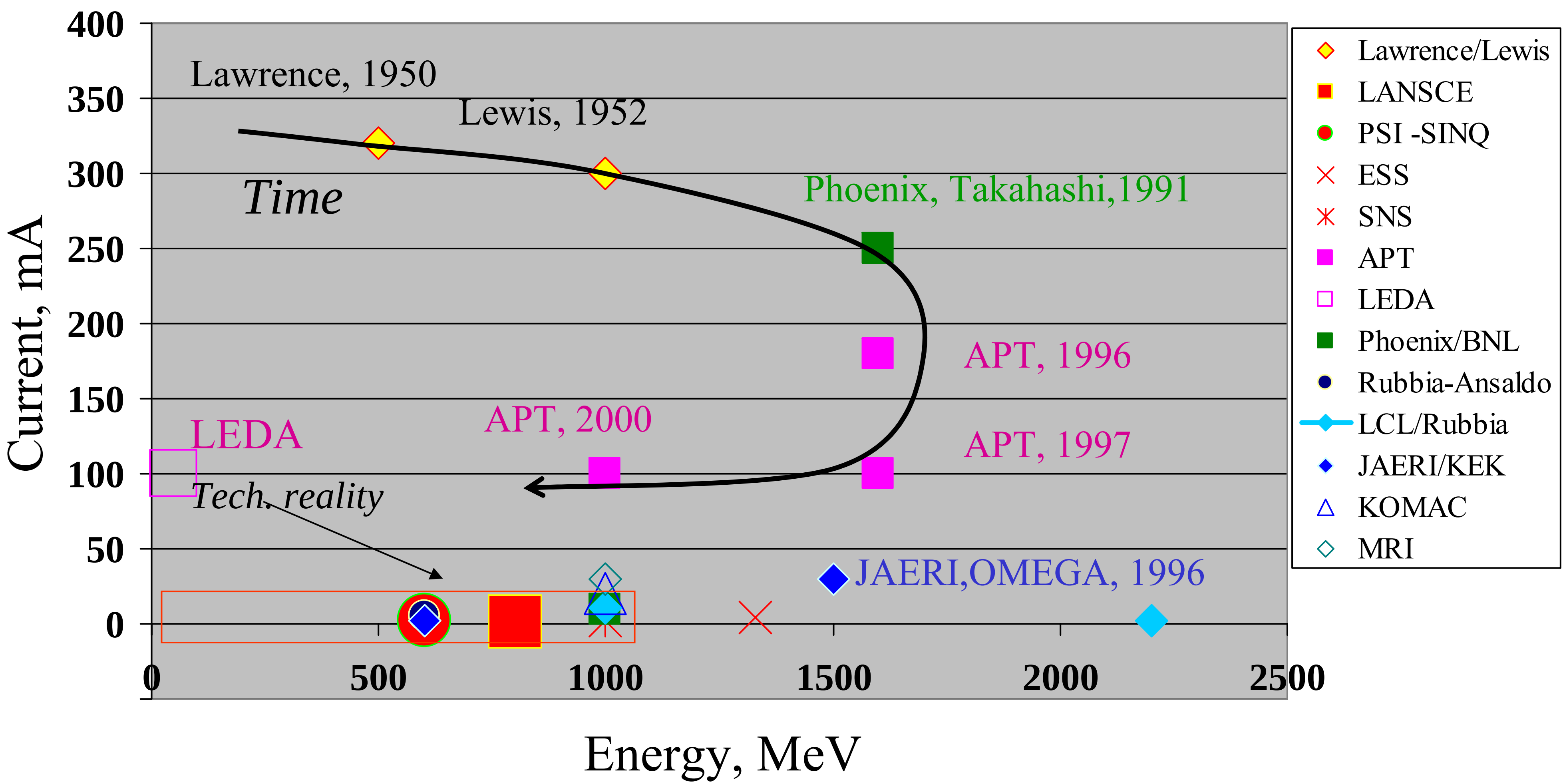
MEGA-PIE EXPERIMENT at PSI

- A child of of Russian target!
- An efficient technology transfer

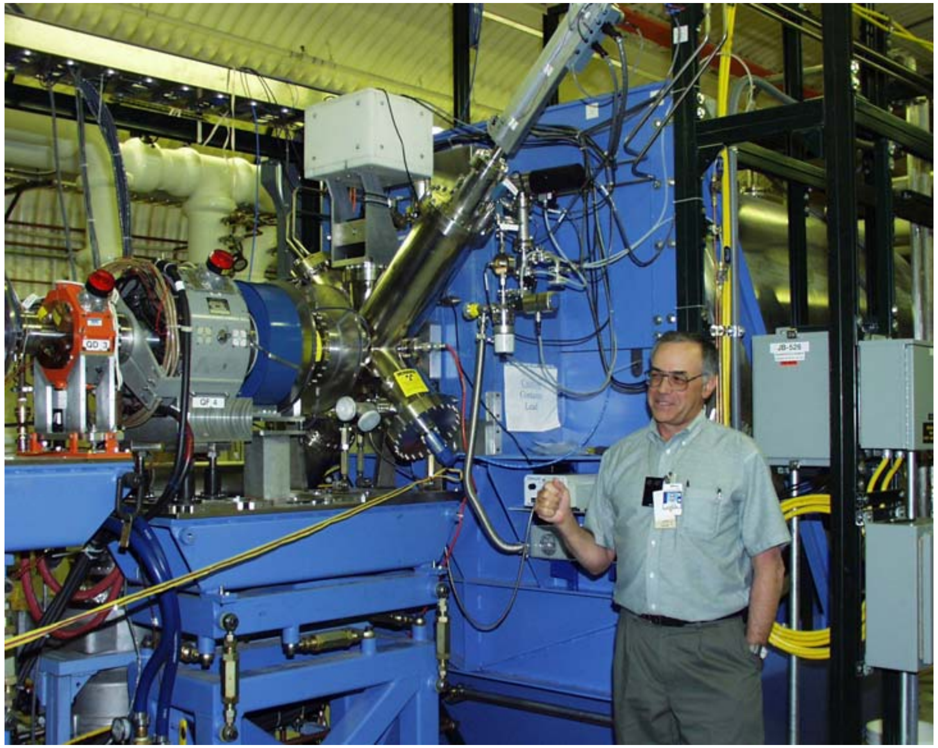
a) b)

Accelerator options

Accelerators proposed for ATW/APT/ADS/Spallation

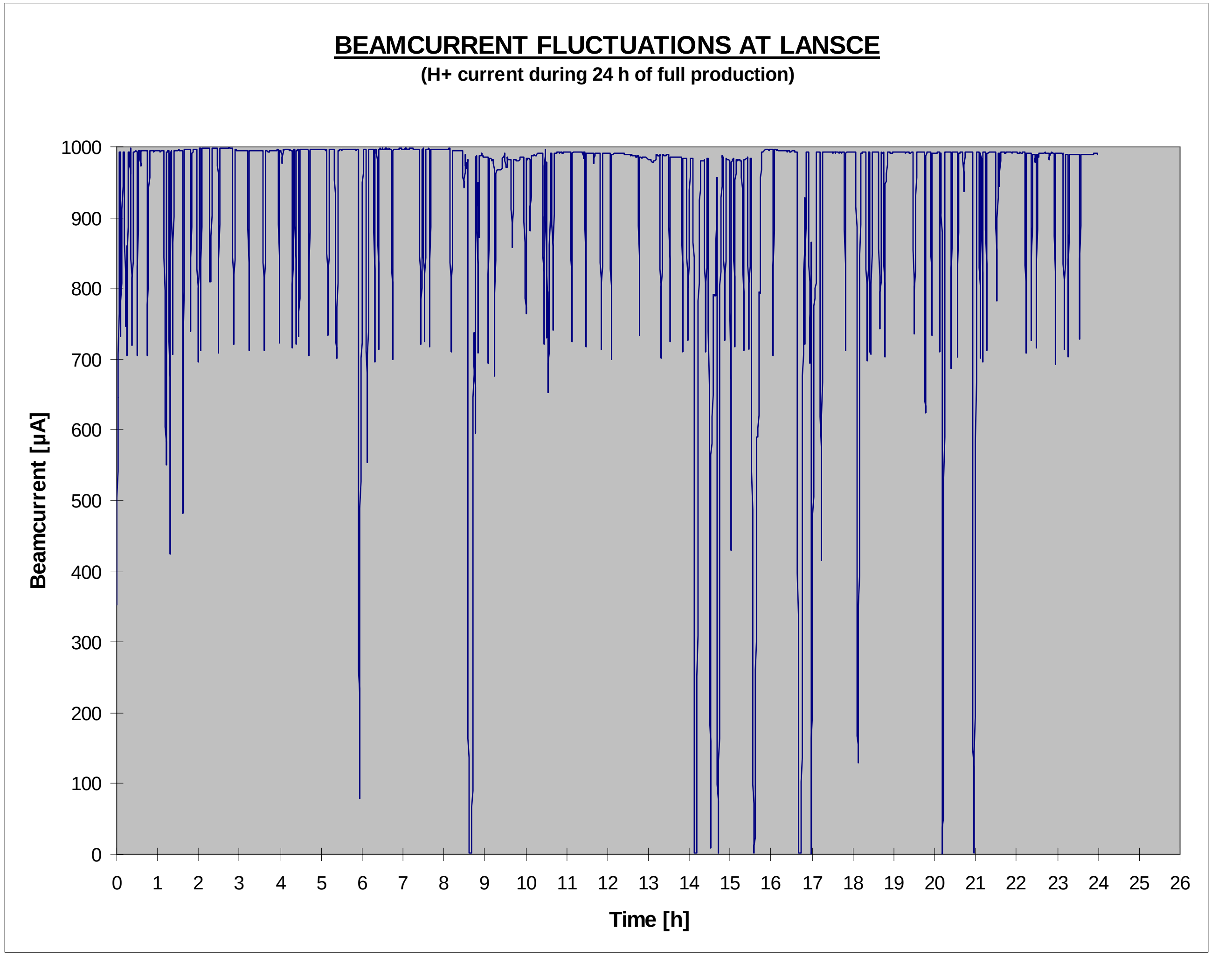


LEDA - facility

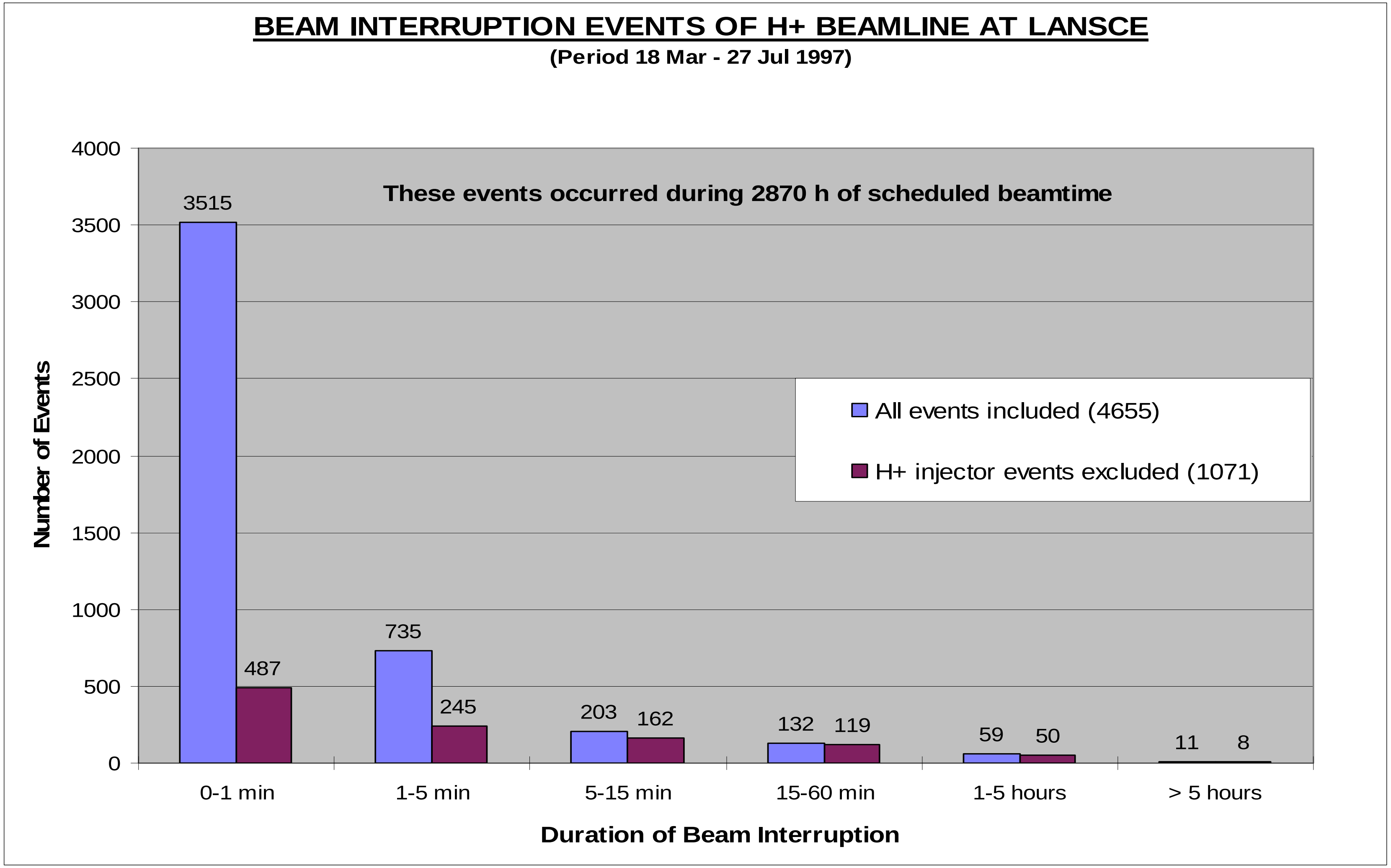


Accelerator performance

must improve

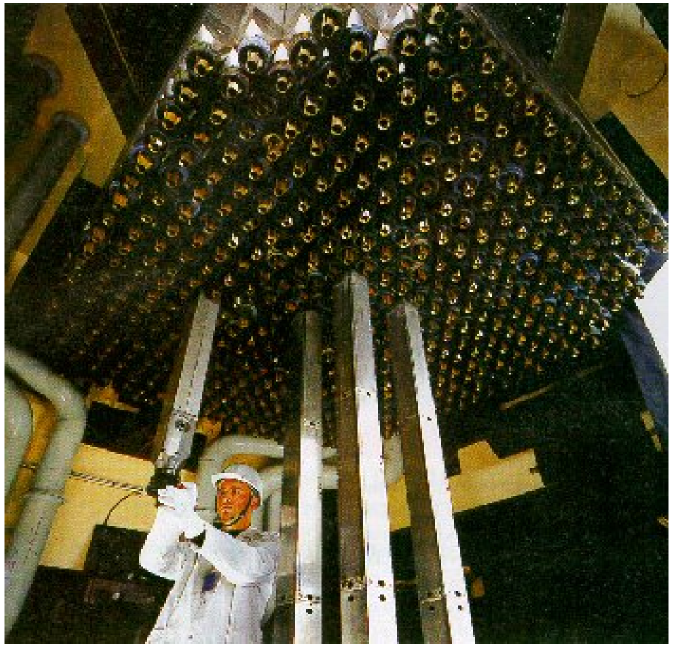


Beam interruption events at LANSCE



Integral experiments - the MASURCA Facility based MUSE project

MASURCA is the experimental reactor at CEA/Cadarache dedicated to studies of ADS, in the framework of the MUSE-program (currently MUSE-4).



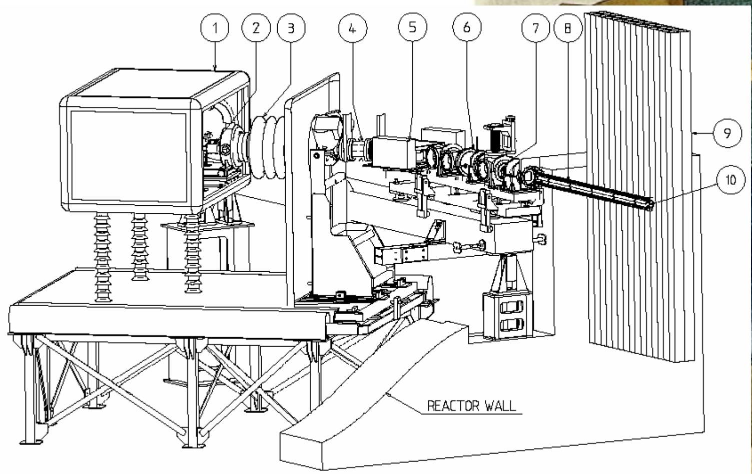
Some Characteristics of MASURCA:

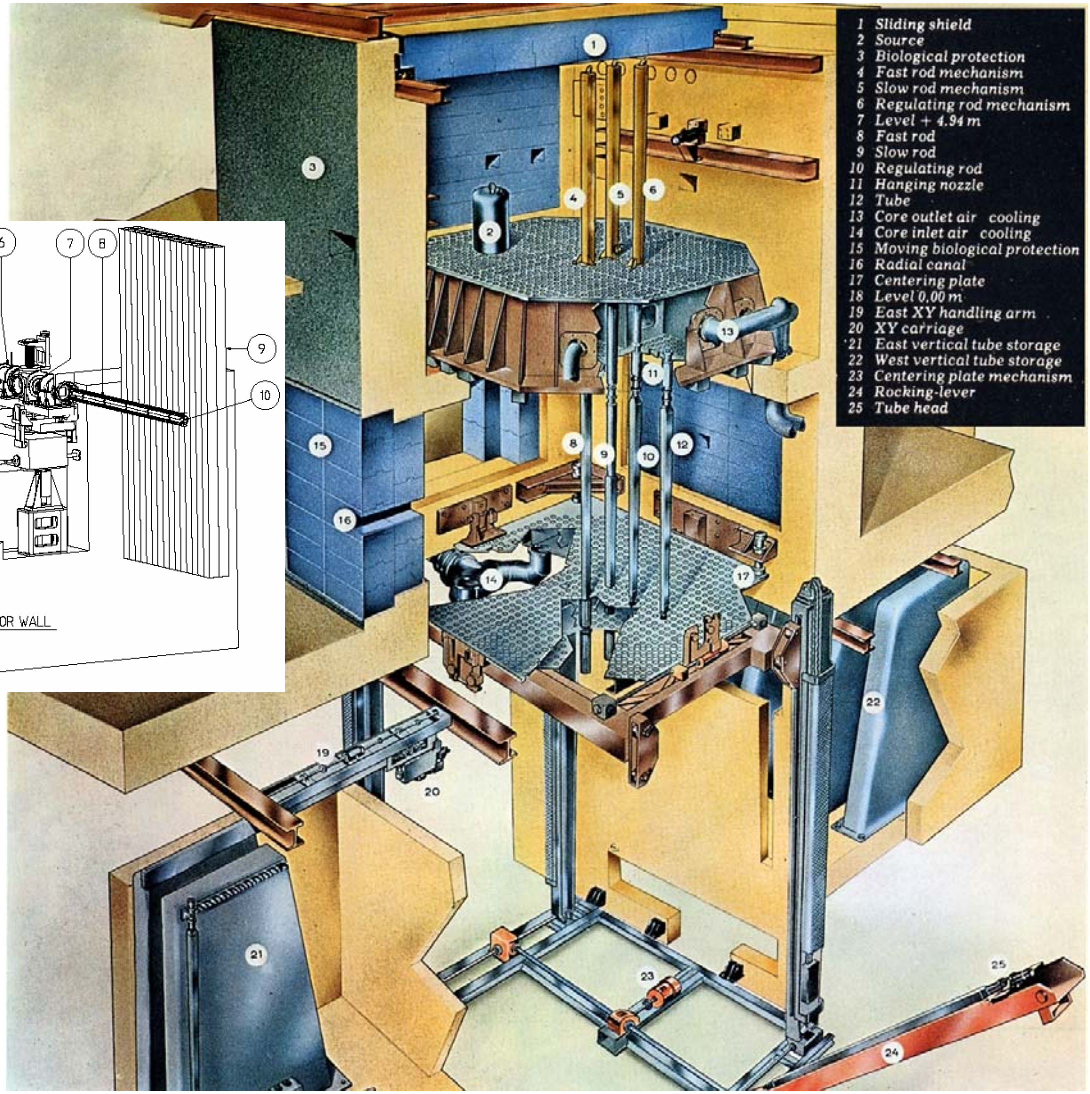
- **Loaded with 10.6*10.6 cm subassemblies**
- **Flexible design**
  - Different fuels can be used (Th, U, Pu)
  - Different coolant media can be simulated (sodium, lead, gas)
  - Different levels of sub-criticality are possible
- **Low Power (air cooled)**

Objectives with the MUSE Experiments:

- ❑ Experimental Validation of the Main Physical Principals of a Sub-critical System:
- ❑ Validation of Nuclear Data and Calculation Codes:

MASURCA & MUSE



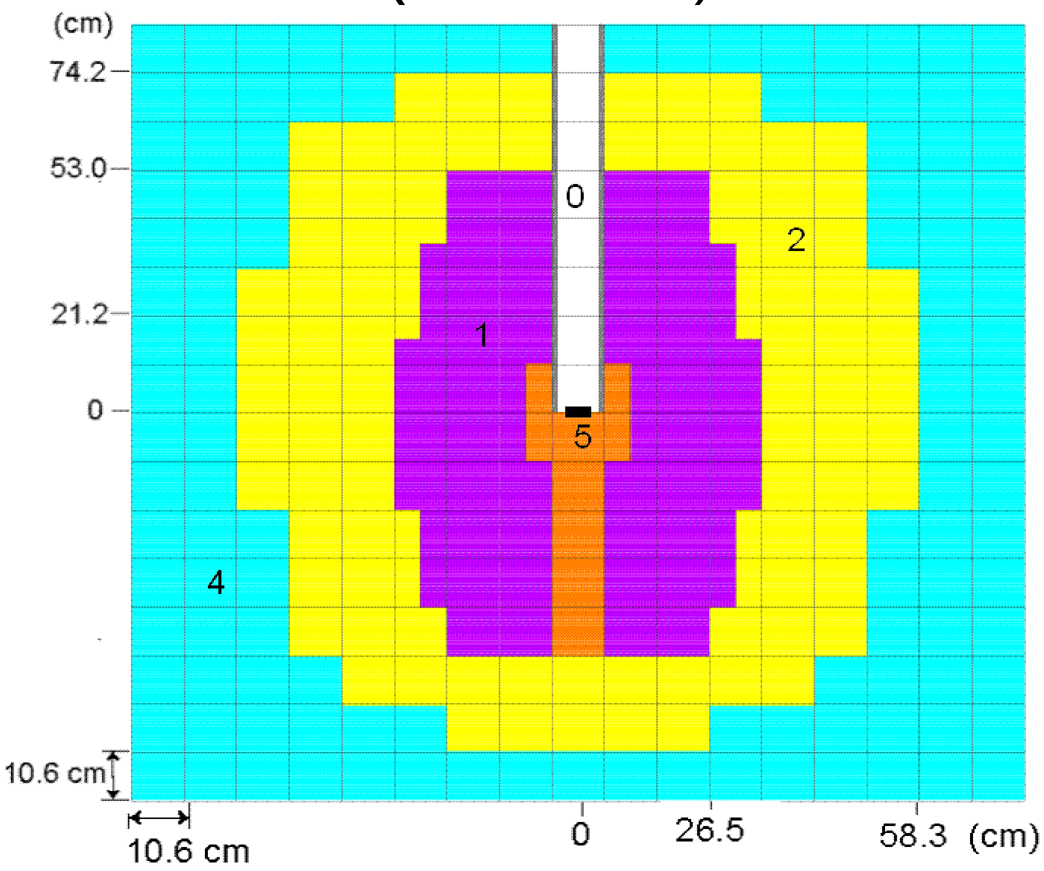


Program of MUSE

- Experimental characterisation of important neutronic properties of a multiplying sub-critical media driven by an external source. The properties can be described in terms of reactivity, external source worth, flux and power distributions and neutron spectra etc.
- **Development of sub-criticality measurements and monitoring.**
- Obtaining a database to validate the predictions of the computing codes.
- Identifying possible deficiencies in the data or the methods.
- Comparing different experimental techniques for incineration of long-lived fission products

The Geometry of MUSE-4

MUSE-4 (k-eff ~ 0.97)



Different parts of MASURCA:

- 0: Accelerator Tube (250 keV deuterons)
- Target (Deuterium or Tritium)
  - (D,D) – E_n ~ 2.0 – 3.1 MeV
  - (D,T) – E_n ~ 13.1 – 15.2 MeV
- 1: Fuel + Na-coolant
- 2: Na/SS Reflector
- 3,4: Radial and Axial Shields
- 5: Lead Buffer

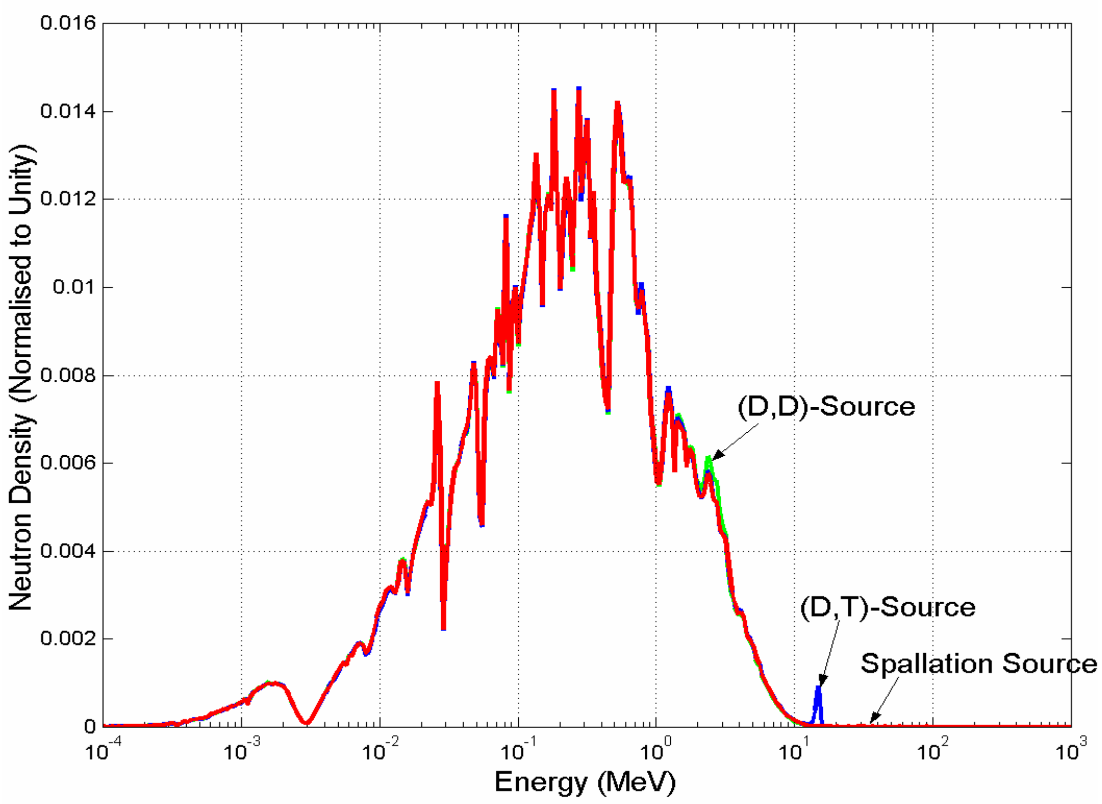
Composition of the Fuel:

- U-238: 72 %
- Pu-239: 21 %
- Pu-240: 5 %
- Small traces of other actinides

Schematic view of the sub-critical MUSE-4 configuration (seen from above). The accelerator tube is introduced horizontally into the core.

Results - Neutron Energy Spectra

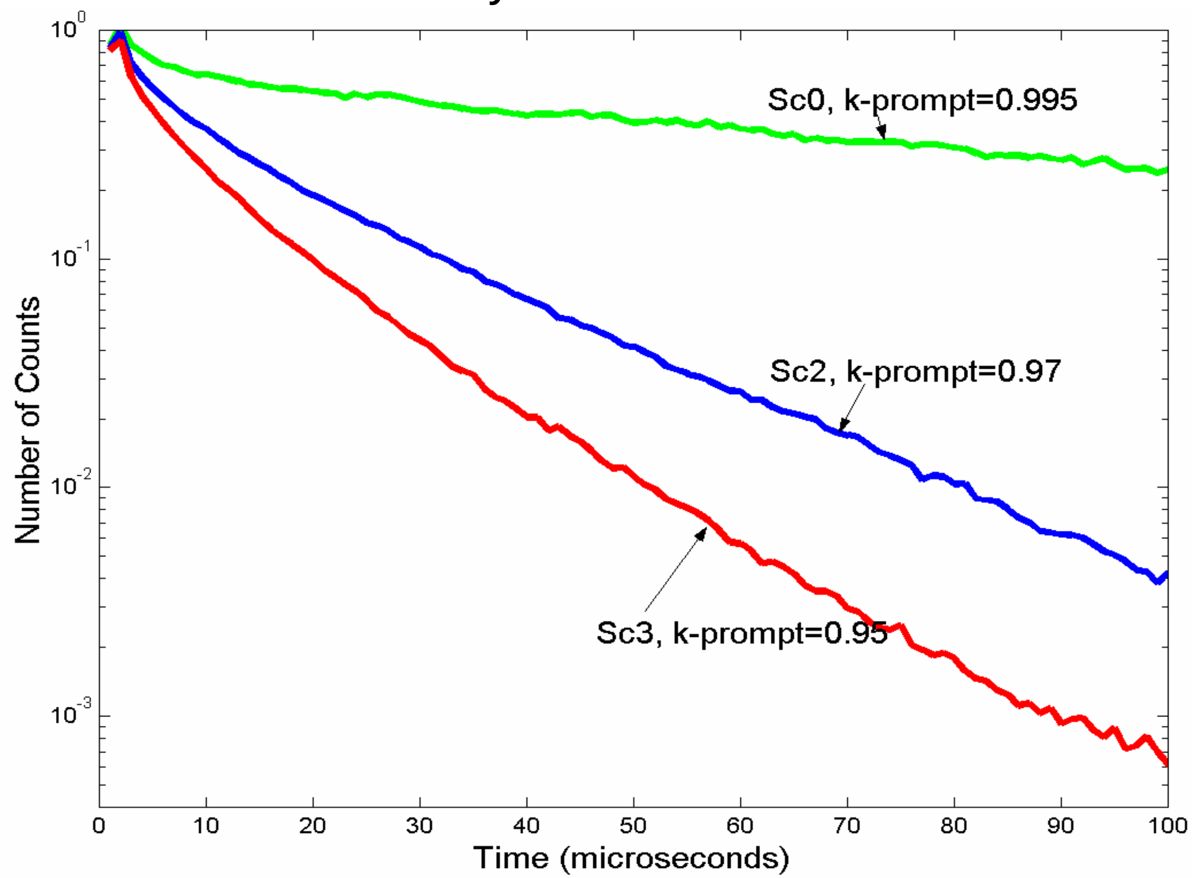
The spectra were found to be largely dominated by the fission multiplication in the fuel and the origin of the external neutron sources are nearly dissapeared.



<u>Conclusion:</u> Considering only the neutron energy spectrum, the presence of the sources can be considered forgotten in the fuel

Results – Time Detector Response

Sub-critical reactivity determination with the "Pulsed Neutron Source Method"



Relation governing the prompt decay process after a neutron pulse:

$$N(t) = N_0 e^{-\alpha t}$$

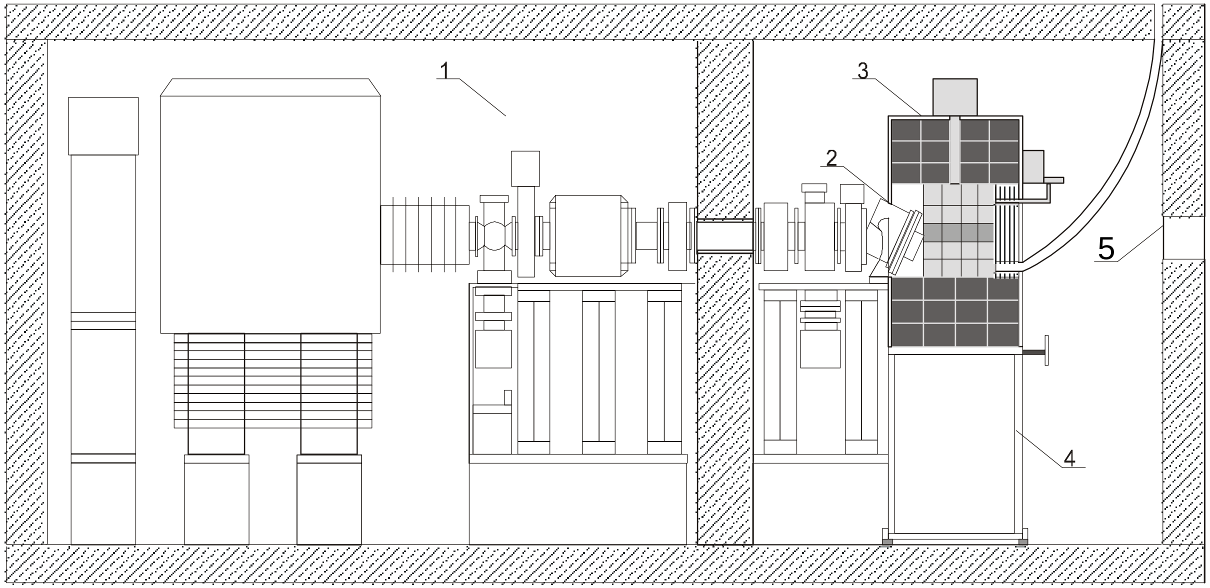
$$k_{prompt} = 1 - \alpha \cdot \Lambda$$

Prompt decay (no delayed neutrons) of a neutron pulse:

Conclusion: Larger sub-criticality → faster decay rate (larger α)

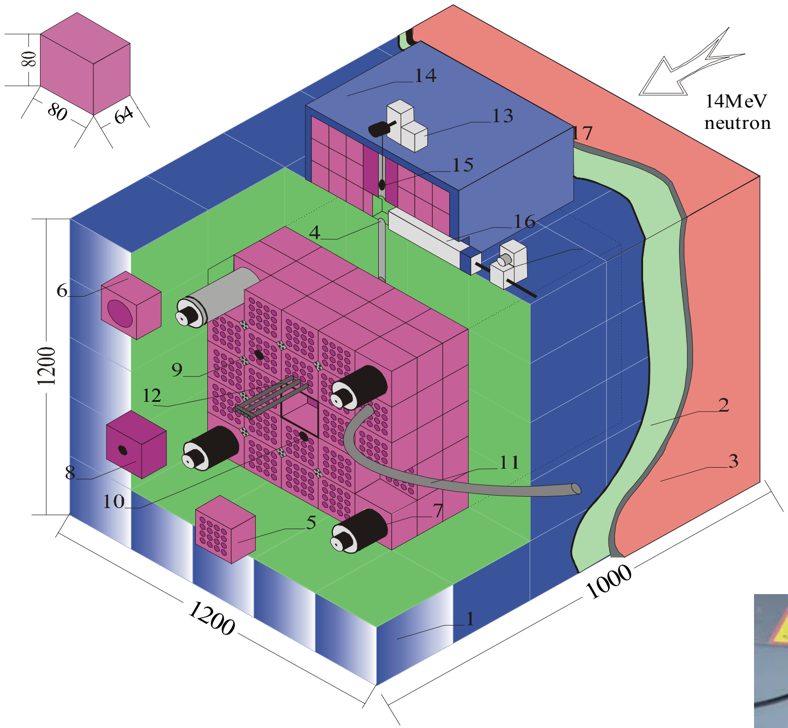
Yalina-experiment

Neutron generator: flux of neutrons (2.0 MeV < E_n < 3.0 MeV) equals approximately to $2 \cdot 10^{10}$ n/s at deuteron current and energy equal to 10 mA and 250 keV resp.
The duration of the neutron pulse τ can be changed from 5 μsec to 100 μsec and the pulse repetition can be changed from 1 to 1 000 Hz.



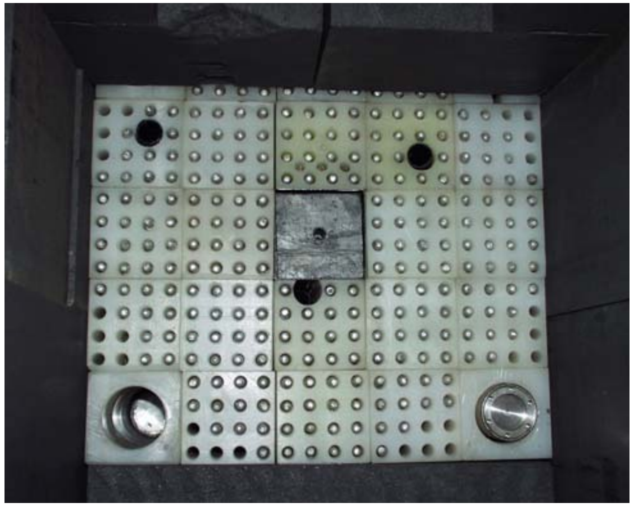
The subcritical facility "Yalina": 1 - neutron generator, 2 - Ti-^{3}H target system, 3 - subcritical assembly, 4 - movable platform, 5 - collimator.

Yalina set-up



Uranium-polyethylene assembly

1 - graphite block
2 - cadmium screen
3 - covering
4 - neutron source channel
5 - polyethylene block
6 - block of control and protection system
7 - neutron sensor
8 - lead target block
9 - fastener
10 - exsperimental channel
11 - rabbit system pipe
12 - compensation rods
13 - servo-motor of neutron source
14 - container with netron source
15 - neutron source
16 - damper
17 - servo-motor damper



Yalina objectives

- Investigation of physics of the subcritical systems driven by a neutron generator,
- Measurements of transmutation rates of the fission products and minor actinides,
- Investigation of spatial kinetics of the subcritical systems with the external neutron sources,
- Validation of the experimental techniques for subcriticality monitoring, neutron spectra measurement etc.
- Investigation of dynamics characteristics of the subcritical systems with the external neutron sources in pulse mode of the neutron generator operation

SAD facility

Subcritical Assembly Driven by Proton Accelerator in Dubna

At **Joint Institute for Nuclear Research**

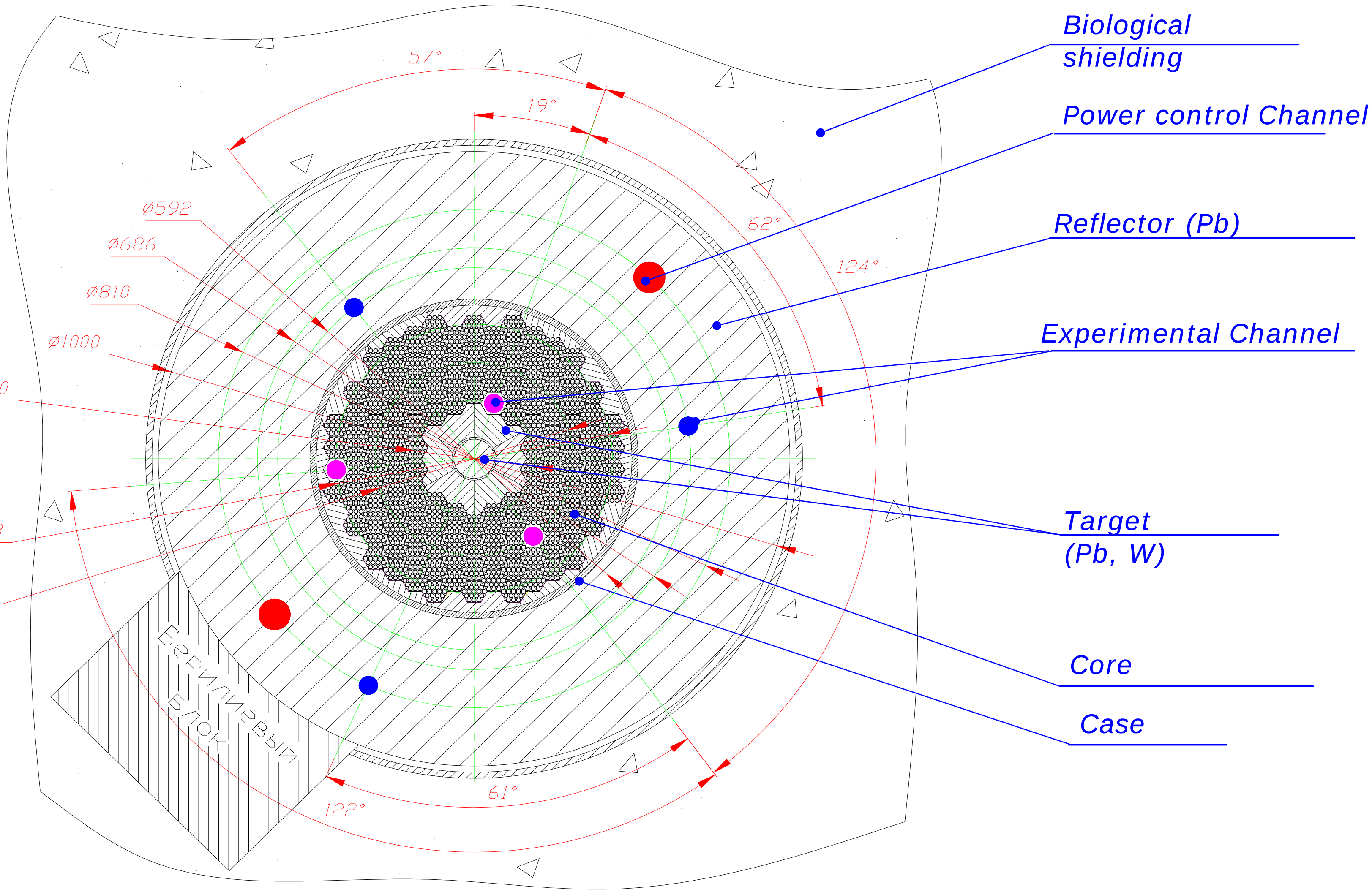
Financing Party: **EC (Sweden, Germany France, Spain)**

Timing: **Physical Startup at 2006 – 2007**

SAD Basic Data

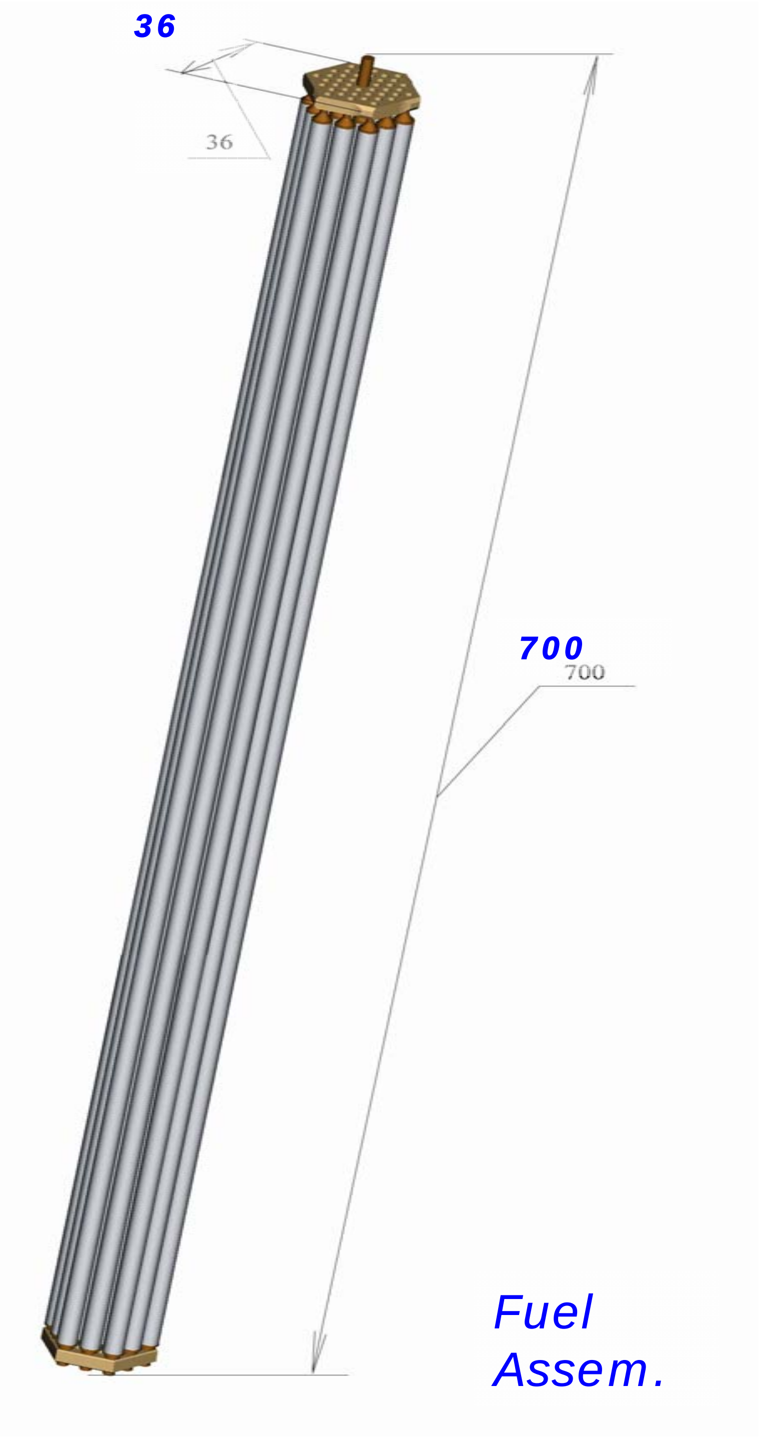
- **Proton beam power: 0.5 – 1 kWt**
- **Core thermal power: 15 – 20 kWt**
- **Proton beam direction on target: vertical**
- **Subcritical MOX blanket with $k_{eff} \leq 0.95$**
- **$PuO_2 + UO_2$ fuel pellets**
- **$PuO_2 \leq 30\%$**
- **^{239}Pu content $\geq 95\%$**
- **Fuel density: 10.2 ± 0.2 g/cm^3**
- **Air cooling**

SAD Core


Biological shielding
Power control Channel
Reflector (Pb)
Experimental Channel
Target (Pb, W)
Core
Case
57°
19°
62°
124°
61°
122°
ø592
ø686
ø810
ø1000
ø190
ø438
ø314
Бериллиевый блок

SAD Core: Fuel Elements

- Number of Fuel Assemblies – 141;
- Number of FE in FA – 19;
- Fuel – (MOX) UO_2 – PuO_2
- Fuel density – 10.2 g/cm^3;
- FEA spacing – 36 mm;
- FE spacing – 7.95mm;
- FE clad tube diameter – 6.9mm;
- Clad tube wall thickness – 0.4mm;
- Fuel pellet diameter – 5.95mm;

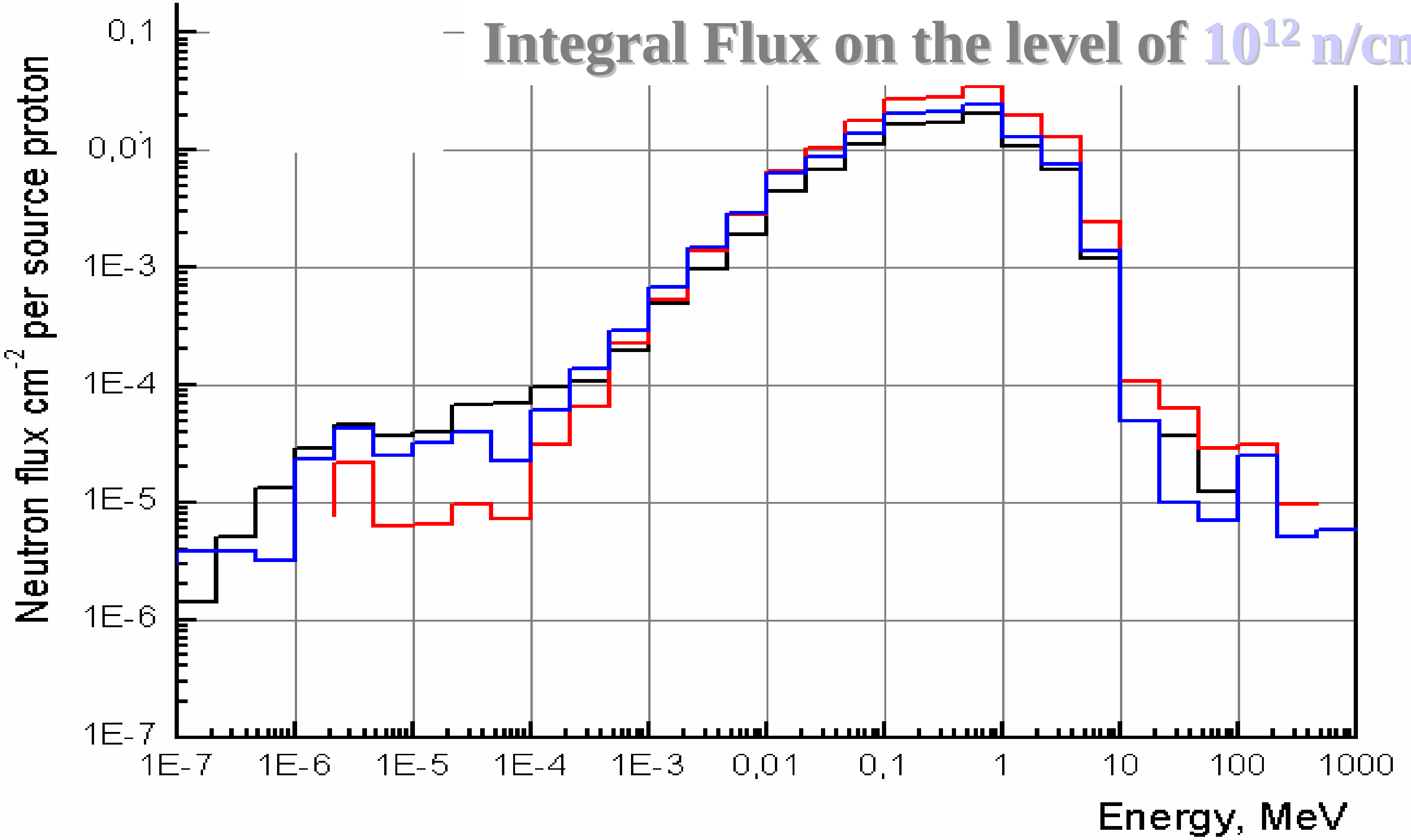


Fuel
Assem.

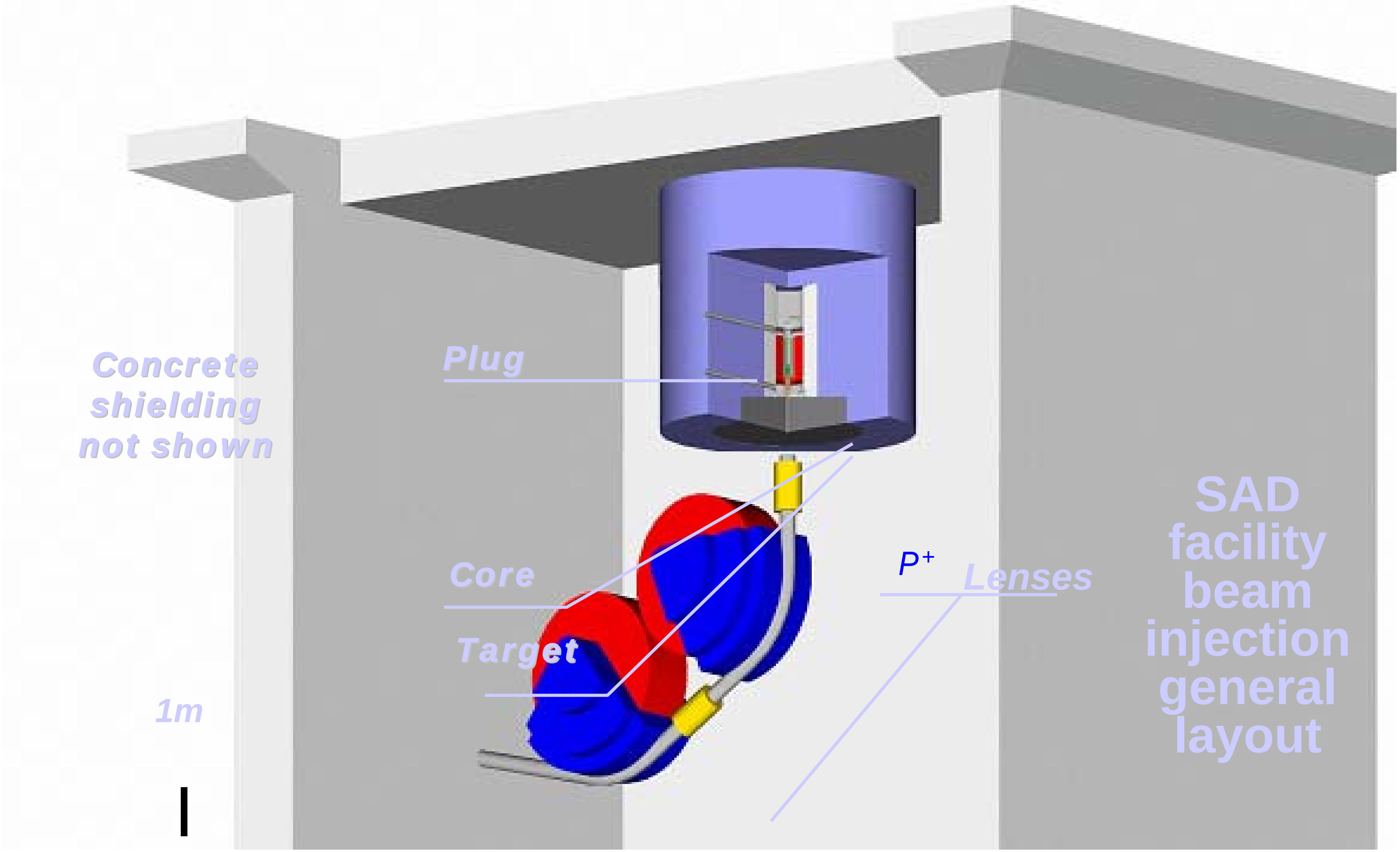
SAD Neutrons

Neutron spectra at the experimental channel in the middle of core (red - in the center, blue, black - tom and bottom correspondingly)

Integral Flux on the level of 10^{12} n/cm^{2}s



SAD general layout



SAD - scientific program

Studying the problems of target and core integration, including influence of the target size and position on main SAD characteristics

Measurements of absolute value of the SAD power gain and reactivity, computer codes validation

Deep subcriticality measurements and monitoring

Measurements of the kinetic properties of the blanket

Measurement of shielding efficiency (especially in a direction of a primary proton beam)

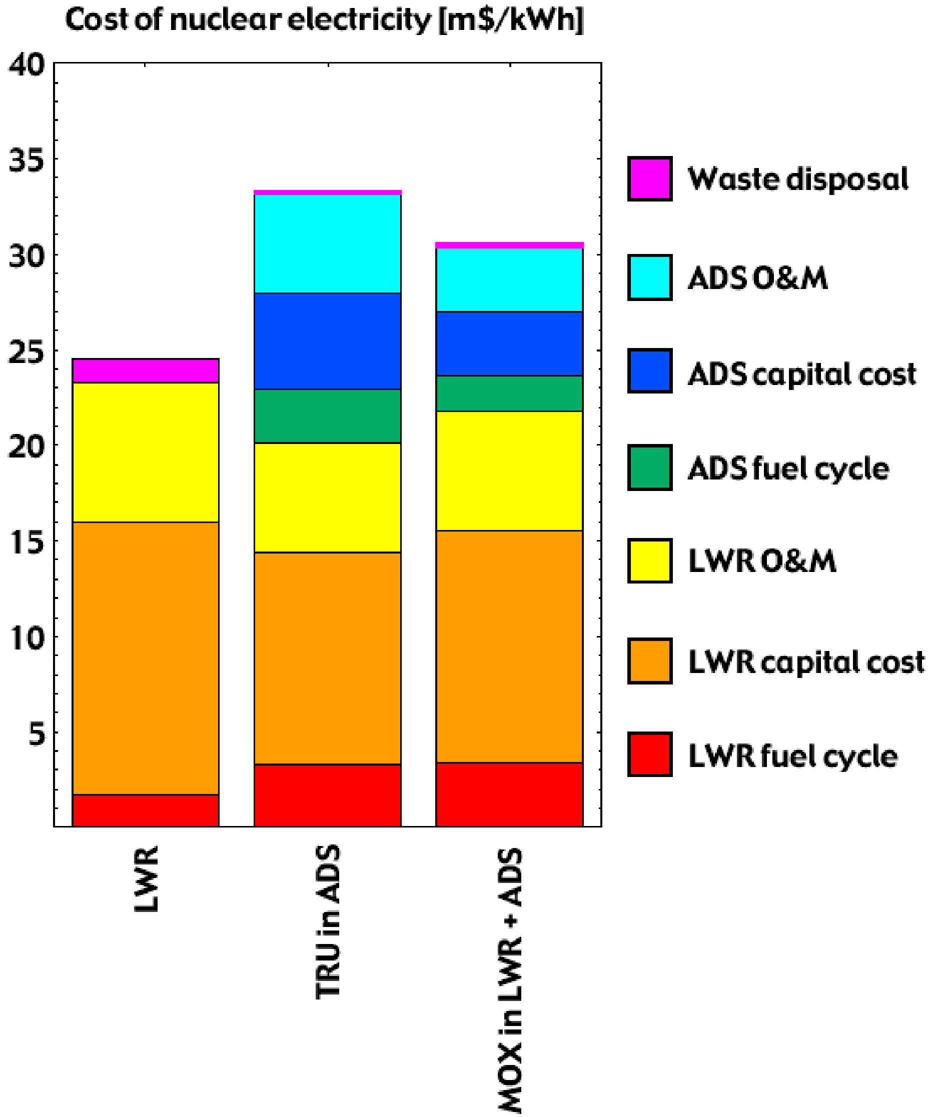
Studying spatial and energy distributions of neutron field in target and fuel blanket

Measurements of transmutation rates for MA and LLFP in different neutron spectra, computer codes validation

Studying the spallation products yields in target using He jet techniques

VALIDATION EXPERIMENT!!

We even do cost/benefit assessments



Conclusions

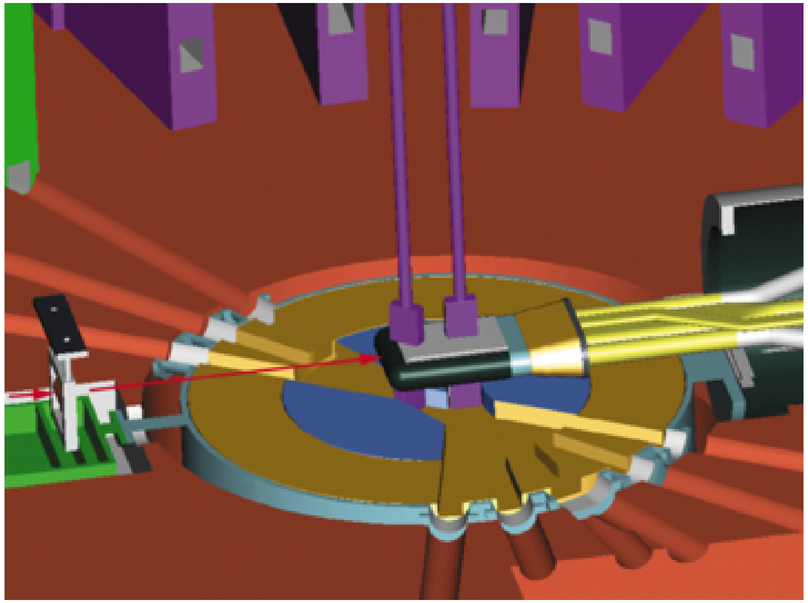
- Instrumental challenges:
  - More basic data on ”exotic” isotopes – ”cross-section” physics with highly radioactive samples and energies upp to GeV
  - Kinetic behaviour of ADS, subcriticality monitering: very sophisticated and difficult neutron flux measurements covering counting rates in 5-6 order of magnitudes with good precision
  - Corellation measurement
  - New shielding problems

Conclusions

- Synergies:
  - With spallation neutron physics: SNS at USA, KEK/JAERI in Japan, ESS- hopefully
  - With reactor physics
    - Reactivity measurements
    - Fuel development
    - Radiation damages and aging
    - Development of new reactors (Generation IV)

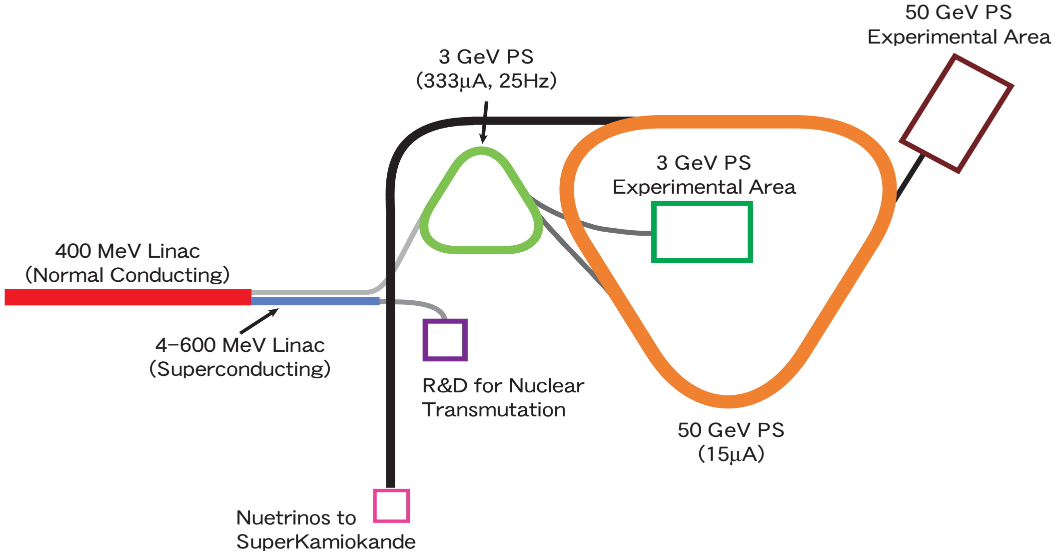
SNS – project in USA

US - THE SPALLATION NEUTRON SOURCE (SNS), $1.3 billion project. Facility under construction at Oak Ridge National Laboratory. Mercury target!



KEK/JAERI – multipurpuse facility

(approved 1.5 G$ project)



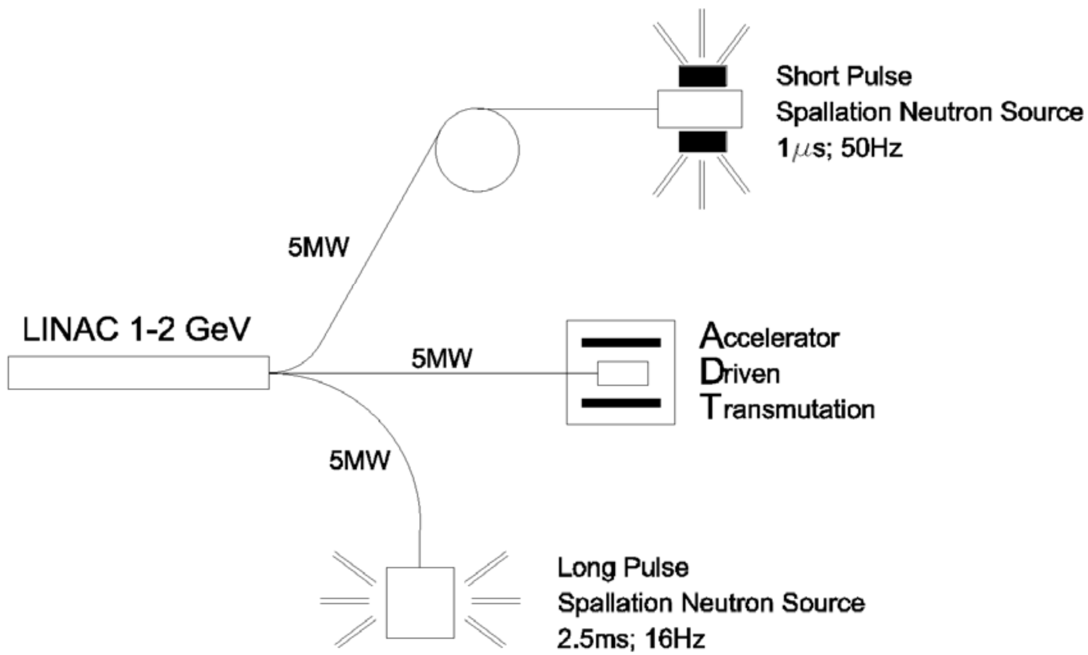
Major Highlights

- Multi-purpose nuclear-, particle-, materials-, and biological sciences plus nuclear technology development. (ADS for nuclear transmutation)
- World-class facility open to international users.

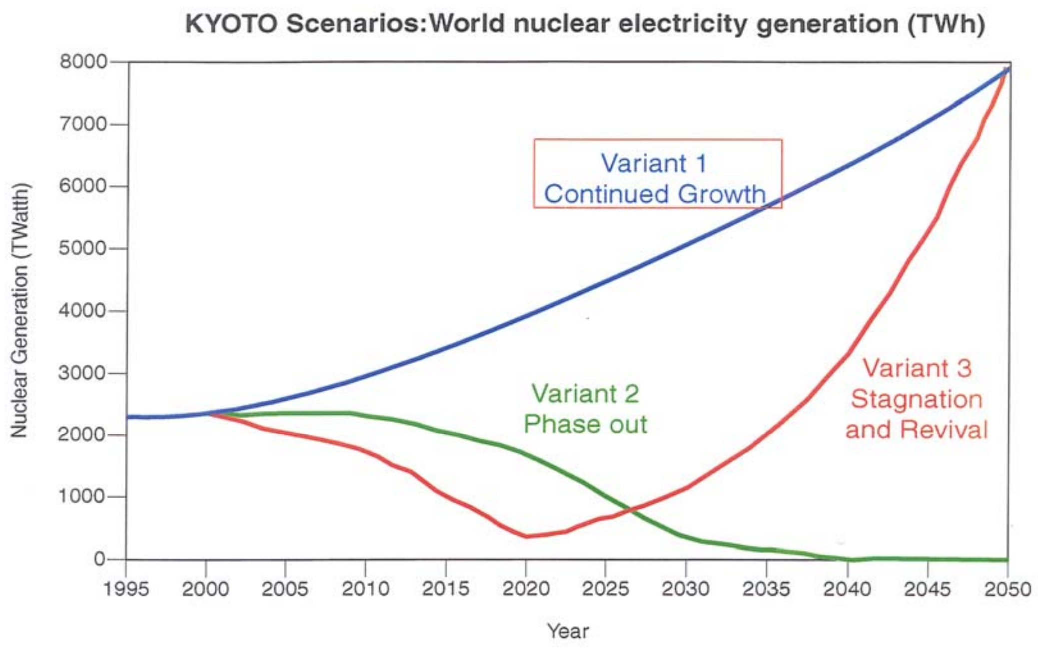
European Spallation Source

European Spallation Source – ESS, recently the ESS Scandinavian initiative

Principle Applications for Spallation Sources



Kärnkraften imorgon?



Om scenario 1 eller 3 gäller i framtiden då kommer vi att behöva i världen 1 Yucca Mountain-liknande förvar (70 000 t av HM) per 8 månader eller ett svenskt-liknande förvar varje månad fr.o.m. 2050!!
Och vilken slösseri av energirikt U-238!!

Obs! Kapacitet av existerande idag bearbetningsanläggningar är ung. 2000 t/år. Bättre teknik MÅSTE utvecklas!!

Reactor Physics – who we are:

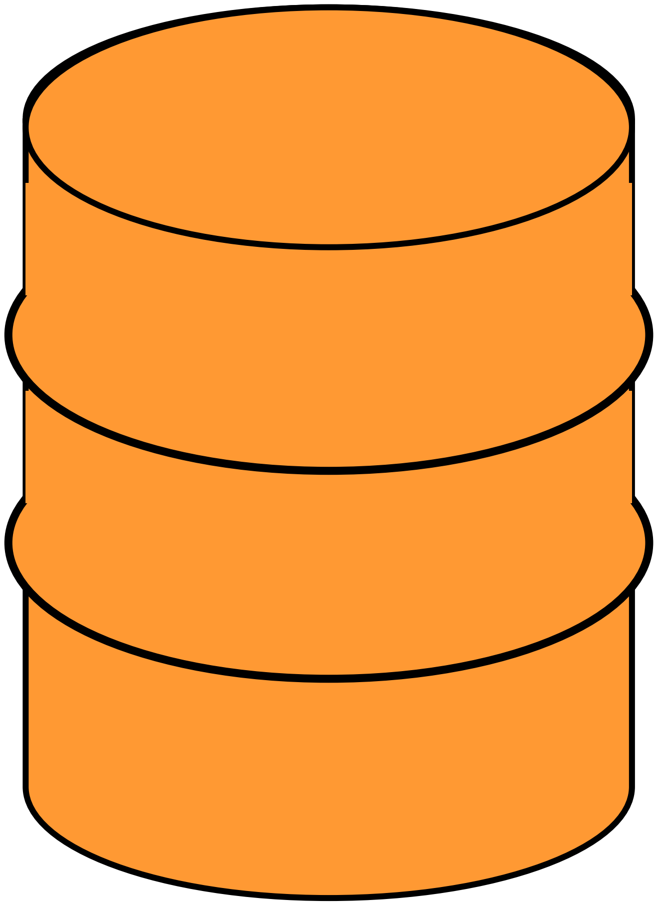
- W. Gudowski, prof.
- Scientists:
  - Janne Wallenius, doc. (nuclear physics)
  - Jerzy Cetnar, PhD (guest researcher ~2 year) (nuclear physicist, code developer) – just left
  - Vasily Arzhanov, post-doc, reactor physics, Monte Carlo
  - Mikael Jollkonnen, chemist
  - Torbjörn Bäck,deltid – reactor sumulator

- PhD students:
  - Johan Carlsson, fluid dynamics, safety of ADS – ready, left now
  - Kamil Tucek, ADS design, optimization of neutronics, lic.
  - Per Seltborg, subcritical studies, neutron source efficiency, experimental activities, lic.
  - Christina Lagerstedt, radiation damage
  - Marcus Ericsson, kinetic of ADS, safety
  - Alberto Talamo, Gas cooled reactors
  - Daniel Westlen, neutronics and economy of ADS
  - Jan Dufek, thermal hydraulics
  - Patrick Isaksson, neutron measurements,

- Students:
  - Jitka Zakova
  - Andre Grisell


STUDSVIK, March 2003

• Non Destructive Assay



Problem:
Nonintrusive determination transuranic alpha-emitters inside the waste drums.

- >7000 drums, 280 litre
- Drum wall: few mm steel and ~ 10 cm concrete
- Search for ^{239}Pu and ^{240}Pu, ^{241}Am, - milligrams!!
- All alpha emitters with different half-lives
- Limit of maximum permitted activities
- Conclusions:
  - Different mass limits for different isotopes

$$M \sim T_{1/2} * \text{Activity}$$

  - Neutrons are the best signals to determine the amount of actinides

- Other complementary measurements to be performed:
  - Verification of gamma activity
  - Drum ”tomography” with hard X-rays or gammas. Neutron-induced gamma-radiation can be easily utilised!
  - Neutron tomography