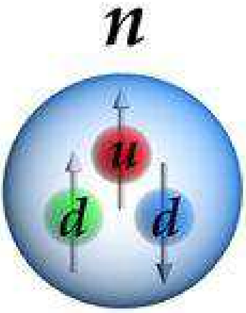
Neutron production

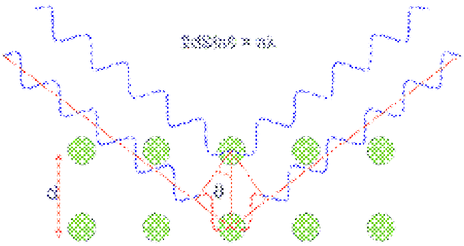
Ulli Köster, ILL

What is a neutron ?

1. a subatomic particle

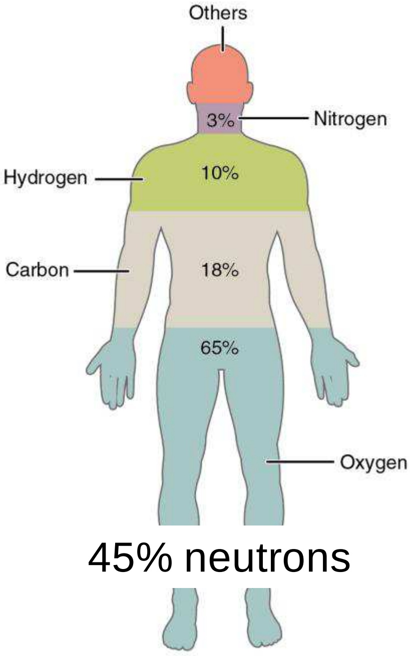


2. a matter wave

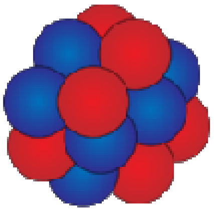


Neutrons are everywhere

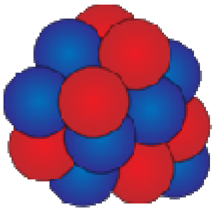
Bound neutrons are everywhere



45% neutrons

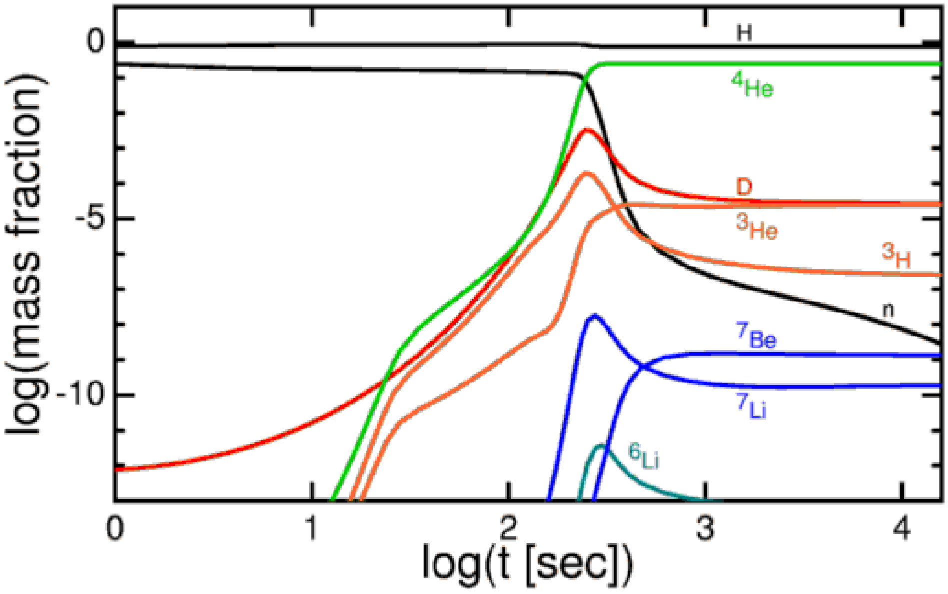


Carbon-12
98.9%
6 protons
6 **neutrons**



Carbon-13
1.1%
6 protons
7 **neutrons**

Big Bang Nucleosynthesis



Free neutrons have become rare

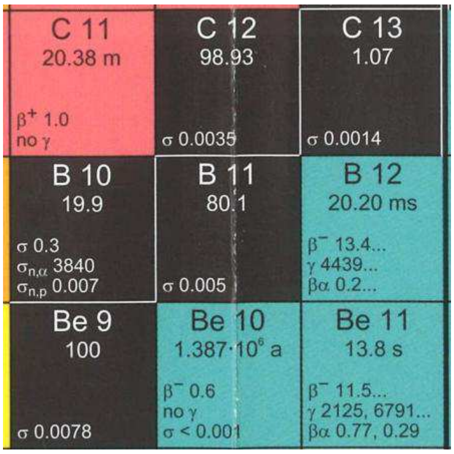
The Neutron’s Circle of Life

1. How neutrons are born
2. How neutrons are conformed to use
3. How neutrons die
4. What neutrons are good for

   (except neutron scattering and nuclear spectroscopy)

How neutrons are born

1. **Alpha-induced reactions:** ^{9}**Be(α,n)**^{12}**C** +5.7 MeV

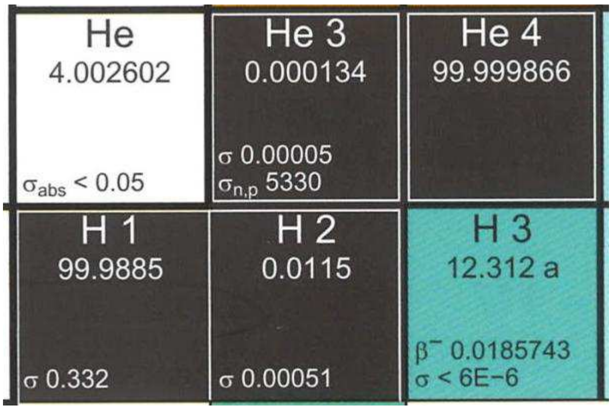


C 11 20.38 m β^+ 1.0 no γ	C 12 98.93 σ 0.0035	C 13 1.07 σ 0.0014
B 10 19.9 σ 0.3 $\sigma_{n,\alpha}$ 3840 $\sigma_{n,p}$ 0.007	B 11 80.1 σ 0.005	B 12 20.20 ms β^- 13.4... γ 4439... $\beta\alpha$ 0.2...
Be 9 100 σ 0.0078	Be 10 $1.387 \cdot 10^6$ a β^- 0.6 no γ $\sigma < 0.001$	Be 11 13.8 s β^- 11.5... γ 2125, 6791... $\beta\alpha$ 0.77, 0.29

How neutrons are born

1. **Alpha-induced reactions:** ^{9}**Be(α,n)**^{12}**C** +5.7 MeV

2. **Deuteron fusion: d(d,n)**^{3}**He** +3.3 MeV**, t(d,n)**^{4}**He** +17.6 MeV



He 4.002602 $\sigma_{abs} < 0.05$	He 3 0.000134 σ 0.00005 $\sigma_{n,p}$ 5330	He 4 99.999866
H 1 99.9885 σ 0.332	H 2 0.0115 σ 0.00051	H 3 12.312 a β^- 0.0185743 $\sigma < 6E-6$

How neutrons are born

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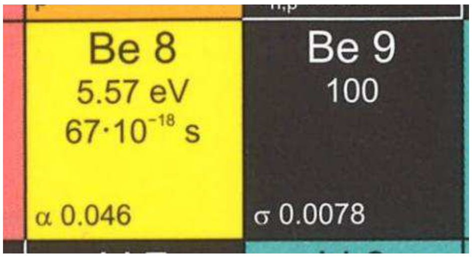
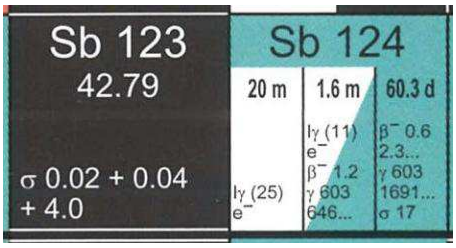
3. **Photo-dissociation:** **$^{9}Be(\gamma,n)2\alpha$** -1.66 MeV



Be 8	Be 9
5.57 eV $67 \cdot 10^{-18}$ s α 0.046	100 σ 0.0078



Sb 123	Sb 124		
42.79	20 m	1.6 m	60.3 d
σ 0.02 + 0.04 + 4.0	Iγ (25) e^-	Iγ (11) e^- β^- 1.2 γ 603 646...	β^- 0.6 2.3... γ 603 1691... σ 17

How neutrons are born

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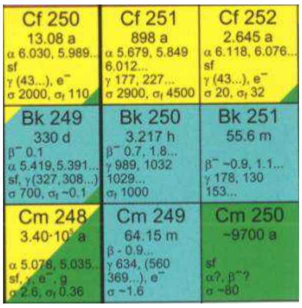
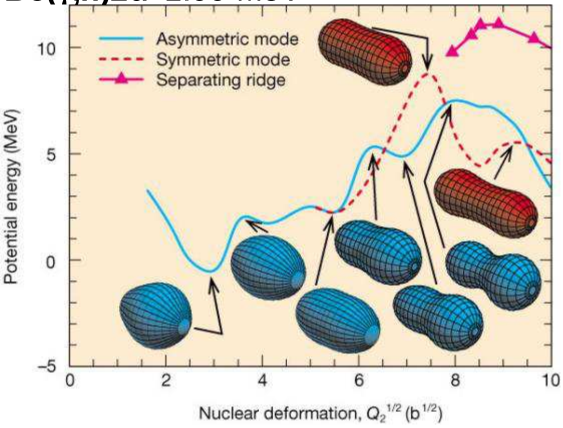
4. **Spontaneous fission:**



Cf 250	Cf 251	Cf 252
13.08 a α 6.030, 5.989... sf γ (43...), e^- σ 2000, σ_f 110	898 a α 5.679, 5.849 6.012... γ 177, 227... σ 2900, σ_f 4500	2.645 a α 6.118, 6.076... sf γ (43...), e^- σ 20, σ_f 32
Bk 249 330 d β^- 0.1 α 5.419, 5.391... sf, γ(327, 308...) σ 700, σ_f ~0.1	**Bk 250** 3.217 h β^- 0.7, 1.8... γ 989, 1032 1029... σ_f 1000	**Bk 251** 55.6 m β^- ~0.9, 1.1... γ 178, 130 153...
Cm 248 3.40·10 a α 5.078, 5.035... sf, γ, e^-, g σ 2.6, σ_f 0.36	**Cm 249** 64.15 m β - 0.9... γ 634, (560 369...), e^- σ ~1.6	**Cm 250** ~9700 a sf α?, β^-? σ ~80



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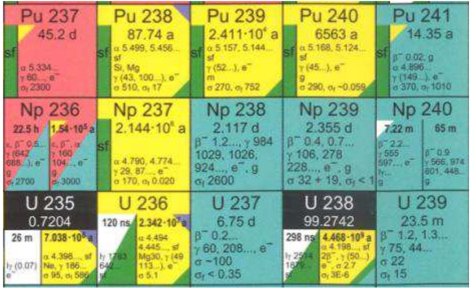
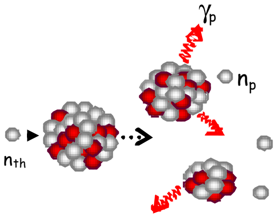


Pu 237 45.2 d sf α 5.334... γ 60..., e^- σ_f 2300	Pu 238 87.74 a sf α 5.499, 5.456... sf Si, Mg γ (43, 100...), e^- σ 510, σ_f 17	Pu 239 $2.411 \cdot 10^4$ a sf α 5.157, 5.144... sf γ (52...), e^- m σ 270, σ_f 752	Pu 240 6563 a sf α 5.168, 5.124... sf γ (45...), e^- g σ 290, σ_f ~0.059	Pu 241 14.35 a sf β^- 0.02, g α 4.896... γ (149...), e^- σ 370, σ_f 1010
Np 236 22.5 h $1.54 \cdot 10^5$ a σ_f 2700 σ_f 3000	Np 237 $2.144 \cdot 10^6$ a sf α 4.790, 4.774... γ 29, 87..., e^- σ 170, σ_f 0.020	Np 238 2.117 d β^- 1.2..., γ 984 1029, 1026, 924..., e^-, g σ_f 2600	Np 239 2.355 d β^- 0.4, 0.7... γ 106, 278 228..., e^-, g σ 32 + 19, σ_f < 1	Np 240 7.22 m 65 m β^- 2.2... γ 555 597..., e^- β^- 0.9 γ 566, 974 601, 448... g
U 235 0.7204 26 m $7.038 \cdot 10^8$ a α 4.398..., sf Ne, γ 186... σ 95, σ_f 586	U 236 120 ns $2.342 \cdot 10^7$ a α 4.494 4.445..., sf Mg30, γ (49 113...), e^- σ 5.1	U 237 6.75 d β^- 0.2... γ 60, 208..., e^- σ ~100 σ_f < 0.35	U 238 99.2742 298 ns $4.468 \cdot 10^9$ a α 4.198..., sf $2\beta^-$, γ (50...) e^-, σ 2.7 σ_f 3E-6	U 239 23.5 m β^- 1.2, 1.3... γ 75, 44... σ 22 σ_f 15



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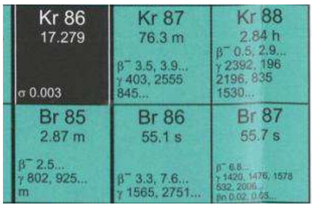
6. **Beta-delayed n emission:** **$^{87}Br(\beta^{-})^{87}Kr^{*} \rightarrow ^{86}Kr+n$** +1.3 MeV



Kr 86 17.279 σ 0.003	Kr 87 76.3 m β^- 3.5, 3.9... γ 403, 2555 845...	Kr 88 2.84 h β^- 0.5, 2.9... γ 2392, 196 2196, 835 1530...
Br 85 2.87 m β^- 2.5... γ 802, 925,... m	Br 86 55.1 s β^- 3.3, 7.6... γ 1565, 2751...	Br 87 55.7 s β^- 6.8... γ 1420, 1476, 1578 532, 2006... βn 0.02, 0.05...

How neutrons are born

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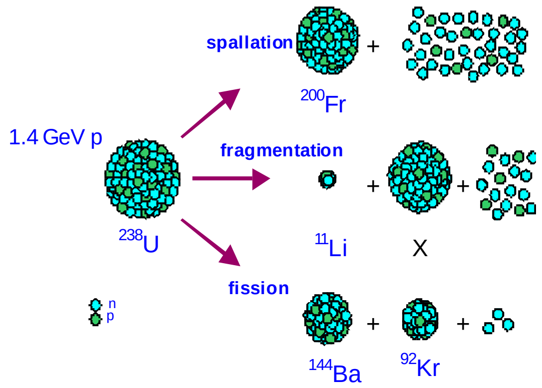
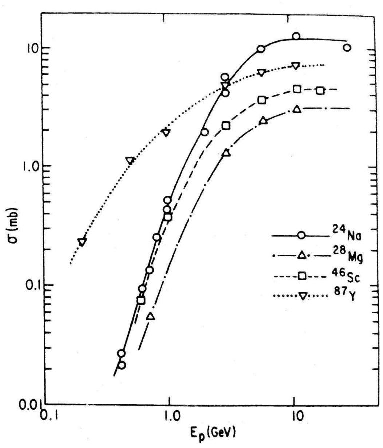
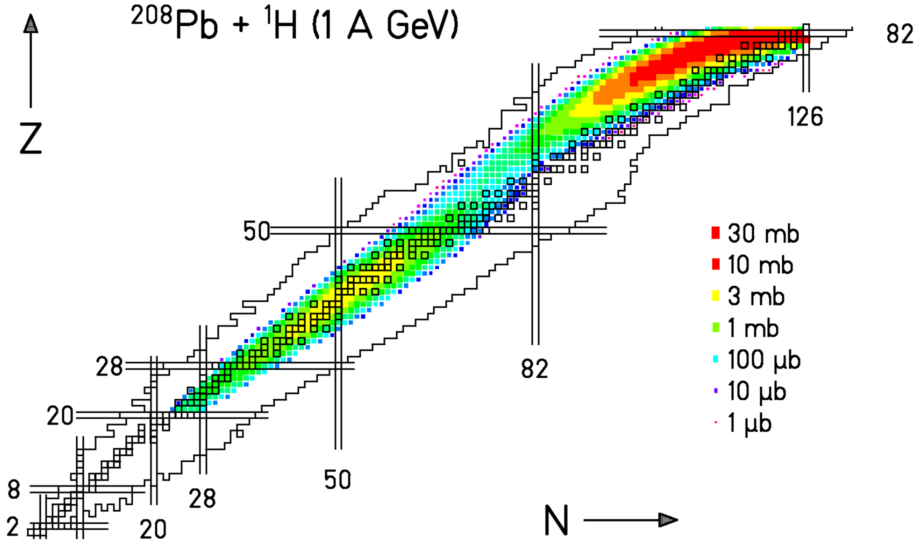
High energy nuclear reactions





Spallation + Fragmentation + Fission



W. Wlazło et al., Phys. Rev. Lett. 84 (2000) 5736.

T. Enqvist et al., Nucl. Phys. A 686 (2001) 481.

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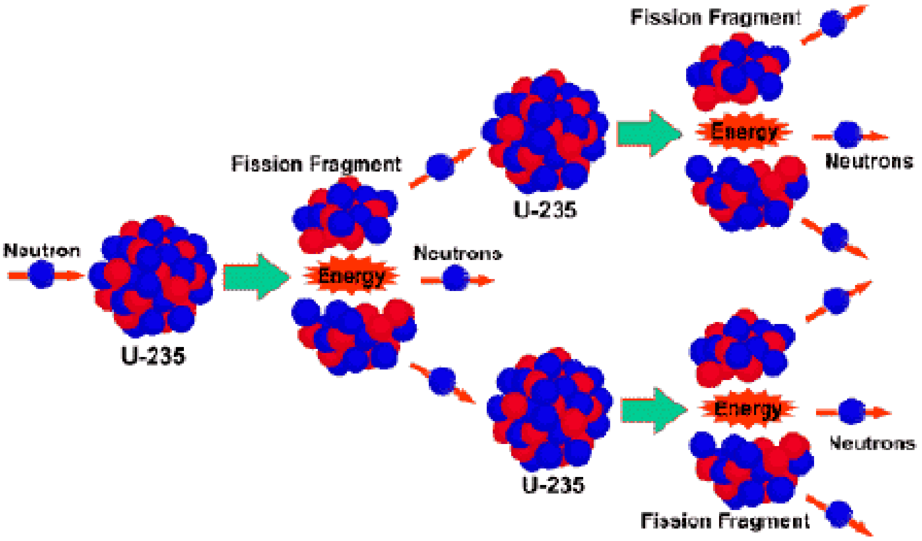
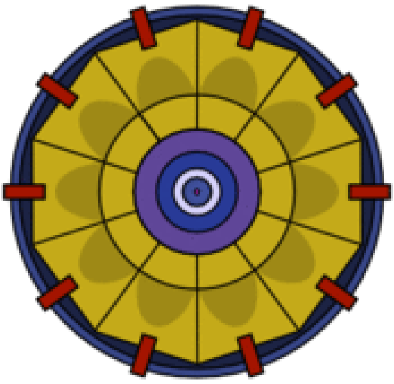
A nuclear chain reaction

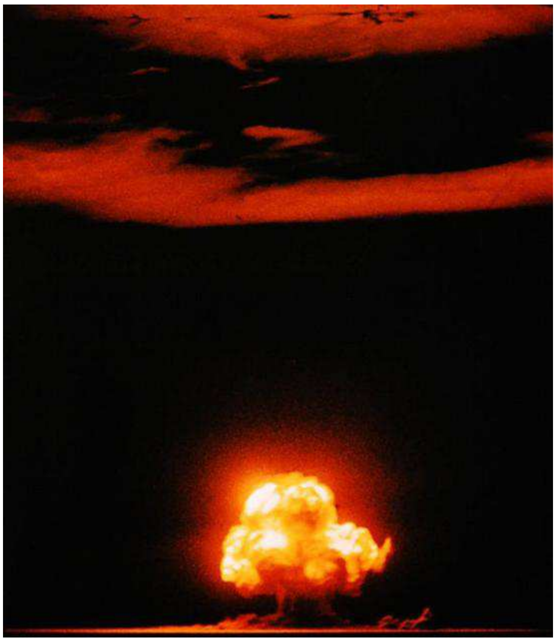


A single-pulse neutron source

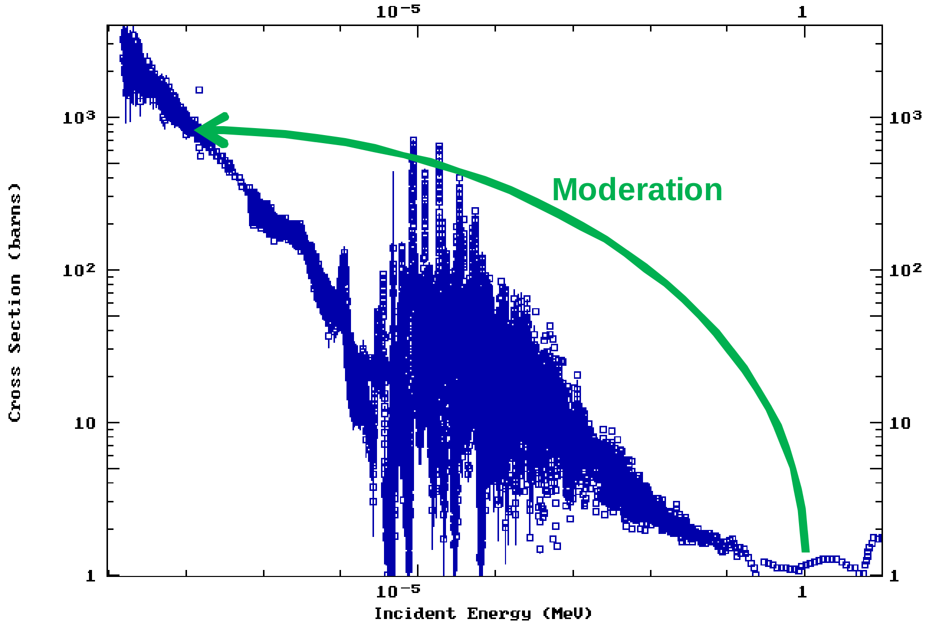


Uncontrolled
chain reaction
of fast-neutron
induced fission

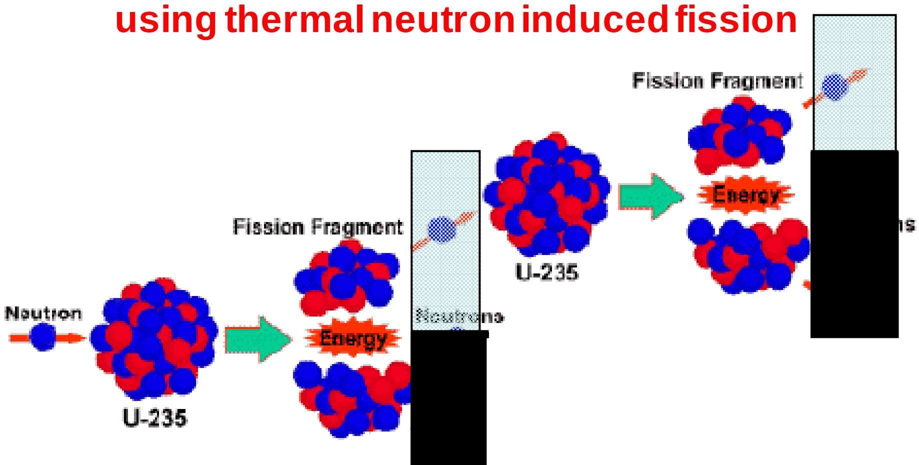
≈25 kg of 93% ^{235}U



^{235}U(n,f) cross-section as function of energy

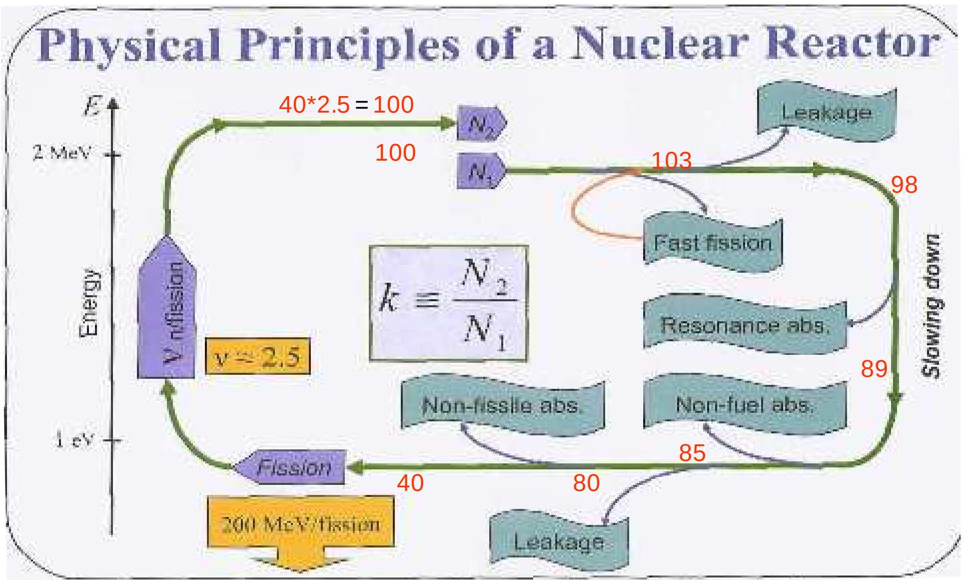


A controlled nuclear chain reaction using thermal neutron induced fission



1. Moderate neutrons
2. Control neutron losses

Physical Principles of a Nuclear Reactor



neutron numbers are given for a typical PWR reactor

≈0.6% of fission neutrons are beta-delayed by 12 s on average

⇒slows down reactor kinetics (Δk = 0.001) from ≈0.05 s to ≈80 s

⇒ essential for reliable control of reactor power

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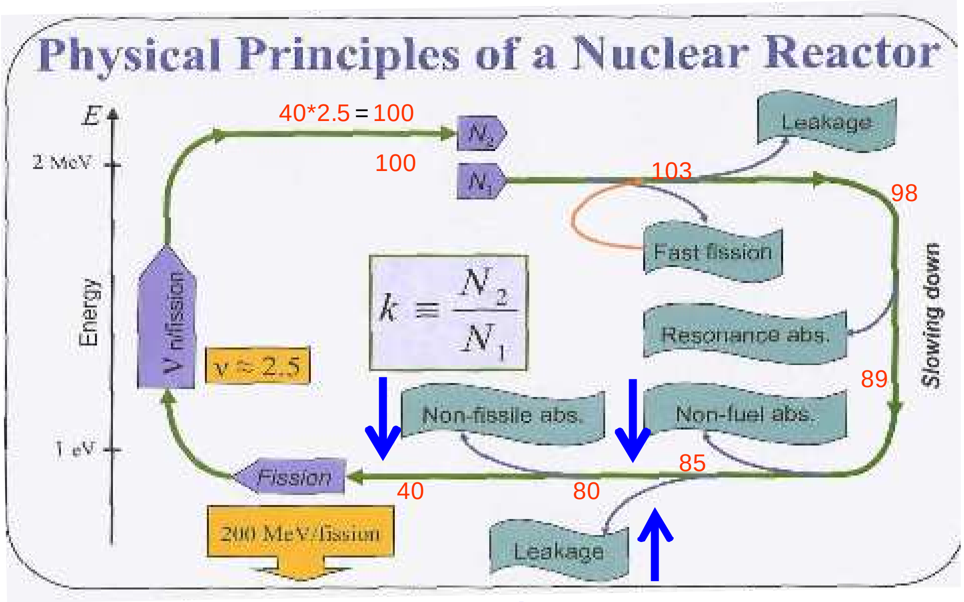


Research reactor

Components of a nuclear reactor

1. Fuel
2. Moderator
3. Control rods
4. Coolant
5. Pressure vessel
6. Containment
7. Steam generator (for power plants) or experimental facilities (for research reactors)

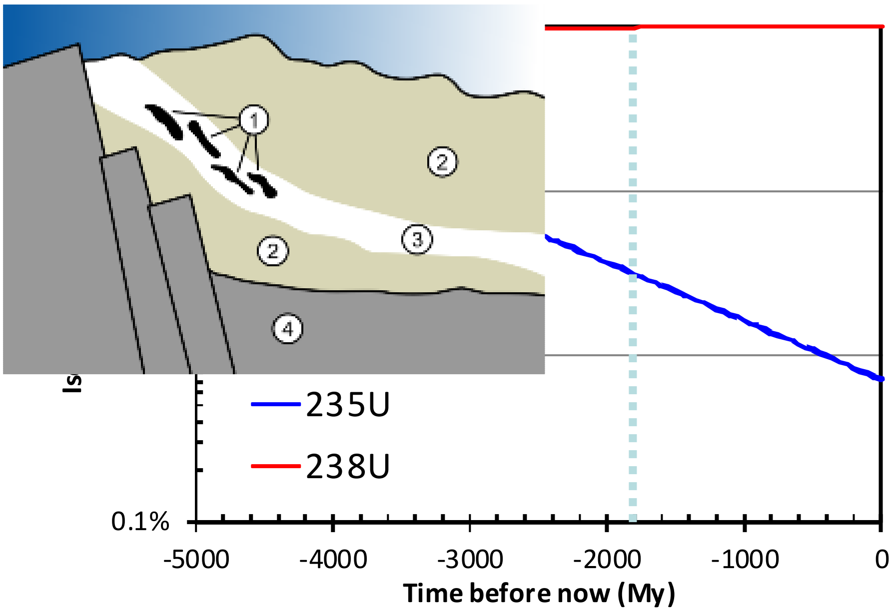
Moderator

elastic collisions with light atoms (mass A):
average energy loss $E_{n+1} - E_n = 2\ E_n\ A/(A+1)^2$

$\ln(E_n) – \ln(E_{n+1}) = \xi = 1 – (A-1)^2/(2A) * \ln[(A+1)/(A-1)]$

	Moderating power: $\xi\Sigma_{scatter}$	Moderating ratio: $\xi\Sigma_{scatter}/\Sigma_{abs.}$
Light water (H_2O)	1.28	58
Heavy water (D_2O)	0.18	21000
Beryllium (Be)	0.16	130
Graphite (C)	0.064	200
Polyethylene $(CH_2)_x$	3.26	122

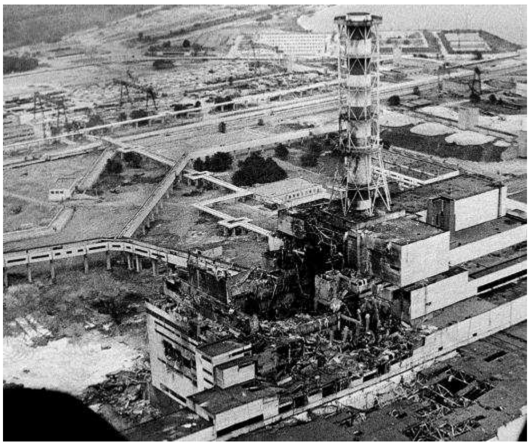
The first nuclear reactor on Earth



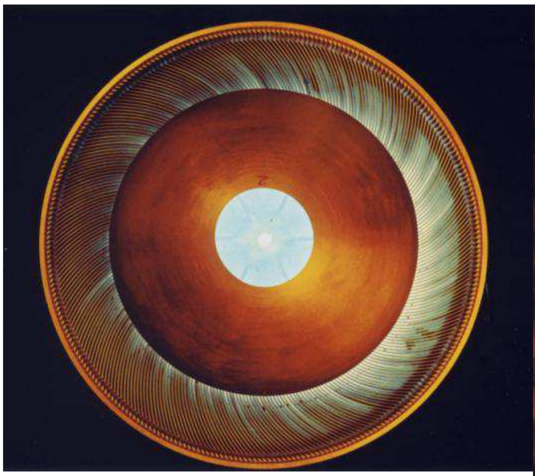
Choice of coolant

coolant = moderator
⇒ passive regulation
⇒ intrinsic safety

RBMK:
graphite moderator
water cooling
⇒ positive void coefficient !

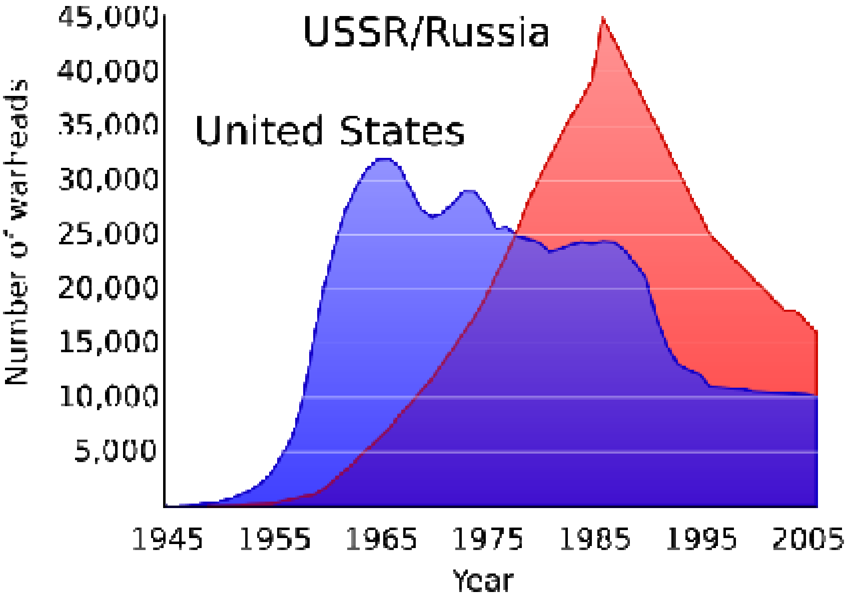


RHF fuel element



8.6 kg ^{235}U, 93% enriched

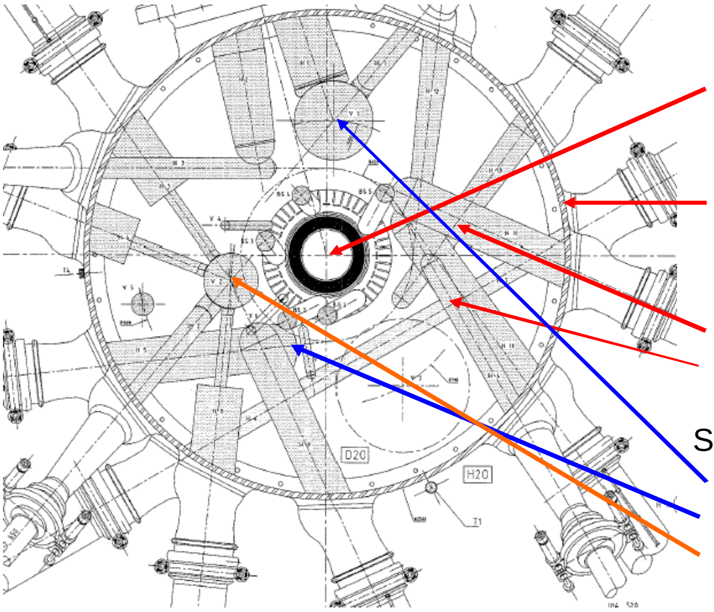
8 December 1987: Intermediate-Range Nuclear Forces Treaty



1 warhead = 25 kg HEU = 3 fuel elements for ILL

The ILL reactor contributes to permanent disarmament!

The reactor core and vessel

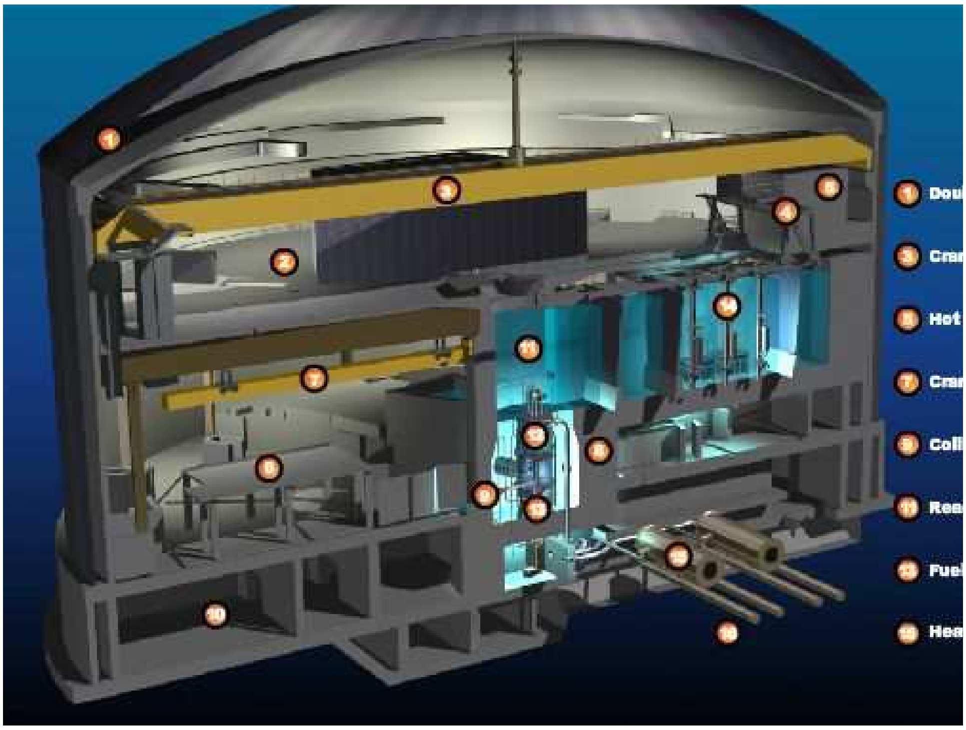


Fuel element :
-R_i = 14 cm
-R_e = 19 cm

Vessel:
- R = 125 cm

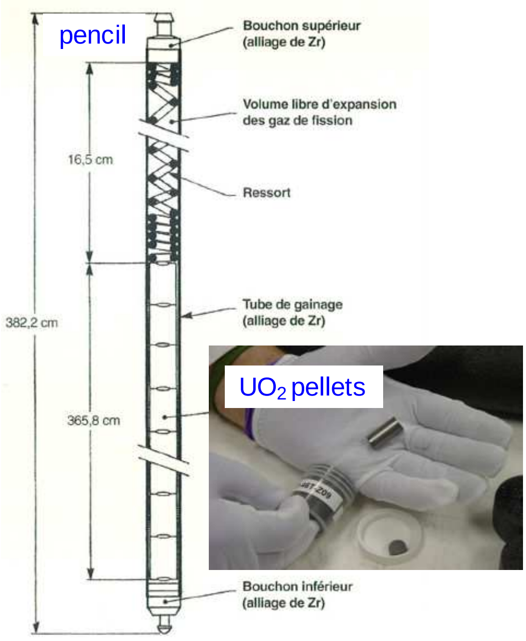
Beam tubes :
-13 Horizontal
-4 inclined

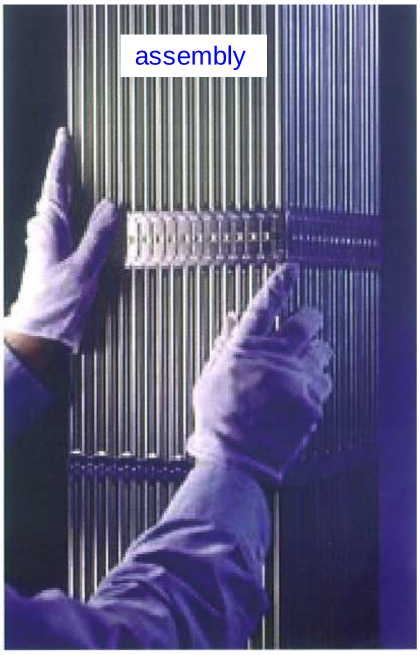
Sources
-VCS
-HCS
-HS



Some comments on recent events…

Reactor fuel elements = 1^{st} barrier

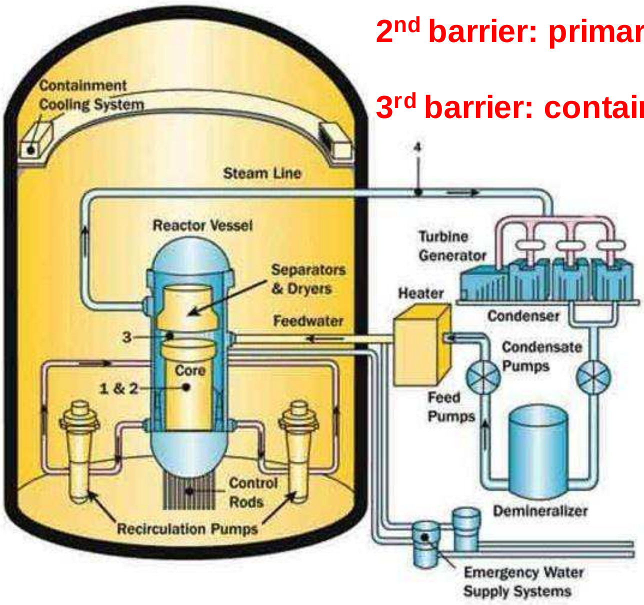




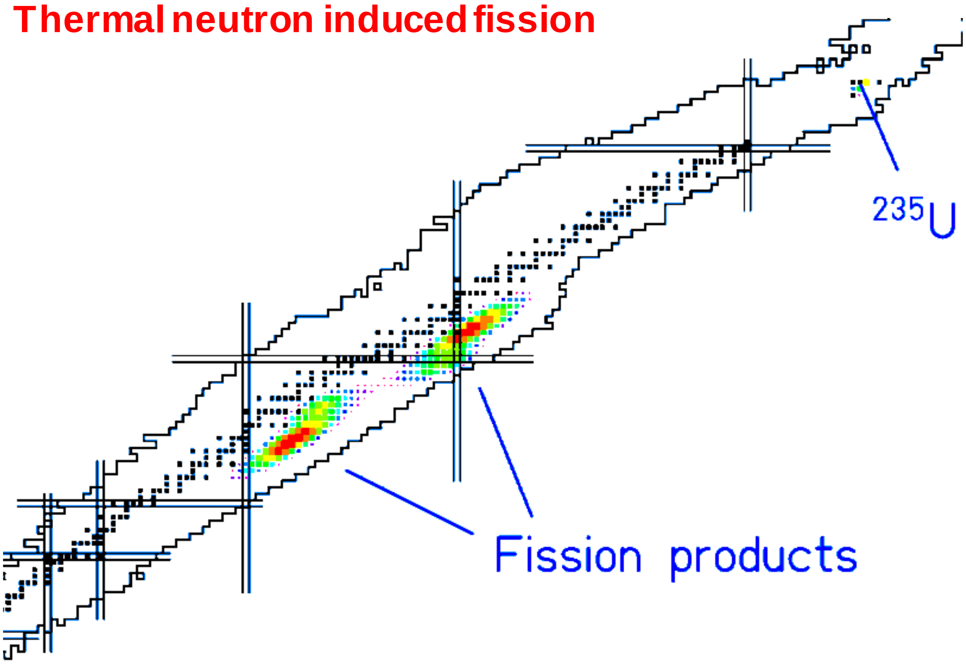
Typical boiling-water reactor

2nd barrier: primary cooling circuit

3rd barrier: containment

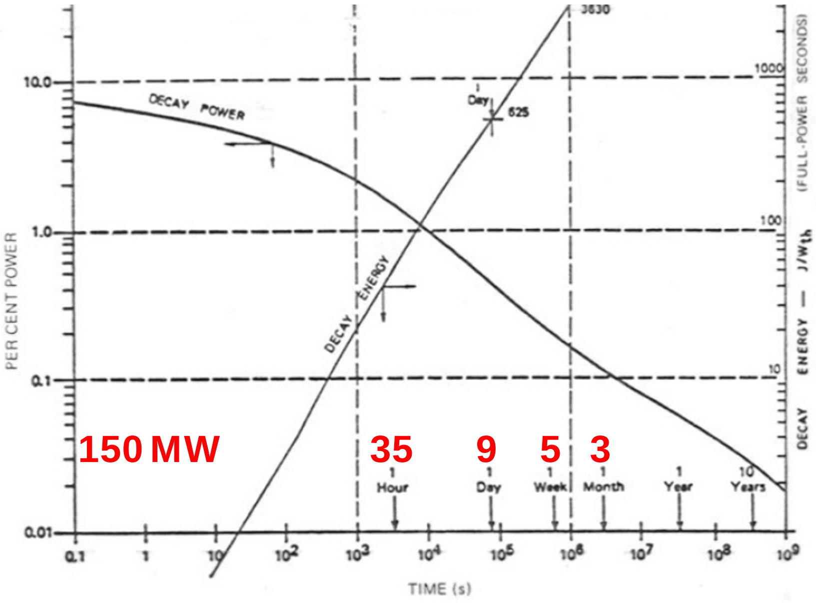


Thermal neutron induced fission



Nuclear decay heat

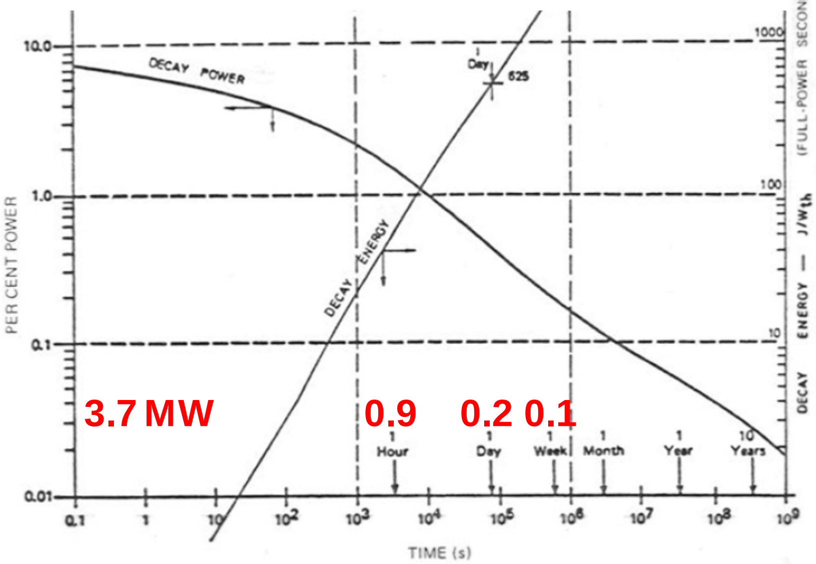
Fukushima 2 and 3: 784 MWe, 2300 MWth



150 MW 35 9 5 3

Nuclear decay heat

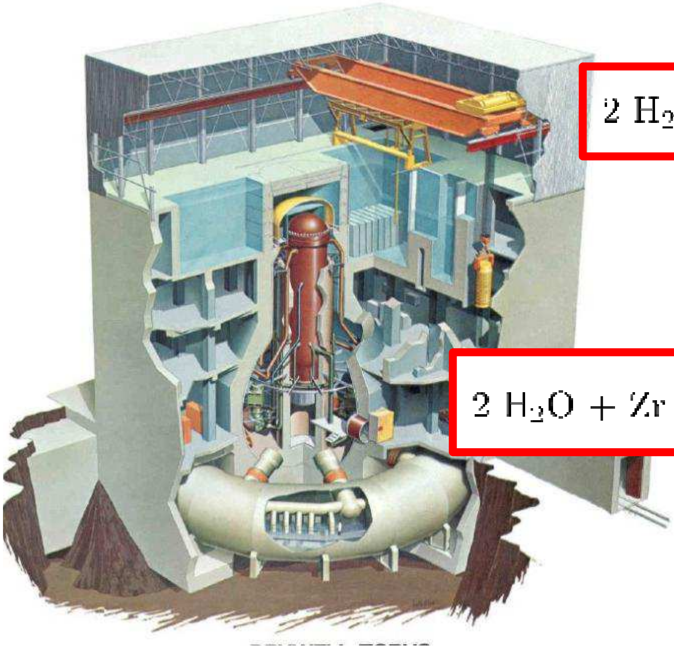
ILL: 57 MWth



3.7 MW 0.9 0.2 0.1

Decay heat can be passively cooled by natural convection!

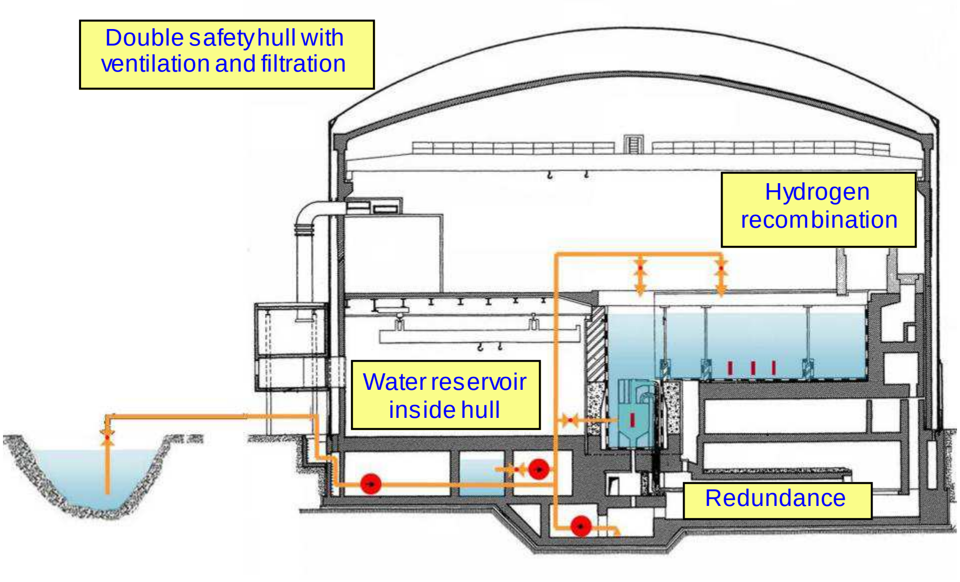
Secondary reactions



$$2\ H_2 + O_2 \rightarrow 2\ H_2O$$

$$2\ H_2O + Zr \rightarrow ZrO_2 + 2\ H_2$$

Safety features of the ILL reactor



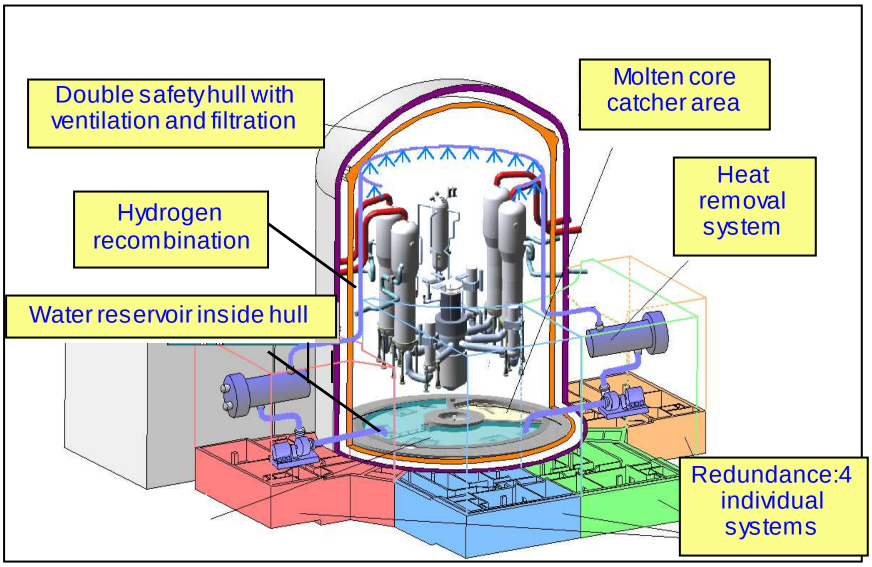
Double safety hull with ventilation and filtration

Hydrogen recombination

Water reservoir inside hull

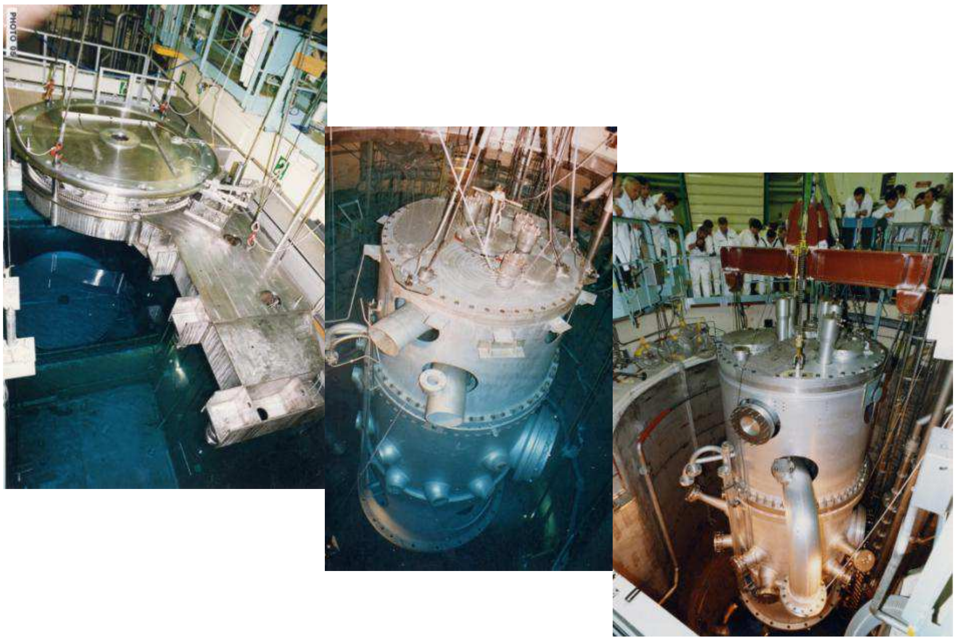
Redundance

Safety features of Generation 3+ reactors (EPR)

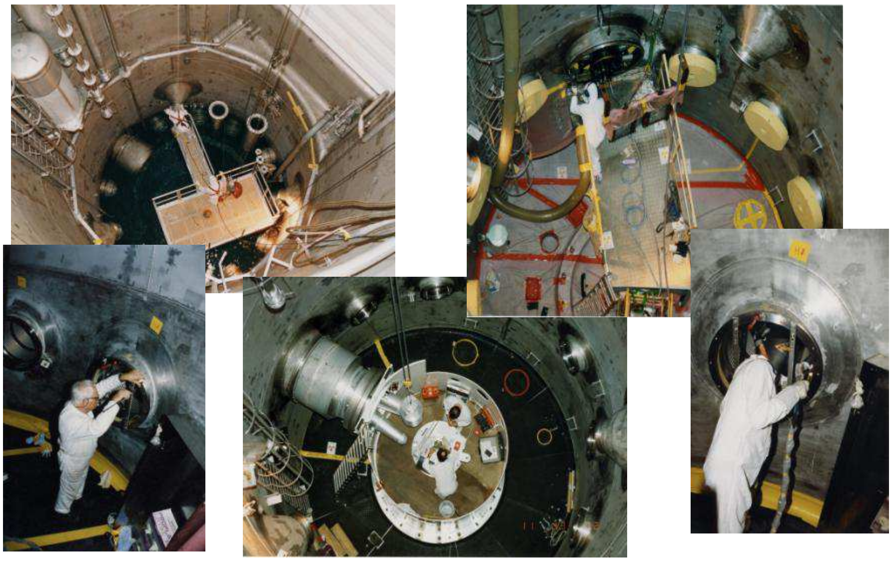


Power reactor ⇔	**Research reactor**
• **heat used to produce electricity**	• **neutrons used for applications**
• **neutrons just to maintain chain reaction**	• **heat not used**
• **needs high power, high temperature and high pressure for good thermal efficiency**	• **operates at lower power, low temperature (ILL 30-48°C) and low pressure (<14 bar)**
• **BWR: 75 bar, 285°C**	
• **PWR: 155 bar, 315°C**	
• **25 cm thick steel pressure vessel ⇒ defines lifetime (40..60 y)**	• **vessel and all inserts made from pure Al-alloy** • **modular and exchangeable ⇒ no finite lifetime**

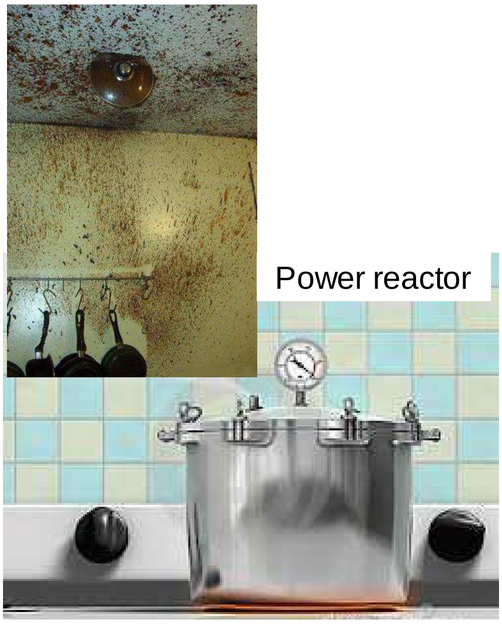
ILL: Replacement of the reactor vessel 1990-94



ILL: Replacement of the reactor vessel 1990-94



The risk profile of power versus research reactors



Spallation neutron source versus reactor

Advantage for reactor
- higher time-averaged flux (for high-flux reactor)
- flux very constant over time
- larger irradiation volume possible (multiple fuel elements)
- much lower electricity bill

Advantage for spallation neutron source
- pulsed operation much easier
- much higher peak flux for TOF applications

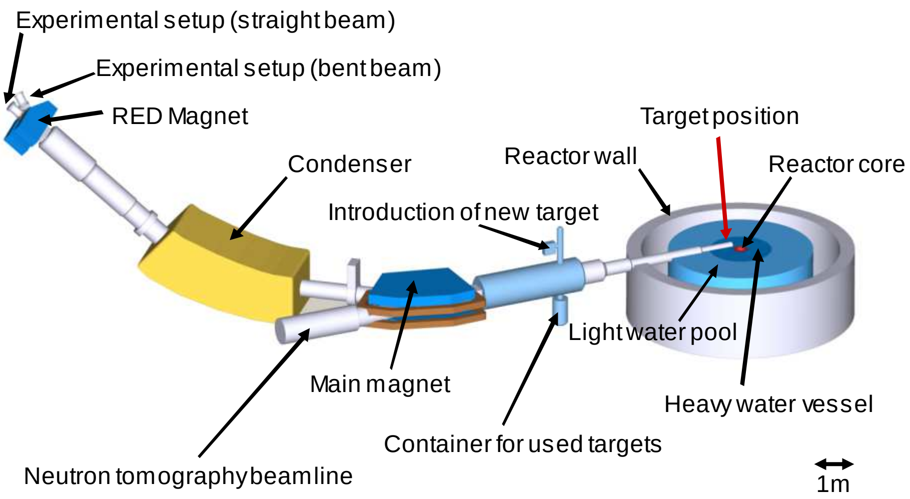
- does not carry “reactor” in its name

Urban legend:
“Reactors are dirty, spallation neutron sources are clean.”

Back to physics:

the neutron as a tool to study fission

LOHENGRIN Setup



Experimental setup (straight beam)

Experimental setup (bent beam)

RED Magnet

Condenser

Introduction of new target

Reactor wall

Target position

Reactor core

Light water pool

Main magnet

Heavy water vessel

Container for used targets

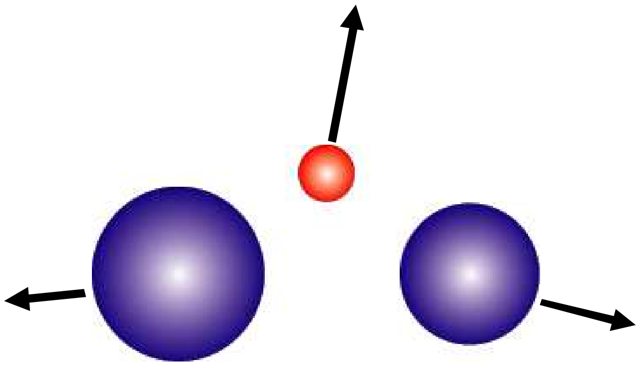
Neutron tomography beamline

1m

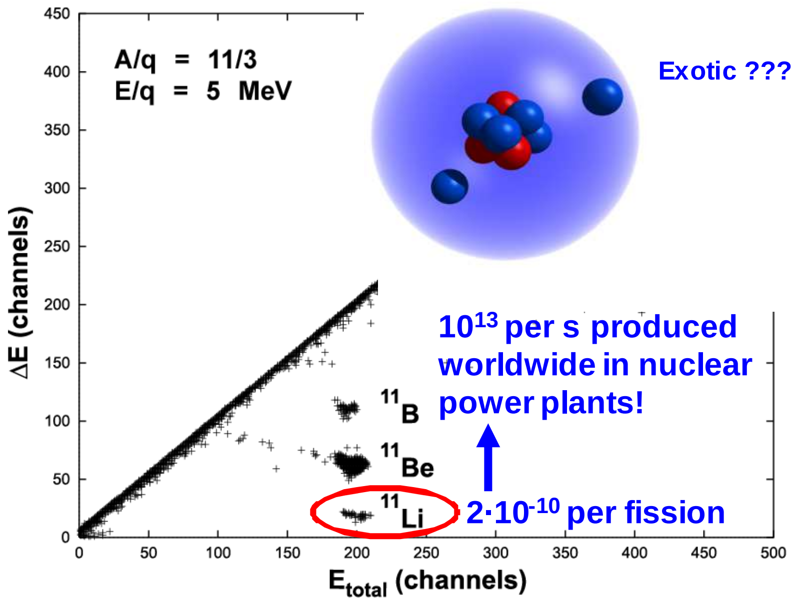
^{11}Li production in thermal neutron induced fission?

	^{235}U	+	n	⇒	^{11}Li	+	^{140}Xe	+	^{85}Br
A	235	+	1	=	11	+	140	+	85
Z	92	+	0	=	3	+	54	+	35
ΔM (MeV)	40.919		8.071		40.801		-72.990		-78.610

Q (MeV) $= (40.919 + 8.071) - (40.801 - 72.990 - 78.610) = +159.8$



Detection of rare ternary particles



Exotic ???

10^{13} per s produced worldwide in nuclear power plants!

$2 \cdot 10^{-10}$ per fission

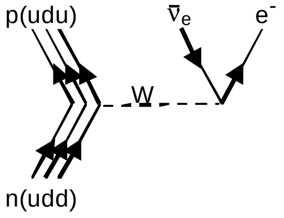
Neutron (particle) physics:

the neutron as an object

Neutron Decay

- **clean semi-leptonic decay**

$$n \longrightarrow p + e^{-} + \overline{\nu}_e$$



- **clear theoretical understanding:**

only **3 free parameters** **in Standard Model**

$$\tau^{-1} \propto \left|V_{ud}\right|^2 \left(1+3\lambda^2\right)$$

- **ratio** of coupling constants
- and its **phase**

$$\lambda = \frac{g_A}{g_V}$$

- **Quark mixing** from the Cabibbo-Kobayashi-Maskawa matrix

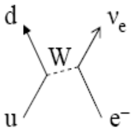
$$V_{ud}$$

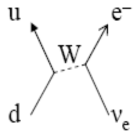
Ideal system for **precision measurements**

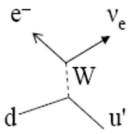
Why is neutron-decay important?

neutron-decay provides key input for many disciplines:
astrophysics, cosmology and particle physics

Primordial element formation	$n + e+ \rightarrow p + \nu'_e$	$\sigma_\nu \sim 1/\tau$
(^{2}H, ^{3}He, ^{4}He, ^{7}Li, …)	$p + e^- \rightarrow n + \nu_e$	$\sigma_\nu \sim 1/\tau$
	$n \rightarrow p + e^- + \nu'_e$	τ
Solar cycle	$p + p \rightarrow {}^2H + e^+ + \nu_e$	
	$p + p + e^- \rightarrow {}^2H + \nu_e$ etc.	$\sim (g_A/g_V)^5$
Neutron star formation	$p + e^- \rightarrow n + \nu_e$	
Pion decay	$\pi^- \rightarrow \pi^0 + e^- + \nu'_e$	
Neutrino detectors	$\nu'_e + p \rightarrow e^+ + n$	
Neutrino forward scattering	$\nu_e + n \rightarrow e^- + p$ etc.	
W and Z production	$u' + d \rightarrow W^- \rightarrow e^- + \nu'_e$ etc.	



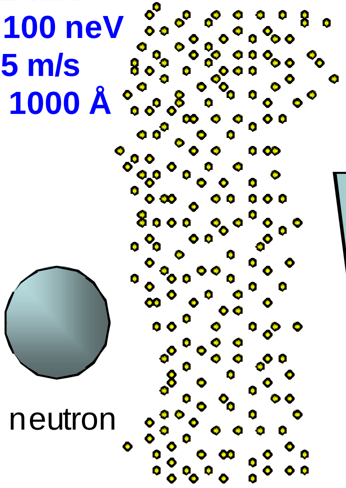




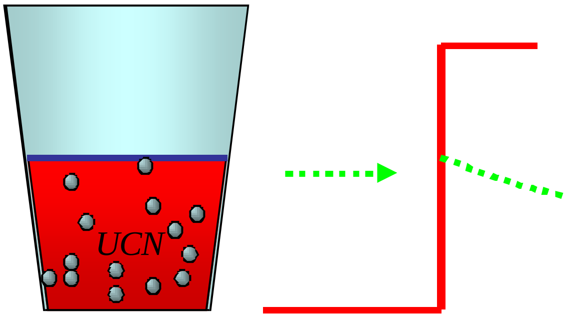
From cold to ultracold neutrons

Ultracold neutrons

$T \approx 2$ mK
$E \approx 100$ neV
$v \approx 5$ m/s
$\lambda \approx 1000$ Å

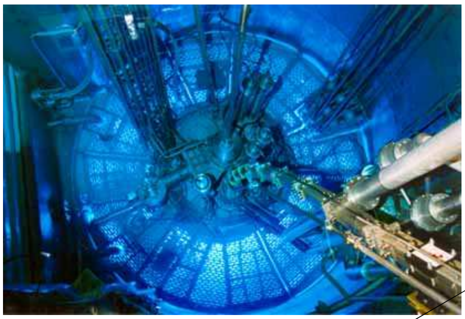


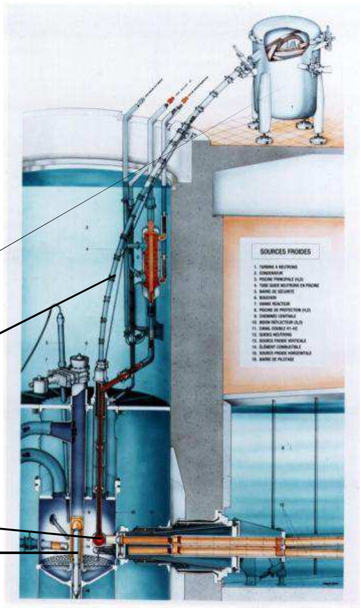
Typically:
99.99 % - elastic reflection
0.009% - inelastic scattering
to the thermal energy range
0.001% - absorption



Total reflection under any angle on suitable materials like Be, Ni, C ⇒ possibility to store for long time

The UCN facility PF2 at ILL





Neutron turbine
A. Steyerl (TUM - 1985)

Vertical guide tube

Cold source

Reactor core

Measurements of the neutron lifetime τ_n

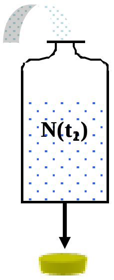
exponential decay law: $N = N_0 e^{-\lambda t}$

or, ultimately, measure the exponential decay directly

Storage experiments with UCN

"counting the surviving neutrons"

"UCN bottle"



$$\frac{1}{\tau_m} = \frac{1}{t_2 - t_1} \cdot \ln\frac{N(t_1)}{N(t_2)}$$

$$\frac{1}{\tau_m} = \frac{1}{\tau_\beta} + \underbrace{\frac{1}{\tau_{\text{wall}}}} + \underbrace{\frac{1}{\tau_{\text{leak}}} + \frac{1}{\tau_{\text{vacuum}}} + \ldots}_{\to 0 \text{ (experiment)}}$$

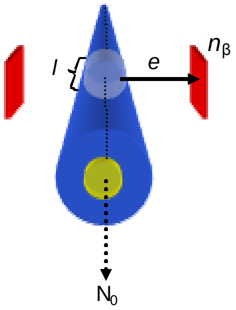
$$\frac{1}{\tau_{\text{wall}}} = \mu \cdot v_{\text{eff}} \to 0 \text{ (extrapolation)}$$

$$\Rightarrow \quad \frac{1}{\tau_m} = \frac{1}{\tau_\beta}$$

Two relative measurements

Beam experiments with cold neutrons

"counting the dead neutrons"



$$n_\beta = \frac{dN}{dt} = -\frac{N_0}{\tau_n} e^{-\frac{l}{v \cdot \tau_n}}$$

Two absolute measurements

A "typical" UCN storage experiment at ILL – MamBo I

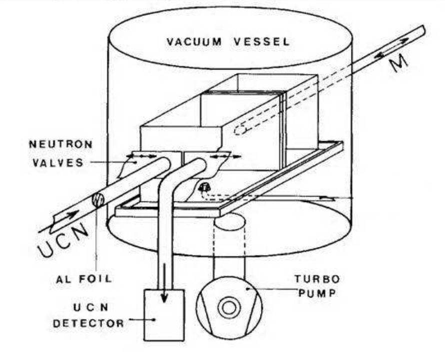


FIG. 1. Sketch of the apparatus.

Glass walls:
H=0.3 m, W=0.4 m
L=0.5m … 0.01 m
(surface *A* and volume *V* sizeable)

$$\frac{1}{\tau_m} = \frac{1}{\tau_\beta} + \frac{1}{\tau_{\text{wall}}} + \ldots$$

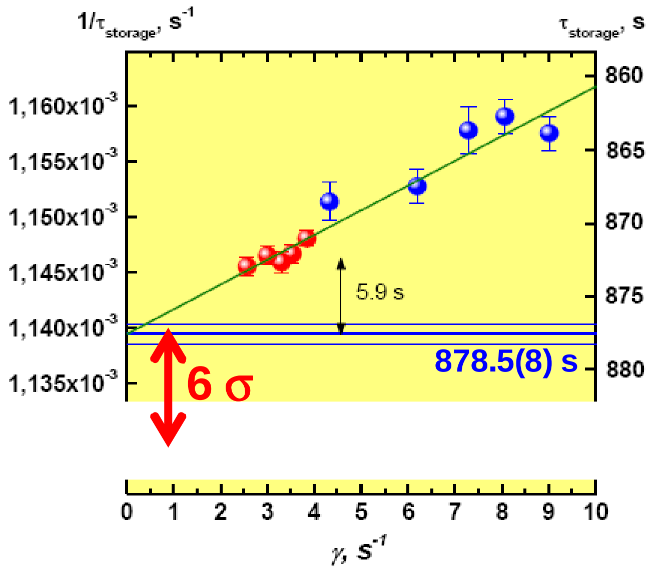
$\tau_{\text{wall}} \to$ number of wall collisions, i.e. mean free path λ

Measure storage lifetime τ_{st} for different volume to surface ratios *V/A* and extrapolate for $V \to \infty$

$$\frac{1}{\tau_{\text{wall}}} \to 0$$

“GRAVITRAP” Neutron Lifetime Experiment at the PF2/MAM beam position in the ILL

Extrapolation to n-lifetime

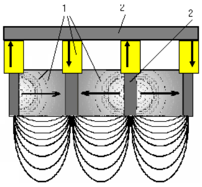


Magnetic confinement

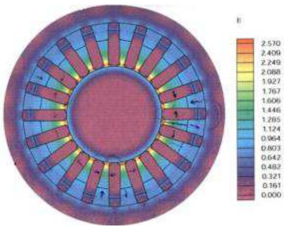
- For μ_n = -60.3 neV/T, a 2T field generates a 120 neV barrier.
- Force due to field gradient, $F = -\mu\,(dB/dz)$, repels only one spin state.
- Use permanent magnets.

- **Step 1: 1D confinement**

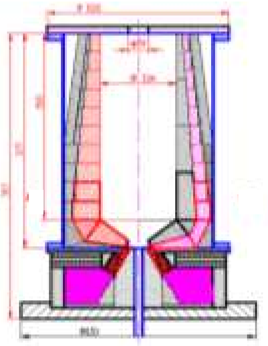
  1 – permanent magnets
  2 – magnetic poles



- **Step 2: 2D confinement**

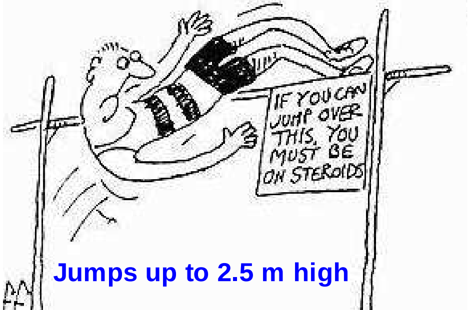


- **Step 3: 3D confinement**
  - top (gravity)
  - bottom (magnetic shutter)



UCN: Particle physics with "human dimensions"

Speed: some m/s

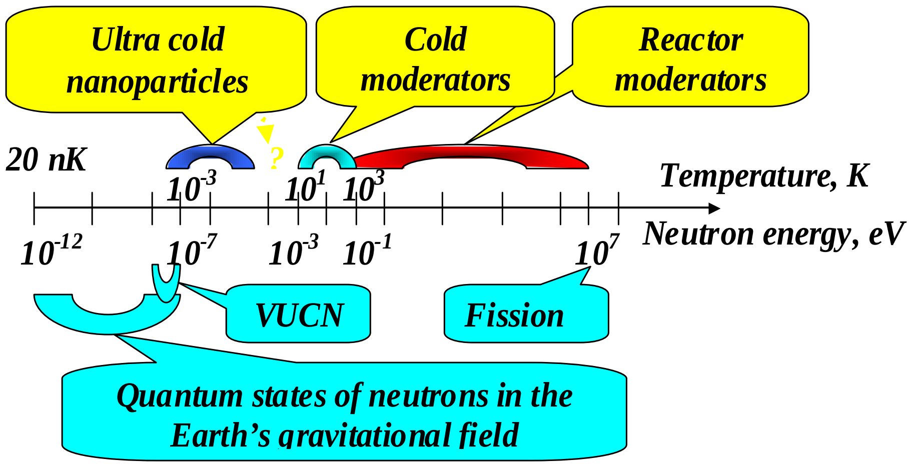


Jumps up to 2.5 m high

Magnetic field:
1 T may repel
UCN 0.6 m high

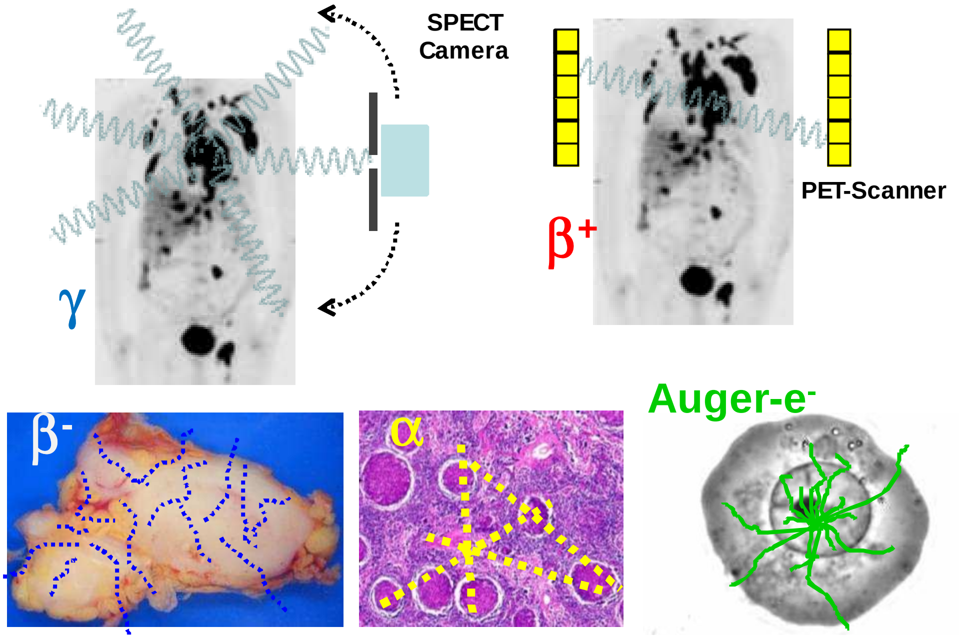
≈10 min half-life
≈15 min lifetime

Scales of temperature and energy in neutron physics



Neutrons as a tool for medicine

The Nuclear Medicine Alphabet



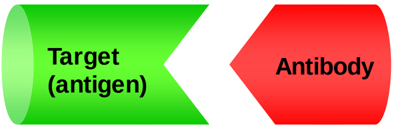
Cancer and efficiency of treatments

At time of diagnosis	Primary tumor	With metastases	Total
Diagnosed	**58%**	**42%**	**100%**
Cured by:			
Surgery	**22%**		
Radiation therapy	**12%**		
Surgery+radiation therapy	**6%**		
All other treatments and combinations incl. chemotherapy		**5%**	
Fraction cured	**69%**	**12%**	**45%**

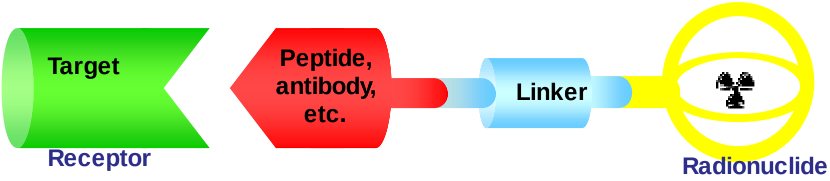
Over one million deaths per year from cancer in EU.

⇒ improve early diagnosis
⇒ improve systemic treatments

Immunology approach



Multidisciplinary collaboration to fight cancer



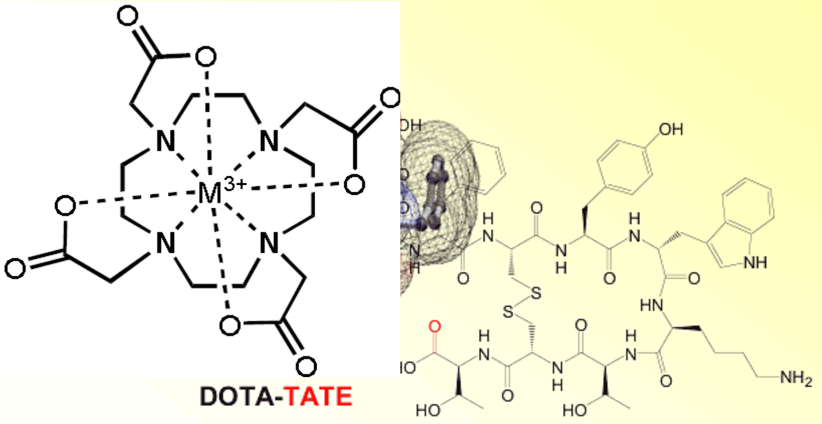
Immunology
Structural biology

Coordination
chemistry

Nuclear physics
and
radiochemistry

Nuclear medicine and medical physics

Structural Formula of DOTA-TOC/TATE



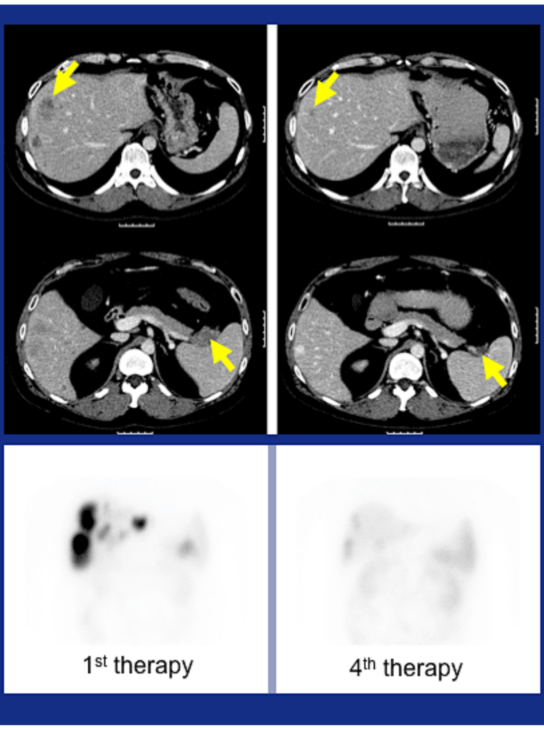
1,4,7,10-tetraazacyclododecantetraacetate

^{111}In ^{90}Y
^{67}Ga ^{177}Lu
^{68}Ga ^{213}Bi

IC$_{50}$ (Y^{III}) = 1.6 ± 0.4 nM

Helmut Maecke, EANM-2007.

Universitätsspital Basel



Male

36 years of age

Small cell pancreatic neuroendocrine tumour

Liver metastases

Ki-67 index 10-15% (liver biopsy)

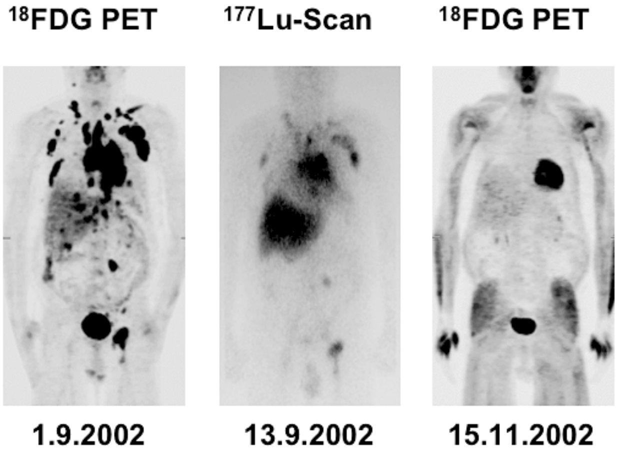
4 cycles with ^{177}Lu-octreotate and capecitabine

Partial remission

Roelf Valkema, EANM-2008.

Lymphoma therapy: RITUXIMAB+^{177}Lu

E.B., 1941 (m): UPN 6



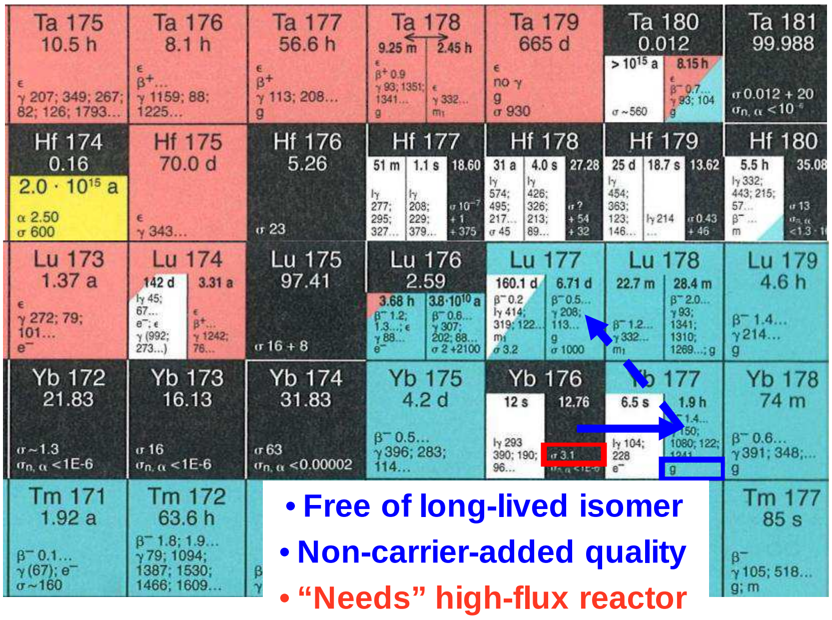
Still in CR

15.9.2009

F. Forrer et al., J Nucl Med 2013;54:1045.

University Hospital Basel, CH

Alternative production route to ^{177}Lu



Ta 175 10.5 h ε γ 207; 349; 267; 82; 126; 1793…	Ta 176 8.1 h ε β+… γ 1159; 88; 1225…	Ta 177 56.6 h ε β+ γ 113; 208… g	Ta 178 9.25 m ↔ 2.45 h ε; β+ 0.9; γ 93; 1351; 1341…; g ε; γ 332…; m₁	Ta 179 665 d ε no γ g σ 930	Ta 180 0.012 > 10^{15} a; σ ~560 8.15 h; ε; β⁻ 0.7…; γ 93; 104; g	Ta 181 99.988 σ 0.012 + 20 σn, α <10^{-6}
Hf 174 0.16 2.0 · 10^{15} a α 2.50 σ 600	Hf 175 70.0 d ε γ 343…	Hf 176 5.26 σ 23	Hf 177 51 m; Iγ 277; 295; 327… 1.1 s; Iγ 208; 229; 379… 18.60; σ 10^{-7} + 1 + 375	Hf 178 31 a; Iγ 574; 495; 217…; σ 45 4.0 s; Iγ 426; 326; 213; 89… 27.28; σ ? + 54 + 32	Hf 179 25 d; Iγ 454; 363; 123; 146… 18.7 s; Iγ 214 … 13.62; σ 0.43 + 46	Hf 180 5.5 h; Iγ 332; 443; 215; 57…; β⁻ …; m 35.08; σ 13; σn, α <1.3 · 1
Lu 173 1.37 a ε γ 272; 79; 101… e⁻	Lu 174 142 d; Iγ 45; 67…; e⁻; ε; γ (992; 273…) 3.31 a; ε; β+…; γ 1242; 76…	Lu 175 97.41 σ 16 + 8	Lu 176 2.59 3.68 h; β⁻ 1.2; 1.3…; ε; γ 88…; e⁻ 3.8·10^{10} a; β⁻ 0.6…; γ 307; 202; 88…; σ 2 +2100	Lu 177 160.1 d; β⁻ 0.2; Iγ 414; 319; 122…; m₁; σ 3.2 6.71 d; β⁻ 0.5…; γ 208; 113…; g; σ 1000	Lu 178 22.7 m; β⁻ 1.2…; γ 332…; m₁ 28.4 m; β⁻ 2.0…; γ 93; 1341; 1310; 1269…; g	Lu 179 4.6 h β⁻ 1.4… γ 214… g
Yb 172 21.83 σ ~1.3 σn, α <1E-6	Yb 173 16.13 σ 16 σn, α <1E-6	Yb 174 31.83 σ 63 σn, α <0.00002	Yb 175 4.2 d β⁻ 0.5… γ 396; 283; 114…	Yb 176 12 s; Iγ 293; 390; 190; 96… 12.76; σ 3.1; σn, α <1E-6	Yb 177 6.5 s; Iγ 104; 228; e⁻ 1.9 h; β⁻ 1.4…; γ 150; 1080; 122; 1241…; g	Yb 178 74 m β⁻ 0.6… γ 391; 348;… g
Tm 171 1.92 a β⁻ 0.1… γ (67); e⁻ σ ~160	Tm 172 63.6 h β⁻ 1.8; 1.9… γ 79; 1094; 1387; 1530; 1466; 1609…	β γ				Tm 177 85 s β⁻ γ 105; 518… g; m

- **Free of long-lived isomer**
- **Non-carrier-added quality**
- **“Needs” high-flux reactor**


The rising star
for therapy

Acknowledgements

Thanks for transparencies from:

Hartmut Abele
Roger Brissot
Bruno Desbriere
Peter Geltenbort
Bastian Maerkisch
Valery Nesvishevsky
Anatoli Serebrov
Oliver Zimmer