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OF TECHNOLOGY

Nuclear Fuel Cycle 2011

Lecture 10: The History of Nuclear Power

Oklo: The first nuclear reactor 1.7×10^9 years ago

It was found that the ^{235}U abundance was 0.44%.
Only possible reason: A natural nuclear reactor

$$N = N_0 e^{-\lambda t}$$

$$N_{\text{U-235}} = 0.720\%, \quad t_{1/2, \text{U-235}} = 7.04 \times 10^8 \text{y}$$

$$N_{\text{U-235}} = 99.2745\%, \quad t_{1/2, \text{U-235}} = 4.47 \times 10^9 \text{y}$$

=> U-235 abundance 1.7×10^9 y ago was 2.9%

Sufficient to permit fission to occur, providing
other conditions are right



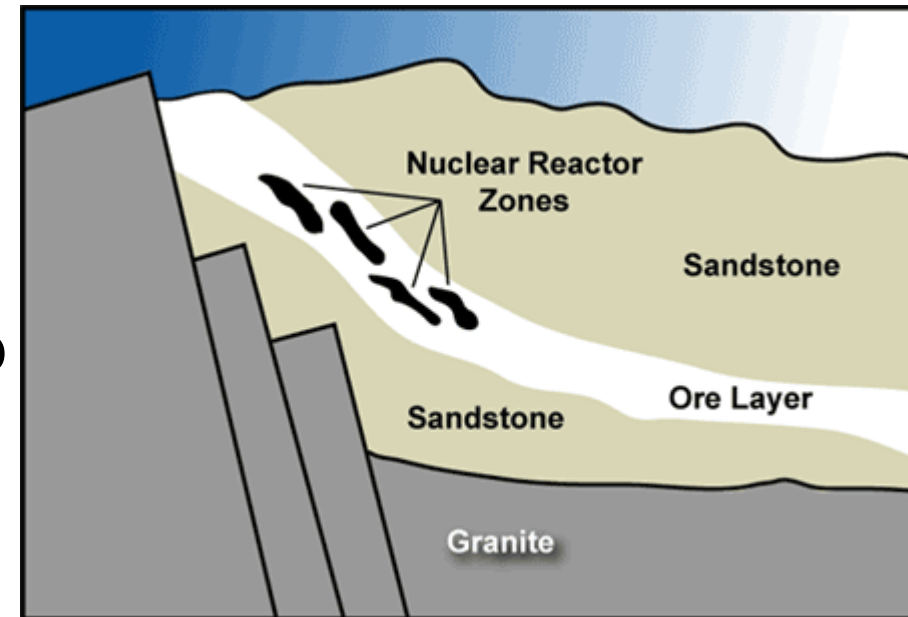
Oklo geology

U was accumulated at a redox-front. The redox-front did not arise until the O_2 -concentration was sufficiently high (c:a 1.7 billion y ago)

Water flowing through the sandstone worked as moderator.

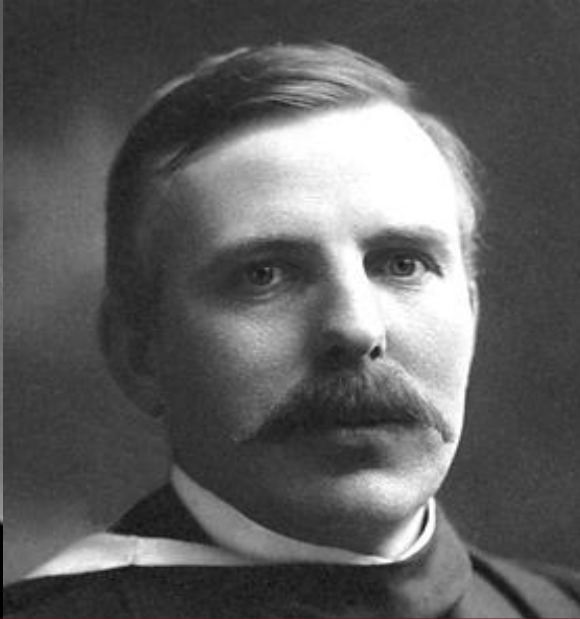
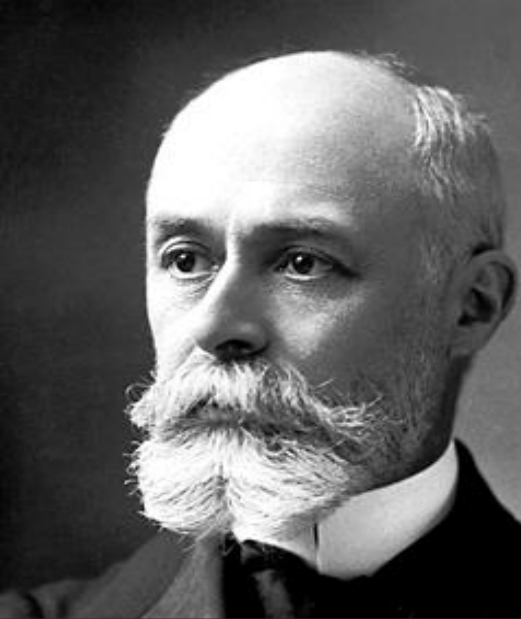
Reactor shut down now and then due to boiling of moderator or neutron poisons

Reactor operated off and on for about 1 000 000 years



Oklo is the best natural analogue for a deep repository of spent nuclear fuel.

Pu has moved less than 10 ft from where it was produced 2 billion years ago



Fundamental nuclear chemistry –
understanding the nature of the atom



Mendeleev's periodic table 1869

Reihen	Gruppe I. — R'O	Gruppe II. — RO	Gruppe III. — R'O ³	Gruppe IV. RH ⁴ RO ²	Gruppe V. RH ³ R'O ³	Gruppe VI. RH ² RO ³	Gruppe VII. RH R'O ²	Gruppe VIII. — RO ⁴
1	II=1							
2	Li=7	Be=9,4	B=11	C=12	N=14	O=16	F=19	
3	Na=23	Mg=24	Al=27,8	Si=28	P=31	S=32	Cl=35,5	
4	K=39	Ca=40	—=44	Ti=48	V=51	Cr=52	Mn=55	Fe=56, Co=59, Ni=59, Cu=63.
5	(Cu=63)	Zn=65	—=68	—=72	As=75	Se=78	Br=80	
6	Rb=86	Sr=87	?Yt=88	Zr=90	Nb=94	Mo=96	—=100	Ru=104, Rh=104, Pd=106, Ag=108.
7	(Ag=108)	Cd=112	In=113	Sn=118	Sb=122	Tc=125	J=127	
8	Cs=133	Ba=137	?Di=138	?Co=140	—	—	—	— — — —
9	(—)	—	—	—	—	—	—	
10	—	—	?Er=178	?La=180	Ta=182	W=184	—	Os=195, Ir=197, Pt=198, Au=199.
11	(Au=199)	Hg=200	Tl=204	Pb=207	Bi=208	—	—	
12	—	—	—	Th=231	—	U=240	—	— — — —

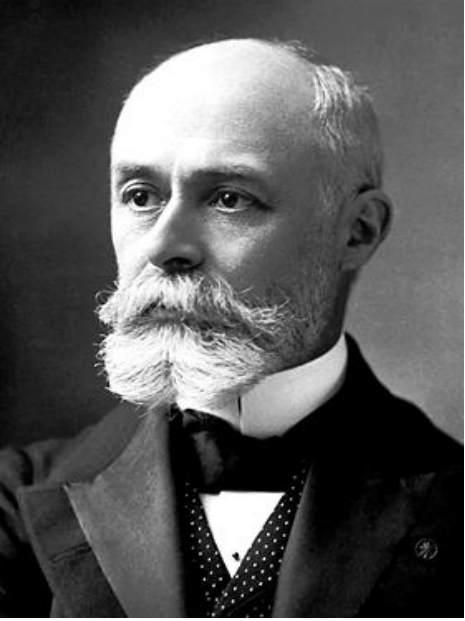


Wilhelm Conrad Röntgen

The Nobel Prize in Physics 1901

"In recognition of the extraordinary services he has rendered by the discovery of the remarkable rays subsequently named after him".

Ionizing radiation was discovered by Röntgen. An electric current passed an evacuated glass tube and X-rays was produced



Henri Becquerel

Becquerel discovered that pitchblende made photographic plates to darken. From the start Becquerel believed that this had something to do with sunlight, but experiments in the dark gave the same results.

β -radiation

α -radiation

Becquerel shared Nobel prize with Pierre and Marie Curie 1903 for his discovery of radiation.

Radioactive decay



Discovery of Po and Ra



Determination of atomic weight of Ra

22 avril

$$\begin{aligned}
 \text{Cl} + \text{CaCl} &= 14,448,05 \\
 \text{id} + \text{RaCl} &= 14,857,3 \quad \text{RaCl} = 0,109,25 \\
 \text{Creuset vide} &= 10,314,65 \\
 \text{Cl} + \text{AgCl} &= 10,421,12 \quad \text{AgCl} = 0,10647
 \end{aligned}$$

$ \begin{array}{r} 7.39270 \\ 7.02723 \\ \hline 2.41993 \end{array} $	$ \begin{array}{r} \text{Cl} = 0.10925 \\ \text{Ra} = 0.08295 \\ \hline 0.02630 \end{array} $	$ \begin{array}{r} 0.0694 \\ 0.2723 \\ \hline 0.2723 \end{array} $
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$ \begin{array}{r} 2.91882 \\ 2.41993 \\ \hline 0.49889 \end{array} $	$ \begin{array}{r} 1.85003 \\ 0.49889 \\ \hline 2.34892 \end{array} $
--	--

$\text{Ra} = \underline{223.3}$

AgCl réduit avec Zn et HCl, lavé
le creuset, pesé

$$\begin{aligned}
 \text{Creuset} + \text{argent} &= 10.3942 \\
 &= 10.31465 \\
 \text{Ag} &= 0.07955 \\
 \text{Cl} &= 0.02630 \\
 \hline
 \text{d'où} \quad \text{AgCl} &= 0.10585
 \end{aligned}$$

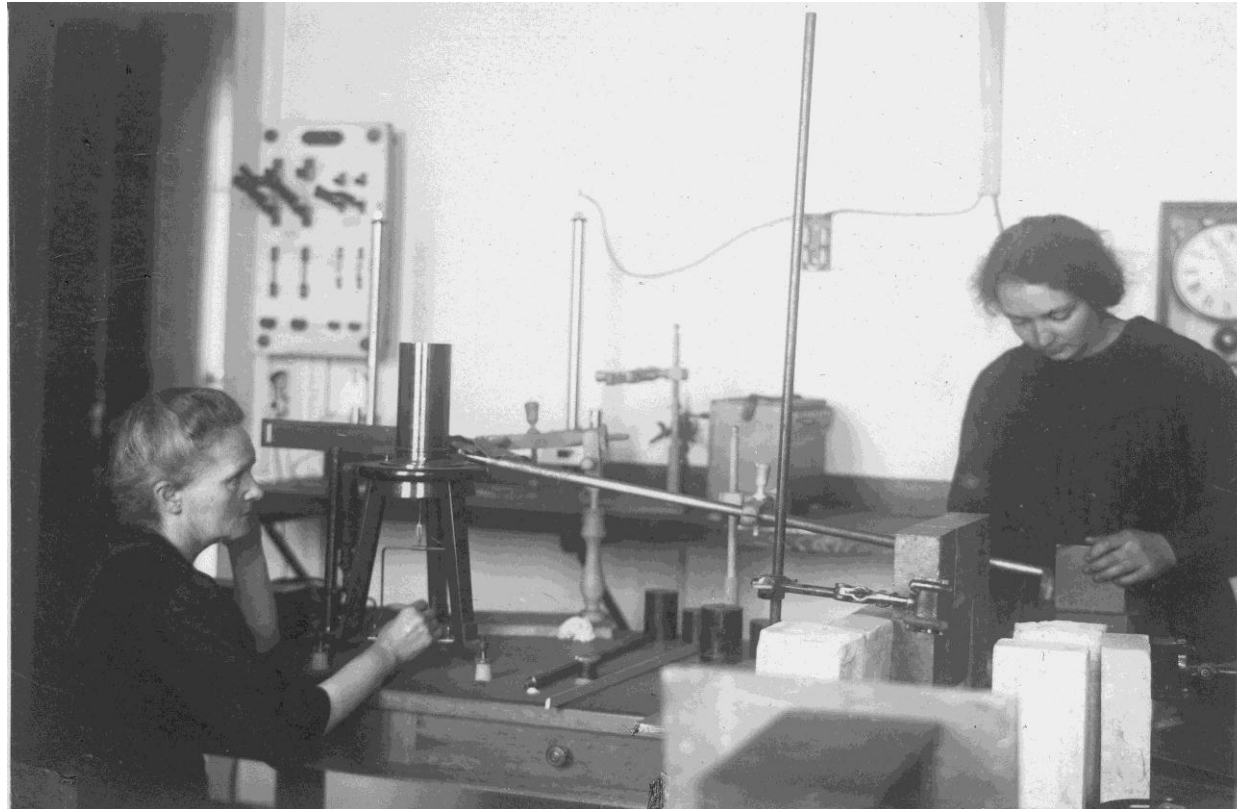
Différence avec AgCl trouvé précédemment
0.00083 m

y a-t-il eu un peu de Ag non adhérent au creuset ?



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Marie and Irène



Applications of Curies research

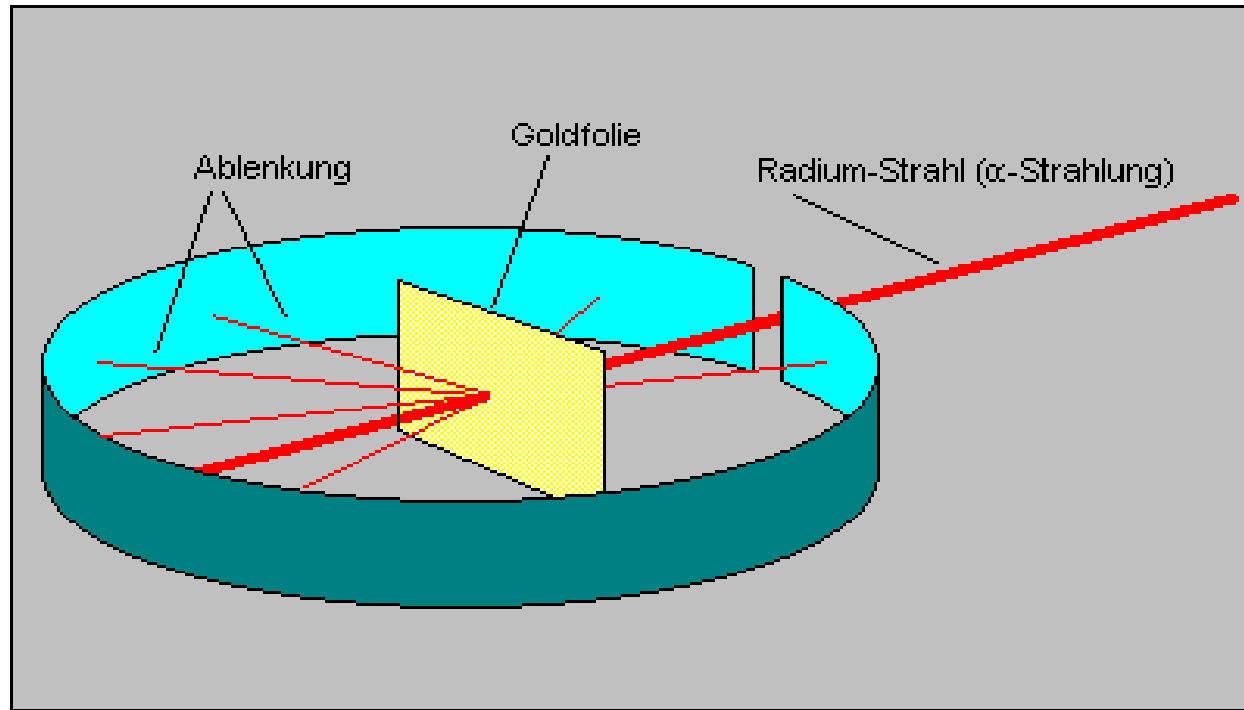
Radiumhemmet 1910



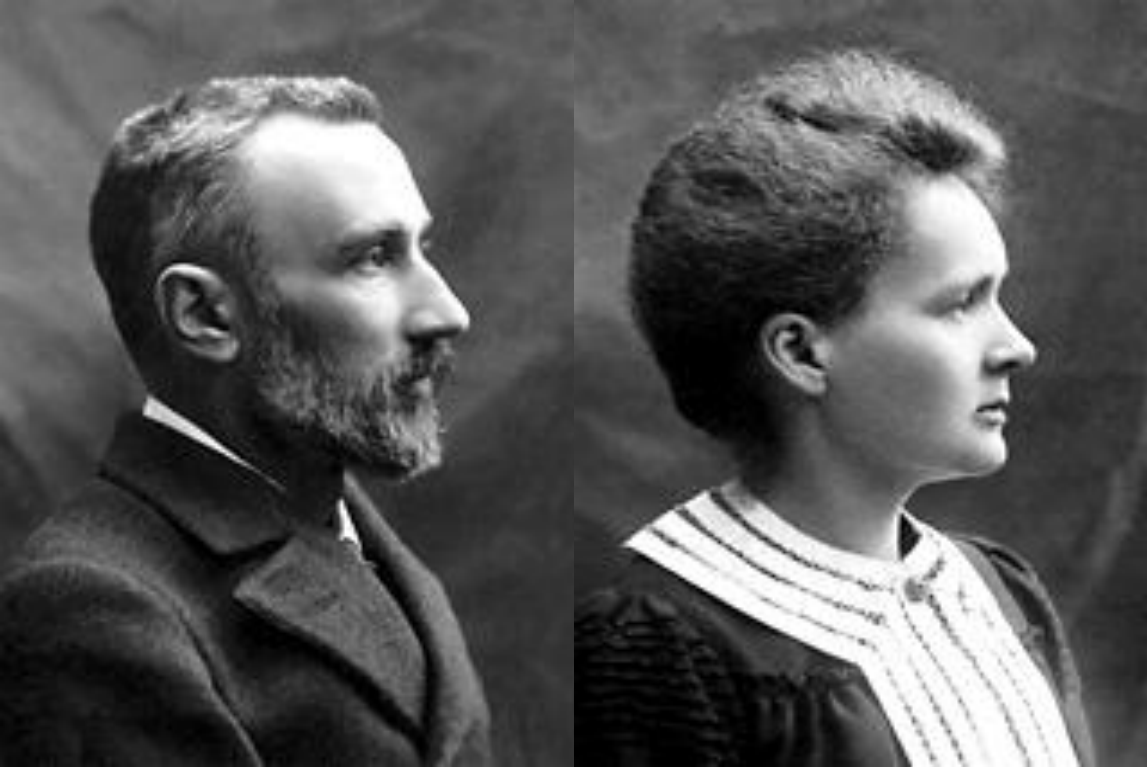
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Rutherford's atomic model



Versuchsanordnung für Rutherford's Streuversuche



Pierre Curie

Marie Curie

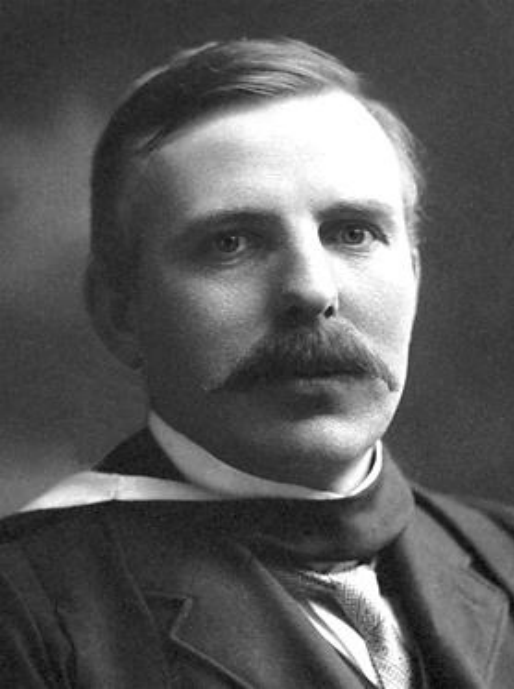
Nobel Prize in Physics 1903 was divided

Antoine Henri Becquerel "in recognition of the extraordinary services he has rendered by his discovery of spontaneous radioactivity",

Pierre Curie and Marie Curie, née Sklodowska "in recognition of the extraordinary services they have rendered by their joint researches on the radiation phenomena discovered by Professor Henri Becquerel".

The Nobel Prize in Chemistry 1911

Marie Curie *"in recognition of her services to the advancement of chemistry by the discovery of the elements radium and polonium, by the isolation of radium and the study of the nature and compounds of this remarkable element"*



The Nobel Prize in Chemistry 1908 was awarded to Ernest Rutherford

"for his investigations into the disintegration of the elements, and the chemistry of radioactive substances".

Rutherford discovered that spontaneous radioactivity gave another element. He also showed that by firing α -particles into $N_{2\text{gas}}$ that O_2 was forming.



The Nobel Prize in Physics 1922

The Nobel Prize in Physics 1922 was awarded to Niels Bohr *"for his services in the investigation of the structure of atoms and of the radiation emanating from them".*



Frederick Soddy

Nobel Prize in Chemistry 1921 "*for his contributions to our knowledge of the chemistry of radioactive substances, and his investigations into the origin and nature of isotopes*".



George de Hevesy

The Nobel Prize in Chemistry 1943 was awarded to "*for his work on the use of isotopes as tracers in the study of chemical processes*".



The Nobel Prize in Physics 1935

James Chadwick *"for the discovery of the neutron"*

Other researchers realized that by bombarding atoms with accelerated protons nuclear transformation took place. Enrico Fermi discovered that a great variety of radionuclides could be formed by bombarding with neutrons instead.



The Nobel Prize in Physics 1938

Enrico Fermi

"for his demonstrations of the existence of new radioactive elements produced by neutron irradiation, and for his related discovery of nuclear reactions brought about by slow neutrons".



The Nobel Prize in Chemistry 1944

Otto Hahn *"for his discovery of the fission of heavy nuclei"*.

1938 Otto Hahn and Fritz Strassman demonstrated fission by bombarding uranium where barium and other lighter elements with around half mass of U.

Lisa Meitner and Otto Frish (working under Nils Bohr) calculated the energy release from fission. Confirmation of $E=mc^2$



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1939

FISSION - RELEASE OF ENERGY
- RELEASE OF NEUTRONS
Chainreaction

U_{235} better than U_{238} = separation /enrichment

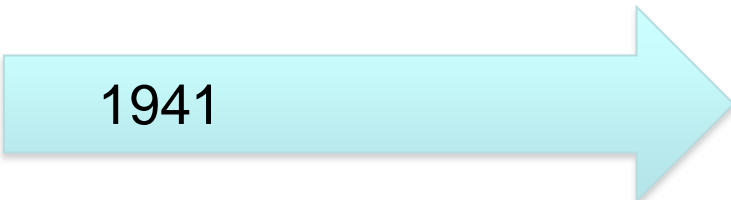
Slow neutrons higher efficiency than fast

England, USA, Russia

Bomb



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1941



-fission cross section of U_{235}

-Planning of a bomb- but was only possible if U_{235} could be highly enriched. Realized that radiation from a bomb would restrict humans to access the bombed areas for a very long time

The first manmade nuclear reactors

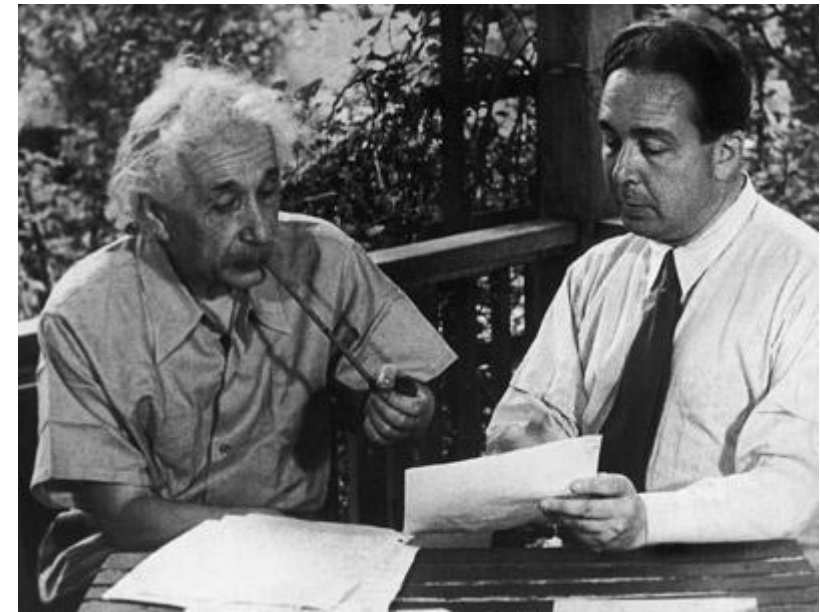
The first nuclear reactors were constructed with one purpose:
To build an atomic bomb.

The Manhattan Project



The beginning

- Researchers in USA knew that it is possible to induce fission by bombarding U with neutrons.
- German scientists were among the leading scientists in this area and the US-based scientists feared that Germany would produce an atomic bomb.
- 1939 Albert Einstein and Leo Szilard wrote letter about their concerns to president Roosevelt.
- The president decided that more research in this area was to begin and started Office of Scientific Research and Development (OSRD) 1941



The beginning

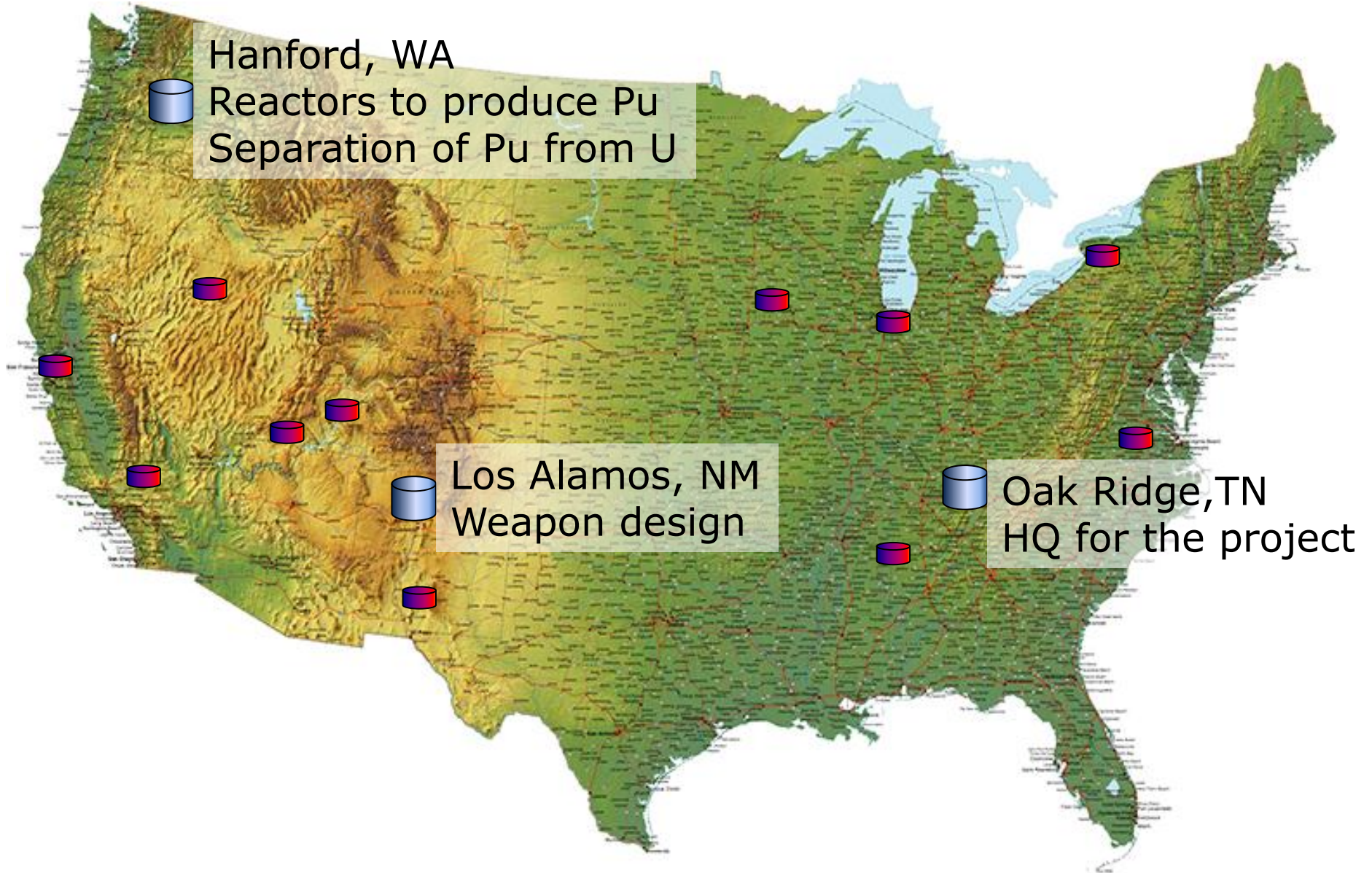


- October 1941 Pres Roosevelt approved atomic program
 - Sept. 1941 was J Rupert Oppenheimer (professor at Berkeley) assigned director of the Manhattan Project, which was debated since he had no Nobel prize and was suspected to have communist sympathies.
-

Tasks to overcome

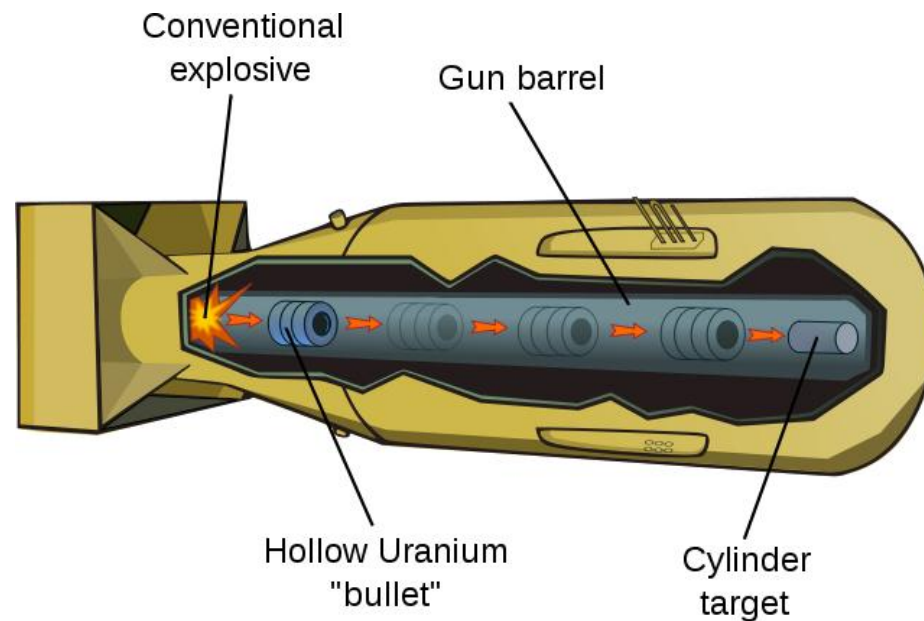
- Enriching U-235 (Separating U-235 and U-238)
 - Electromagnetic separation (Berkeley, CA)
 - Gaseous diffusion (University of Columbia)
 - Thermal diffusion (Carnegie Inst. of Washington)
 - Centrifugation (University of Columbia)
 - Reactor Technology
 - Heavy Water with graphite as moderator
 - Plutonium (was discovered 1941!) in reactor
 - Bomb design (how to get criticality when they wanted)
-

The Facilities



Making the Uranium bomb

- All proposed methods of isotope separation were developed and actually used during the project.
- Eventually they had a production line producing weapon grade U-235 (85%)



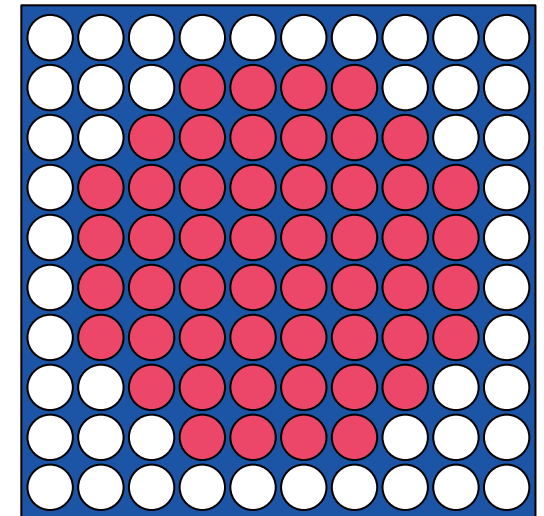
“Little Boy”

Plutonium Bomb: Construction

- The construction at Hanford started March 1943
 - 554 buildings, including 3 reactors and 3 Pu-processing facilities.
 - The lay-out of the reactors were decided, but the actual design was design-while-constructing
 - The first reactor: Construction started Aug-43 and went critical sept-44!!
-

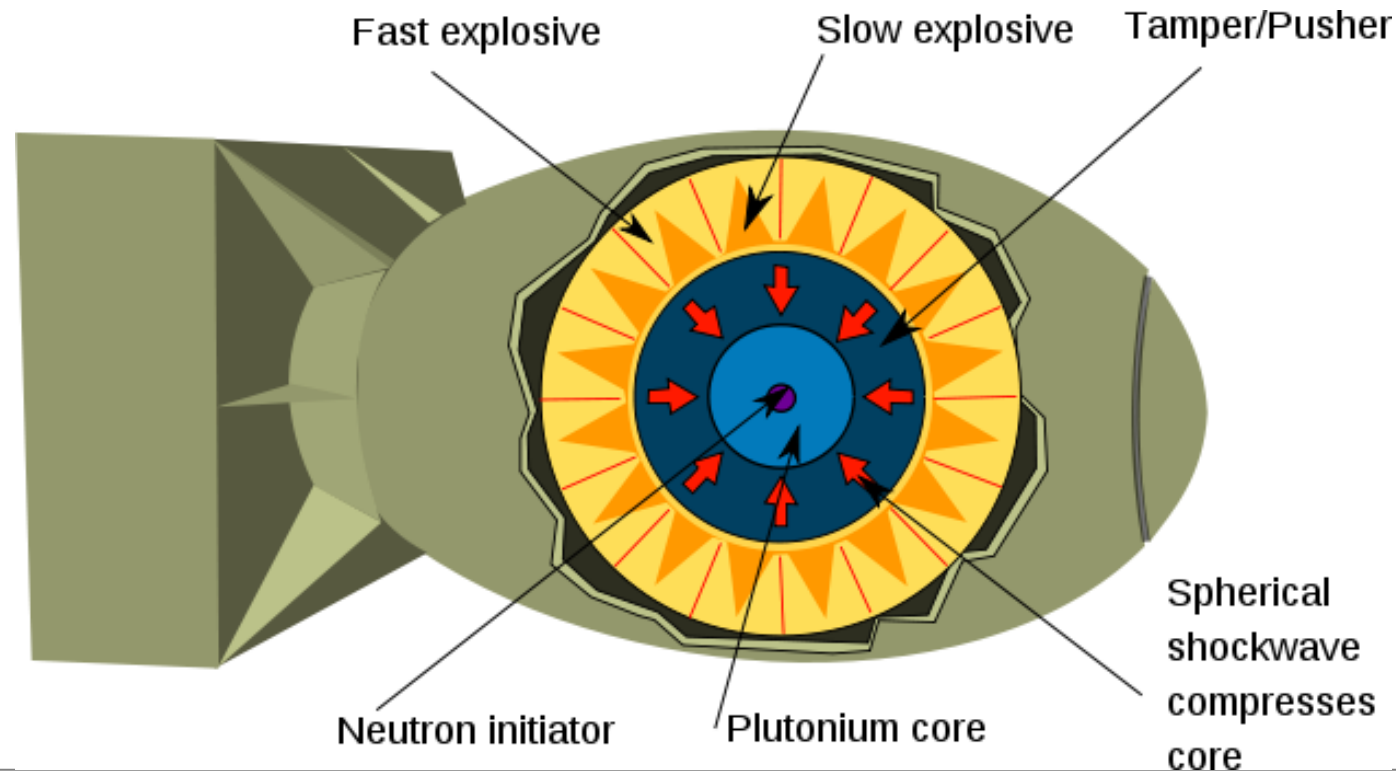
Problems

- In the beginning big problems with neutron poison (Xe-135)
Luckily enough, the engineers had put in extra tubes to fill the voids (was considered waste of time & money by scientists)
- When they finally got the first grams of Pu it turned out it contained more than 5 times more Pu-240 than expected
- Re-design of bomb to implosion bomb



Implosion bomb

- Metallurgy problems since properties of Pu was not known
- Find a proper neutron initiator (finally Po-Be succeeded)



"Fat Man"

Mayak: History

The Mayak plant was built in 1945–48, in a great hurry and in part of the Soviet Union's nuclear weapon program.

Production of Pu for weapons

Later reprocessing plant for Pu

Now also production of tritium and radioisotopes



- Mayak built in 1945-1948
- Production of Pu
- Lack of knowledge
=> Low safety, many accidents



Used river water for cooling water...

Radioactivity directly out in Techa river which ends up in Ob in the magnitude of tons. Exposure of the workers during in particular in the beginning of Mayak.

Hanford and Mayak sites comparable in contamination...

Accidents at Majak site

1957 Failure of cooling system for a tank with tons of dissolved nuclear waste. EXPLOSION spread 2×10^6 Curie over a very large area. Hundred of persons died of radiation sickness, ten of thousands were evacuated from their homes and hundreds of thousands were exposed to radiation. Rated as 6 on the INES scale

1992 the accident became official. Already information in the west.

Immediate health effects.

Accidents at Mayak site

1968 Reached criticality in a vessel when Pu was decanted in an organic solution in a vessel.

196? Heavy rains released radioactivity in the dried Lake Karachay to be released into surrounding waters

1967 Contaminated dust from Lake Karachay spread by the wind over parts of the Ozersk, where many hundred thousands of people got exposed.

The potential for groundwater contamination from the site is very high



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Development of nuclear power for energy production

Realization that the heat released in the fission could be used both for direct use and for generating electricity

Nuclear power by year

1951 first reactor in Idaho (experimental breeder reactor)

1953 Atom for peace program in USA : research towards electricity production

1954 First nuclear reactor in Obninsk

1960 first commercial reactor in Yankee Rowe, USA

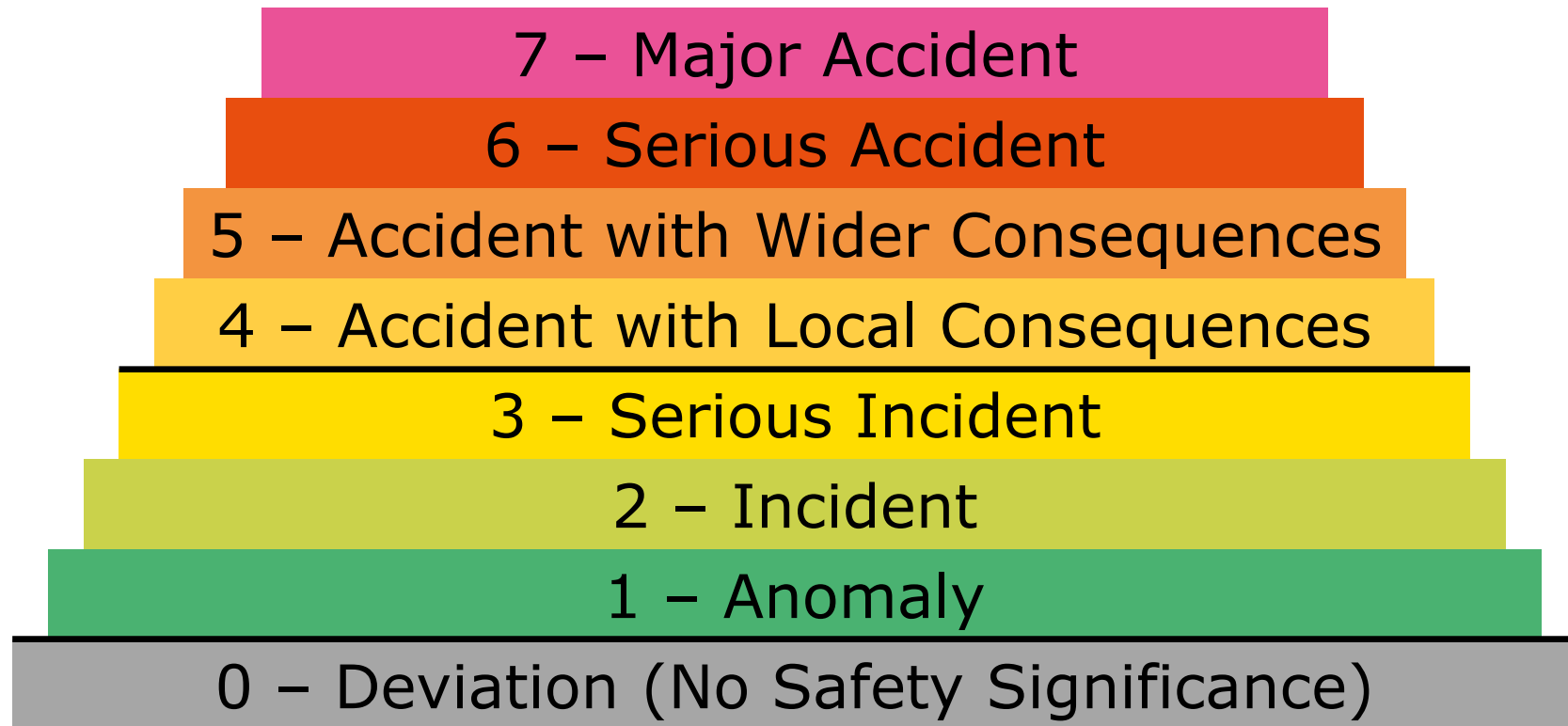
1960 Commercial BWR in Dresden

1959 First commercial plant France

1970-2002 –

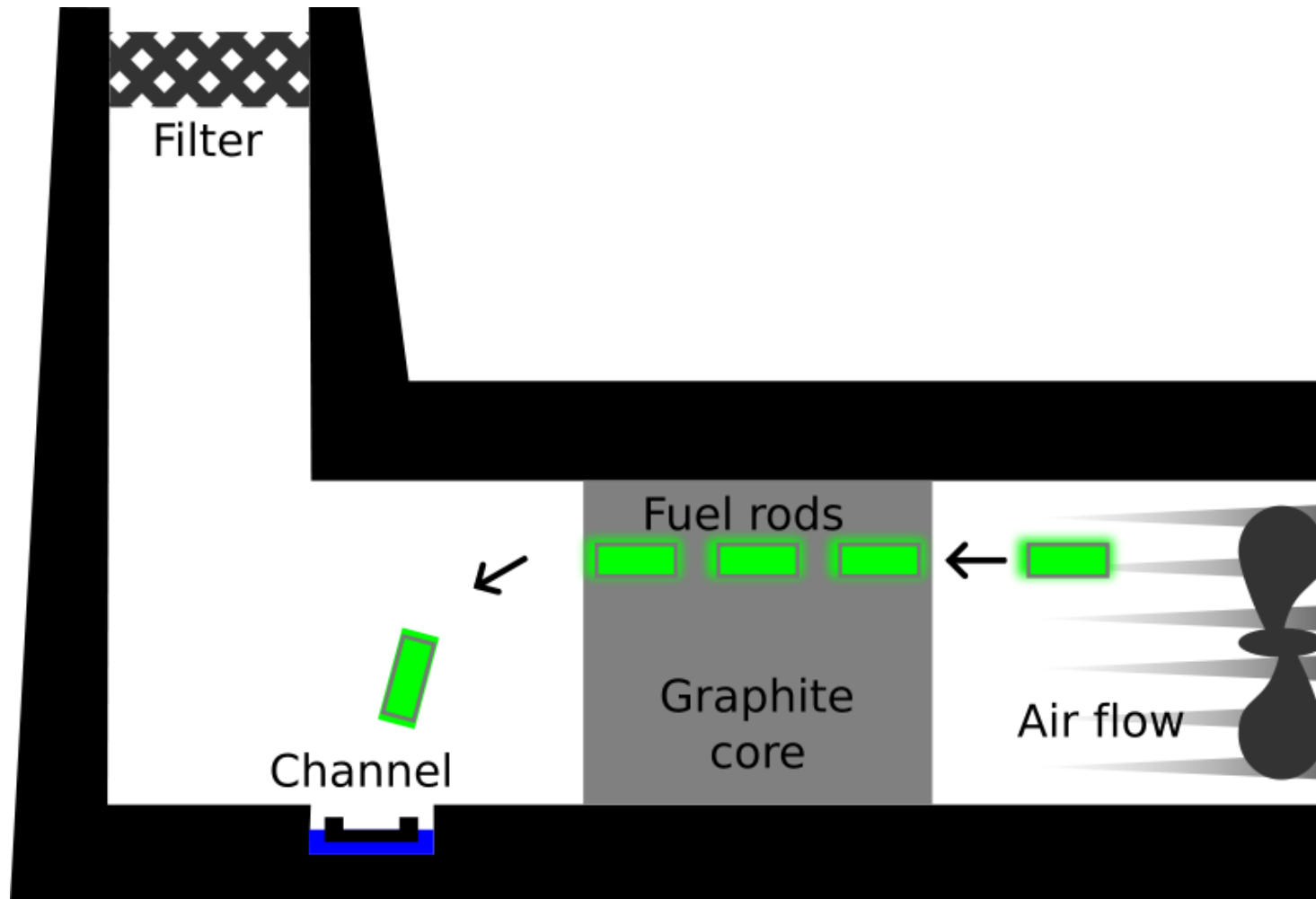
Nuclear again

International Nuclear Event Scale INES



1957: Sellafield (Windscale)

- Facility to produce weapon Pu



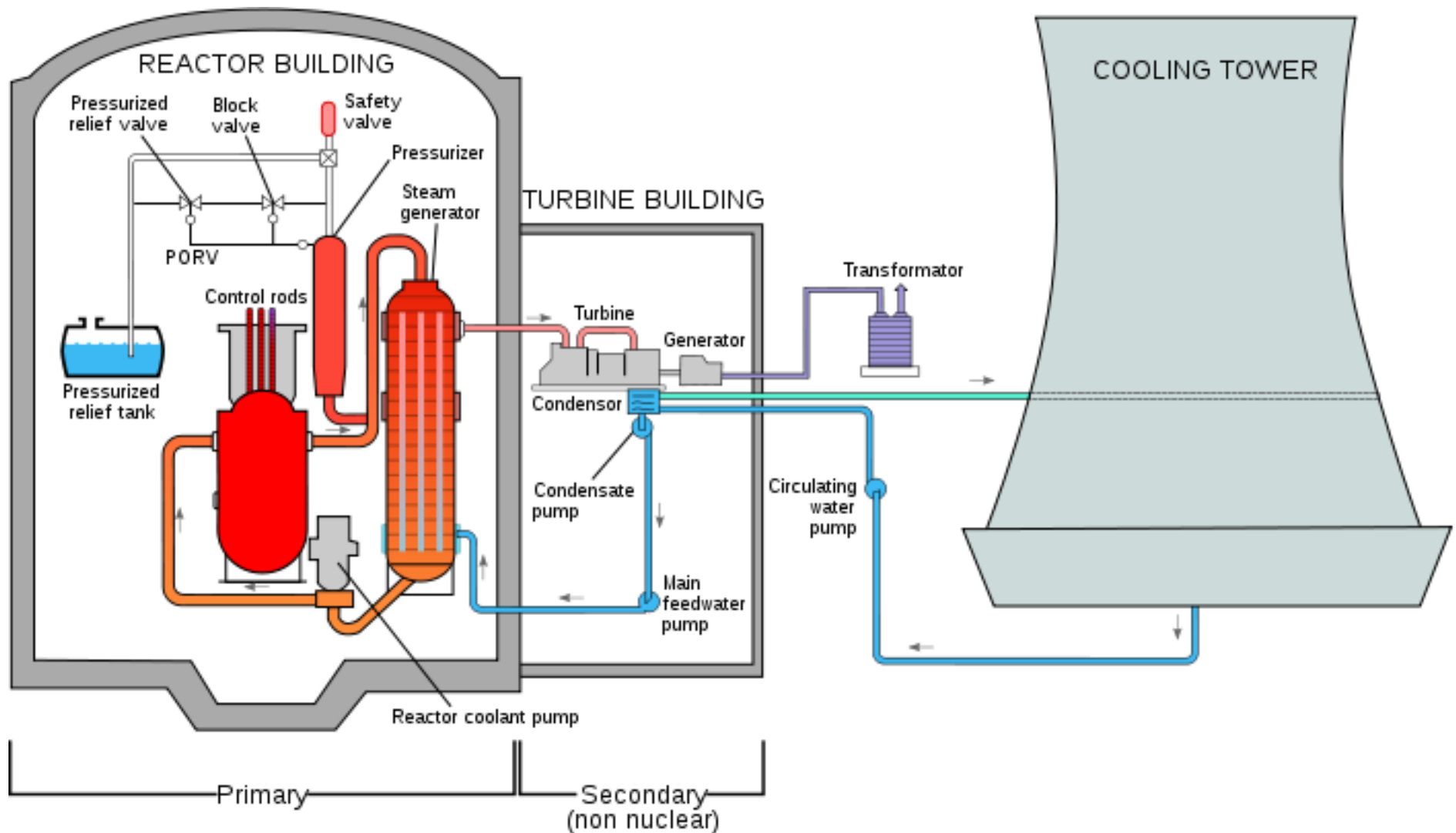


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The Sellafield fire

- US recently had detonated a thermo nuclear bomb and England wanted one as well.
 - A thermo nuclear bomb requires tritium
 - The Windscale reactor was modified, but had to work at higher temperatures => The size of the cooling fins was reduced. Safety factor reduced but tritium could be produced.
 - With new design thermo couples were no longer positioned at the hottest spots and the reactor was hence operated in the wrong way.
 - This eventually lead to that the fuel elements caught fire. At one spot the temperature was measured to be 1 300°C
 - No direct deaths. Estimated 240 cases of cancer
-

1979: Three Mile Island (Harrisburg)



1986: Chernobyl, the worst accident ever

An experiment was to be performed:

- the reactor was to be running at a low power, between >700 MW & 800 MW (normal: 3 GW)
 - the steam turbine was to be run up to full speed
 - when these conditions were achieved, the steam supply was to be closed off
 - the turbines would be allowed to freewheel down
 - generator performance was to be recorded to determine whether it could provide the bridging power for coolant pumps
-

Chernobyl

- A local power station went down and the Chernobyl director was asked to postpone the experiment.
 - The less experienced night-shift personnel performed the experiment.
 - 700MW thermal was reached but reactor continued to go down due to Xe-135
 - Reactor went down to 30MW
 - Staff decided to extract control rods
 - The low effect operation increased Xe-135 levels
 - More rods were extracted to counteract
-

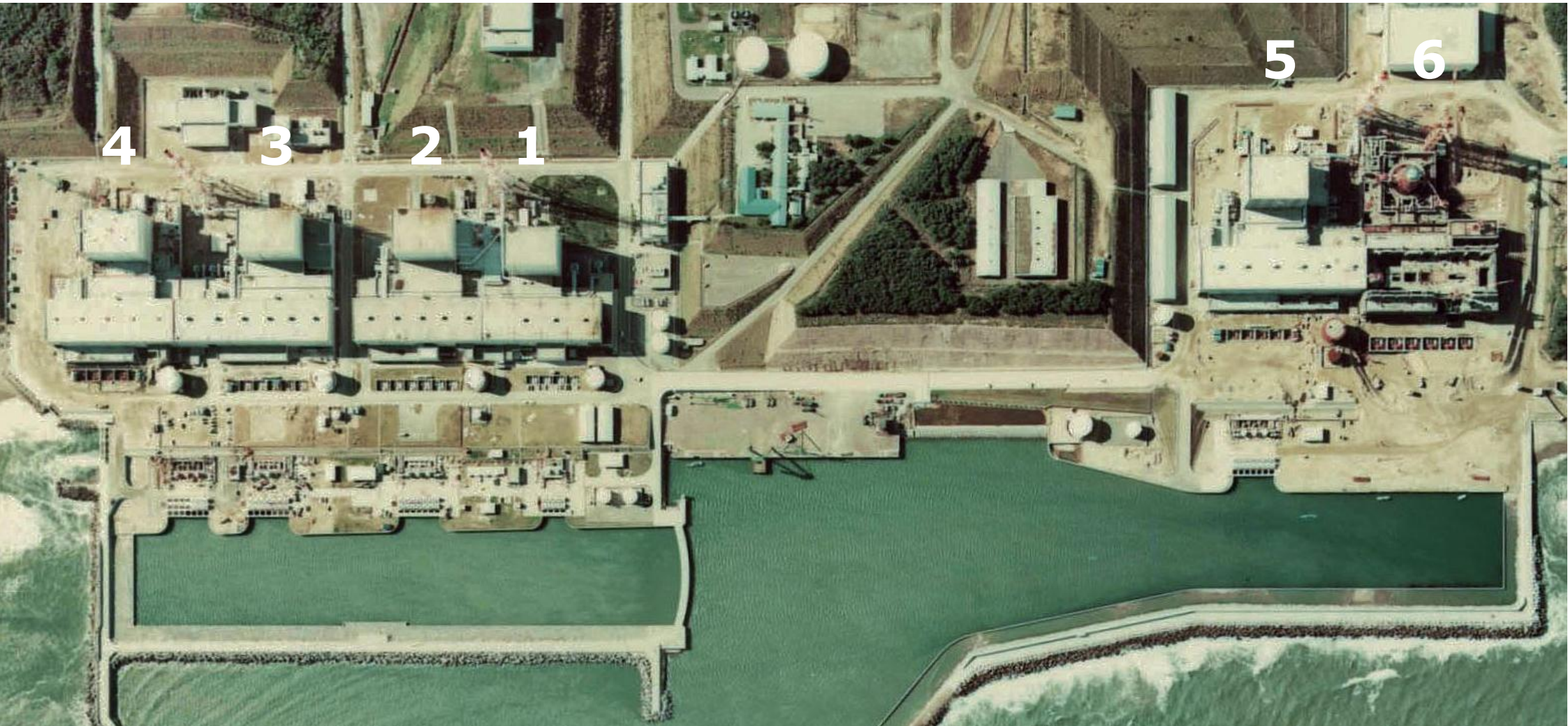
Experiment => Disaster

- The experiment started: turbines turned off
 - As a consequence the pumps slowed down and the flow was lowered
 - Formation of voids in the core, effect increased
 - This was automatically counteracted by lowering control rods
 - Someone pushed the emergency button
 - All control rods started to slowly go down in core
 - Not fast enough; graphite tip on rods started moderating neutrons
 - ⇒ Explosion
 - ⇒ The explosion destroyed rods so they couldn't go down
-

Fukushima Daiichi



Fukushima Daiichi, reactors



Fukushima Daiichi

- 11 March 2010 an earthquake occur with epicenter close to Honshu
 - Reactors 4, 5 & 6 were not operating due to periodic inspection
 - Reactors 1, 2 & 3 closed down automatically when the earthquake struck -> stopping normal power to the plant
 - Emergency diesel generators started power cooling system (2 generators per reactors 1-5 and 3 for reactor 6)
-

Fukushima Daiichi

- A 13-15 meter high tsunami arrived 50 min after the earth quake to the Fukushima Daiichi nuclear power plant (seawall was 5.7 m.)
 - The basement of turbine basement was flooded, disabling emergency generators
 - Without cooling the energy release from the radioactivity of the fuel increased the temperature
 - Eventually full meltdown of all three reactors
 - Several hydrogen explosions
 - At several occasions the pressure in the reactors needed to be released causing release of radionuclides
-

Fukushima Daiichi: Casualties

- The released activity is 1/10 of Chernobyl
 - No immediate deaths due to radiation exposure
 - At least six workers have exceeded life-time radiation dose
 - More than 300 have received significant radiation doses
 - Future cancer deaths in Fukushima area estimated to 100-1000
-
- The tsunami from the Tōhoku earthquake:
 - 15 850 deaths, 6 011 injured, 3 287 people missing
-

Other accidents

- 1977 Jaslovské Bohunice, CZE: Fuel rods were replaced when reactor was active (standard procedure). The humidity absorbers covering the rods were not removed, causing local overheating of fuel. Active zone damaged and both primary & secondary circuits were contaminated
 - 1999 Tokaimura: 3 workers prepared small batch of fuel to an experiment breeder reactor in a bucket. The highly enriched U reached critical mass.
 - 2004 Mihama Nuclear Power Plant: Hot water and steam from a broken pipe killed 5 workers
-

Major nuclear accidents

- INES 7: Chernobyl and Fukushima
 - INES 6: Mayak (1957)
 - INES 5:
 - Windscale fire
 - Harrisburg
 - Chalk River (1952): reactor core damaged
 - Lucens, Switzerland(1969): Test reactor lost coolant, partial core meltdown
 - Goiânia (Brazil): A Cs-137 source was stolen from an abandoned hospital and sold to a scrapyard. 300 contaminated, 11 deaths
-