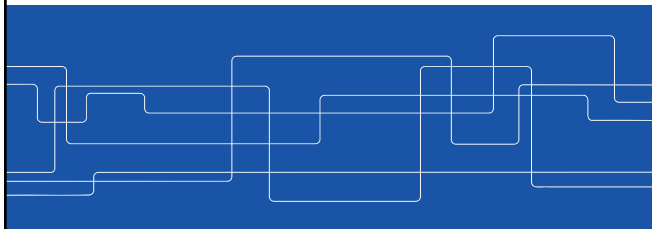




## Nuclear Fuel Cycle 2013

### Lecture 9: The History of Nuclear Power




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### Oklo: The first nuclear reactor

$1.7 \times 10^9$  years ago

It was found that the  $^{235}\text{U}$  abundance was 0.44% (normally it is 0.72%).  
Only possible reason: A natural nuclear reactor

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

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

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

$\Rightarrow$  U-235 abundance  $1.7 \times 10^9$  y ago was 2.9%

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




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### Oklo geology

U was accumulated at a redox-front. The redox-front did not arise until the  $\text{O}_2$ -concentration was sufficiently high (c.a 1.7 billion y ago)

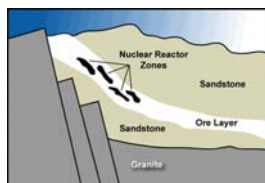
Water flowing through the sandstone worked as moderator.

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




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Mendeleev's periodic table 1869

Period	Gruppe I. R <sup>0</sup>	Gruppe II. R <sup>0</sup>	Gruppe III. R <sup>0</sup>	Gruppe IV. R <sup>0</sup>	Gruppe V. R <sup>0</sup>	Gruppe VI. R <sup>0</sup>	Gruppe VII. R <sup>0</sup>	Gruppe VIII. R <sup>0</sup>
1	Li=7	Be=9,4	B=11	C=12	N=14	O=16	F=19	
2	Na=23	Mg=24	Al=27,3	Si=28	P=31	S=32	Cl=35,5	
3	K=39	Ca=40	—=44	Ti=48	V=51	Cr=52	Mn=55	Fe=56, Co=59, Ni=59, Cu=63.
4	(Ca=63)	Zn=65	—=68	—=72	As=75	Se=78	Br=80	
5	Rb=85	Sr=87	Yt=88	Zr=90	Nb=94	Mo=96	—=100	Ru=104, Rh=104, Pd=106, Ag=108.
6	(Ag=108)	Cd=112	In=113	Sn=118	Sb=122	Te=125	J=127	
7	Ce=133	Ba=137	?Di=138	?Ce=140	—	—	—	—
8	(—)	—	—	—	—	—	—	—
9	—	—	?Er=178	?La=180	Ta=182	W=184	—	Os=195, Ir=197, Pt=195, Au=199.
10	(Au=199)	Hg=200	Tl=204	Pb=207	Bi=208	—	—	—
11	—	—	—	Th=231	—	U=240	—	—
12	—	—	—	—	—	—	—	—

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### 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

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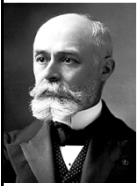
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## Henri Becquerel

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.

$\beta$ -radiation  
 $\alpha$ -radiation

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

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## Radioactive decay



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## Discovery of Po and Ra



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### Determination of atomic weight of Ra



### Marie and Irène



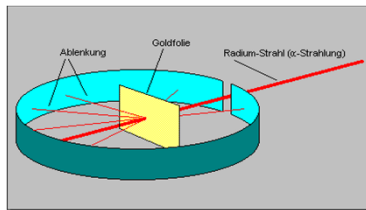
### Applications of Curies research

#### Radiumhemmet 1910





### Rutherfords atomic model



Versuchsanordnung für Rutherfords Streuversuche

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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 Skłodowska** "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"

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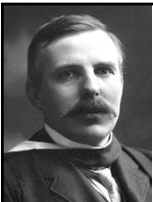
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The Nobel Prize in Chemistry 1908 was awarded to **Ernest Rutherford**

"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  $\alpha$ -particles into  $N_{2gas}$  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".

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**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".

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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.

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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".

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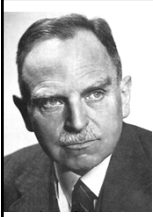
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The Nobel Prize in Chemistry 1944

**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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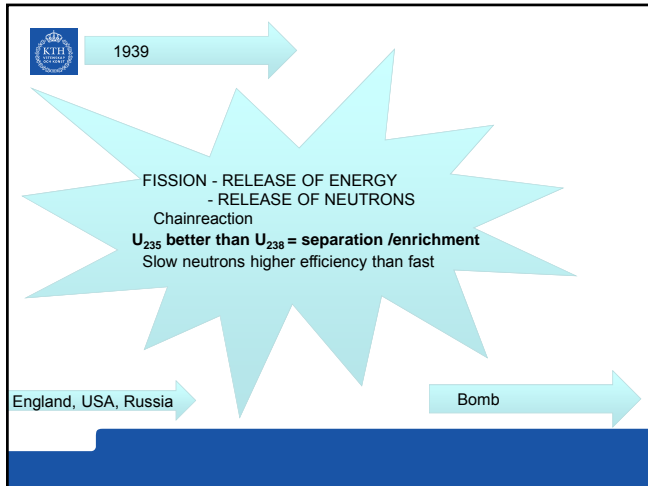
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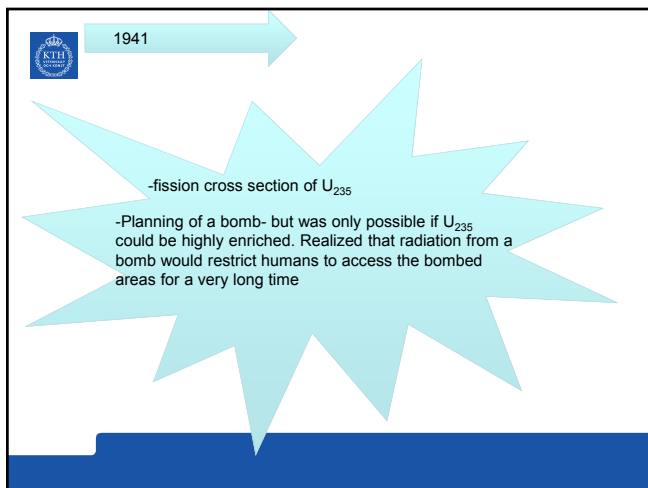
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### The first manmade nuclear reactors

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

#### The Manhattan Project




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### 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

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### The beginning



October 1941 President **Roosevelt** approved the atomic program

Sept. 1941 was **J. Robert 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.

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### 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)

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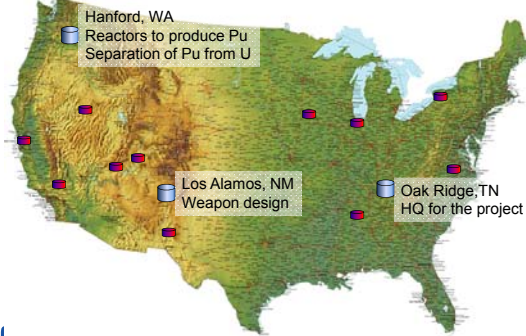
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### The Facilities




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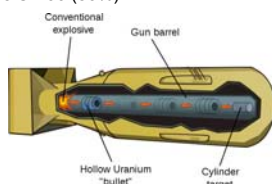
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### Making the Uranium bomb

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%)



"Fat Man"

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## Plutonium Bomb: Construction

The construction at Hanford started March 1943

554 buildings, including 3 reactors and 3 Pu-processing facilities.

(After WWII another 6 was built at the site)

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!!

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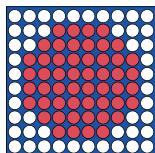


## Problems

In the beginning big problems with neutron poison (Xe-135)  
Lucky 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




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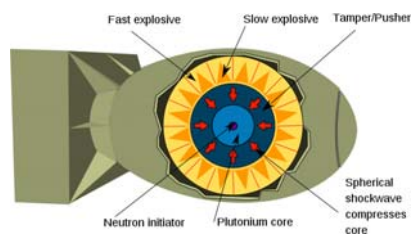
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## Implosion bomb

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



"Fat Man"

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## 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 (5 reactors)

Later reprocessing plant for Pu

Today production of tritium and radioisotopes

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

A lot of waste dumped into the Techa River  
(in the order of 100 PBq);  
Lake Karachay 4 Ebq ( $4 \times 10^{18}$  Bq)




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## 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  $0.8 \times 10^{18}$  Bq 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

**1976** the accident became official. Already information in the west.

Immediate health effects.

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## Accidents at Mayak site

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

**1967** 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



## More Mayak accidents

- 08/02/1993 - Accident at the plant used to treat liquid waste, this incident was associated with the depressurization of the pipeline and caused 2 m<sup>3</sup> of radioactive slurry to the surface of the earth (about 100 m<sup>2</sup> of contaminated surface). Depressurization of the pipeline led to leak to the surface of the pulp radioactive activity of about 0.3 Ci. Radioactive trace was localized, contaminated soil removed.
- 12/27/1993 - Incident at radiololope plant where the replacement of a filter resulted in the release into the atmosphere of radioactive aerosols. Emissions were on the α-activity of 0.033 Ci, and β-activity of 0.36 mCi.
- 04/02/1994 - Recorded increased release of radioactive aerosols: the β-activity of 2-day levels of Cs-137 subsistence levels, the total activity of 7.15 mCi.
- 30/03/1994 - Recorded excess daily release of Cs-137 in 3, β-activity - 1.7, α-activity - by 1.9 times. In May 1994 the ventilation system of the building of the plant spewed activity 10.4 mCi β-aerosols. Emission of Cs-137 was 83% of the control level.
- 07/07/1994 - The control plant detected a radioactive spot area of several square decimeters. Exposure dose was 500 mR / s. The spot was formed by leaking sewage.
- 31/08/1994 - Registered an increased release of radionuclides to the atmospheric pipe building reprocessing plant (238.8 mCi, with the share of Cs-137 was 4.36% of the annual emission limit of this radionuclide). The reason for the release of radionuclides was depressurization of VVER-440 fuel elements during the operation segments idle all SFA (spent fuel assemblies) as a result of an uncontrollable arc.
- 24/03/1995 - Recorded excess of 19% of normal loading apparatus plutonium, which can be regarded as a dangerous nuclear incident.
- 15/09/1995 - High-level liquid radioactive waste (LRW) was found in flow of cooling water. Operation of a furnace into the regulatory regime has been discontinued.
- 21/12/1995 - Cutting of a thermometric channel exposed four workers (1.69, 0.99, 0.45, 0.34 rem). The reason for the incident - a violation of the company's employees process procedures.
- 24/07/1995 - Cs-137 aerosols released, the value of which amounted to 0.27% of the annual value of MPE for the enterprise. The reason - the fire filter cloth.



## 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 energy again?

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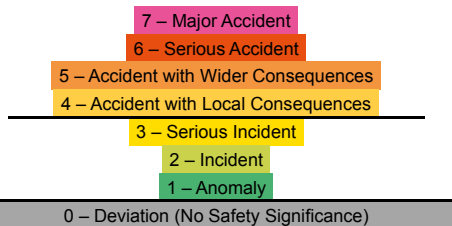
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### International Nuclear Event Scale INES




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### INES Levels

**7 Major accident:** Major release of radioactive material with widespread health and environmental effects requiring implementation of planned and extended countermeasures

**6 Serious accident:** Significant release of radioactive material likely to require implementation of planned countermeasures

**5 Accident with wider consequences:** Limited release of radioactive material likely to require implementation of some planned countermeasures.  
 Several deaths from radiation

**4 Accident with local consequences:** Minor release of radioactive material unlikely to result in implementation of planned countermeasures other than local food controls.  
 At least one death from radiation.

**3 Serious incident:** Exposure in excess of ten times the statutory annual limit for workers. Non-lethal deterministic health effect (e.g., burns) from radiation.

**2 Incident:** Exposure of a member of the public in excess of 10 mSv.  
 Exposure of a worker in excess of the statutory annual limits.

**1 Anomaly:** Overexposure of a member of the public in excess of statutory annual limits.  
 Minor problems with safety components with significant defence-in-depth remaining.  
 Low activity lost or stolen radioactive source, device or transport package.

**0 Deviation:** No safety significance

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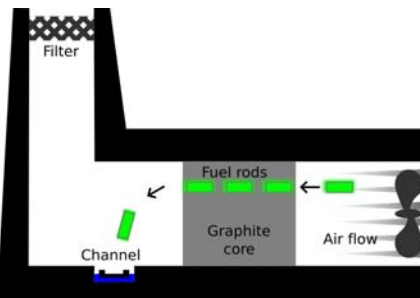
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### 1957: Sellafield (Windscale)

Facility to produce weapon Pu



### The Sellafield fire

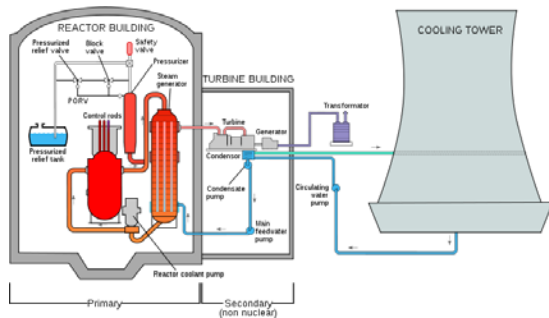
US recently had detonated a thermo nuclear bomb and England wanted one as well.

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)





### Harrisburg accident continued

#### Main reasons for accident:

Poor design of "safety interface"

Insufficient education of staff working in control room

#### Aftermath:

Radioactive Iodine was released

No cases of cancer have been linked to the accident

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### 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

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### 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

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## 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)

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## 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

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## 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

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### 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

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### Major nuclear accidents

INES 7: Chernobyl and Fukushima

INES 6: Mayak (1957)

INES 5:

- Windscale fire (Sellafield)
- 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

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