Radioactivity

Lecture 21

Radioactivity and Long Term Storage

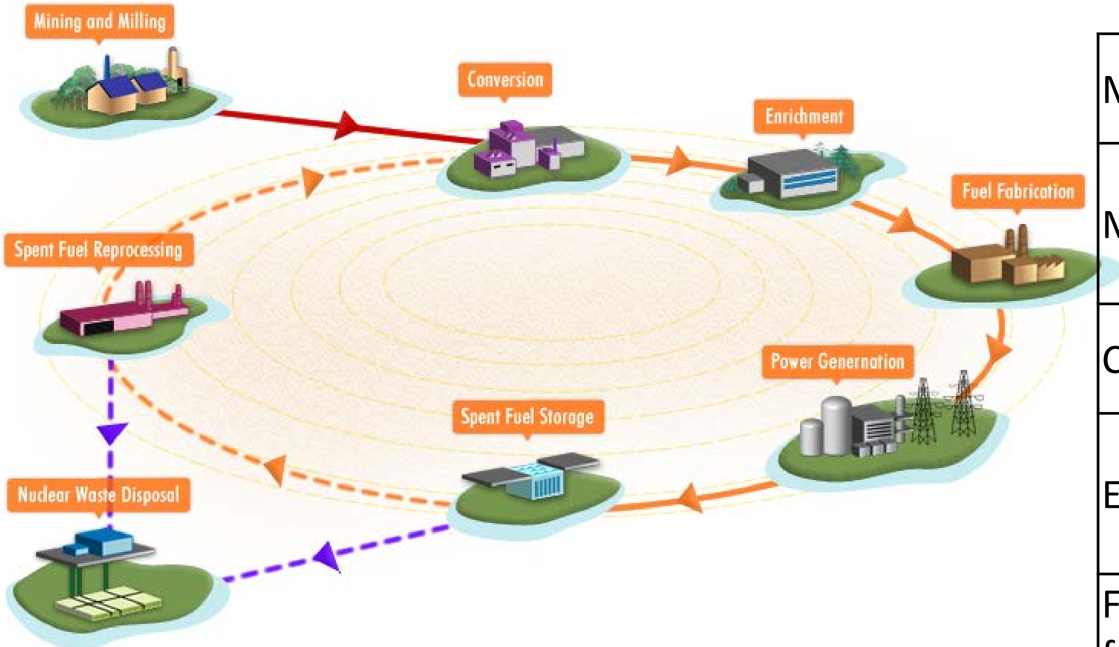
Radioactive Waste

of the Nuclear and other Industries

- Radioactive waste is associated with all aspects of the nuclear fuel cycle
- Uranium (and Thorium) harvesting
- Fuel pellet processing
- Reactor operation and possible failures
- Regenerating fuel rods and storing waste material
- Short time storage concerns
- Long time storage needs
- Alternative plans via waste transmutation

Nuclear Fuel Cycle

Material amounts for the annual operation of a 1000 MWe nuclear power reactors, using 4.5% enriched fuel with 45 GW/t burn-up.



Mining	Anything from 20,000 to 400,000 tons of uranium ore
Milling	249 tons of uranium oxide concentrate (which contains 211 tons of uranium)
Conversion	312 tons of uranium hexafluoride, UF_6 (with 211 tons of Uranium)
Enrichment	35.9 tons of enriched UF_6 (containing 24.3 t enriched U @ 4.5%) – balance is 'tails' @ 0.22%
Fuel fabrication	27.6 tons UO_2 (with 24.3 t enriched U)
Reactor operation	8760 million kWh (8.76 TWh) of electricity at 100% output, hence 24 tons of natural U per TWh
Used fuel	27.6 tons containing 280 kg transuranics (mainly plutonium), 26 t uranium oxide (<1.0% U-235), 1 ton fission products.

Substantial radioactive foot print

Uranium Mining

Fuel rod production

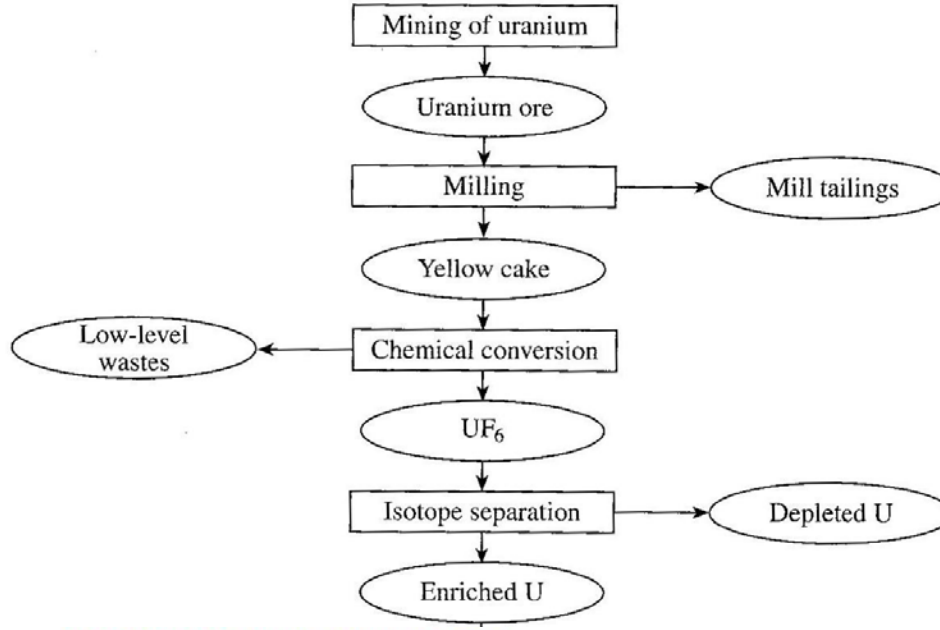
Reactor operation

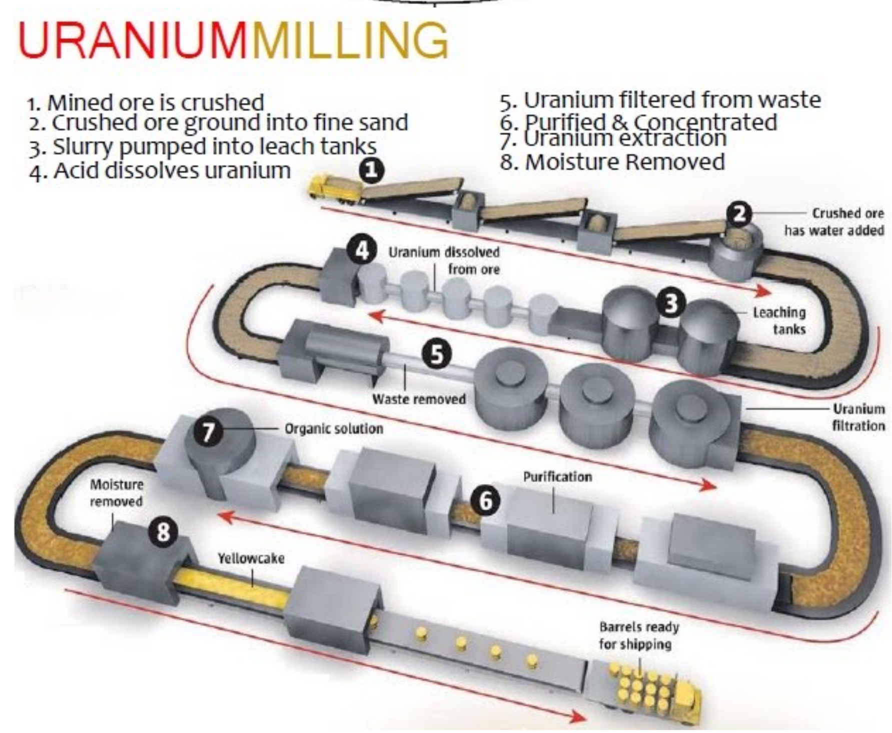
Fuel rod storage

Fuel rod reprocessing

Nuclear waste disposal

Fuel Cycle Steps





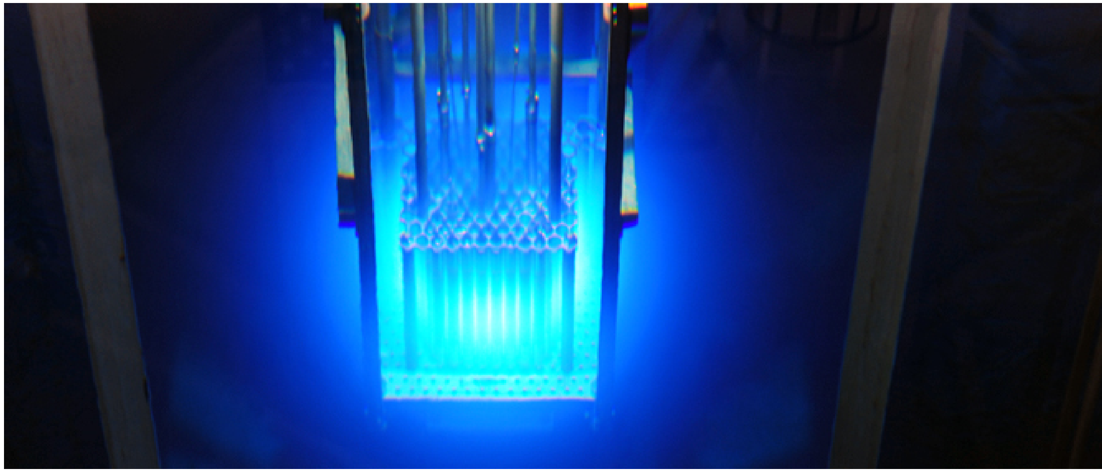
Uranium Mining

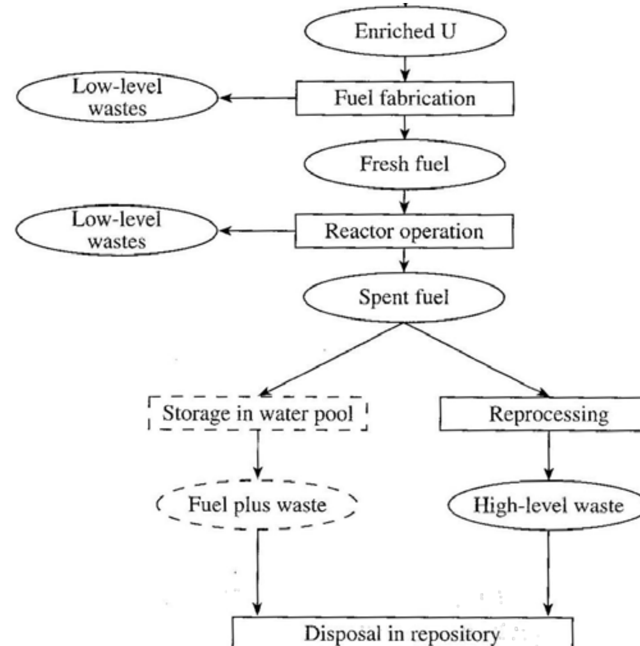
The world's ten largest uranium mines.

Mine	Country	Main owner	Type	Production (tU)	% of world
McArthur River	Canada	Cameco	underground	7356	13
Tortkuduk & Moinkum	Kazakhstan	Katco JV/Areva, Kazatomprom	ISL	4322	8
Olympic Dam	Australia	BHP Billiton	by-product/ underground	3351	6
SOMAIR	Niger	Areva	open pit	2331	5
Budenovskoye 2	Kazakhstan	Karatau JV/Kazatomprom, Uranium One	ISL	2084	4
South Inkai	Kazakhstan	Betpak Dala JV/Uranium One, Kazatomprom	ISL	2002	4
Priagunsky	Russia	ARMZ	underground	1970	4
Langer Heinrich	Namibia	Paladin	open pit	1947	4
Inkai	Kazakhstan	Inkai JV/Cameco, Kazatomprom	ISL	1922	3
Central Mynkuduk	Kazakhstan	JSC Ken Dala, Kazatomprom	ISL	1790	3
Top 10 total				**29,075**	**54%**

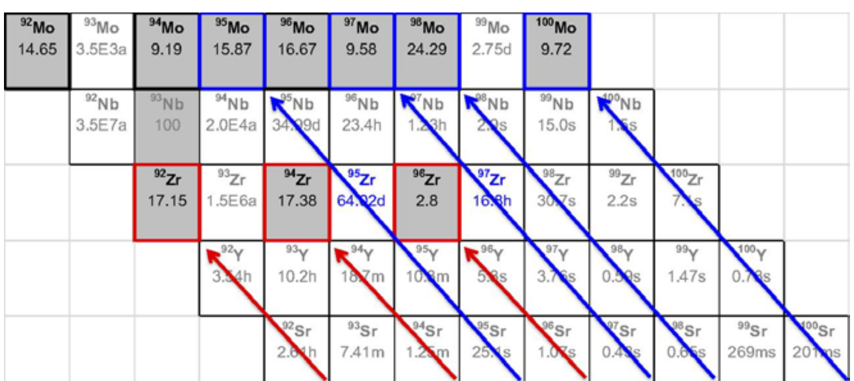
The environmental impact is enormous because of uranium rich tailings, prior to dissolving those by leaching! ISL means "In Situ Leach", where the minerals are dissolved underground and pumped to surface.

Converting fuel to waste

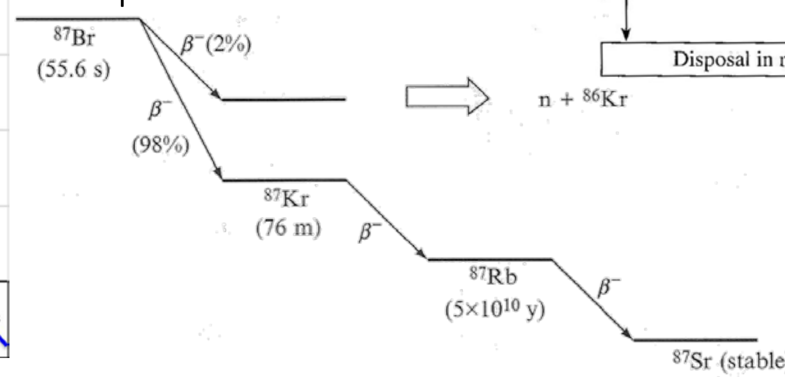




Fission converts ^{235}U into fission product distribution. The neutron-rich fission products decay back to stability. The decay products can be very long-lived! They remain with the unprocessed uranium (of other fission fuel) in the fuel rod contaminating the rod, which has to be reprocessed or stored.



Example ^{87}Br:



Radioactive Fission Products

By yield

Yield	Element	Isotope	Halflife
6.7896%	Caesium	$^{133}Cs \rightarrow {}^{134}Cs$	2.065 y
6.3333%	Iodine, Xenon	$^{135}I \rightarrow {}^{135}Xe$	6.57 h
6.2956%	Zirconium	^{93}Zr	1.53 My
6.1%	Molybdenum	^{99}Mo	65.94 h
6.0899%	Caesium	^{137}Cs	30.17 y
6.0507%	Technetium	^{99}Tc	211 ky
5.7518%	Strontium	^{90}Sr	28.9 y
2.8336%	Iodine	^{131}I	8.02 d
2.2713%	Promethium	^{147}Pm	2.62 y
1.0888%	Samarium	^{149}Sm	virtually stable
0.9%	Iodine	^{129}I	15.7 My
0.4203%	Samarium	^{151}Sm	90 y
0.3912%	Ruthenium	^{106}Ru	373.6 d
0.2717%	Krypton	^{85}Kr	10.78 y
0.1629%	Palladium	^{107}Pd	6.5 My
0.0508%	Selenium	^{79}Se	327 ky
0.0330%	Europium, Gadolinium	$^{155}Eu \rightarrow {}^{155}Gd$	4.76 y
0.0297%	Antimony	^{125}Sb	2.76 y
0.0236%	Tin	^{126}Sn	230 ky
0.0065%	Gadolinium	^{157}Gd	stable
0.0003%	Cadmium	^{113m}Cd	14.1 y

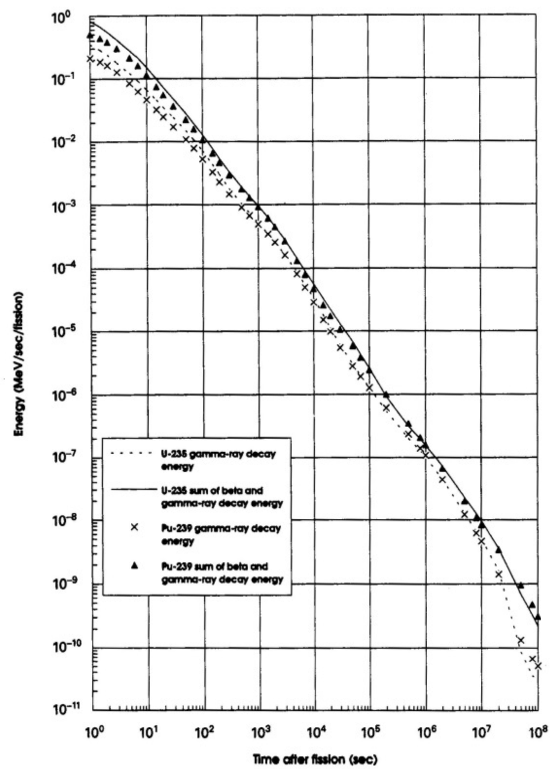
By half-life

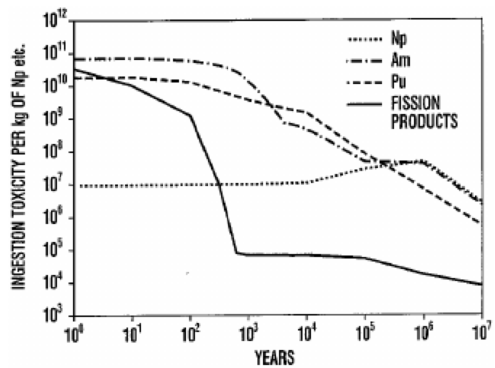
Yield	Halflife
2.8336%	8.02d
0.3912%	373.6d
6.7896%	2.065y
2.2713%	2.62y
0.0297%	2.76y
<0.0330%	4.76y
0.2717%	10.78y
<0.0003%	14.1y
5.7518%	28.9y
6.0899%	30.17y
<0.4203%	90y
6.0507%	211ky
0.0236%	230ky
0.0508%	327ky
6.2956%	1.53My
<6.3333%	2.3My
0.1629%	6.5My
0.6576%	15.7My
<1.0888%	nonradioactive
<0.0065%	nonradioactive

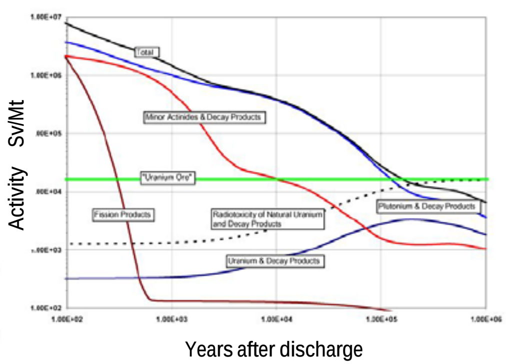
Decay of Radioactivity

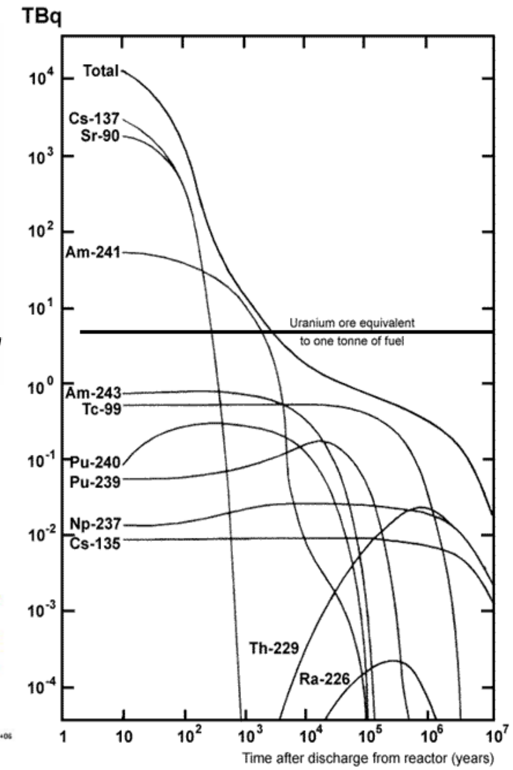
Development of activity is dominated by the radioactive isotope with the largest decay constant or shortest life time at each moment of time.

$$A(t) = \sum_i \lambda_i \cdot N_i = \lambda_1 \cdot N_1 + \lambda_2 \cdot N_2 + \lambda_3 \cdot N_3 + \lambda_4 \cdot N_4 + \ldots = \sum_i \frac{\ln 2}{T_{1/2_i}} \cdot N_i = \frac{\ln 2}{T_{1/2_1}} \cdot N_1 + \frac{\ln 2}{T_{1/2_2}} \cdot N_2 + \frac{\ln 2}{T_{1/2_3}} \cdot N_3 + \frac{\ln 2}{T_{1/2_4}} \cdot N_4 + \ldots$$





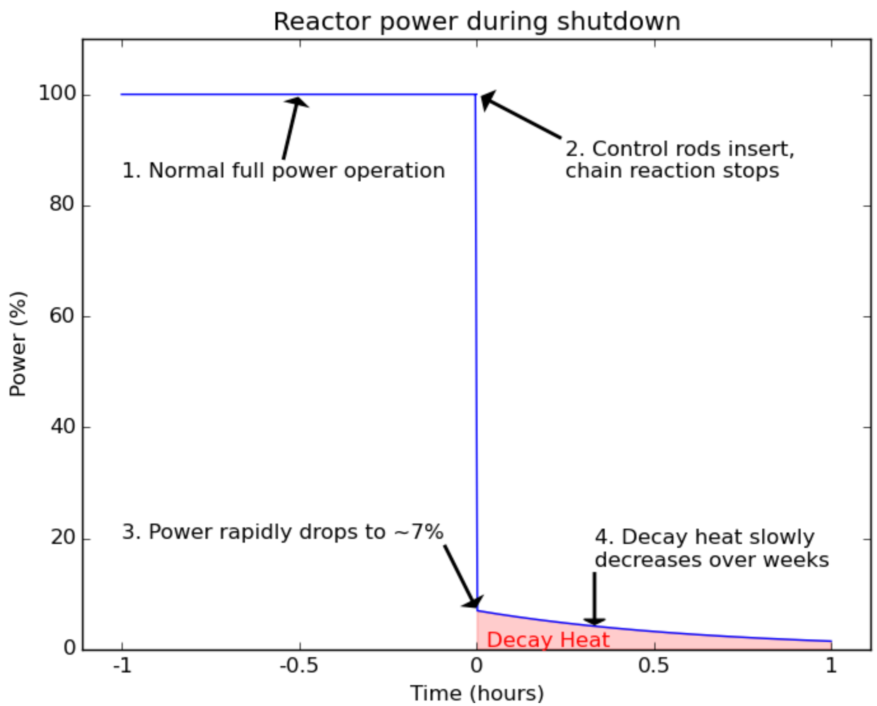




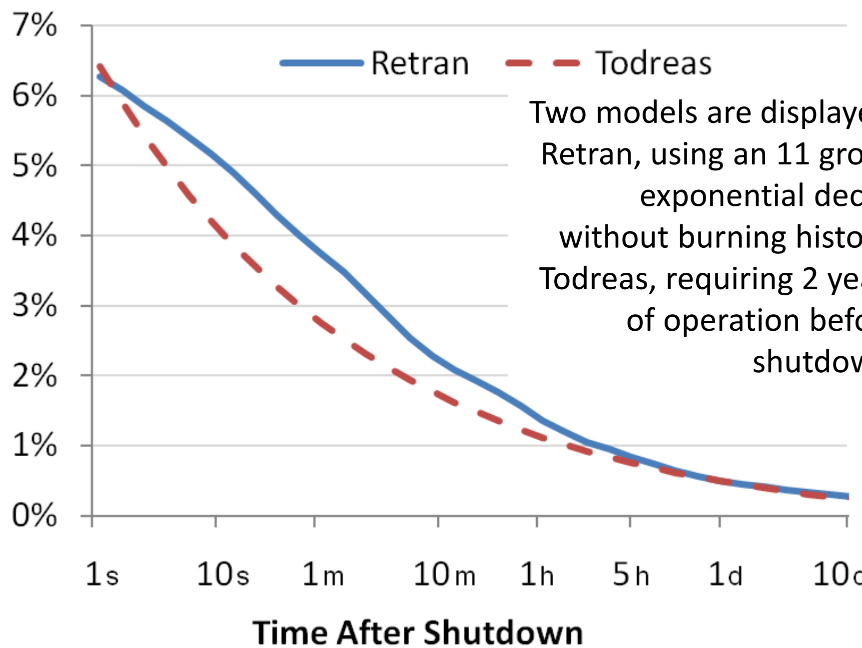
Activity of high-level waste from one tonne of spent fuel

The initial activity in fuel rods is initially extremely high, short-lived radioactivity decreases by nine orders of magnitude during the first year. Material needs to be stored for that time before further processed. It takes another thousand years until activity reaches natural Uranium levels.

Decay heat after reactor shut-down

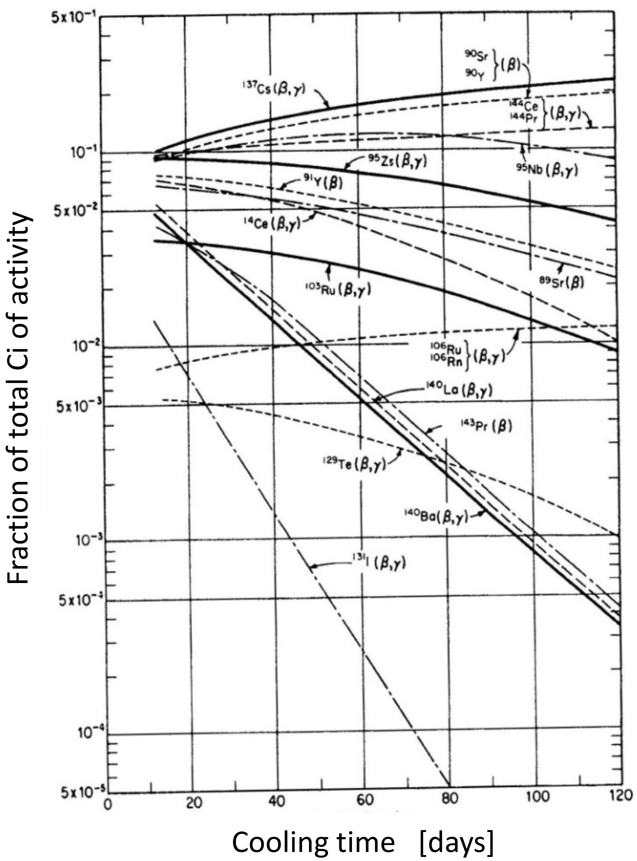


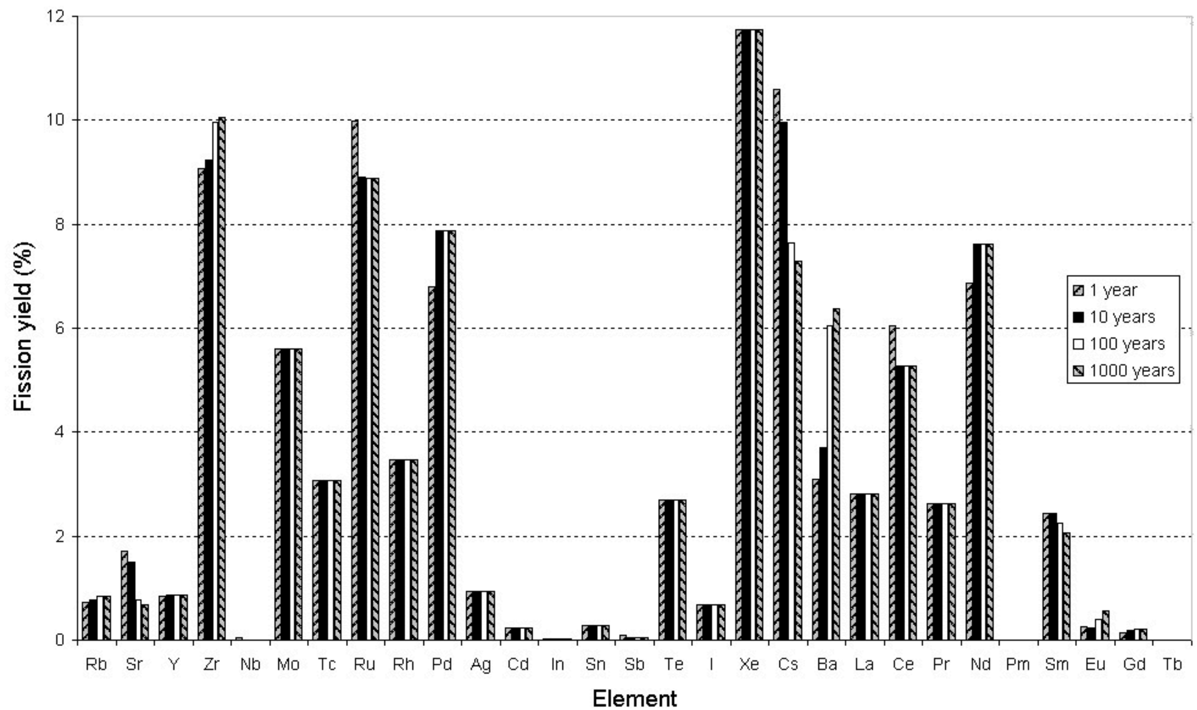
The heat released from the decay of the fission products requires cooling the reactor for about 10 days after shut-down. Decay heat is primarily released from the decay of short-lived nuclei, long-lived nuclei have less activity $Q \sim A \sim 1/T_{1/2}$!



Two models are displayed, Retran, using an 11 group exponential decay, without burning history; Todreas, requiring 2 years of operation before shutdown.

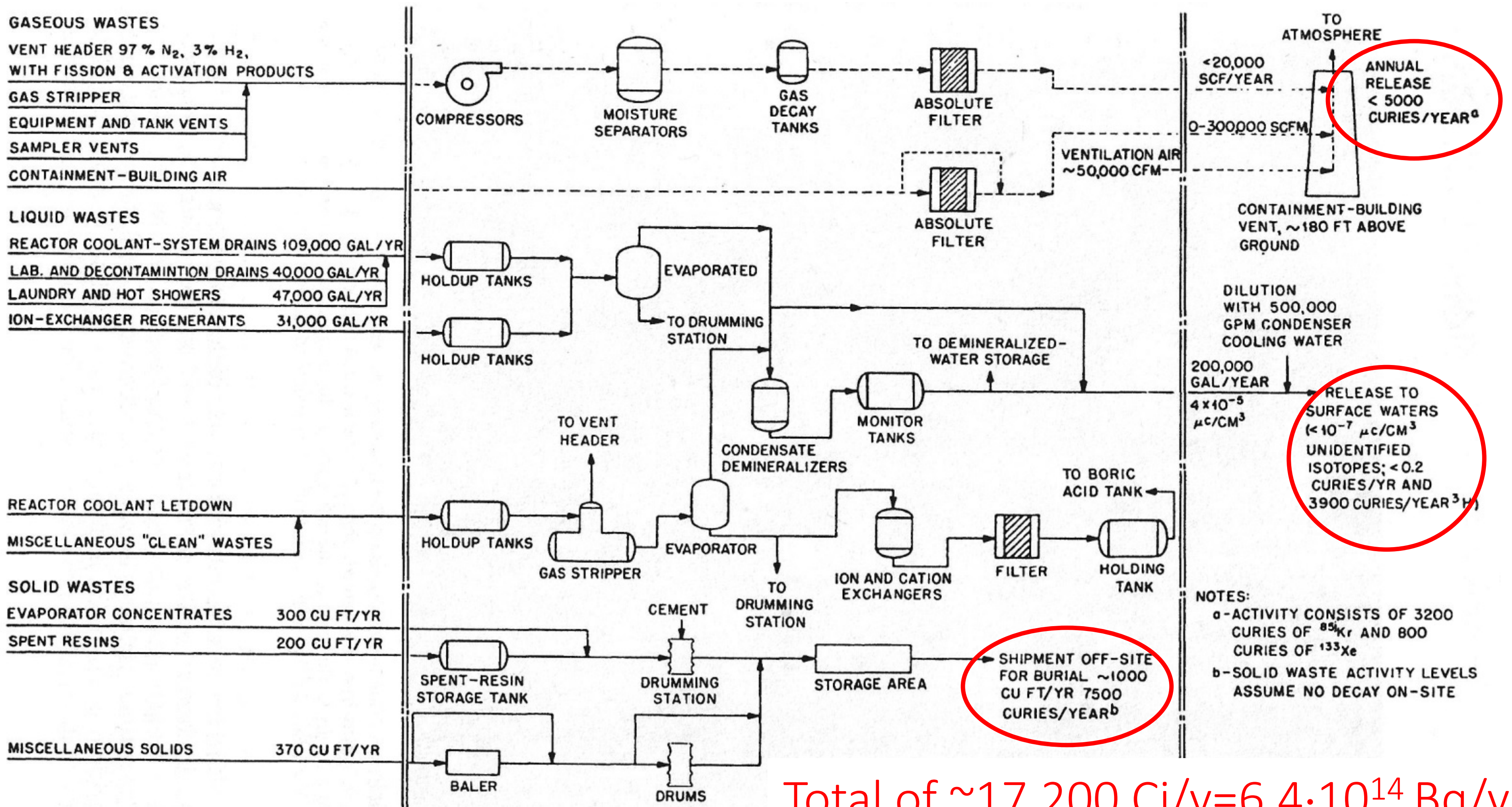
Decay of fission products in fuel rod





Yields at $10^{0,1,2,3}$ years after fission of Pu-239, not considering later neutron capture, fraction of 100% . Beta decay Kr-85→Rb, Sr-90→Zr, Ru-106→Pd, Sb-125→Te, I-129 →Xe, Cs-137→Ba, Ce-144→Nd, Sm-151→Eu, and Eu-155→Gd visible.

Waste management plan for a light water reactor

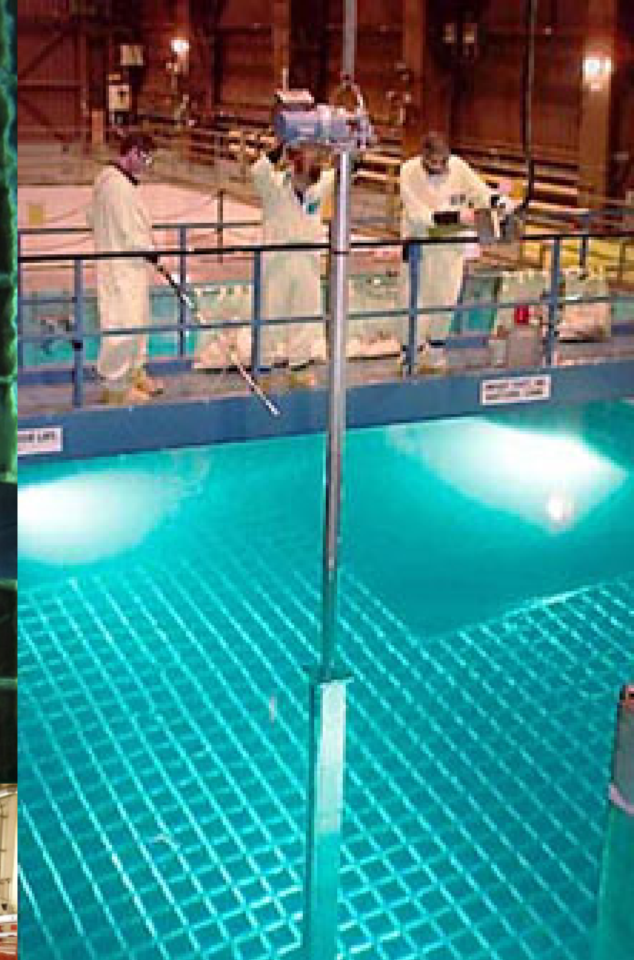


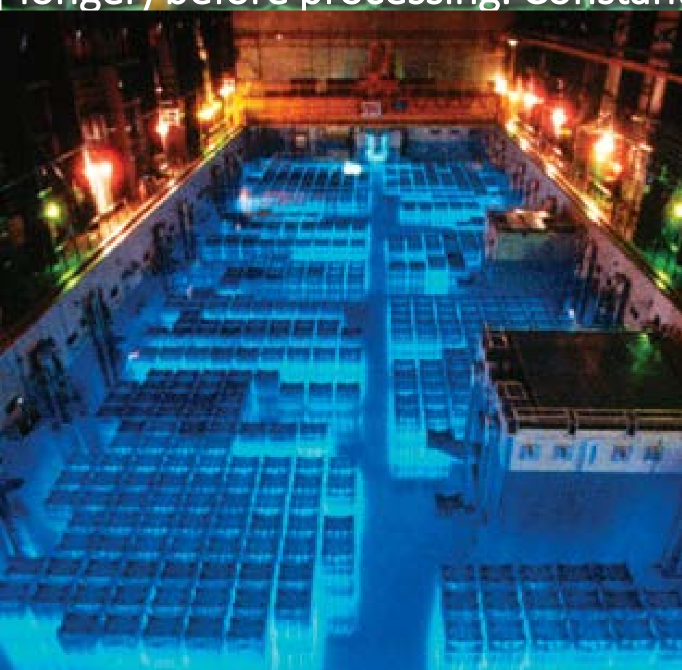
Total of ~17,200 Ci/y=$6.4\cdot10^{14}$ Bq/y

1 Ci=$3.7\cdot10^{10}$ Bq

Intermediary Storage in Spend Fuel Pool

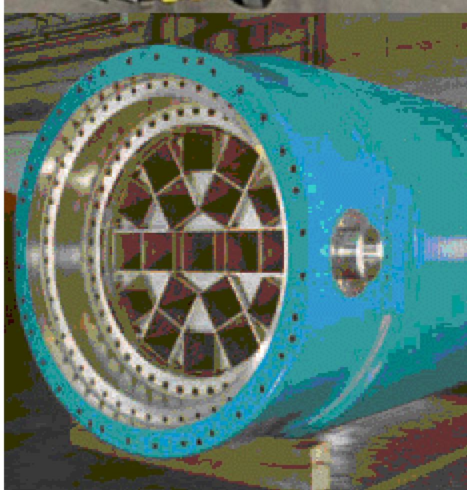
The rods are removed from reactor after the efficiency declines, due to decline in Uranium content and enrichment in neutron poisons, long-lived radioactive fission products ^{90}Se, ^{137}Cs, and non fissile radioactivity ^{241}Am. About a quarter to a third of the total fuel load of a reactor is removed from the core every 12 to 24 months and replaced with fresh fuel (maximum use 4 years). The material is stored in the Spend Fuel Pool SFP for a year (or longer) before processing. Constant cooling must remain assured!





Transport and Storage of spent fuel rods

Transport in a Castor-Container ("cask for storage and transport of radioactive material") to a re-processing plant



The Castor V/19

Reprocessing of spent fuel rods

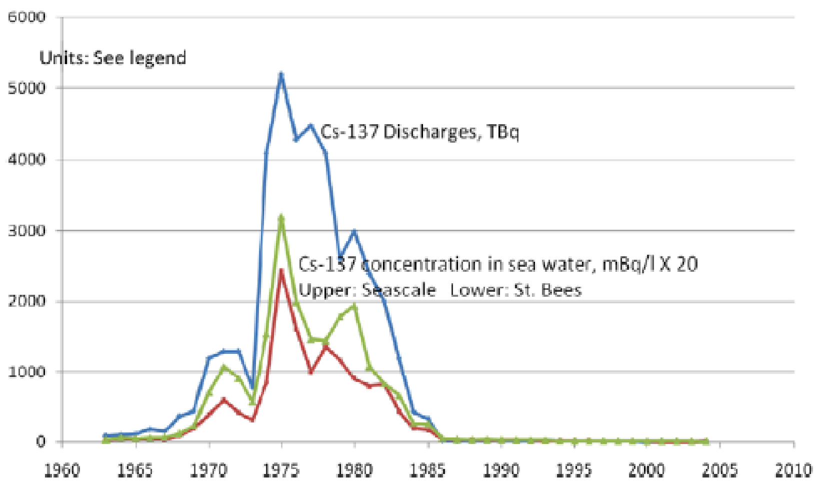
NRC statement: Reprocessing refers generally to the processes necessary to separate spent nuclear reactor fuel into material that may be recycled for use in new fuel and material that would be discarded as waste. There are no reprocessing facilities currently operating in the United States, but there are facilities operating in foreign countries. There exist only two reprocessing plants worldwide, La Hague near Cherbourg in France and Sellafield in the UK.


La Hague in France

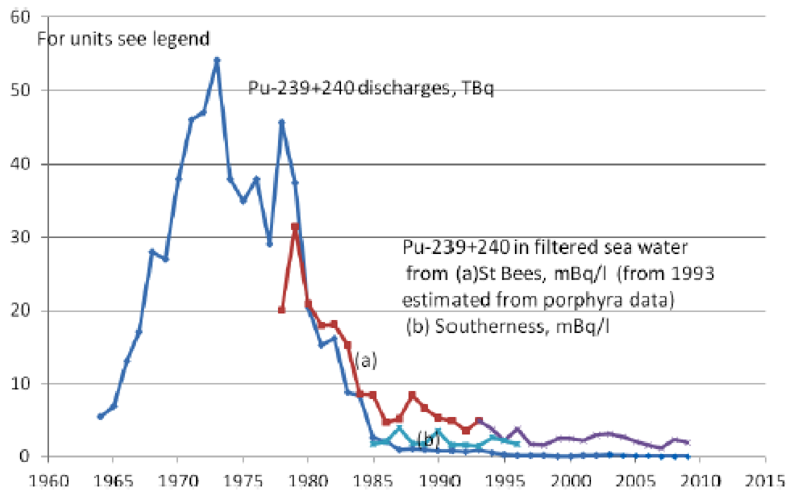

Sellafield in the UK

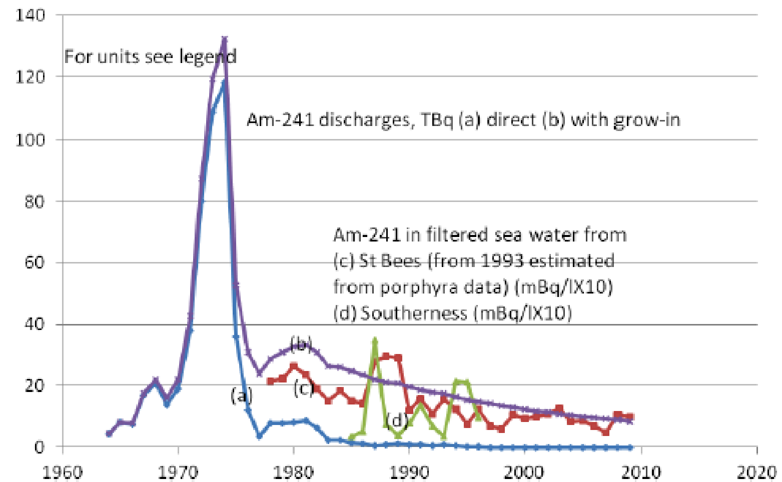
In reprocessing plants, the fuel rods are being disassembled, the pellets are cleaned and the remaining non-fissile radioactivity chemically removed and replaced with fresh Uranium-fluoride. A fuel rod can be reprocessed only one or two times. For that reason, cost arguments and political considerations most countries send their spent fuel rods directly to long term storage facilities.

Radiation Impact



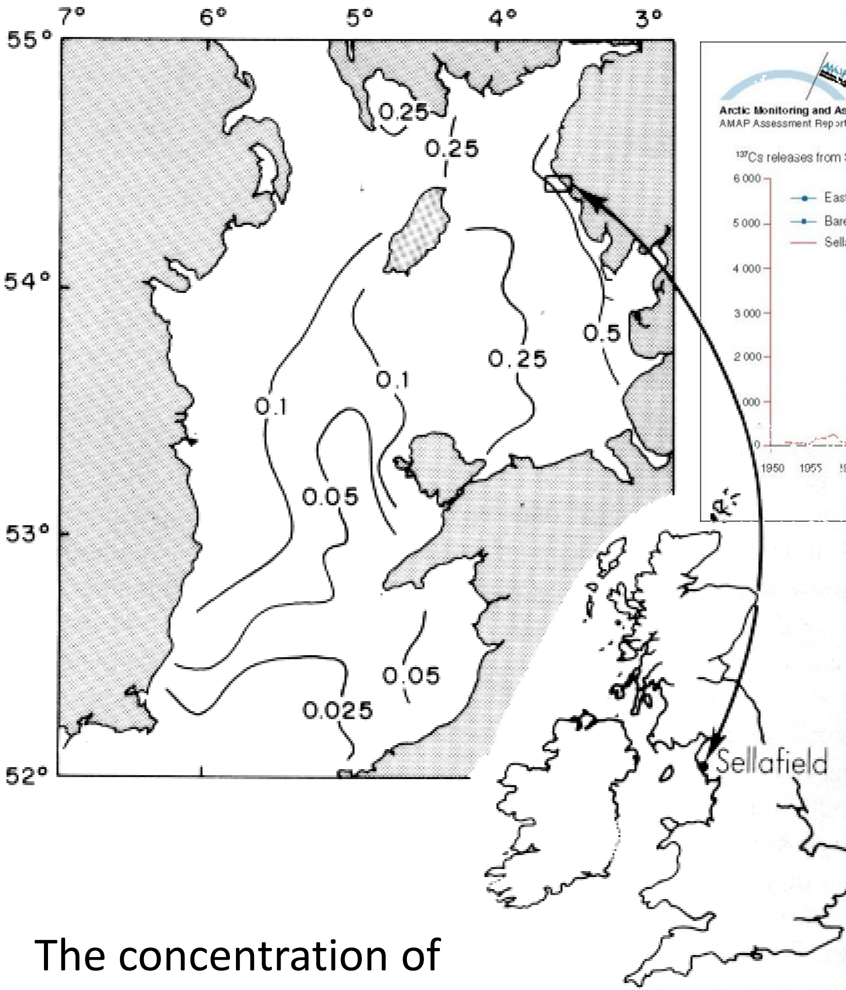
Low-level radioactive waste has been discharged into the Irish Sea as part of operations at Sellafield since 1952. The rate of discharge began to accelerate in the mid- to late 1960s, reaching a peak in the 1970s and generally declining significantly since then. As an example of this profile, discharges of ^{137}Cs as fission fragment and ^{239,241,241}Pu as fuel product peaked in 1973 at 2,755 TBq falling to 8.1 TBq by 2004.

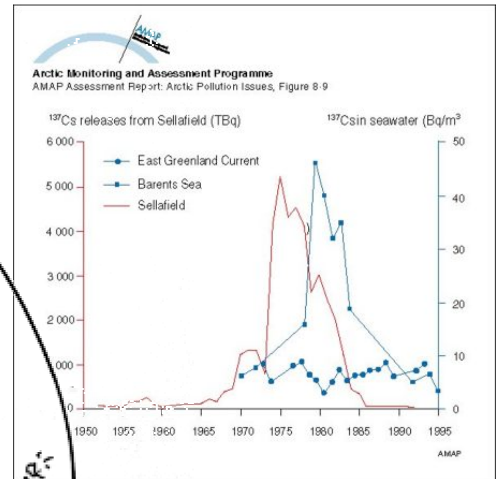


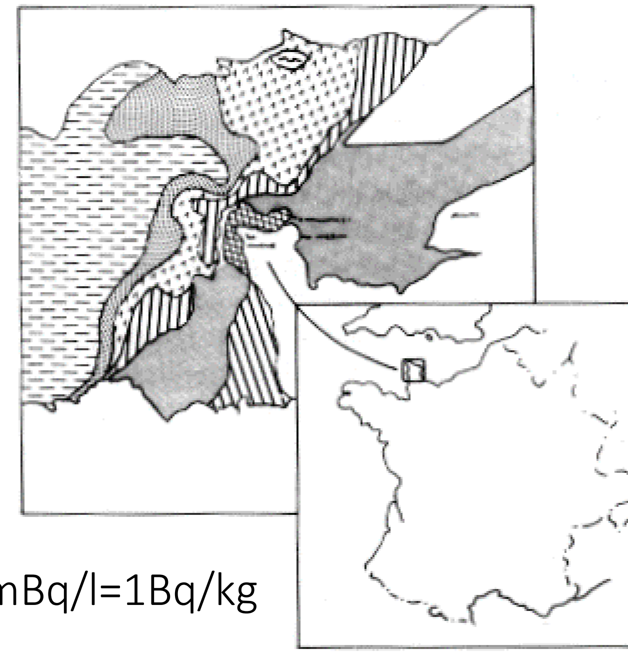


^{241}Pu $T_{1/2}$=14.35 y β-decays to ^{241}Am $T_{1/2}$=432.2 y α decays to ^{237}Np $T_{1/2}=2.14\cdot10^6$ y α decays to ^{233}Pa …

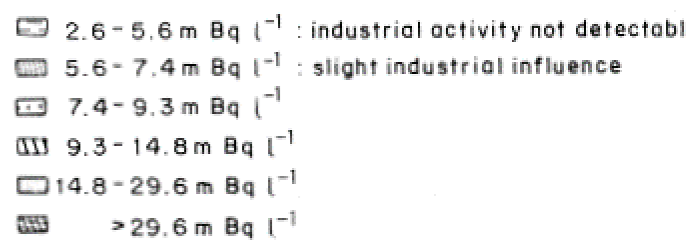
Water contamination from reprocessing





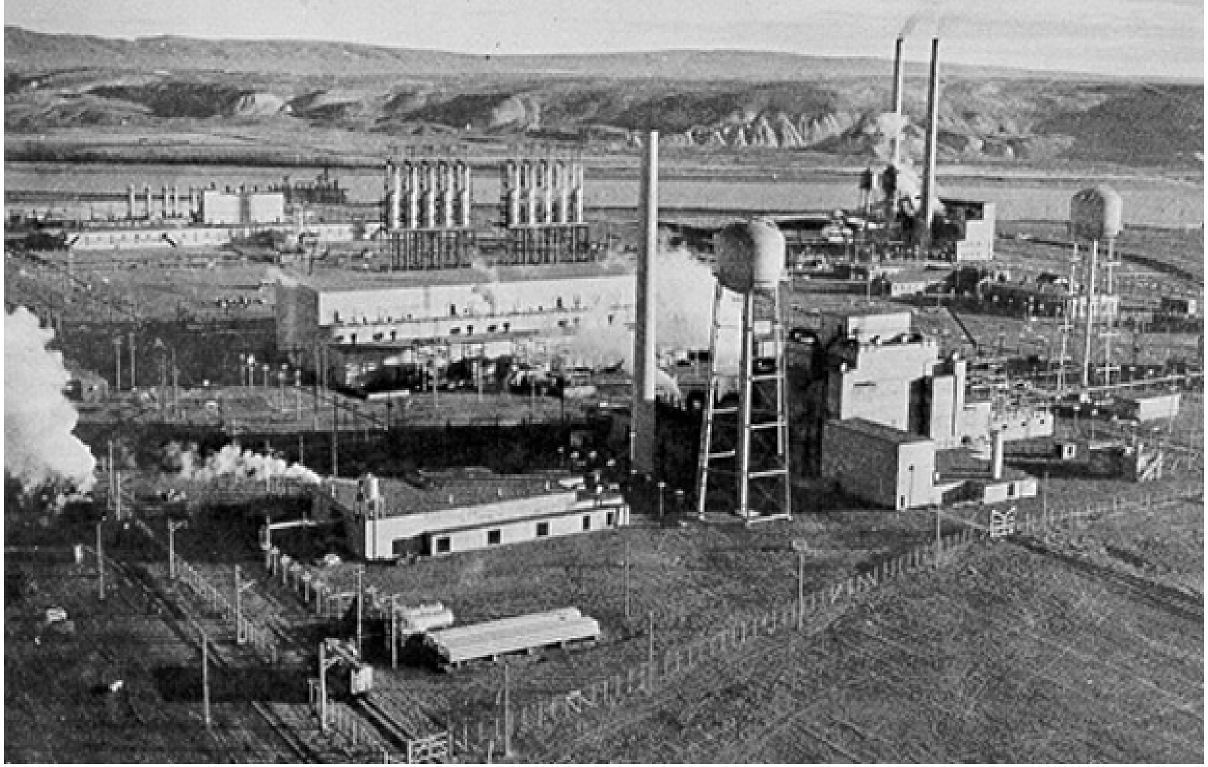


1mBq/l=1Bq/kg



The concentration of ^{137}Cs (Bq kg^{-1}) in filtered water from the Irish Sea, April 1987

The distribution of ^{137}Cs in the locality of La Hague in 1983



Hanford

Constructed in 1943 as follow up on X-10 in Oak Ridge as main site for industrial plutonium production shut-down in 1963! Represents a major nuclear waste problem!

Secret City on the Columbia River in Washington State.

- A series of 9 nuclear reactors were designed to produce plutonium.
- A chemical plant to process material and purify plutonium
- Storage site for the resulting nuclear waste

Nuclear Waste Treatment at Hanford

Solid waste:
burial grounds

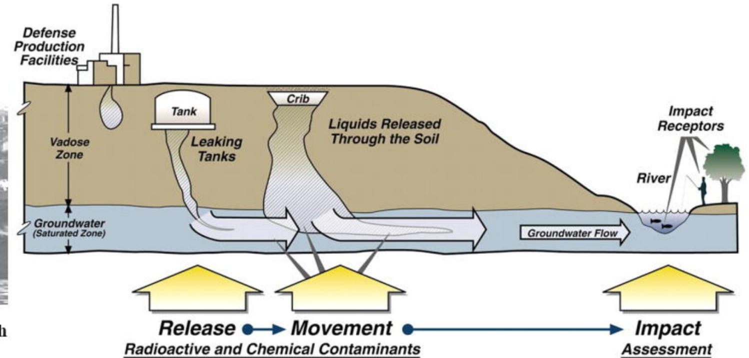
Liquid waste:
retention basins,
reverse wells,
underground tanks
Columbia river

Gaseous waste:
($^{14}N(n,p)^{14}C$
toxic fumes)
ventilation and
exhaust into the
atmosphere

The 618-10 Burial Ground… *"consisted of trenches and rows of burial caissons known as "pipe fields." The caissons were made of 5 to 6 open-bottomed 55-gallon drums welded together and buried upright. From the mid-1950s until about 1960, solid radioactive wastes were collected from operations buildings in cardboard containers and then stored in lead pans known as "gunk catchers" and transported to 300 North [618-10] in shielded "load luggers." The cardboard waste containers then were dropped from the gunk catchers down the caissons, and the holes were filled with sand and dirt until radiation levels declined to a safe or "tolerance" reading. If radiation levels could not be reduced to tolerance ranges, concrete was poured down the hole until such levels were achieved."* - Gerber 1993a, p. 59

Water Contamination at Hanford

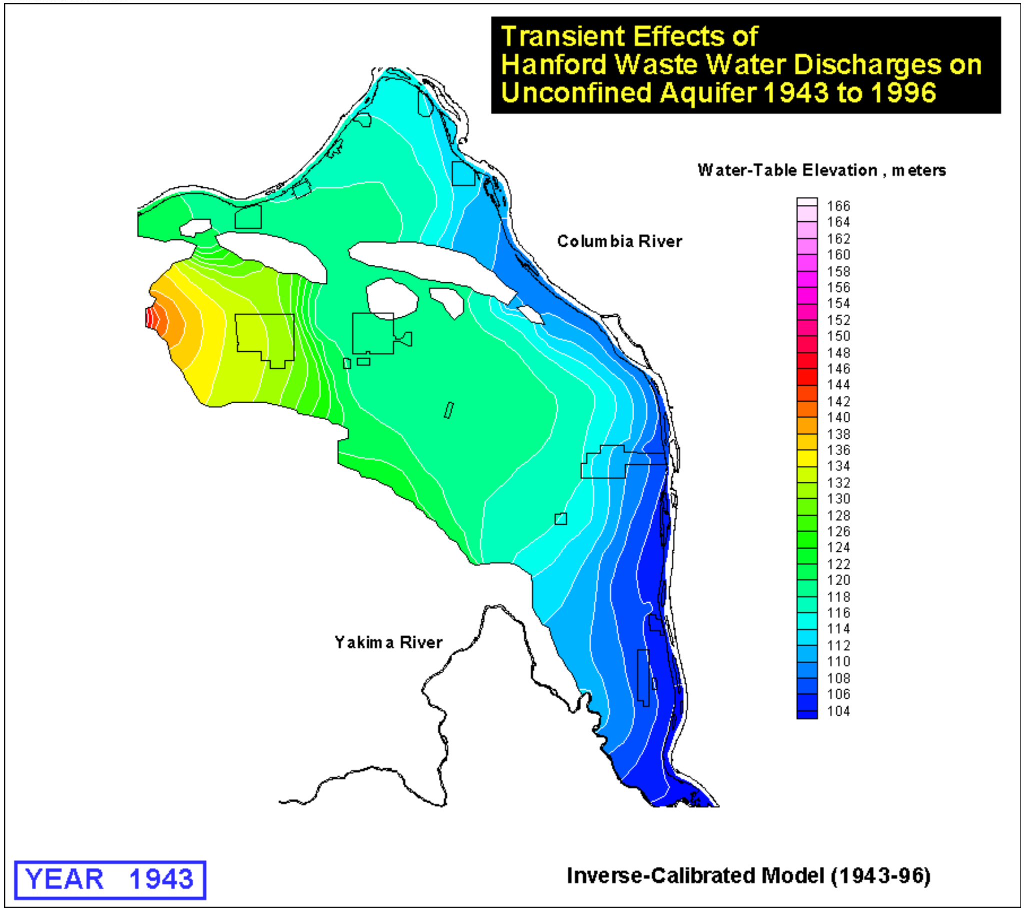
The Hanford Reservation which produced plutonium for decades, is situated along 50 miles of Columbia River. Both upstream and down, the Columbia's watershed has been contaminated with Hanford's deadly fission products.



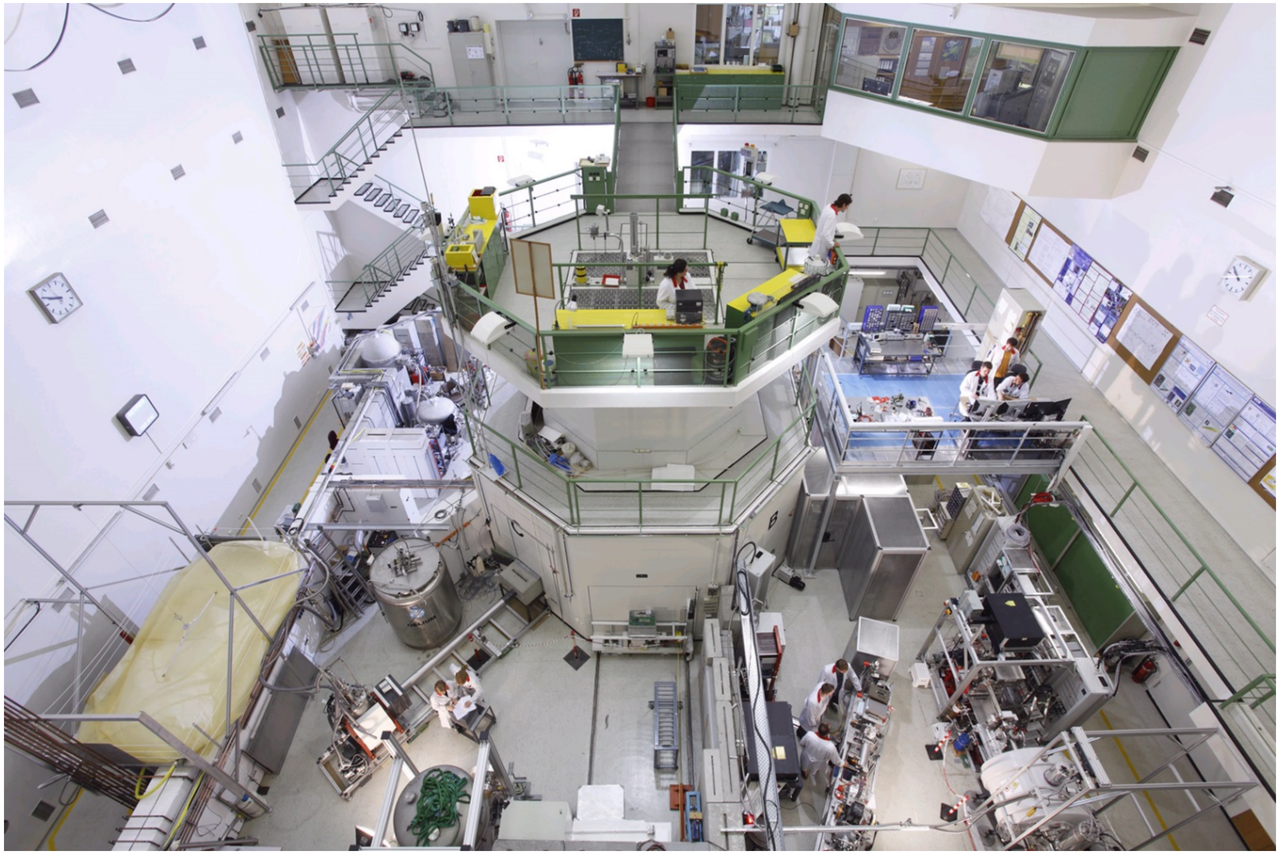
Estimates of Radiation released into the Columbia River by Hanford, 1944-1971

Radionuclide	Amount Released (curies)[11]	half-life
Sodium-24	13,000,000	15 hours
Phosphorus-32	230,000	14 days
Scandium-46	120,000	84 days
Chromium-51	7,200,000	28 days
Manganese-56	80,000,000	
Zinc-65	490,000	245 days
Gallium-72	3,700,000	14 hours
Arsenic-76	2,500,00	26 hours
Yttrium-90	450,000	64 hours
Iodine-131	48,000	8 days
Neptunium-239	6,300,000	2.4 day

114 GCi=$4.22 \cdot 10^{18}$ Bq=4.22 ExaBq



Radiation Exposure

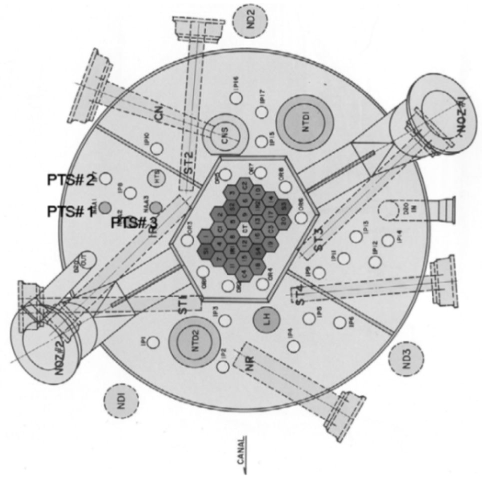


Plutonium-239 contamination in workforce of NUKEM in business for the design of spherical fuel elements for the pebble bed modular reactor project in South Africa.

P B M R

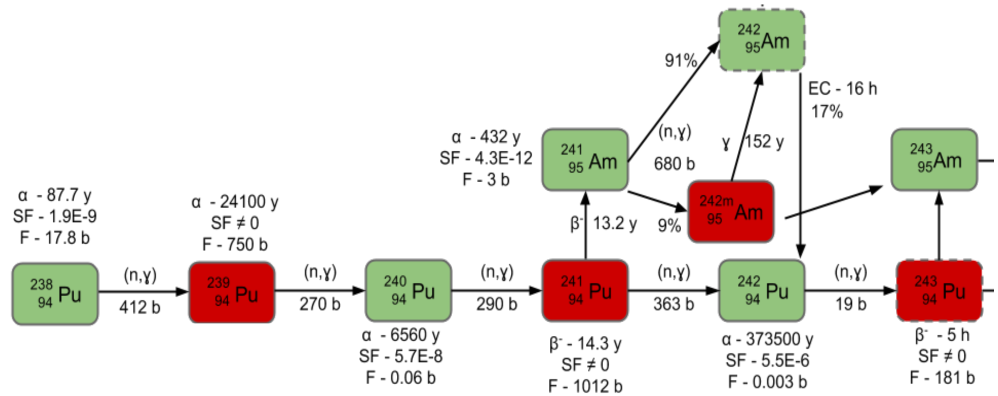
Urine sample analysis for ^{239}Pu contamination; $^{239}\mathrm{Pu}(n,\gamma)^{240}\mathrm{Pu}(\beta^{-}\nu)^{240}\mathrm{Am}(\gamma)$
E_γ = 889 MeV and 986 MeV, easy to detect with Germanium detectors

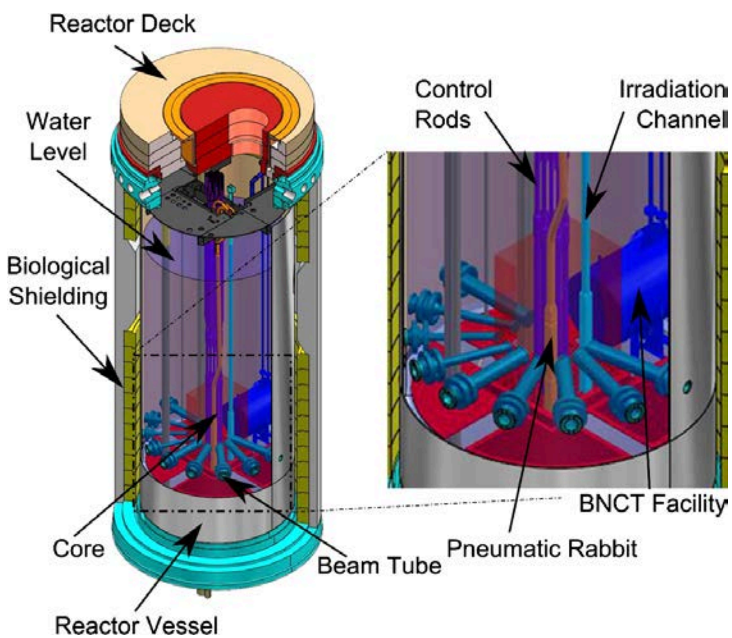
Pneumatic Tube System Activation

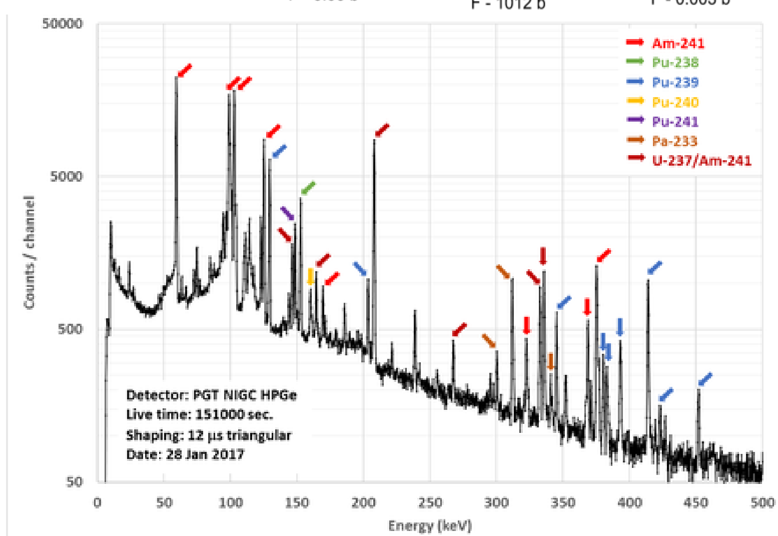




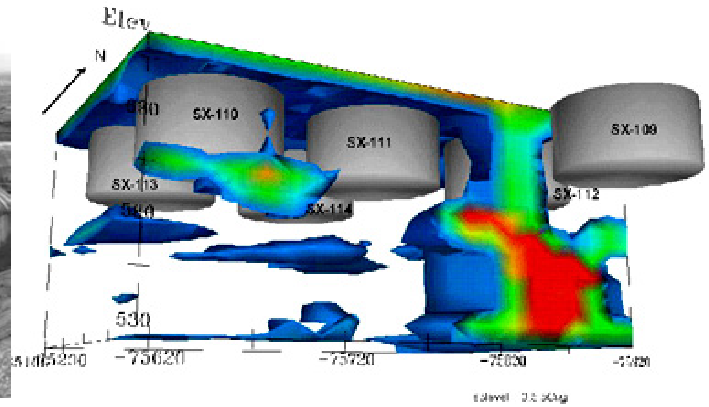
Pneumatic Tube System PTS or rabbit system for fast neutron activation.







Left-overs

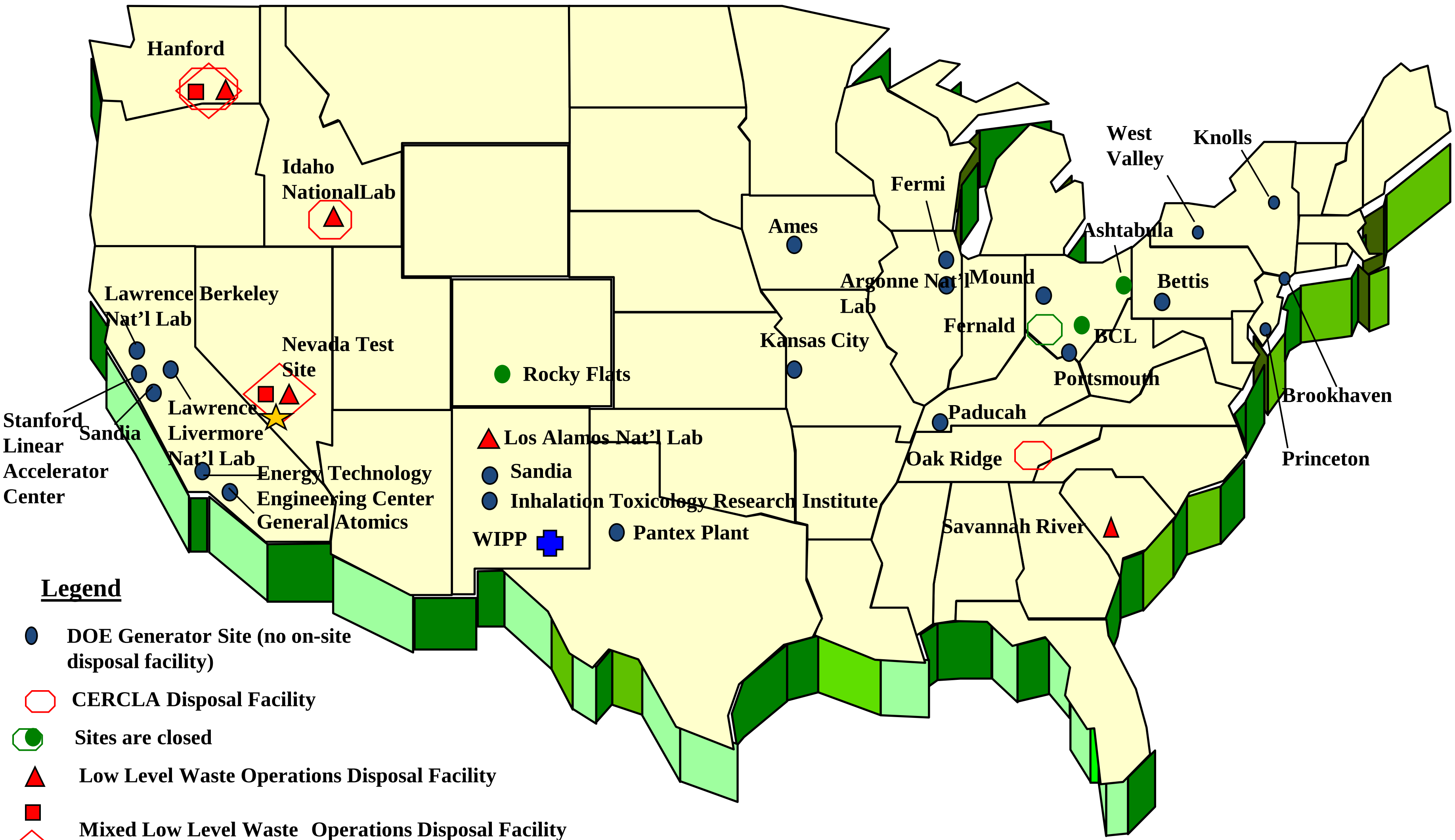


Hanford is arguably the most contaminated site in North America. Cleanup costs are projected in the tens of billions of dollars, and requiring a fifty-year effort.

The Hanford Nuclear Site in southeastern Washington state stores 54 million gallons of dangerous high-level radioactive waste containing hundreds of millions of curies from the nation's nuclear weapons production process.

The issue remains where to put it?

DOE's Waste Disposal Complex



Legend

DOE Generator Site (no on-site disposal facility)

CERCLA Disposal Facility

Sites are closed

Low Level Waste Operations Disposal Facility

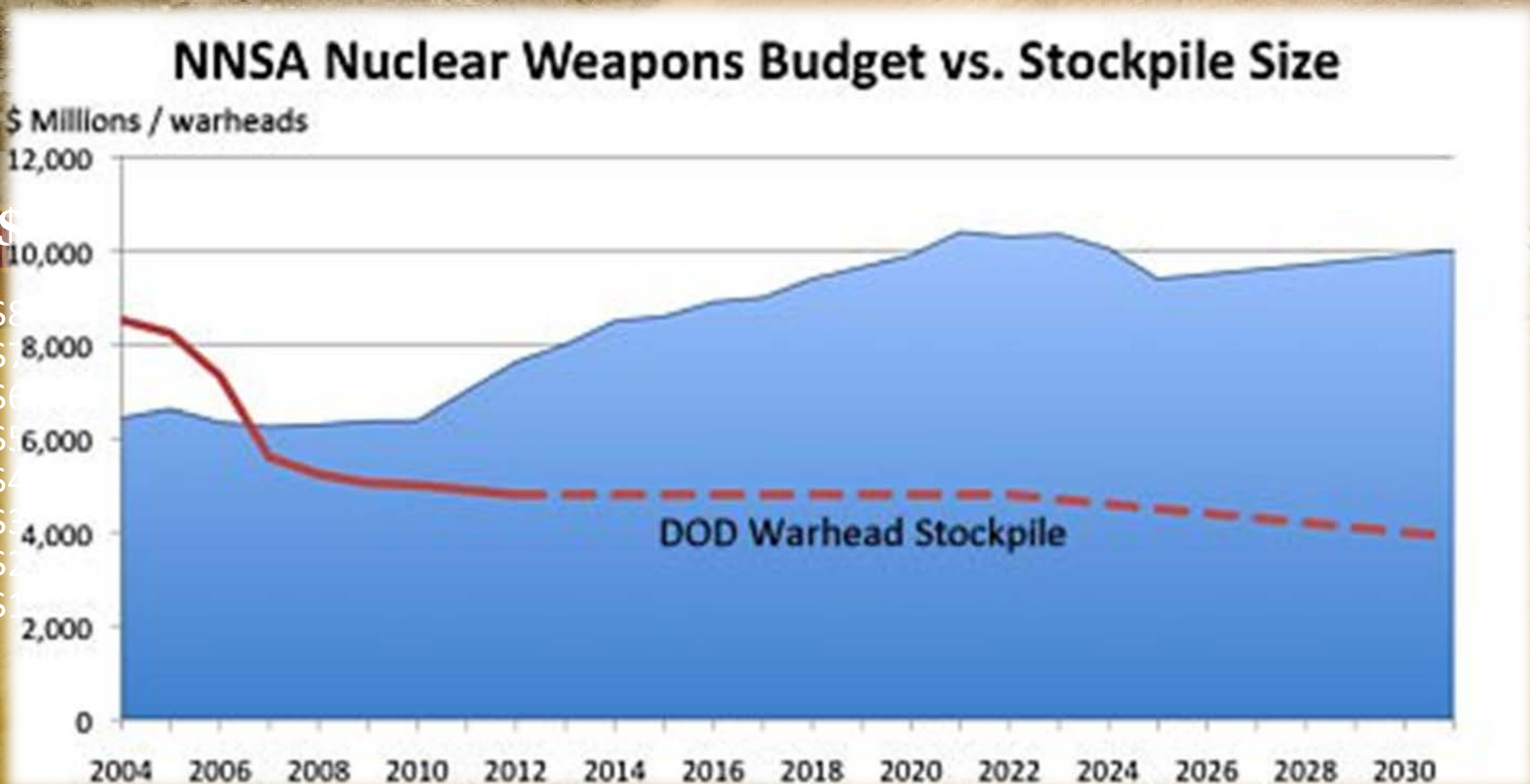
Mixed Low Level Waste Operations Disposal Facility

Regional Low Level Waste Disposal Facility

Yucca Mountain repository for High Level Waste/Spent Nuclear Fuel disposal

Waste Isolation Pilot Plant (WIPP) for Transuranic waste disposal

Stockpile Stewardship Program

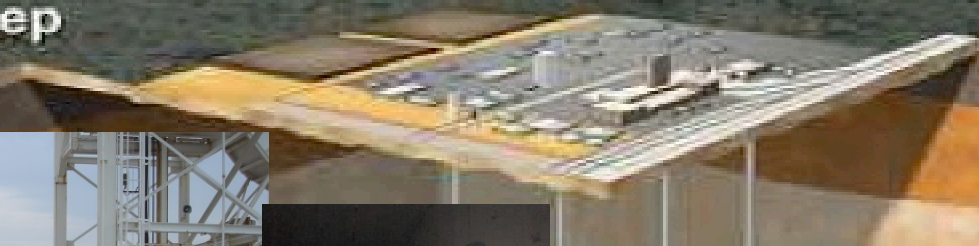


Waste Isolation Pilot Plant

U.S. Department of Energy facility

Designed for permanent disposal of transuranic radioactive waste

2,150 feet deep



Concerns

Barrel Explosion at WIPP
Waste Isolation Pilot Plant
Carlsbad, NM
Feb. 14, 2014

WIPP operations interrupted for at least three years

Hundreds of experiments run to simulate chemical reaction

Over 200-page explanation report assembled by chemical experts

About $ 0.5 billion spent in clean-up

Reopening planned for January 2017

Storage in ultra-pure salt mine environment with floating salt encapsulating the radioactive waste material within 70 years.

Explosion and Radioactivity Release inside WIPP

WIPP Barrel Explosion Parameters
January 10, 2015

The fundamental principle underlying this analysis was recognized on June 10, 2014, results are based entirely on informed consideration of public information.

Barrel actinide isotopic components	^{239}Pu	54%
	^{240}Pu	30%
	^{241}Am	16%
Barrel actinide loading	^{241}Am	38 grams
	^{239}Pu	129 grams
	^{240}Pu	70 grams
Barrel bursting pressure	48 psig	
Barrel temperature prior to bursting	23 C	
Main gas generated	O_2, H_2, CO	
Initial drum gas (air)	96 liters	
Gas generated by alpha particles	222 liters	
Gas generation rate	2.8 liters per day	
Barrel bursting energy from gas pressure	12 megajoules (6.6 lbs. of H. E.)	
Burned gas explosion yield	3 megajoules	
Subsequent barrel content burn yield after explosion	2260 megajoules	
Total radioactivity release	213 curies (86% ^{241}Am)	

Waste probably from ^{238}Pu production for space power

The role of organic material

A Physicist's Perspective on the Barrel Explosion

$$2C_6H_{10}O_5 + \text{alpha particles} \rightarrow 10H_2 + 3O_2 + 4CO + 8C$$

α Emission from Actinides in the stored material disintegrates plastic material in the containers that in turn leads to hydrogen and oxygen build-up, causing a hydrogen gas explosion. Still under investigation!

+	+
Spark or heat	**Spark or heat**
H_2O gas or liquid	**CO_2 gas**

Long Term Concerns

Waste Disposition Plans Affected by Explosion

The 500,000 55-gallon barrels of byproduct waste now in WIPP

Six tons of W-Pu on plutonium pucks stored mostly at Savannah River

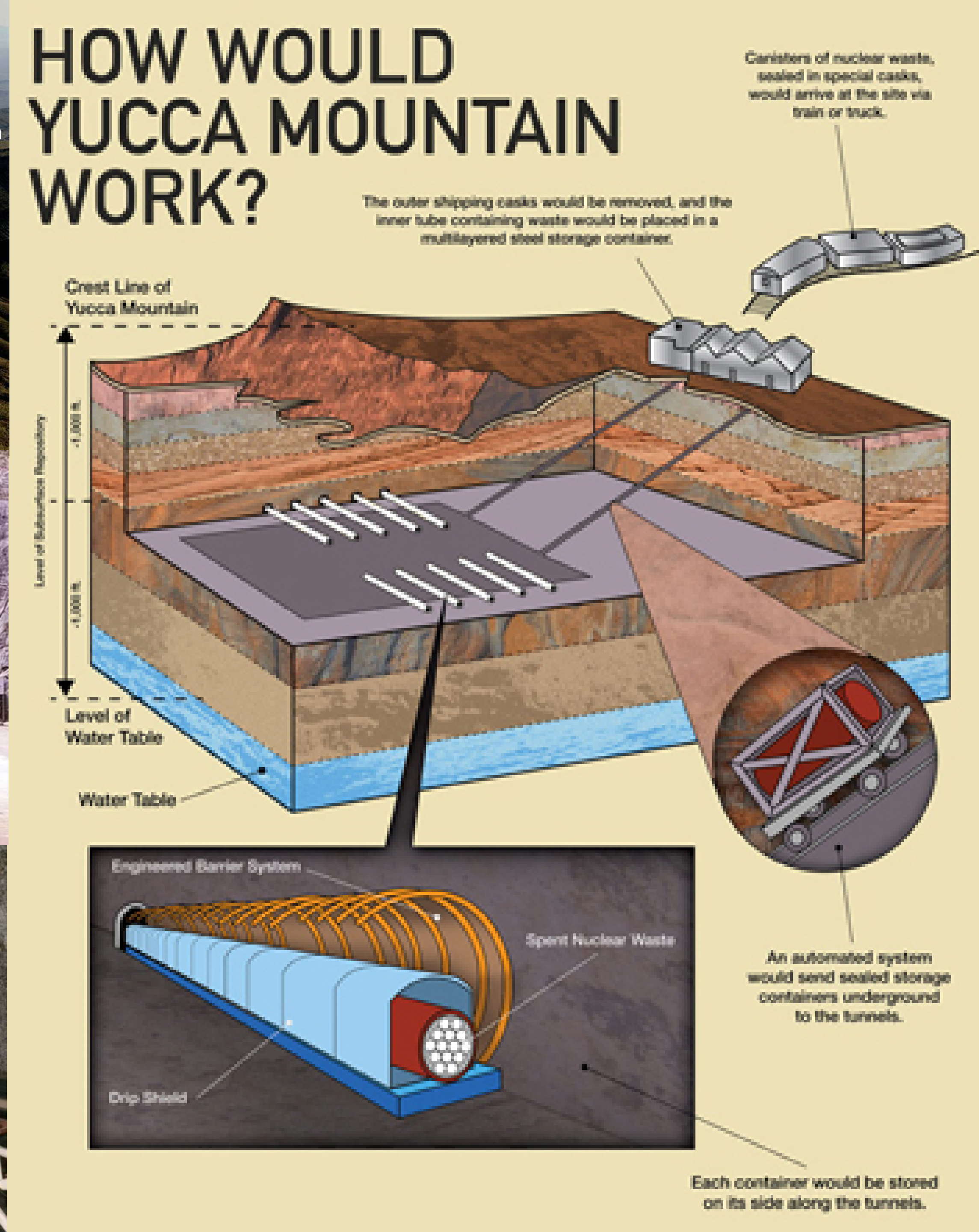
Highly enriched uranium from foreign research reactors temporarily stored at Savannah River Site

Japanese plutonium to be transferred to Savannah River

Additional byproduct waste now at Hanford, Idaho, Los Alamos, Oak Ridge, etc.

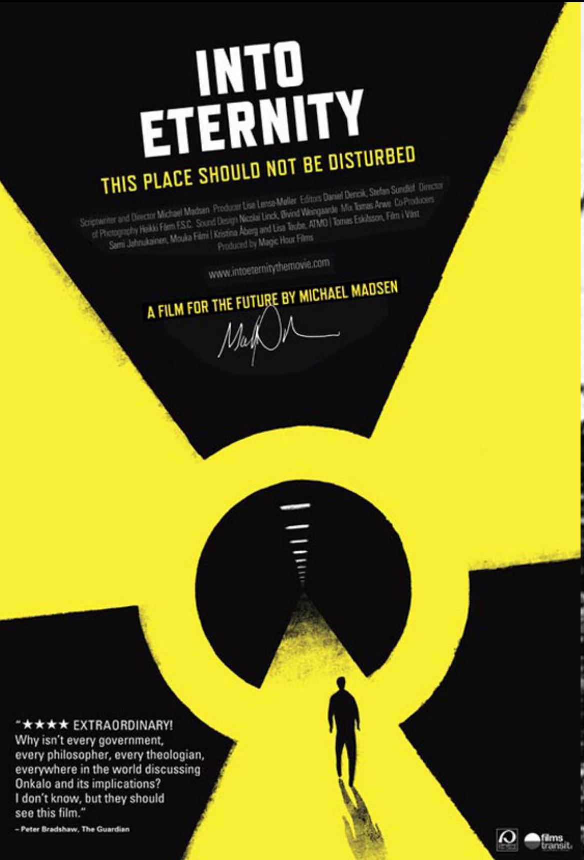
Continued low level operational waste generated in Stockpile maintenance and Replacement

Long-Term Storage



Onkalo – into Eternity

The Great Pyramid of Giza was completed around 4,500 years ago, the transition from nomadic hunter gathering to farming and permanent settlement occurred between 7-10,000 years ago, the last ice age was 20,000 years ago, our Homo sapien ancestors only reached Europe 40,000 years ago, where Neanderthals did not become extinct until 30,000 years ago and the great original Homo sapien migration out of Africa took place between 125-60,000 years ago!

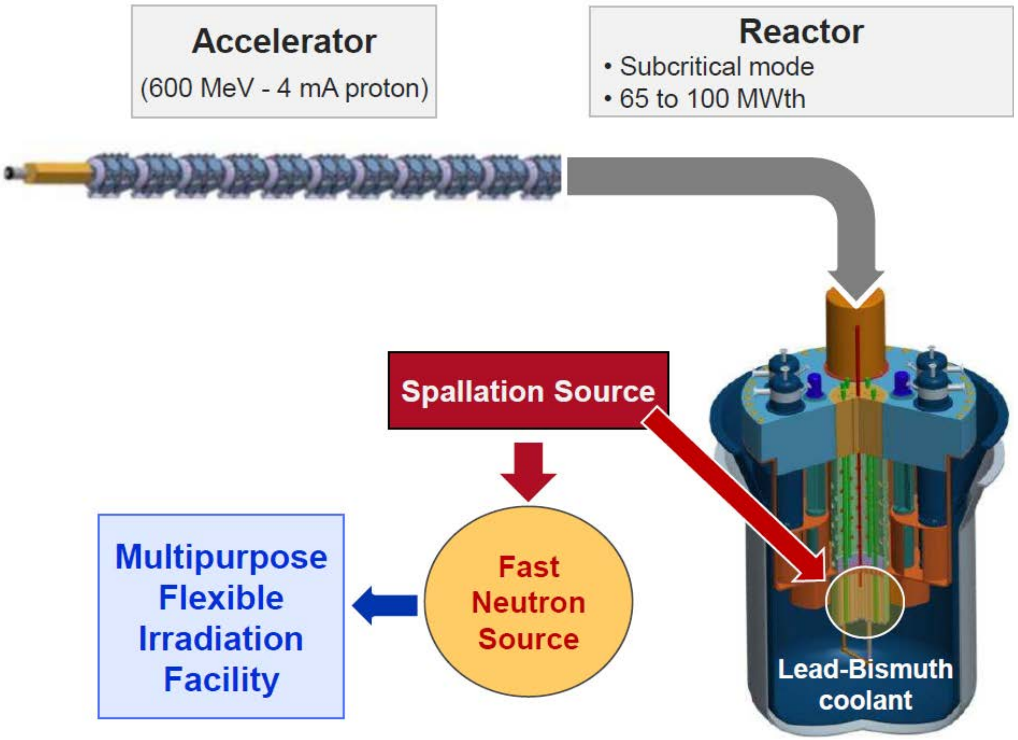


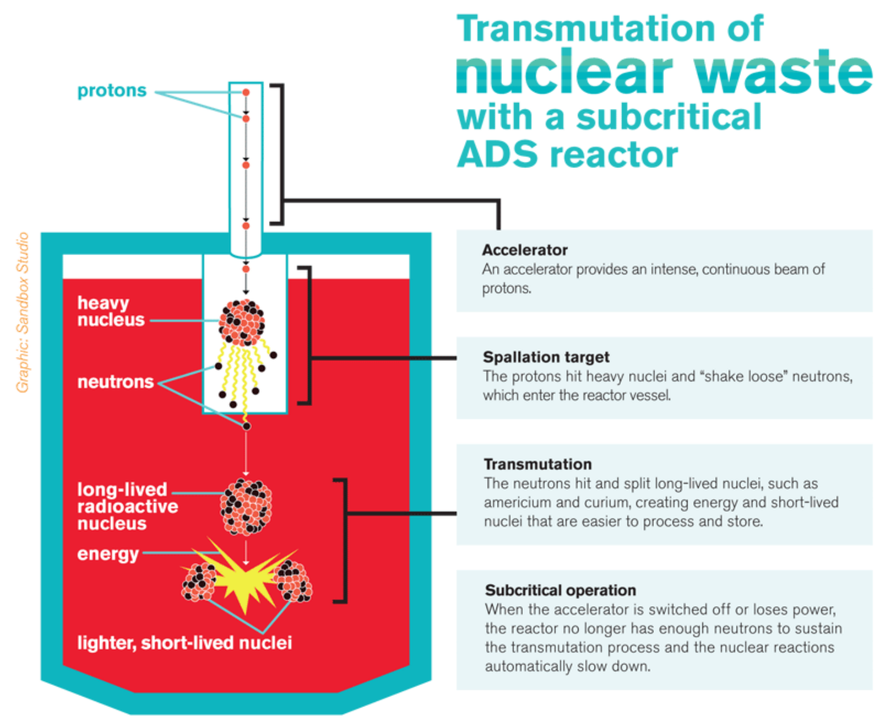
https://www.youtube.com/watch?v=wke01P0rz2Q

Alternative Plans: accelerator driven systems

This is based on the idea of converting long-lived radioactive elements by irradiation with neutron beams or spallation with high energy proton beams into short-lived isotopes and use the resulting decay heat from the short-lived high activity as energy source. Such transmutation concepts are under development at Los Alamos by the US and in Belgium by the European nuclear industry.

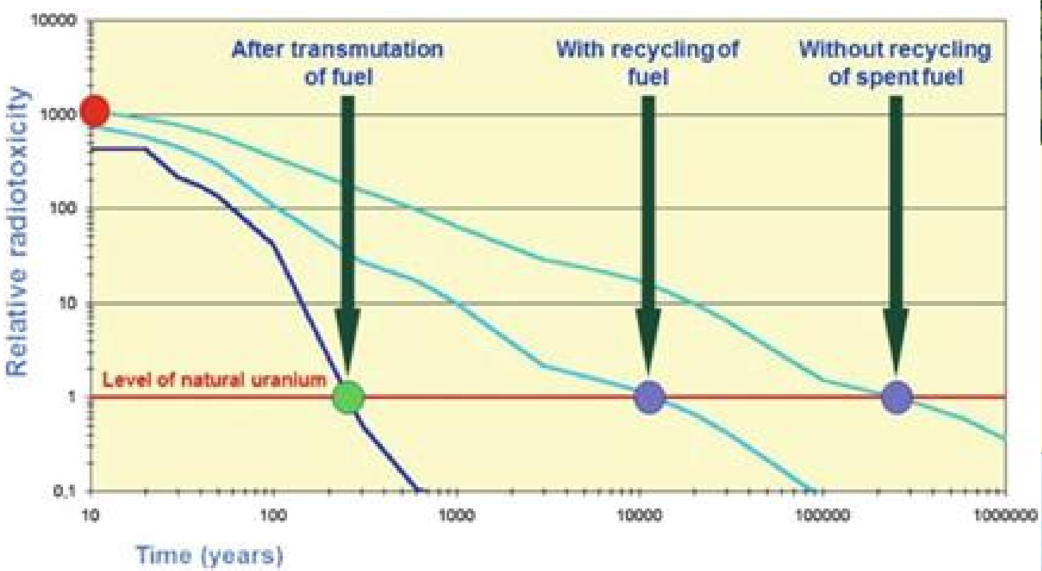
Spallation neutron source for complementing sub-critical reactors and for turning the long-lived radioactive isotopes into short-lived ones.

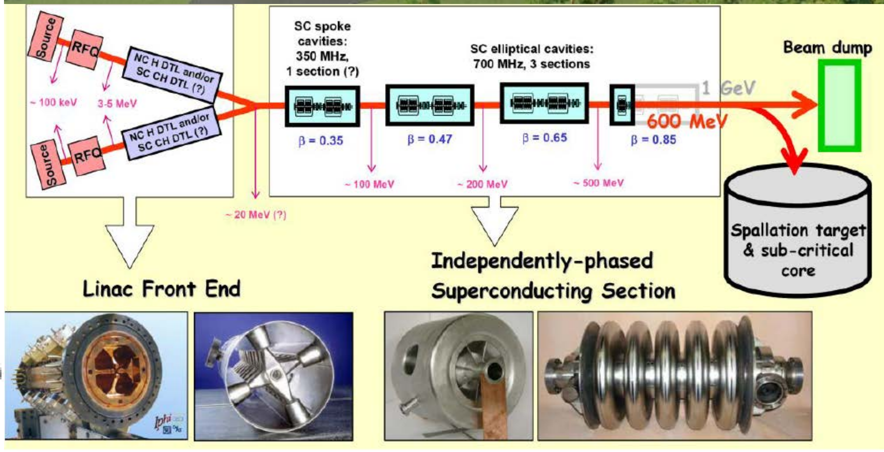




Myrrha, an accelerator driven system for subcritical reactors and transmutation

Myrrha is the mother of Adonis in Greek mythology. She was transformed into a myrrh tree after having had intercourse with her father and gave birth to Adonis. In the nuclear world Myrrha is a European project where neutrons from a spallation source are guided either onto radioactive waste material, to convert the long-lived actinide species by neutron capture into short-lived radioactivity that decays on a much shorter timescale, generating decay heat as an additional benefit.





Contaminated top soil layer from Fukushima