

Radioactivity

Lecture 21

Radioactivity and Long Term Storage

Radioactive Waste

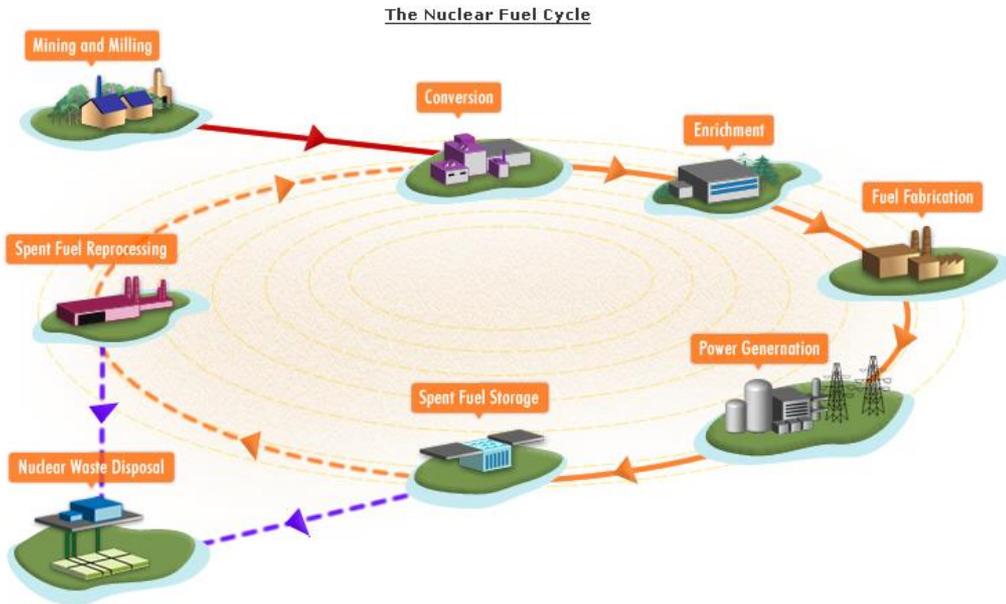
of the Nuclear and other Industries

- 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 reactor, using 4.5% enriched fuel and with 45 GW/t burn-up.



Substantial radioactive foot print

Uranium Mining

Fuel rod production

Reactor operation

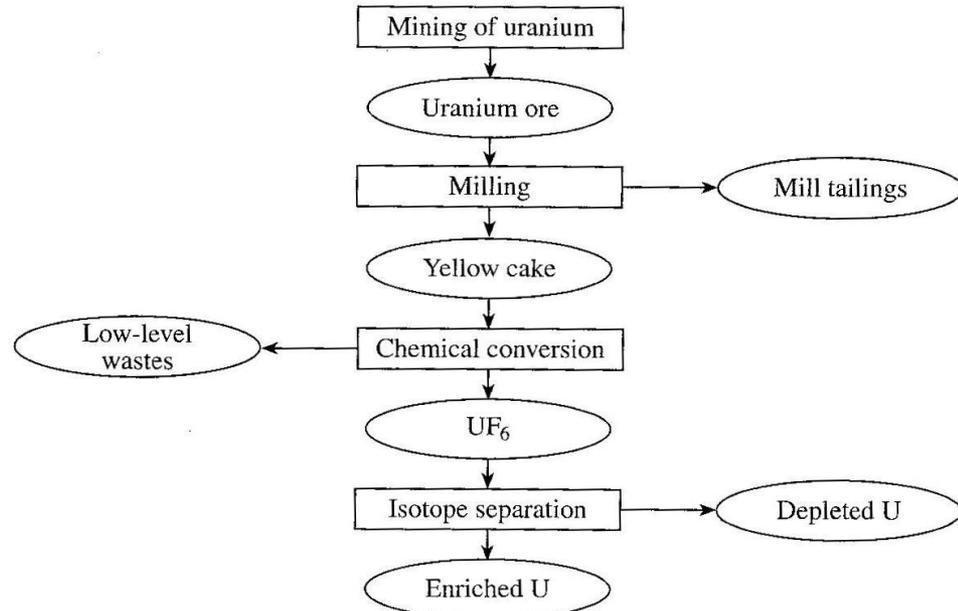
Fuel rod storage

Fuel rod reprocessing

Nuclear waste disposal

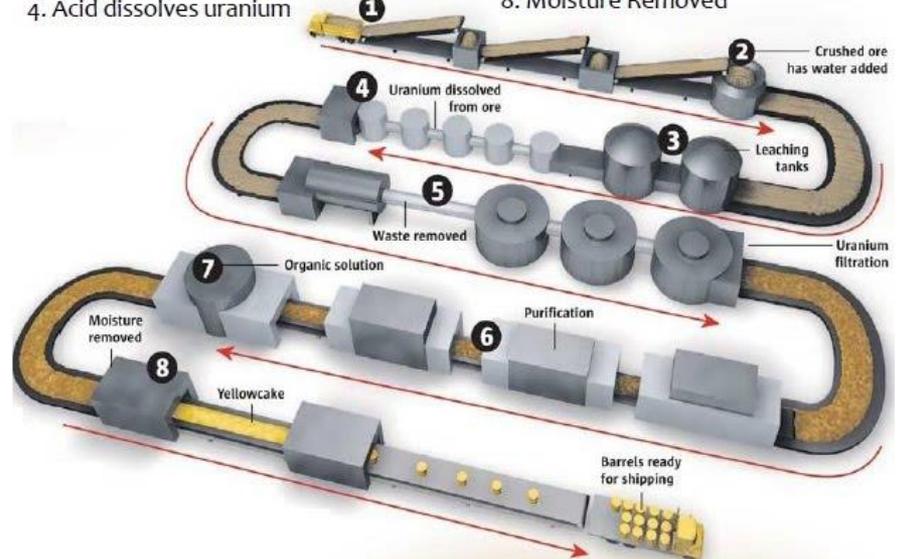
Mining	Anything from 20,000 to 400,000 tons of uranium ore
Milling	249 tons of uranium oxide concentrate (which contains 211 tons of uranium)
Conversion	312 tons of uranium hexafluoride, UF_6 (with 211 tons of Uranium)
Enrichment	35.9 tons of enriched UF_6 (containing 24.3 t enriched U @ 4.5%) – balance 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.

Fuel Cycle Steps



URANIUM MILLING

1. Mined ore is crushed
2. Crushed ore ground into fine sand
3. Slurry pumped into leach tanks
4. Acid dissolves uranium
5. Uranium filtered from waste
6. Purified & Concentrated
7. Uranium extraction
8. Moisture Removed



Uranium Mining

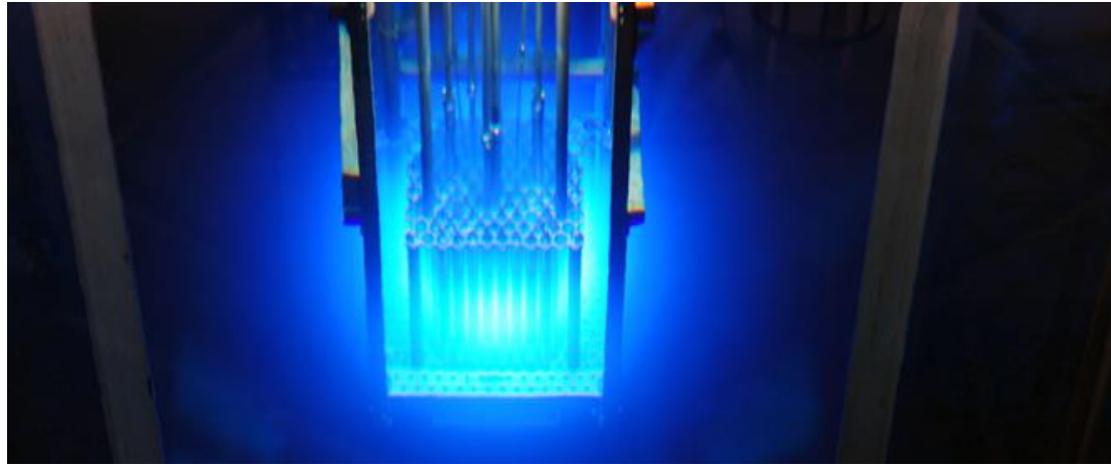
The world's ten largest uranium mines.

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%

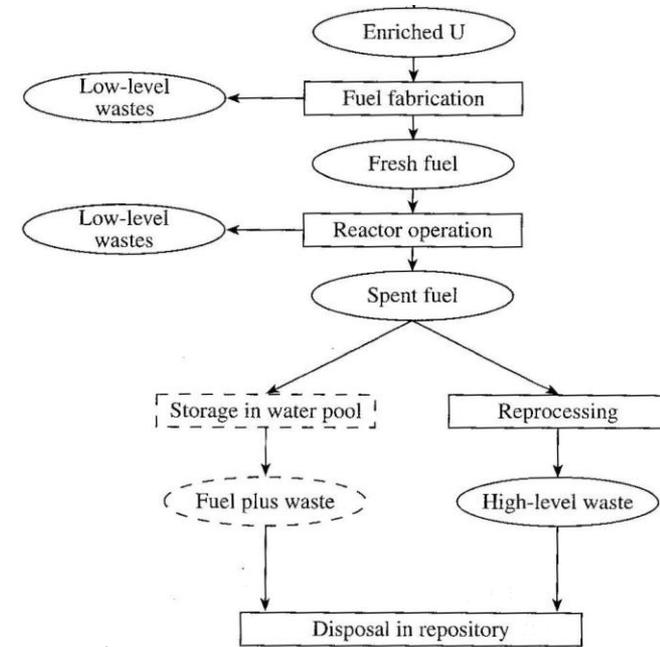
The environmental impact is comparable to what has been discussed in terms of Rare Earth harvesting!
ISL means "In Situ Leach"



Converting fuel to waste

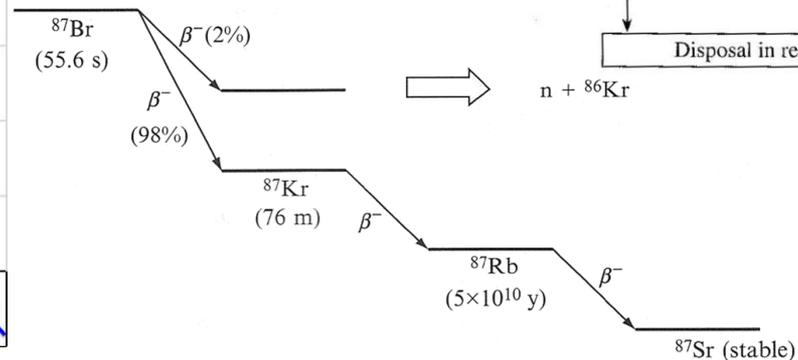


Fission converts ^{235}U into fission product distribution. The neutron-rich fission products decay back to stability. The decay products can be very long-lived! They remain with the unprocessed uranium (of other fission fuel) in the fuel rod contaminating the rod, which has to be reprocessed or stored.



^{92}Mo 14.65	^{93}Mo 3.5E3a	^{94}Mo 9.19	^{95}Mo 15.87	^{96}Mo 16.67	^{97}Mo 9.58	^{98}Mo 24.29	^{99}Mo 2.75d	^{100}Mo 9.72													
	^{92}Nb 3.5E7a	^{93}Nb 100	^{94}Nb 2.0E4a	^{95}Nb 34.99d	^{96}Nb 23.4h	^{97}Nb 1.23h	^{98}Nb 2.2s	^{99}Nb 15.0s	^{100}Nb 1.5s												
		^{92}Zr 17.15	^{93}Zr 1.5E6a	^{94}Zr 17.38	^{95}Zr 64.92d	^{96}Zr 2.8	^{97}Zr 16.8h	^{98}Zr 30.7s	^{99}Zr 2.2s	^{100}Zr 7.3s											
			^{92}Y 3.54h	^{93}Y 10.2h	^{94}Y 18.7m	^{95}Y 10.9m	^{96}Y 5.8s	^{97}Y 3.75s	^{98}Y 0.59s	^{99}Y 1.47s	^{100}Y 0.73s										
				^{92}Sr 2.61h	^{93}Sr 7.41m	^{94}Sr 1.25m	^{95}Sr 257s	^{96}Sr 1.07s	^{97}Sr 0.43s	^{98}Sr 0.65s	^{99}Sr 269ms	^{100}Sr 207ms									

Example ^{87}Br :



Radioactive Fission Products

By yield

Yield	Element	Isotope	Halflife
6.7896%	Caesium	¹³³Cs → ¹³⁴Cs	2.065 y
6.3333%	Iodine, Xenon	¹³⁵I → ¹³⁵Xe	6.57 h
6.2956%	Zirconium	⁹³Zr	1.53 My
6.1%	Molybdenum	⁹⁹Mo	65.94 h
6.0899%	Caesium	¹³⁷Cs	30.17 y
6.0507%	Technetium	⁹⁹Tc	211 ky
5.7518%	Strontium	⁹⁰Sr	28.9 y
2.8336%	Iodine	¹³¹I	8.02 d
2.2713%	Promethium	¹⁴⁷Pm	2.62 y
1.0888%	Samarium	¹⁴⁹Sm	virtually stable
0.9% ^[2]	Iodine	¹²⁹I	15.7 My
0.4203%	Samarium	¹⁵¹Sm	90 y
0.3912%	Ruthenium	¹⁰⁶Ru	373.6 d
0.2717%	Krypton	⁸⁵Kr	10.78 y
0.1629%	Palladium	¹⁰⁷Pd	6.5 My
0.0508%	Selenium	⁷⁹Se	327 ky
0.0330%	Europium, Gadolinium	¹⁵⁵Eu → ¹⁵⁵Gd	4.76 y
0.0297%	Antimony	¹²⁵Sb	2.76 y
0.0236%	Tin	¹²⁶Sn	230 ky
0.0065%	Gadolinium	¹⁵⁷Gd	stable
0.0003%	Cadmium	^{113m}Cd	14.1 y

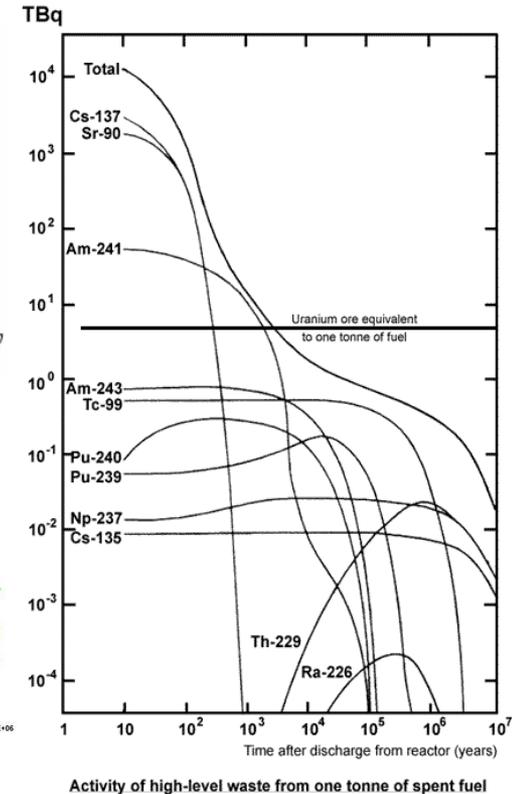
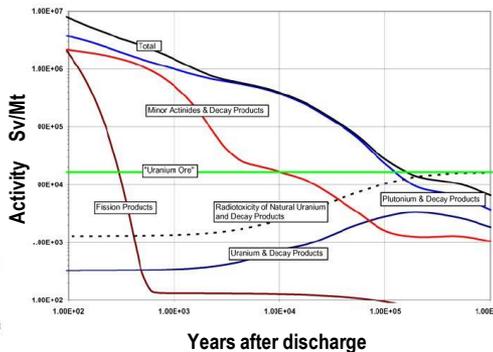
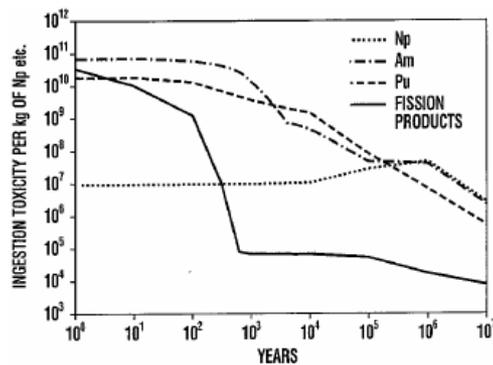
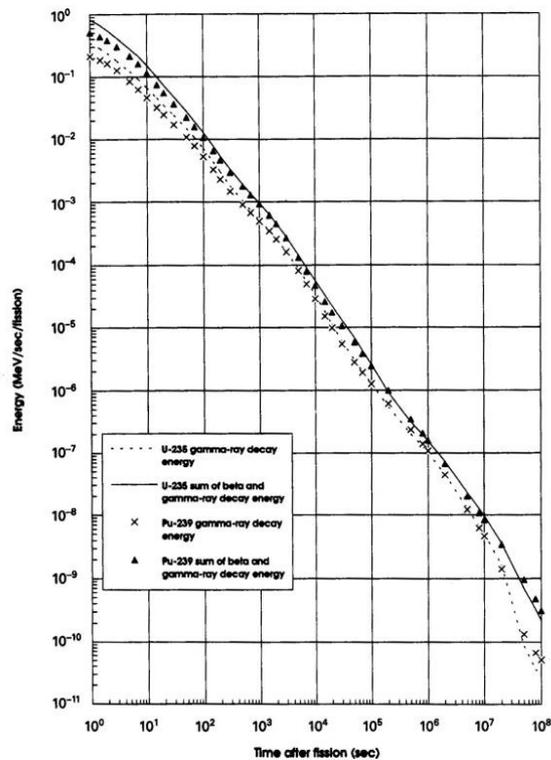
By half-life

Yield	Isotope	Halflife
2.8336%	¹³¹I	8.02d
0.3912%	¹⁰⁶Ru	373.6d
6.7896%	¹³³Cs → ¹³⁴Cs	2.065y
2.2713%	¹⁴⁷Pm	2.62y
0.0297%	¹²⁵Sb	2.76y
<0.0330%	¹⁵⁵Eu → ¹⁵⁵Gd	4.76y
0.2717%	⁸⁵Kr	10.78y
<0.0003%	^{113m}Cd	14.1y
5.7518%	⁹⁰Sr	28.9y
6.0899%	¹³⁷Cs	30.17y
<0.4203%	¹⁵¹Sm	90y
6.0507%	⁹⁹Tc	211ky
0.0236%	¹²⁶Sn	230ky
0.0508%	⁷⁹Se	327ky
6.2956%	⁹³Zr	1.53My
<6.3333%	¹³⁵Cs	2.3My
0.1629%	¹⁰⁷Pd	6.5My
0.6576%	¹²⁹I	15.7My
<1.0888%	¹⁴⁹Sm	nonradioactive
<0.0065%	¹⁵⁷Gd	nonradioactive

Decay of Radioactivity

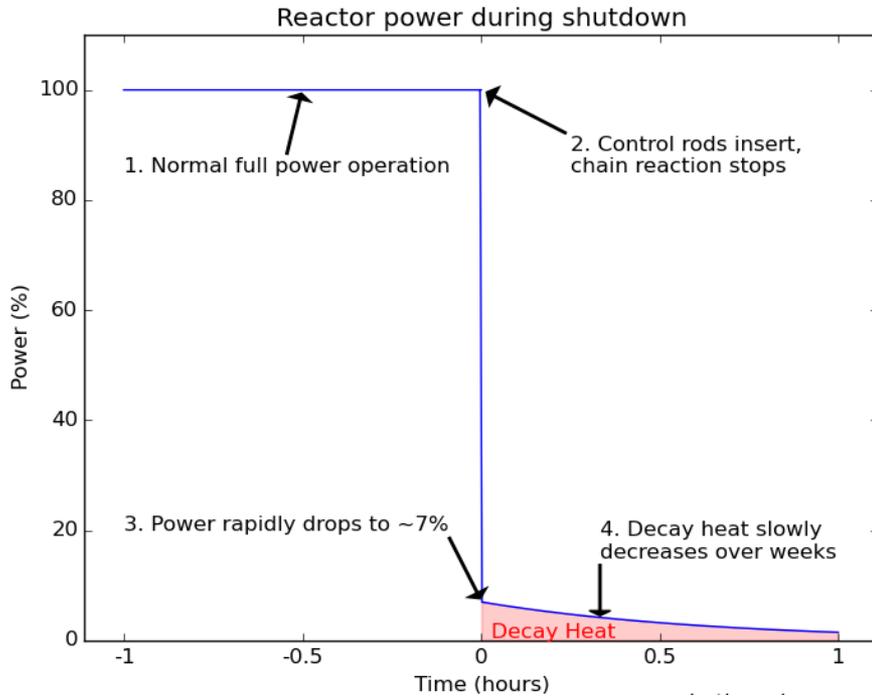
Development of activity is dominated by the radioactive isotope with the largest decay constant or shortest life time at each moment of time.

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

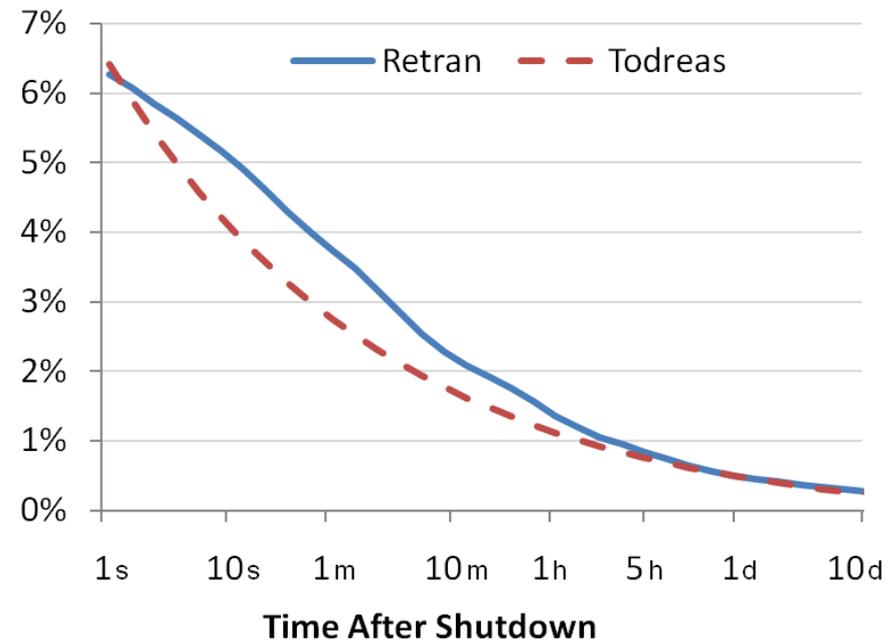


The initial activity in fuel rods is initially extremely high, short-lived radioactivity decreases by nine orders of magnitude during the first year. Material needs to be stored for that time before further processed. It takes another thousand years until activity reaches natural Uranium levels.

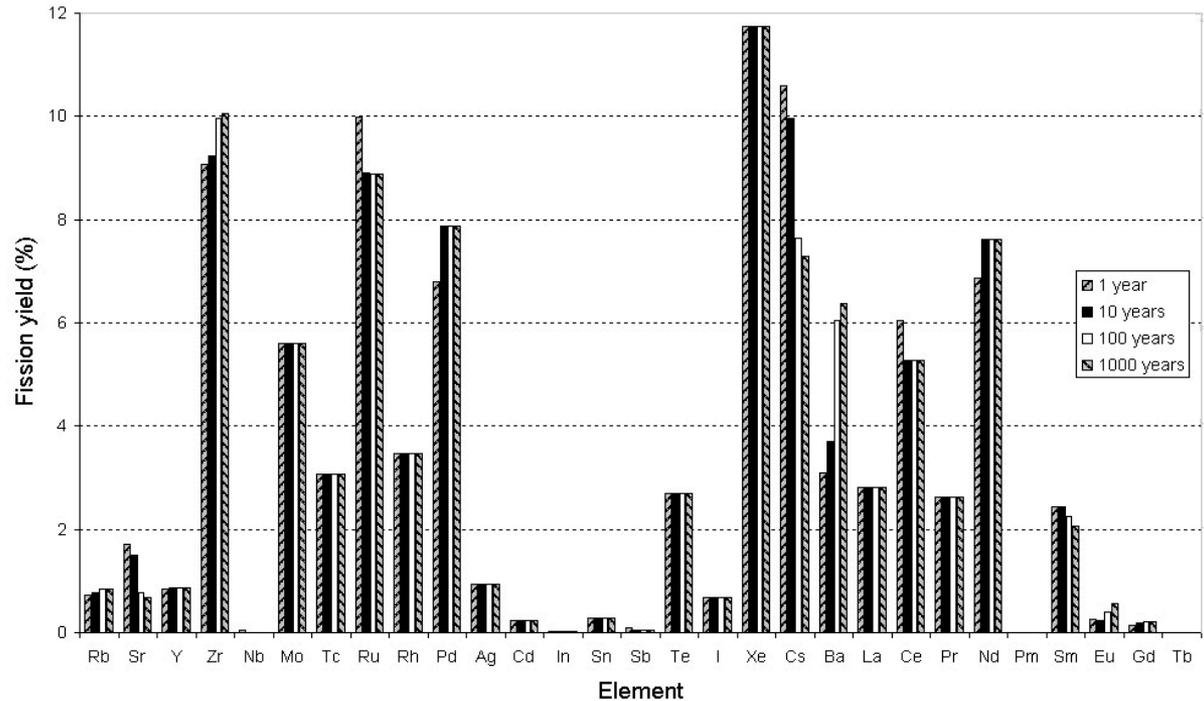
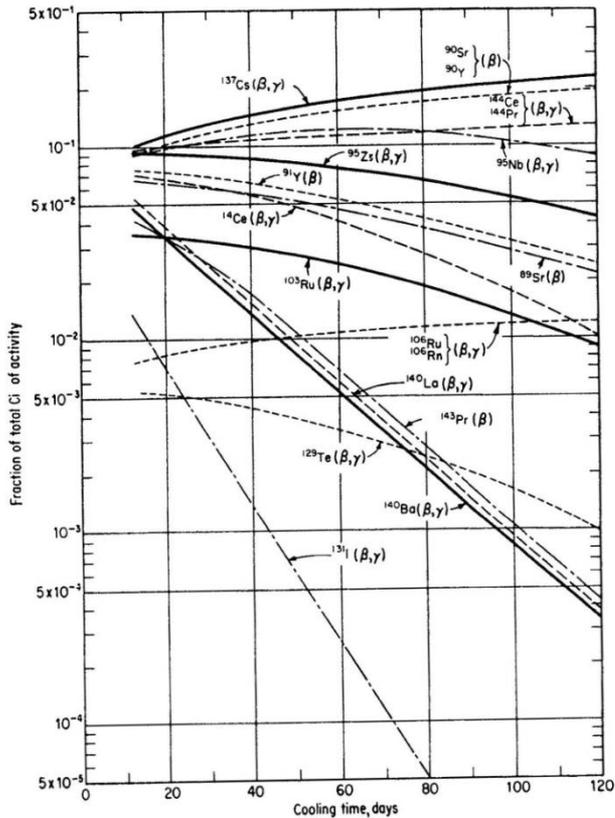
Decay heat after reactor shut-down



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 $Q \sim A \sim 1/T_{1/2}$!

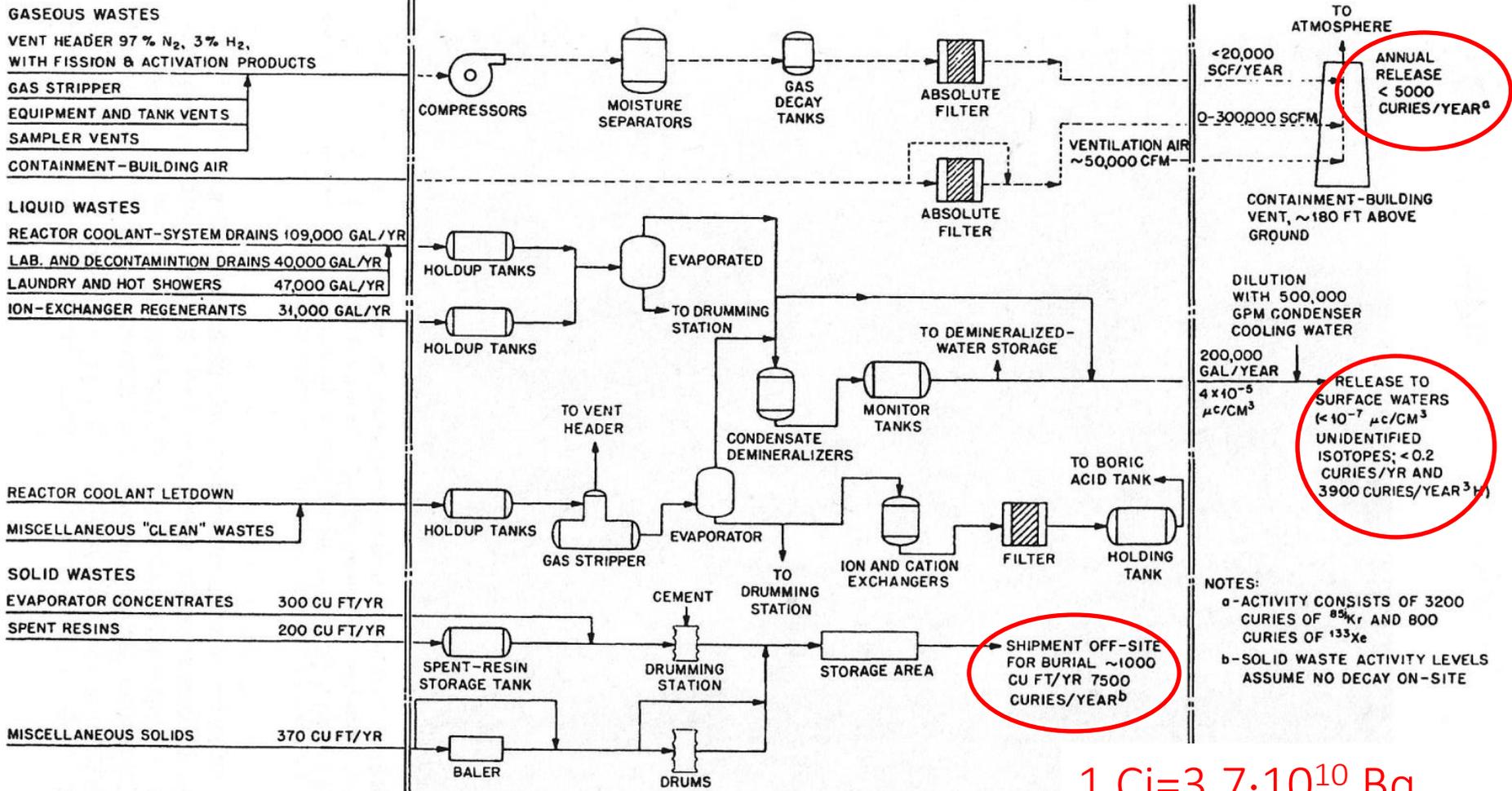


Decay of fission products in fuel rod



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

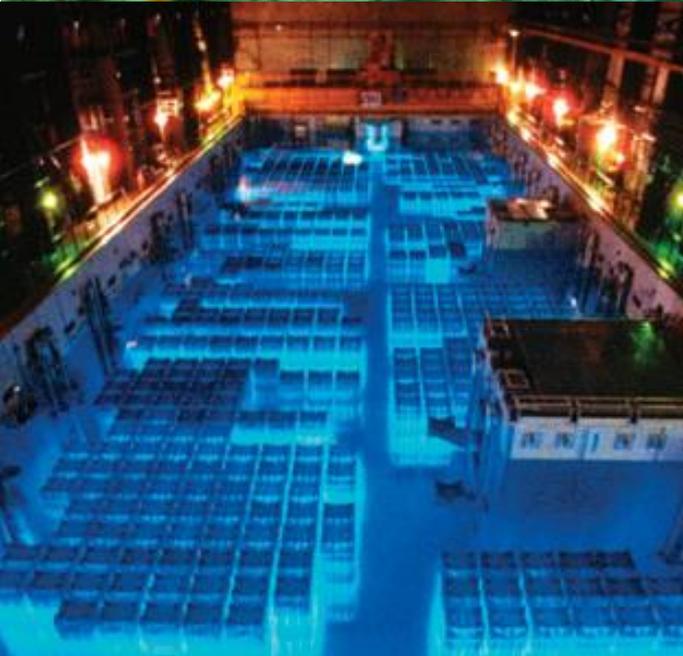
Waste management plan for a light water reactor



$1 \text{ Ci} = 3.7 \cdot 10^{10} \text{ Bq}$

Intermediary Storage in Spent Fuel Pool

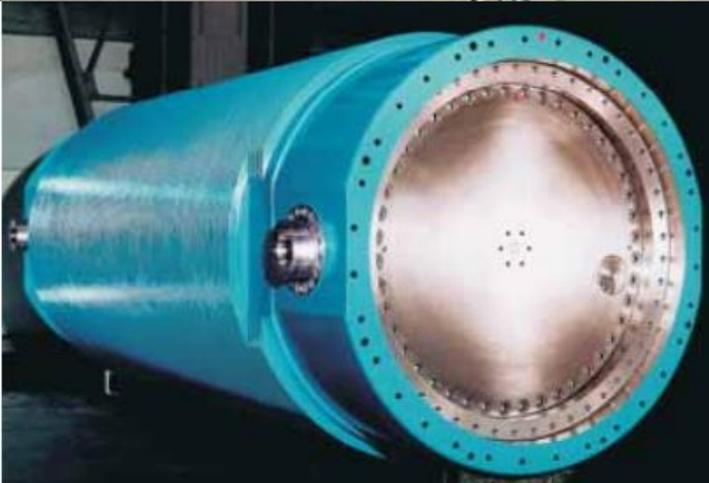
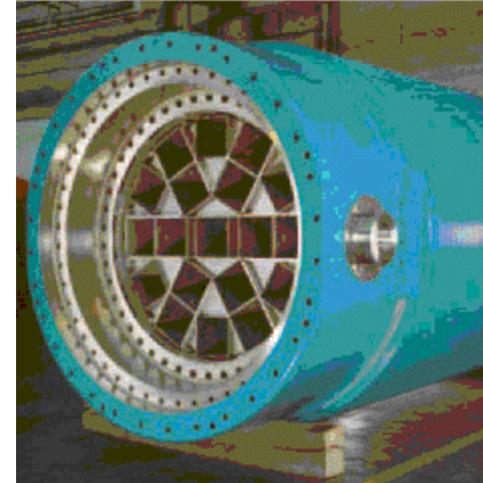
The rods are removed from reactor after the efficiency declines, due to decline in Uranium content and enrichment in neutron poisons, long-lived radioactive fission products ^{90}Se , ^{137}Cs , and non fissile radioactivity ^{241}Am . About a quarter to a third of the total fuel load of a reactor is removed from the core every 12 to 24 months and replaced with fresh fuel. The material is stored in the Spent 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.



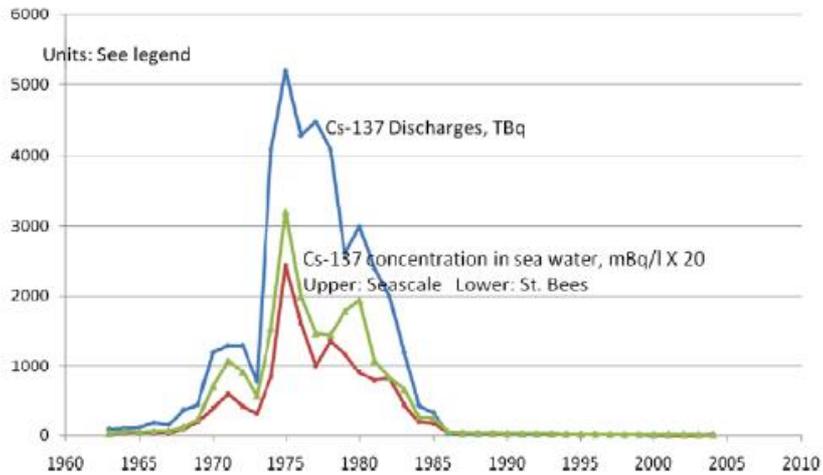
La Hague in France



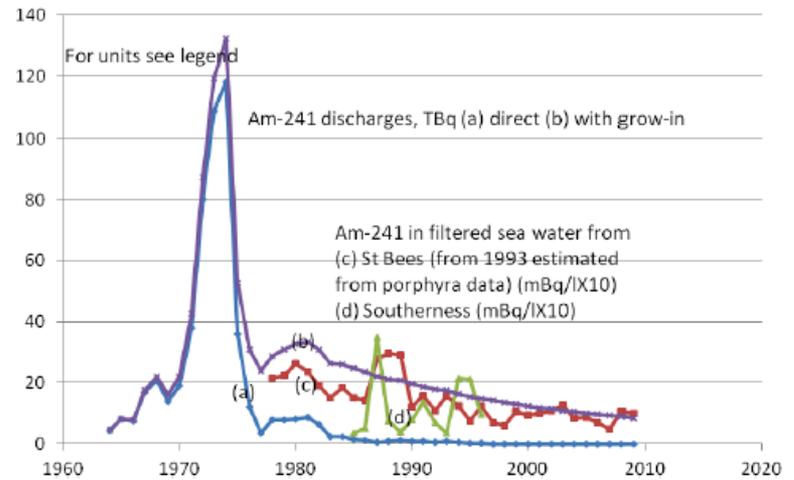
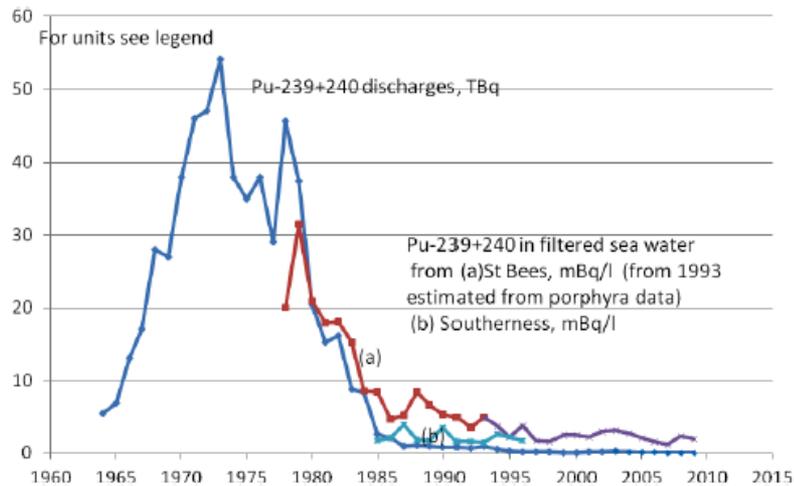
Sellafield in the UK

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

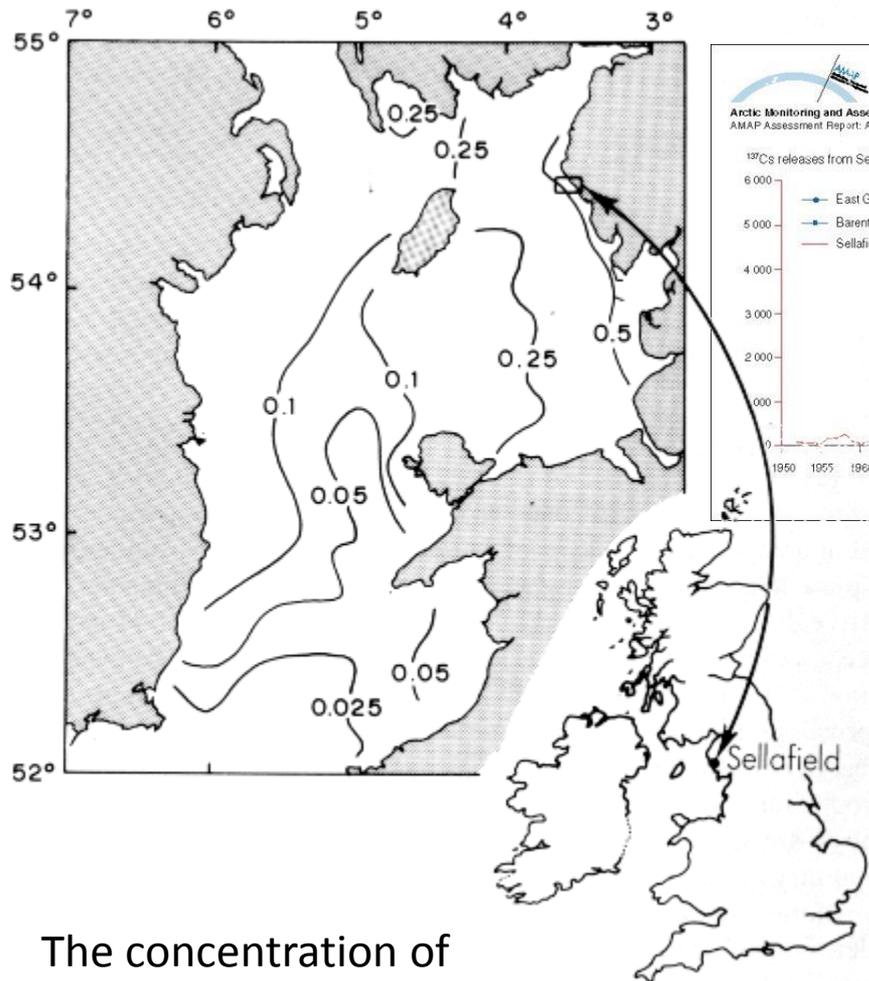


Low-level radioactive waste has been discharged into the Irish Sea as part of operations at Sellafield since 1952. The rate of discharge began to accelerate in the mid- to late 1960s, reaching a peak in the 1970s and generally declining significantly since then. As an example of this profile, discharges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

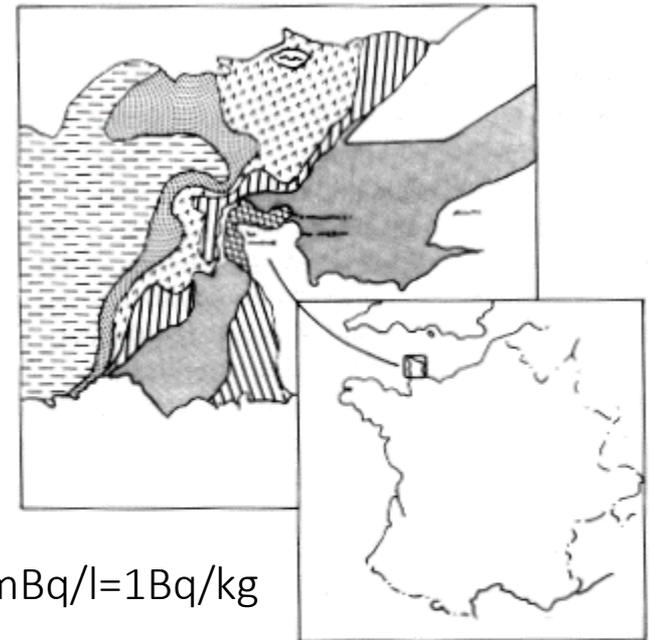
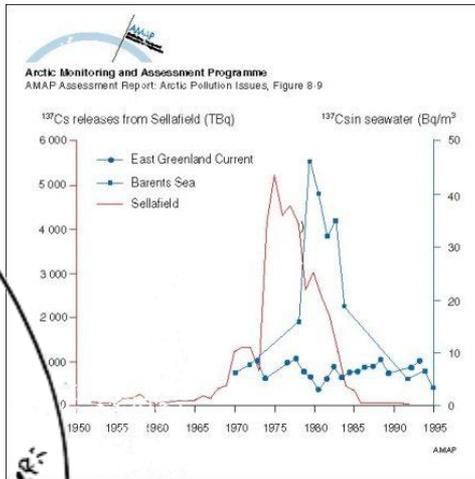


^{241}Pu $T_{1/2} = 14.35$ y β -decays to ^{241}Am $T_{1/2} = 432.2$ y α decays to ^{237}Np $T_{1/2} = 2.14 \cdot 10^6$ y α decays to ^{233}Pa ...

Water contamination from reprocessing



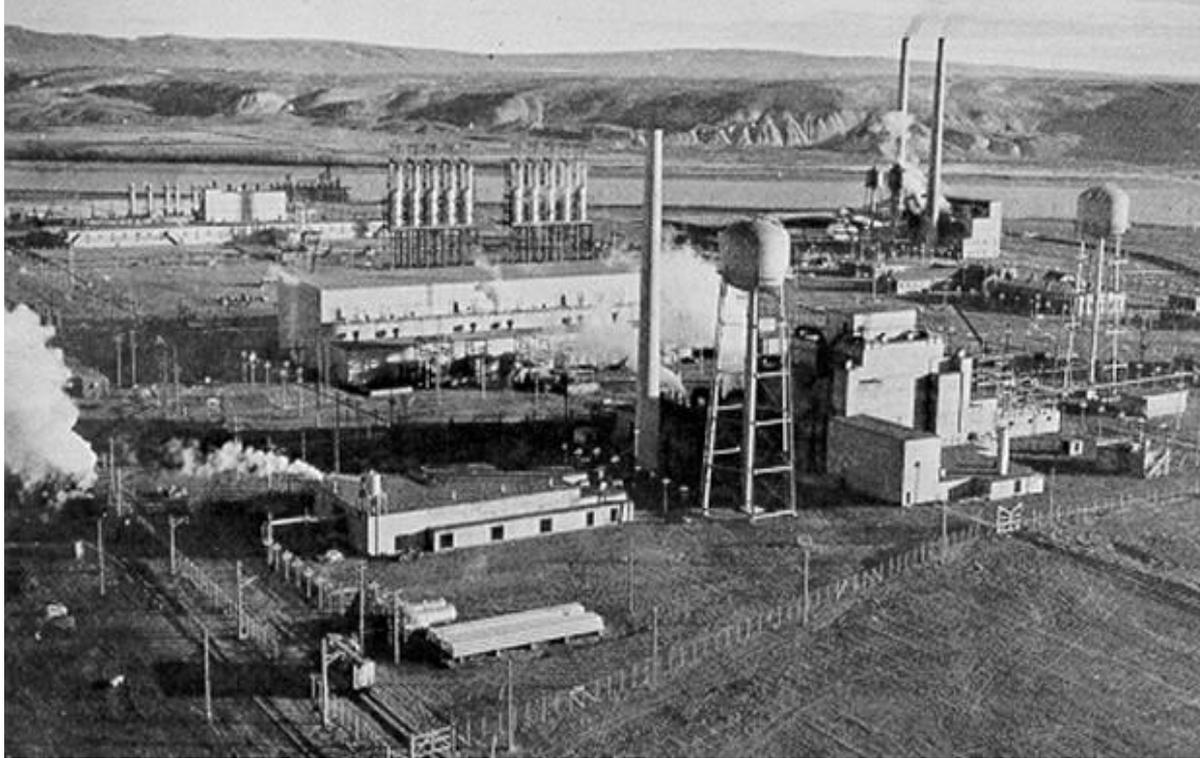
The concentration of ^{137}Cs (Bq kg^{-1}) in filtered water from the Irish Sea, April 1987



$1\text{mBq/l} = 1\text{Bq/kg}$

- $2.6 - 5.6 \text{ m Bq l}^{-1}$: industrial activity not detectable
- $5.6 - 7.4 \text{ m Bq l}^{-1}$: slight industrial influence
- $7.4 - 9.3 \text{ m Bq l}^{-1}$
- $9.3 - 14.8 \text{ m Bq l}^{-1}$
- $14.8 - 29.6 \text{ m Bq l}^{-1}$
- $> 29.6 \text{ m Bq l}^{-1}$

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

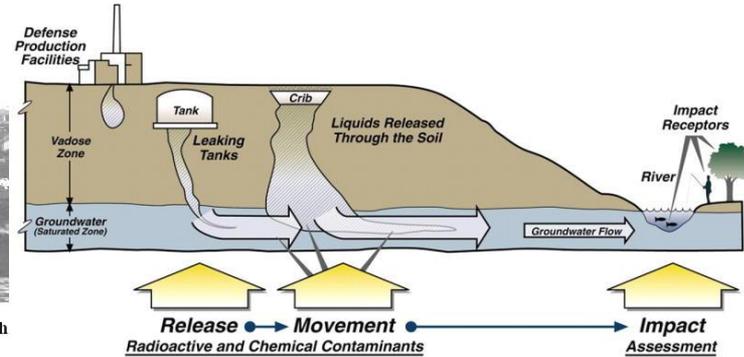
Gaseous waste:
($^{14}\text{N}(n,p)^{14}\text{C}$
toxic fumes)
ventilation and
exhaust into the
atmosphere

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

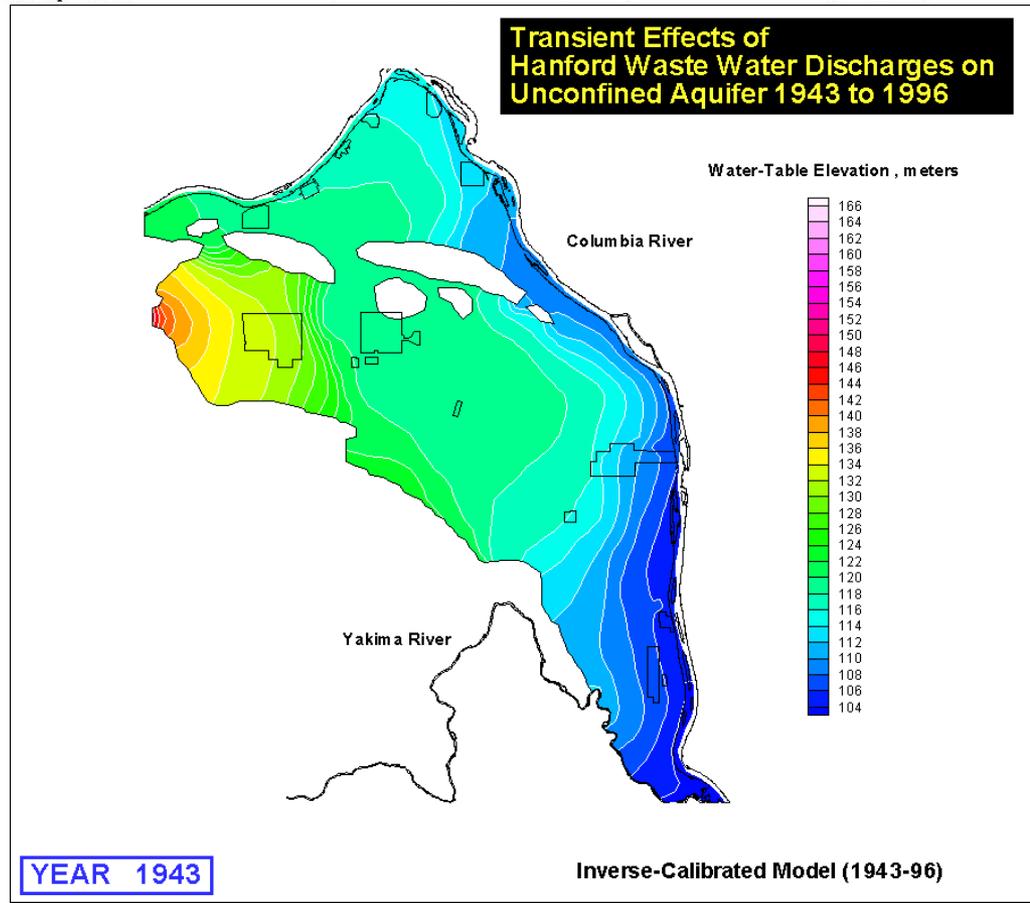


The Hanford Reservation which produced plutonium for decades, is situated along 50 miles of Columbia River. Both upstream and down, the Columbia's watershed has been contaminated with Hanford's deadly fission products.

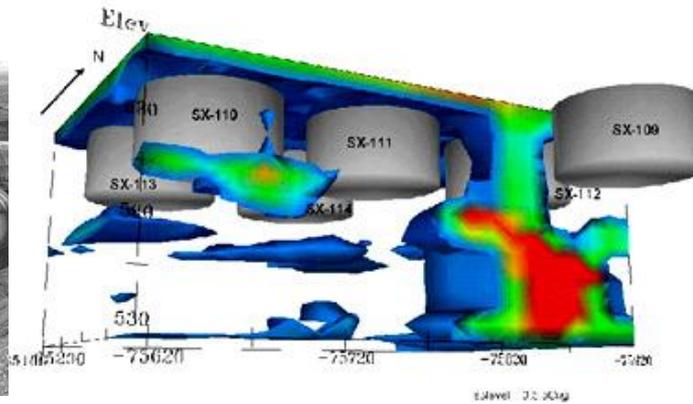


Estimates of Radiation released into the Columbia River by Hanford, 1944-1971

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



Left-overs



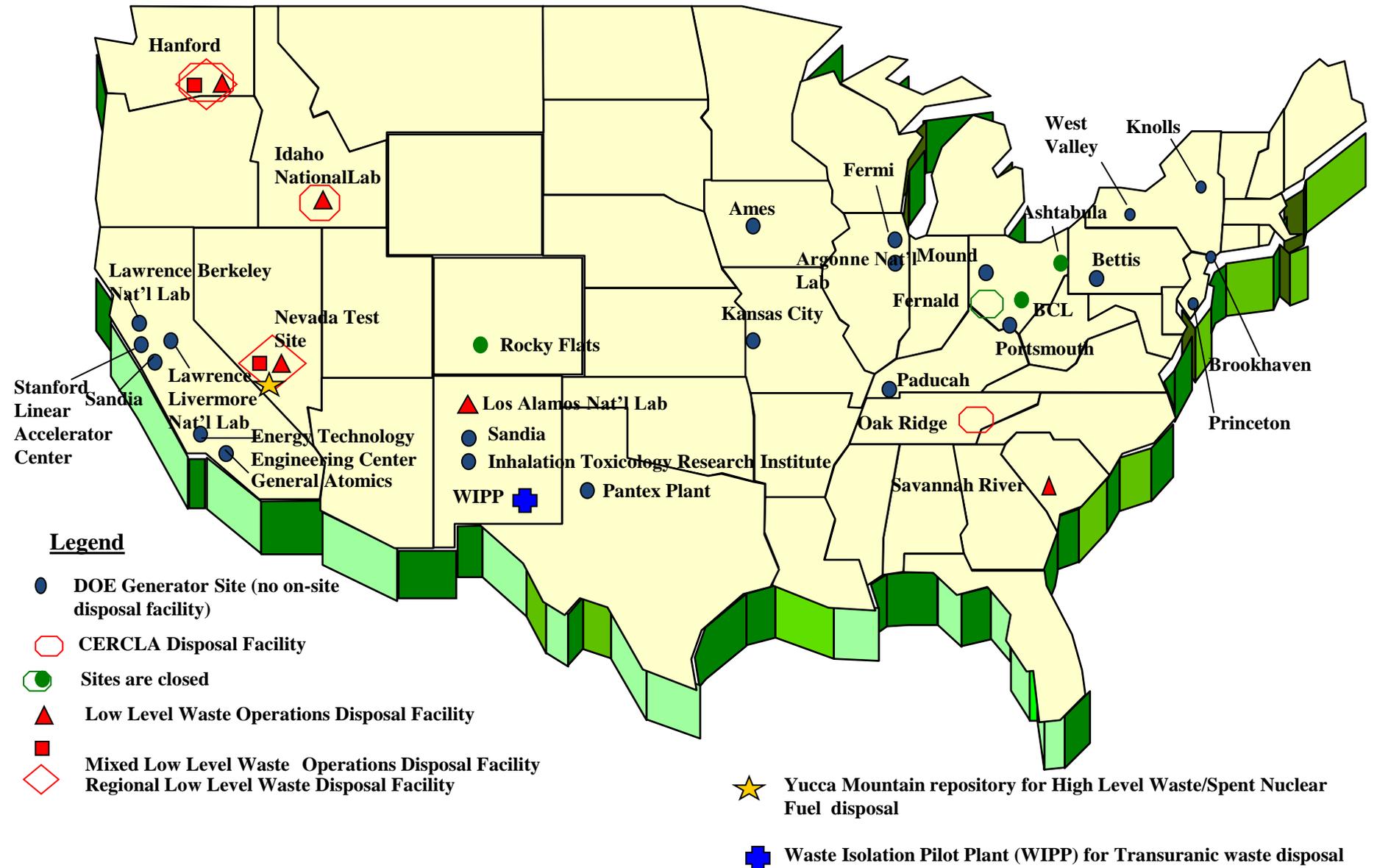
Hanford is arguably the most contaminated site in North America. Cleanup costs are projected in the tens of billions of dollars, and requiring a fifty-year effort.

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



Stockpile Stewardship Program

\$ in billions



Waste Isolation Pilot Plant

U.S. Department of Energy facility
Designed for permanent disposal
of transuranic radioactive waste
2,150 feet deep

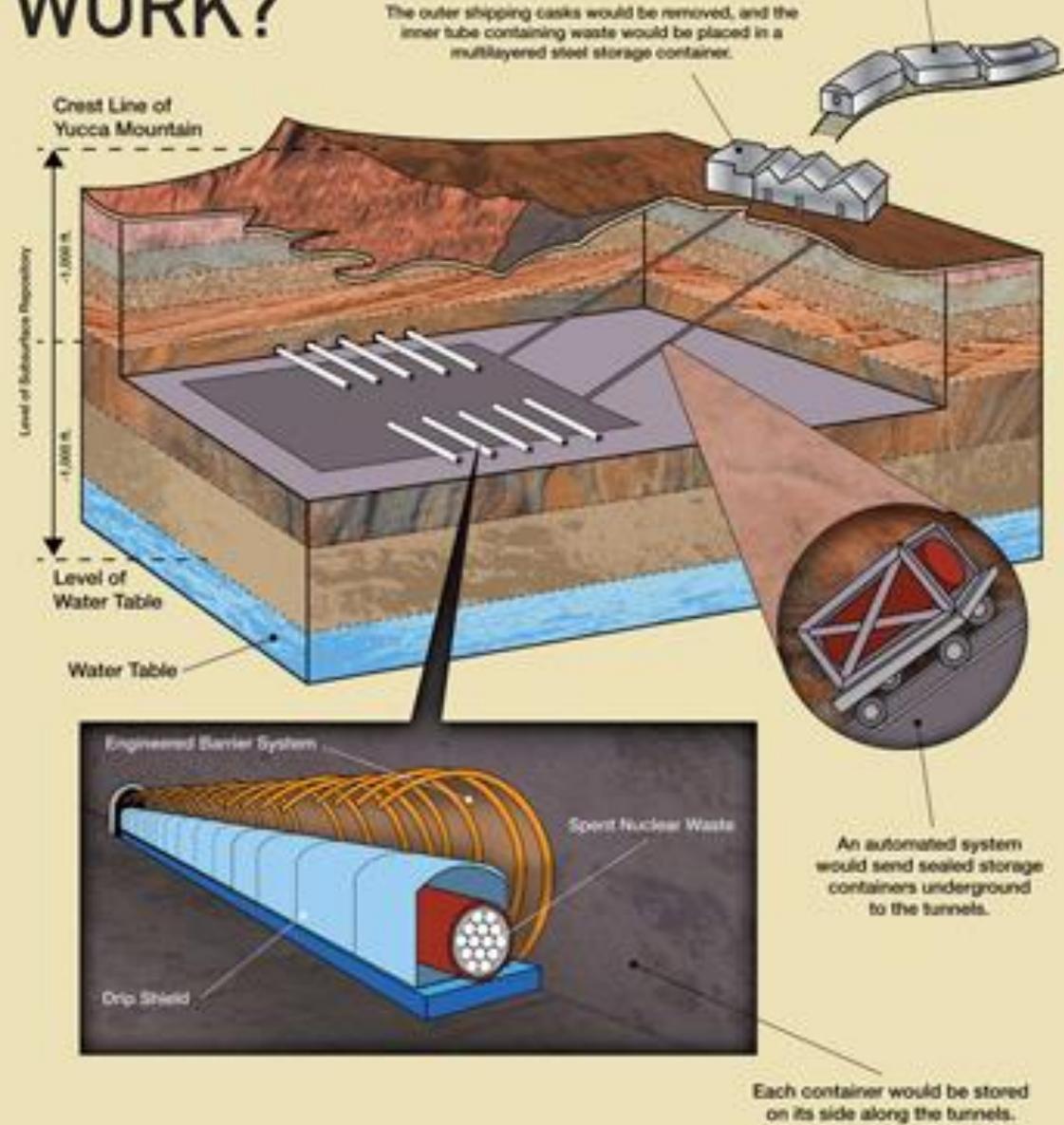




Long-Term Storage

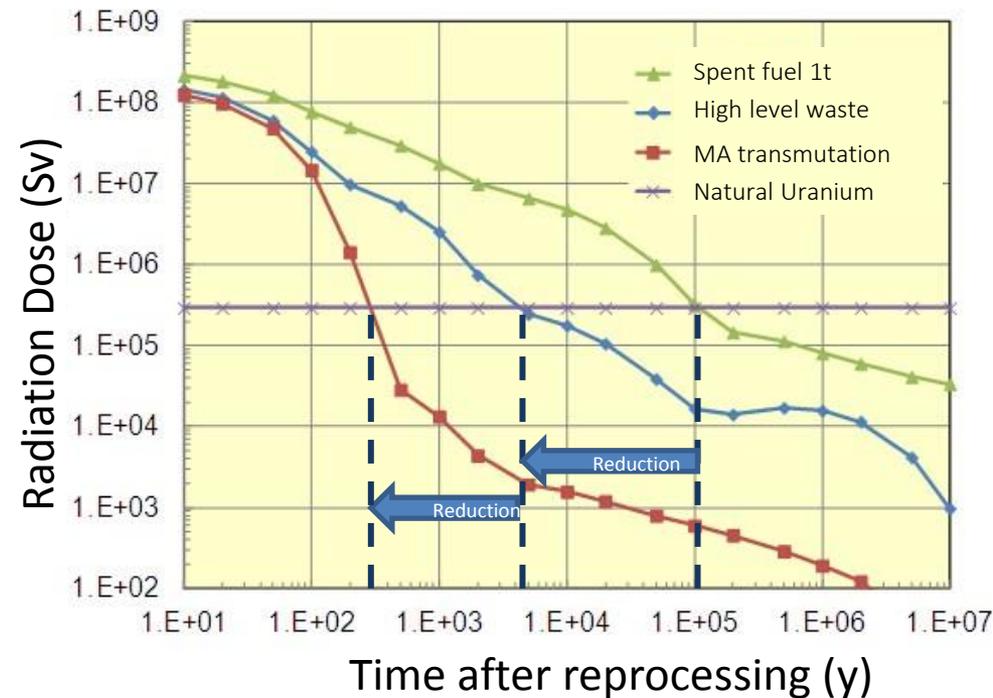
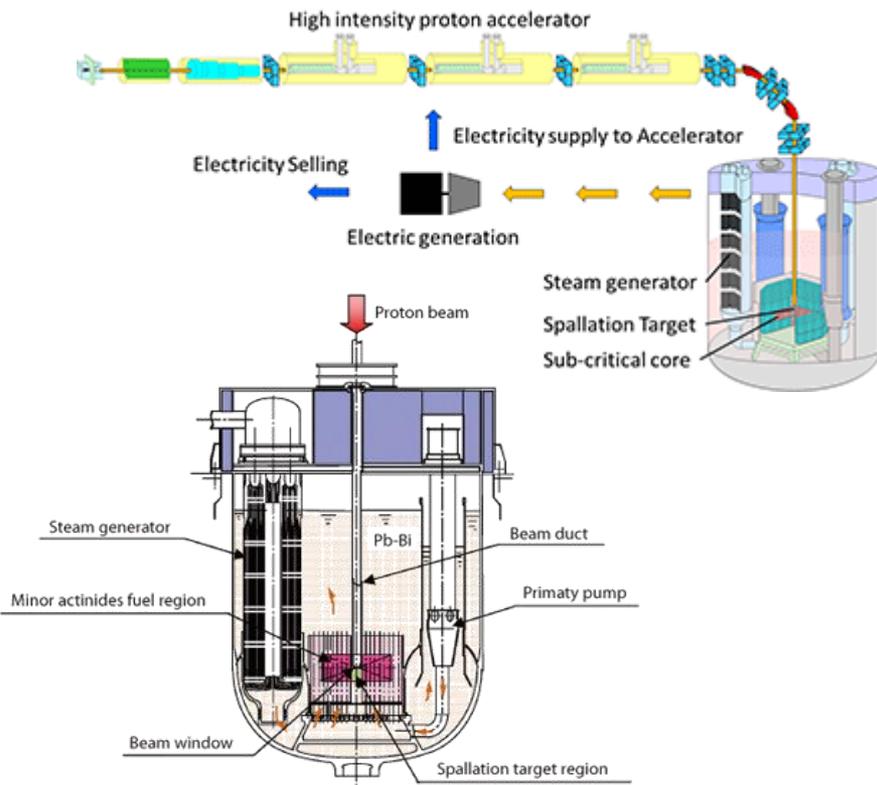


HOW WOULD YUCCA MOUNTAIN WORK?



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.



Concerns

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

Consequences at WIPP

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

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, and the 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)	

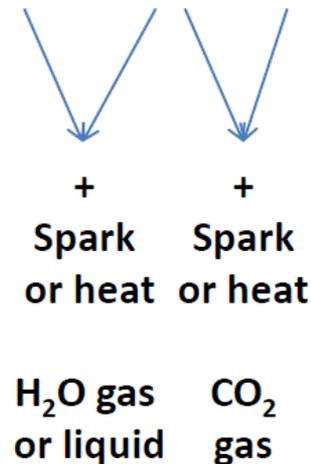
Waste probably from ^{238}Pu production for space power

The role of organic material

A Physicist's Perspective on the Barrel Explosion



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!



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**