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Title: Nuclear Reactor Forensics

Author(s): Scott, Mark R.

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Nuclear Reactor Forensics

DHS Nuclear Forensics Lecture Series at UNLV

July 12, 2013

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Acronyms and Questions

May use lots of acronyms, please stop and ask!

Questions and comments are welcomed!

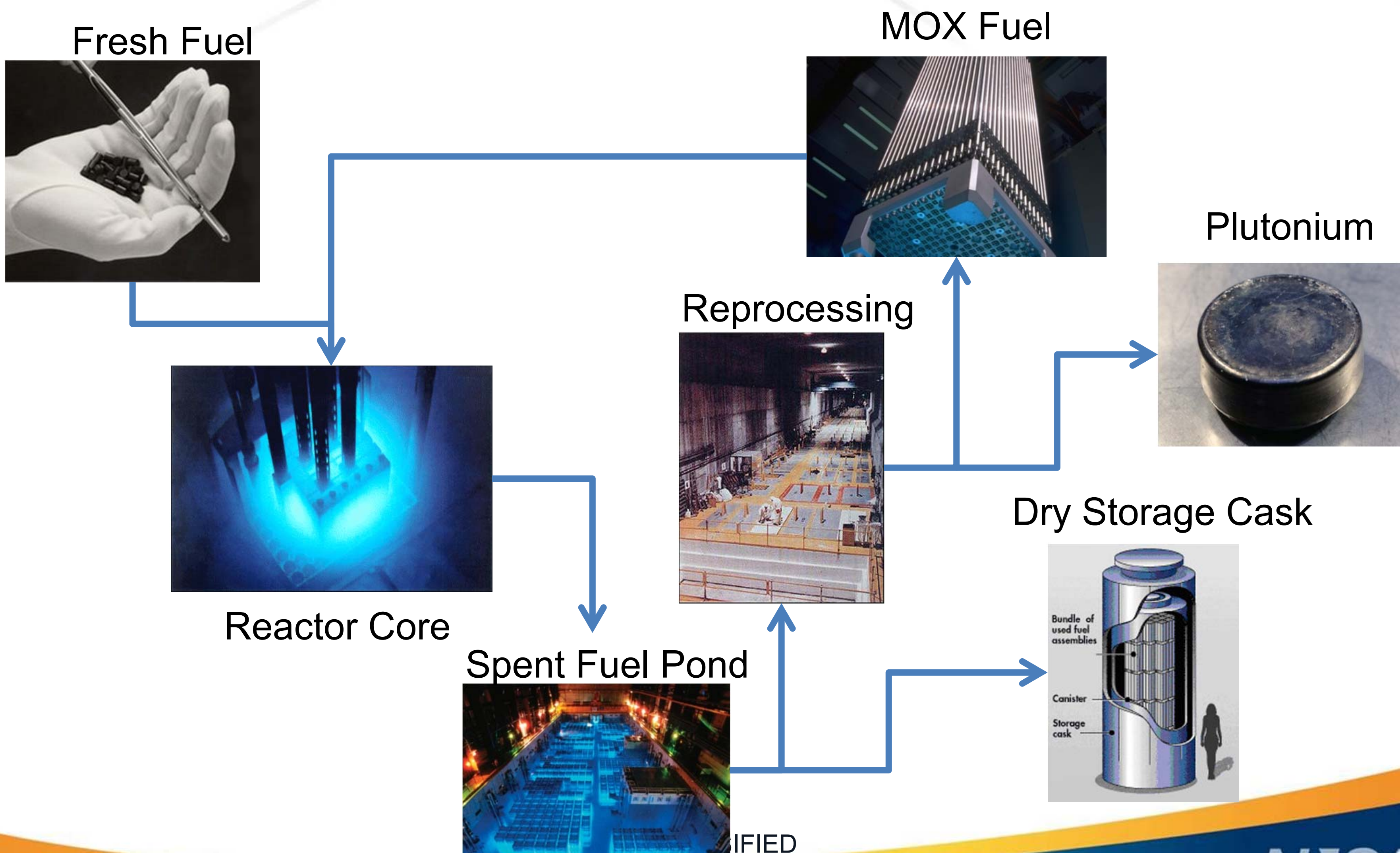
What is Reactor Forensics and Attribution?

- Set of scientific methods that help reveal historical facts and events such as:
  - Nuclear reactor operating history
  - Nuclear reactor design
  - Material reactor irradiation history
  - Material post-irradiation history

Who Cares About Reactor Forensics?

- Law Enforcement (FBI)
  - Determine who committed a crime
  - Evidence to be used in a court of law
- Safeguards Community (IAEA)
  - Monitor countries for compliance with the non-proliferation treaty (NPT)
- National Security (US Government)
  - Detect threats against our country
  - Help determine the actor behind nefarious events
  - Prevent the unauthorized production of plutonium

Nuclear Reactor Fuel



Weapons Created From Nuclear Fuel

- Atomic bomb or Nuclear Weapon
  - Requires highly enriched uranium (HEU) or plutonium.
- Radiological Dispersion Device (RDD)
  - Uses conventional explosives (C4) to disperse radioactive material.
  - Used or spent nuclear fuel is one of the most deadly sources of radioactive material for RDDs.

Spent Nuclear Fuel in the US

"Spent nuclear fuel, the used fuel removed from nuclear reactors, is one of the most hazardous substances created by humans. Commercial spent fuel is stored at reactor sites; about 74 percent of it is stored in pools of water, and 26 percent has been transferred to dry storage casks. The United States has no permanent disposal site for the nearly 70,000 metric tons of spent fuel currently stored in 33 states."

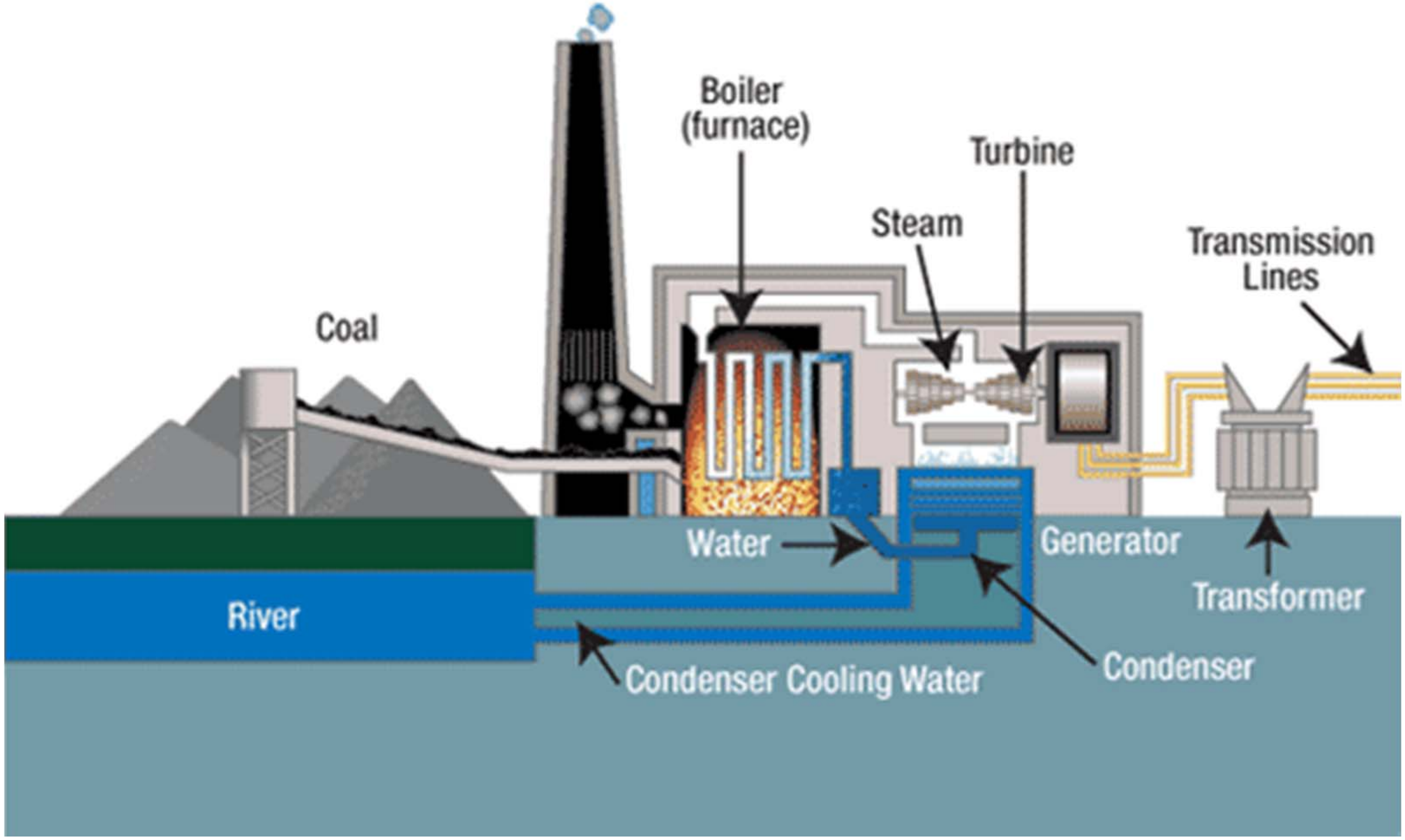
Government Accountability Office

How Would Reactor Forensics be Used

- Pre-detonation of an IND
  - Characterize the special nuclear material in a improvised nuclear device (IND) to help determine the origin of the material.
- Pre or post detonation of an RDD
  - Characterize the spent fuel in an RDD to determine the reactor type, burnup, and age of the fuel to help pinpoint the source reactor.
- Reactor past operation
  - Help determine how a reactor has operated and characterize the spent fuel produced.

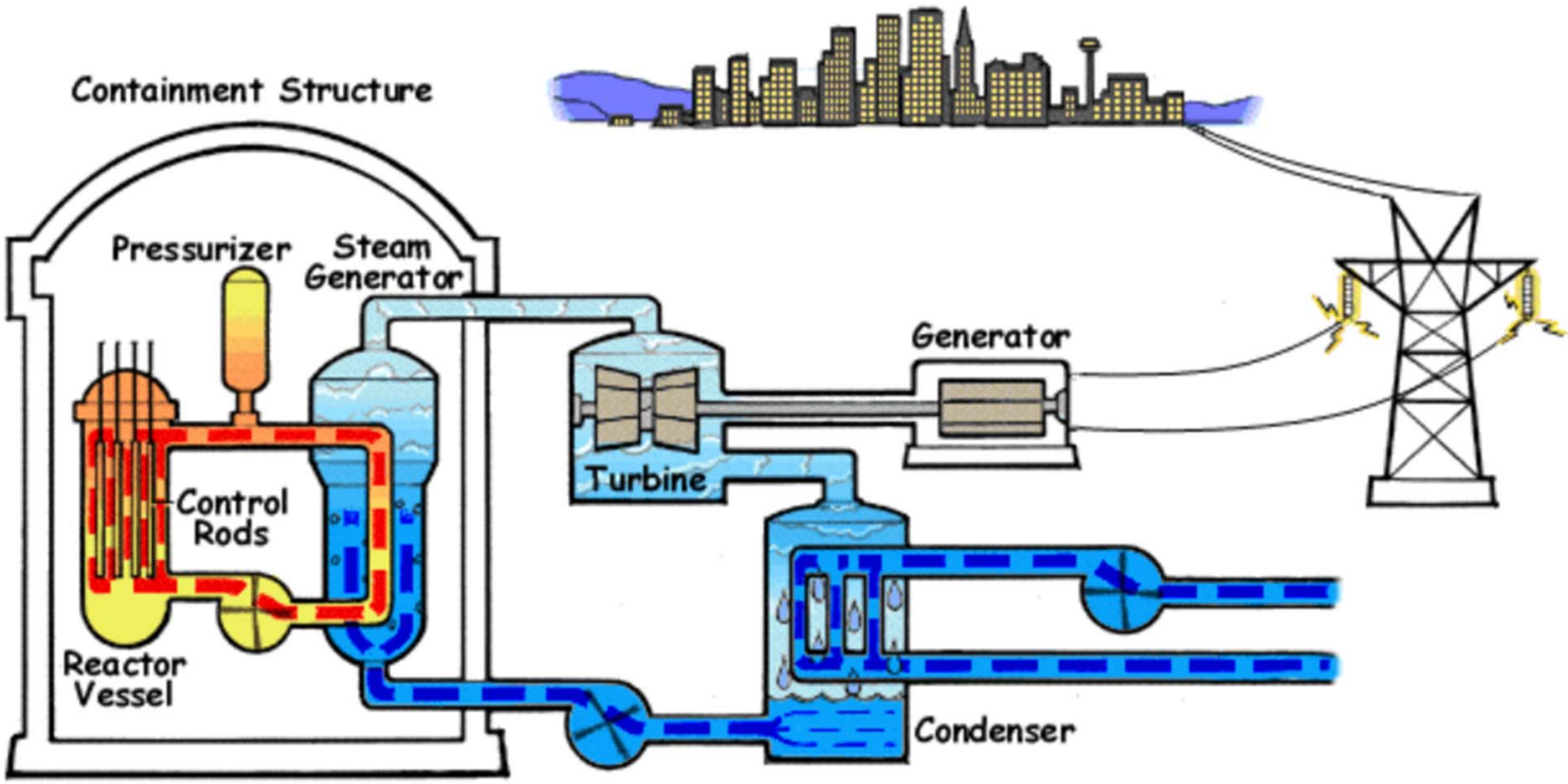
Clip from Sum of All Fears

Electric Power Generation



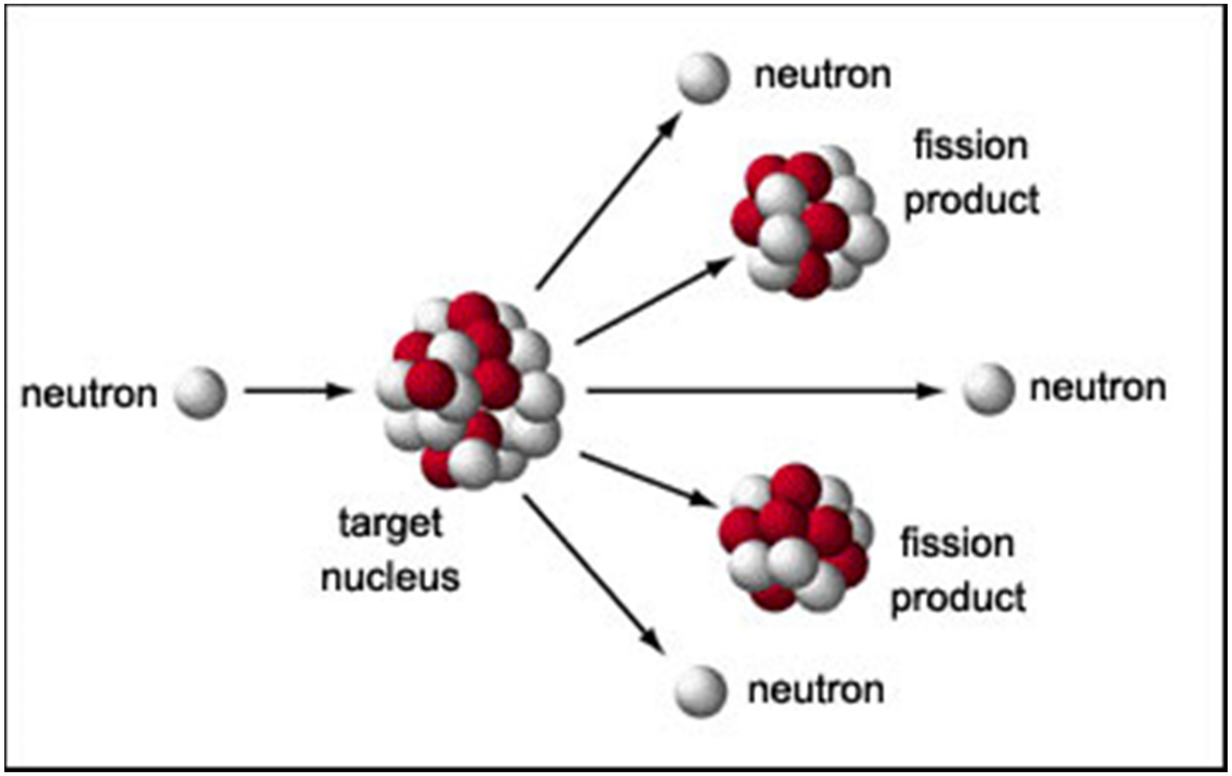
Coal-fired power plant, diagram from Tennessee Valley Authority

Pressurized Water Reactor (PWR)



Pressurized Water Reactor from the NRC website

Nuclear Fission Reaction



Fission Reaction from the Atomic Archive website

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Neutron Interactions

- Scattering
  - Elastic (think billiard balls)
  - Inelastic (think basketballs)
- Absorption

Outgoing Particle(s)
γ (Gamma Rays)
Protons
Alpha
Beta
Neutrons (2n,3n,4n)
Fission products

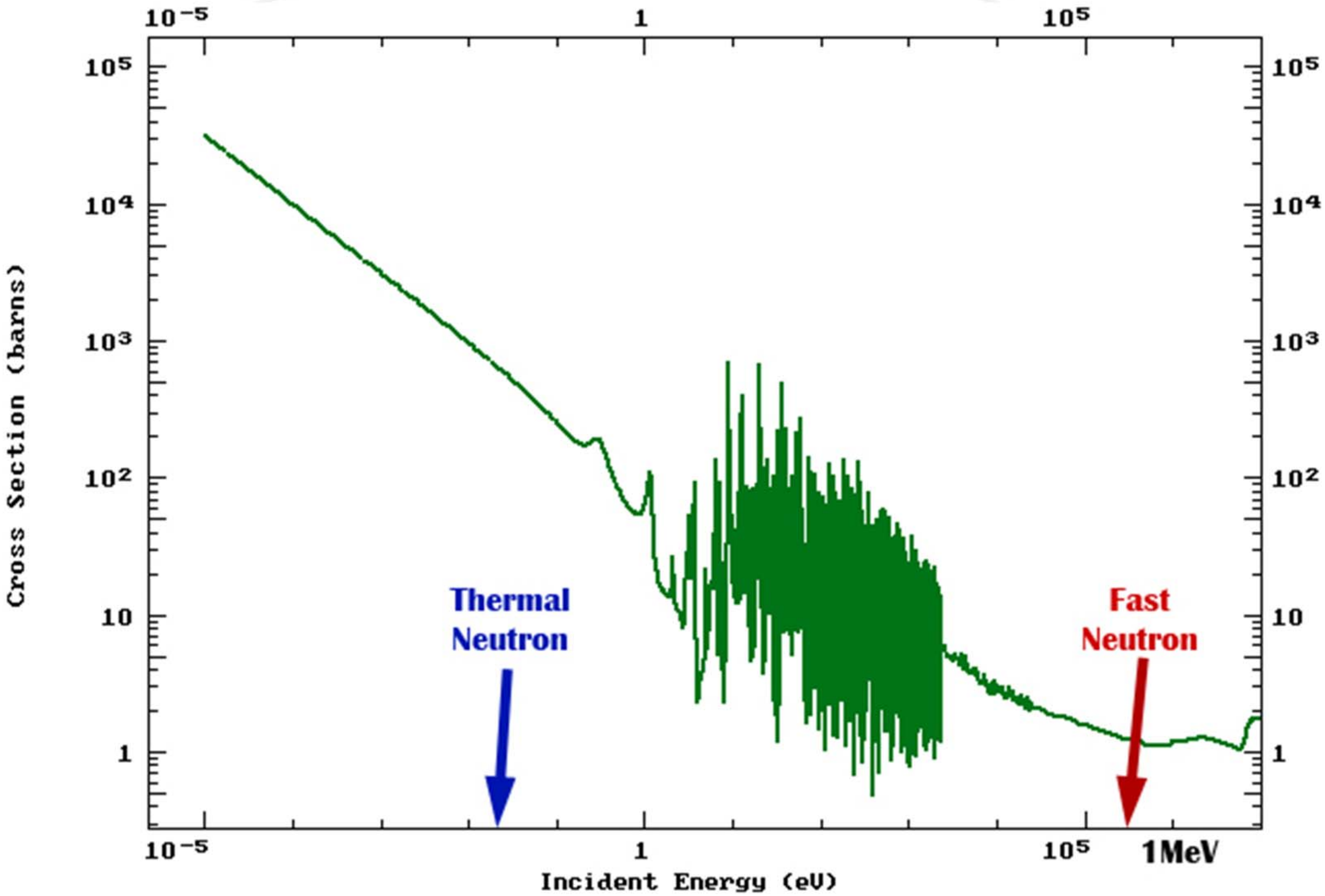
Nuclear Cross Sections

- The probability of a particular event occurring between a neutron and a nucleus is expressed through the concept of the cross section.
- The term barn as a cross section unit came from American physicists describing the uranium nucleus as “big as a barn.” American physicists hoped the whimsical name would obscure any reference to the study of the nuclear structure during WW II.

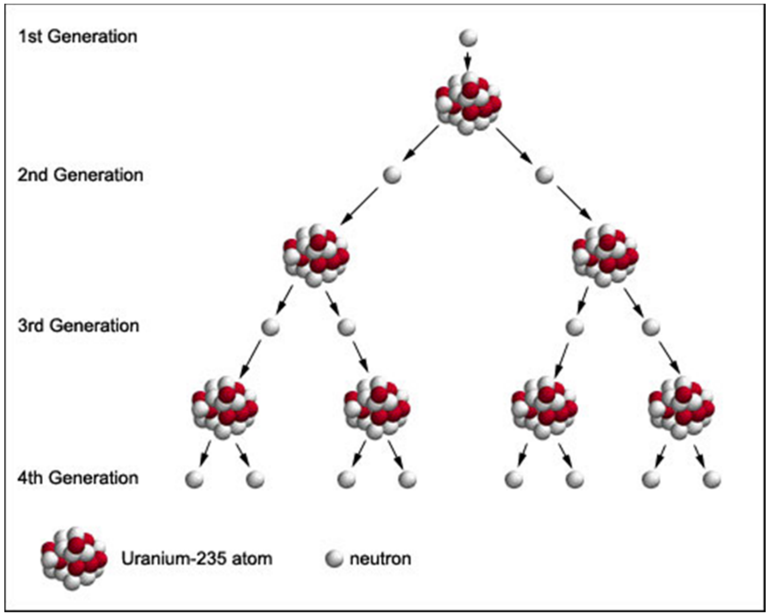
Splitting a Coconut

Increasing the velocity of the hammer striking the coconut has what kind of effect on the probability it will split?

U-235 Neutron Fission X-Section



Energy Release From Each Fission



Fission Reaction from the Atomic Archive website

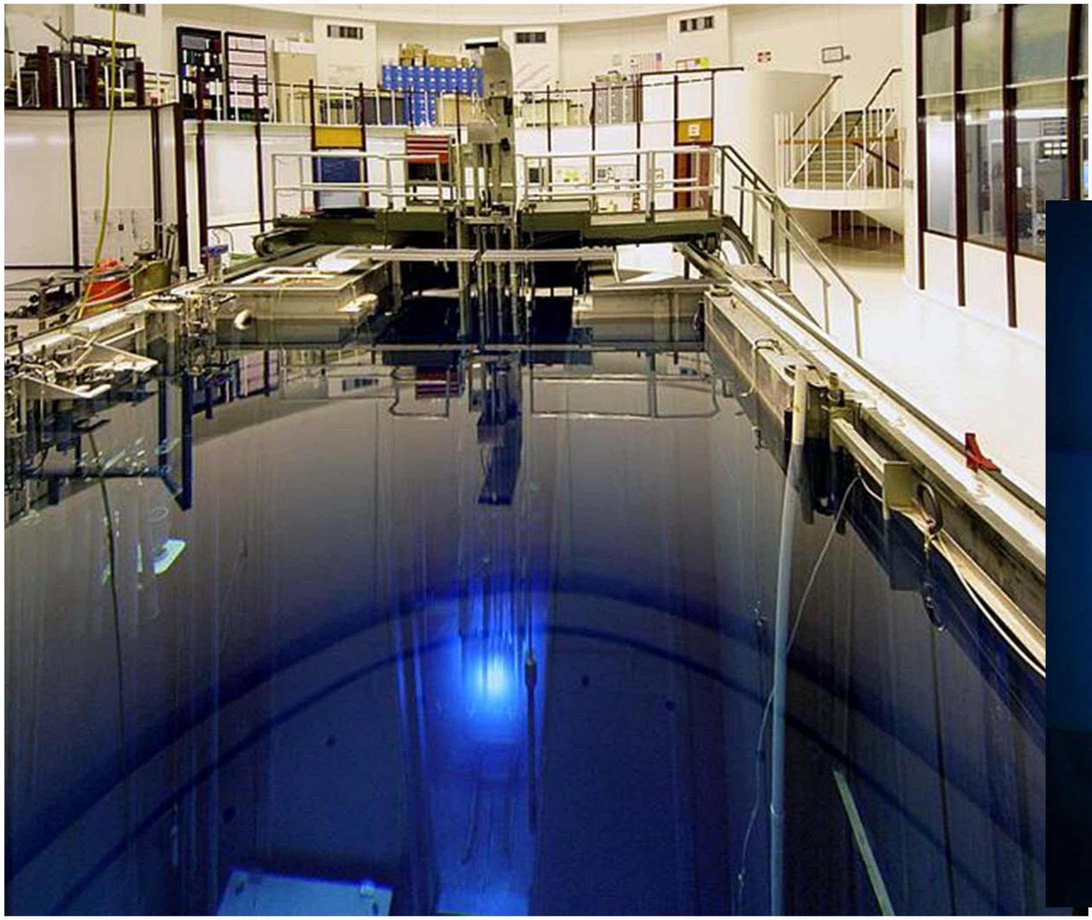
165 MeV	~ kinetic energy of fission products
7 MeV	~ gamma rays
6 MeV	~ kinetic energy of the neutrons
7 MeV	~ energy from fission products
6 MeV	~ gamma rays from fission products
9 MeV	~ anti-neutrinos from fission products
200 MeV	

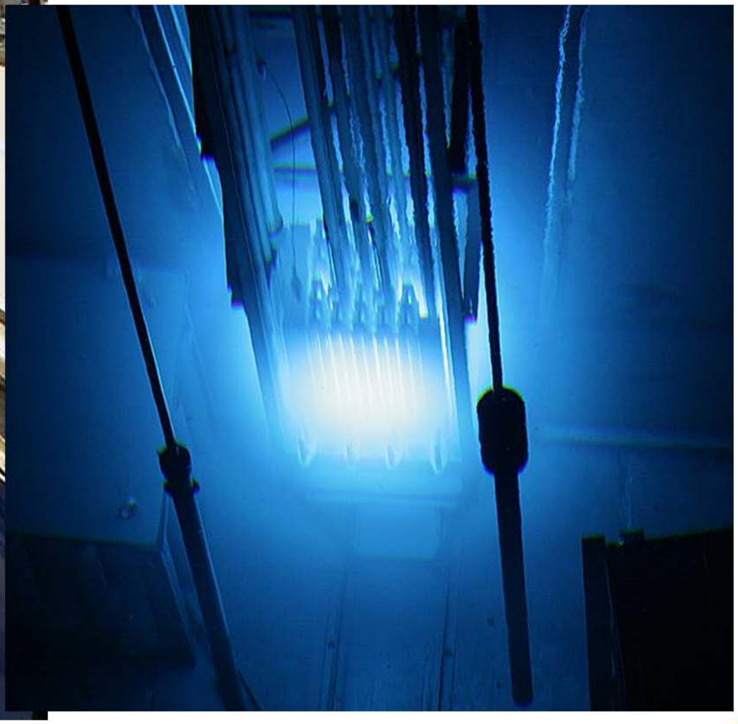
Energy of neutrons produced by fission is about 2.5MeV (fast neutrons)

How to Slow Down a Neutron

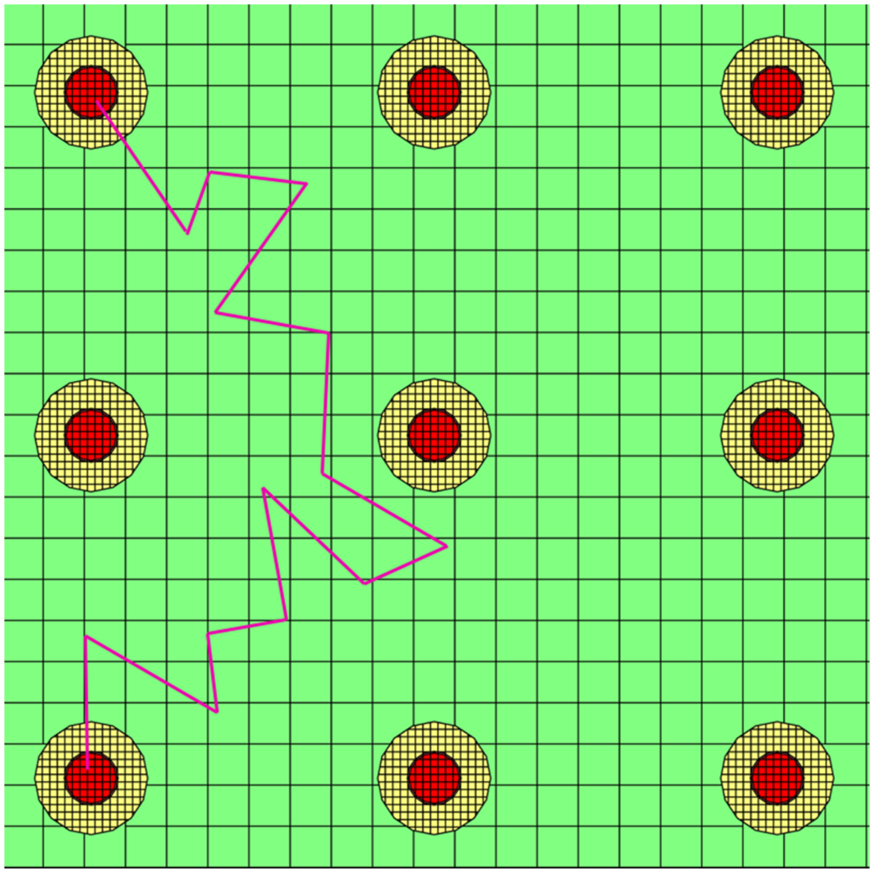
- Ideal neutron moderator
  - Low Z Material
  - High scattering x-section
  - Low absorption x-section
- Examples of neutron moderators
  - Water
  - Heavy water
  - Concrete
  - Graphite
  - Polyethylene

Texas A&M TRIGA Reactor





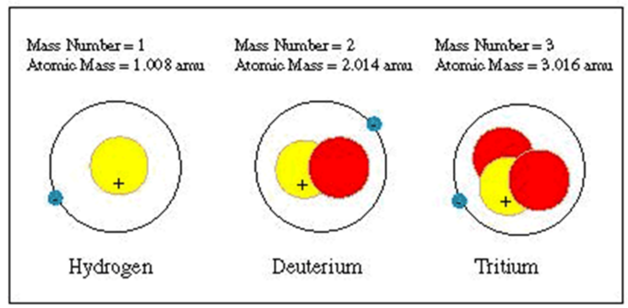
Water as a Moderator and Coolant



On average water (H_2O) will moderate a neutron after 16 collisions.

Heavy water (D_2O) requires approximately 29 collisions to moderate a neutron.

Why is heavy water preferred?



Hydrogen isotope diagram from NASA JPL

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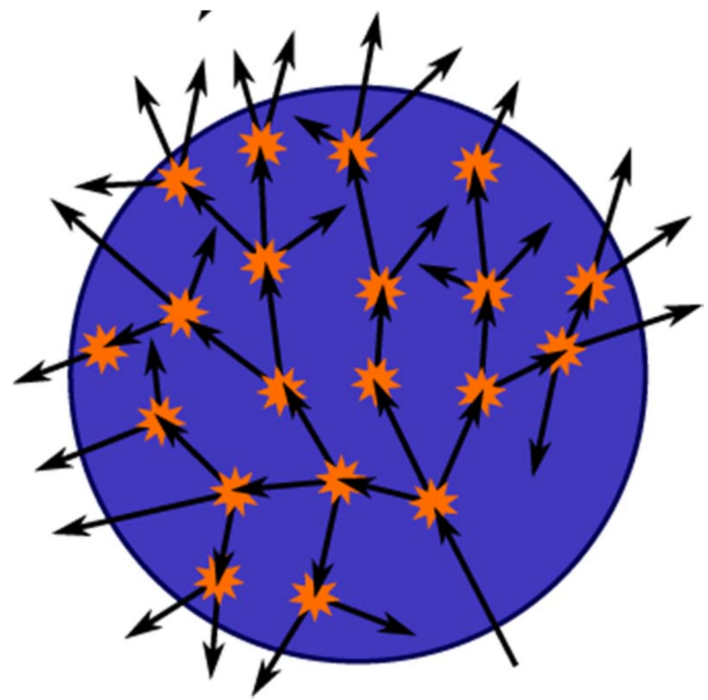
Criticality

- Critical: k=1
- Sub-critical: k<1
- Super critical: k>1
- Super prompt critical?

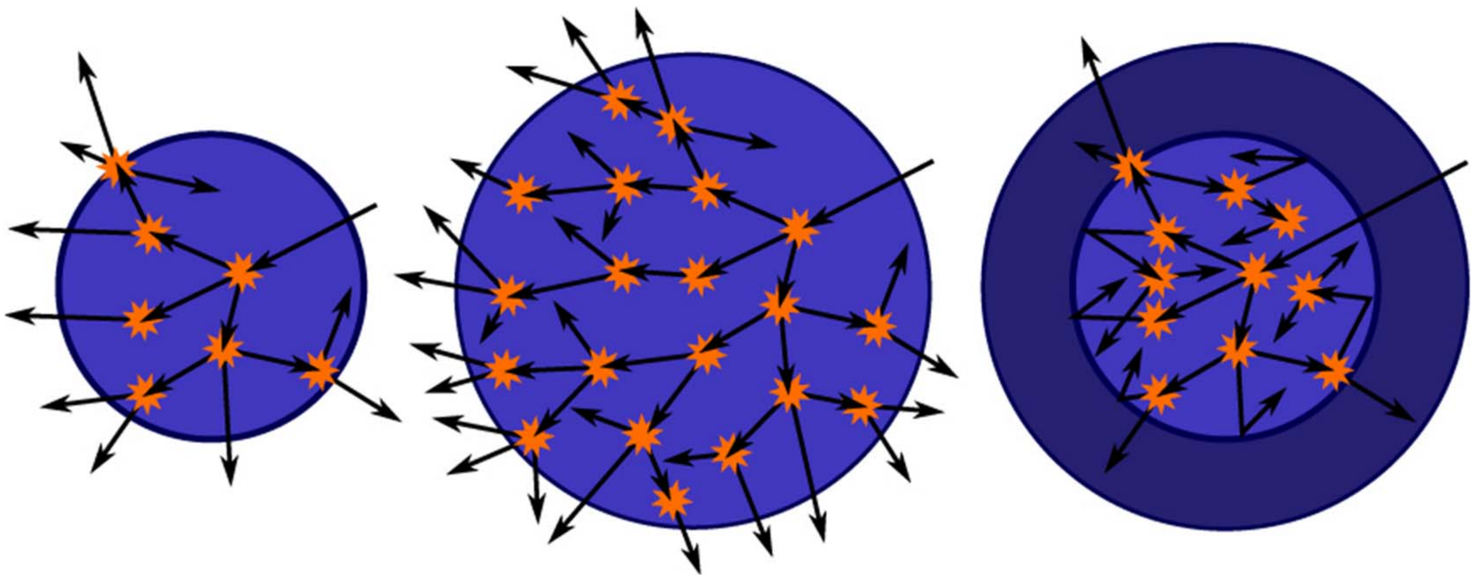


80 generations equate to $6x10^{23}$ (mole) of fissions. How much time does it take to for 80 generations to pass?

A mole of fissions releases over a 1,000 metric tons of TNT

Critical Mass

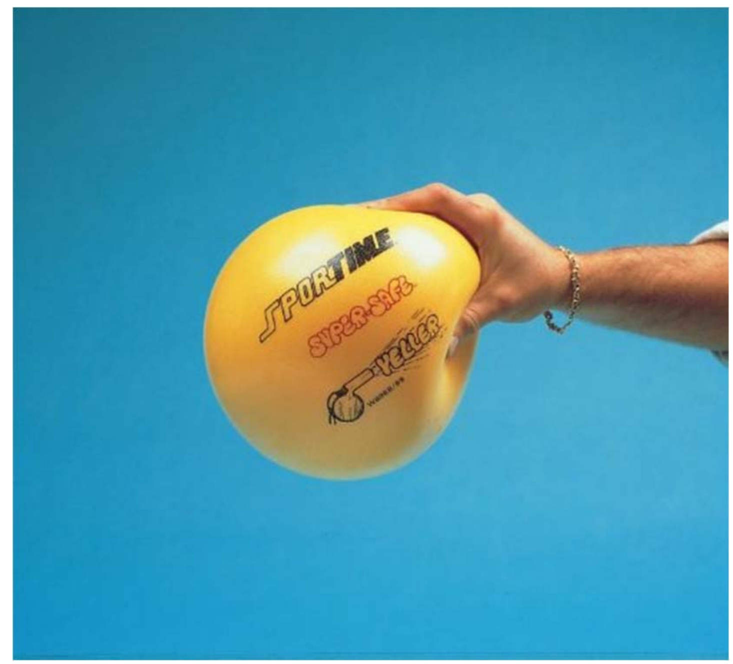
A critical mass is the least amount of fissile material needed for a sustained nuclear chain reaction. A sphere is the most efficient shape.



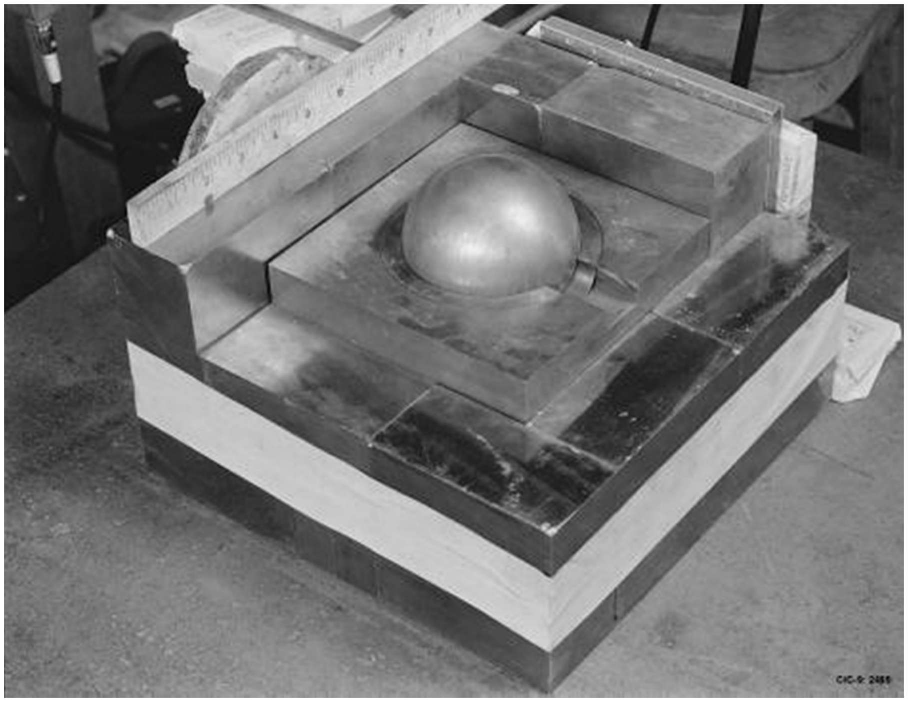
Critical Mass of a Bare Sphere

Pu-239 Critical Mass:
10kg at 9.9 cm

U-235 Critical Mass:
52kg at 17 cm



Criticality Accidents

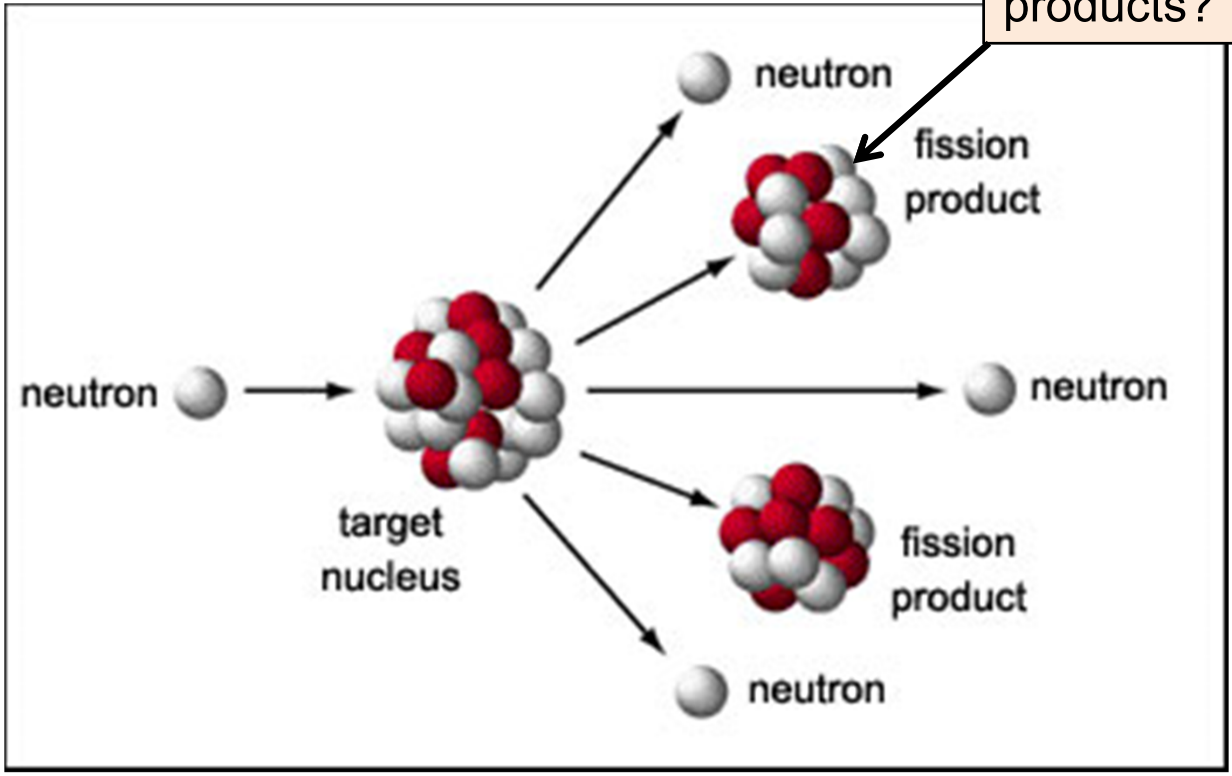


On 4 June 1945, An experiment by Los Alamos scientist John Bistline went critical when water leaked into the box holding HEU.

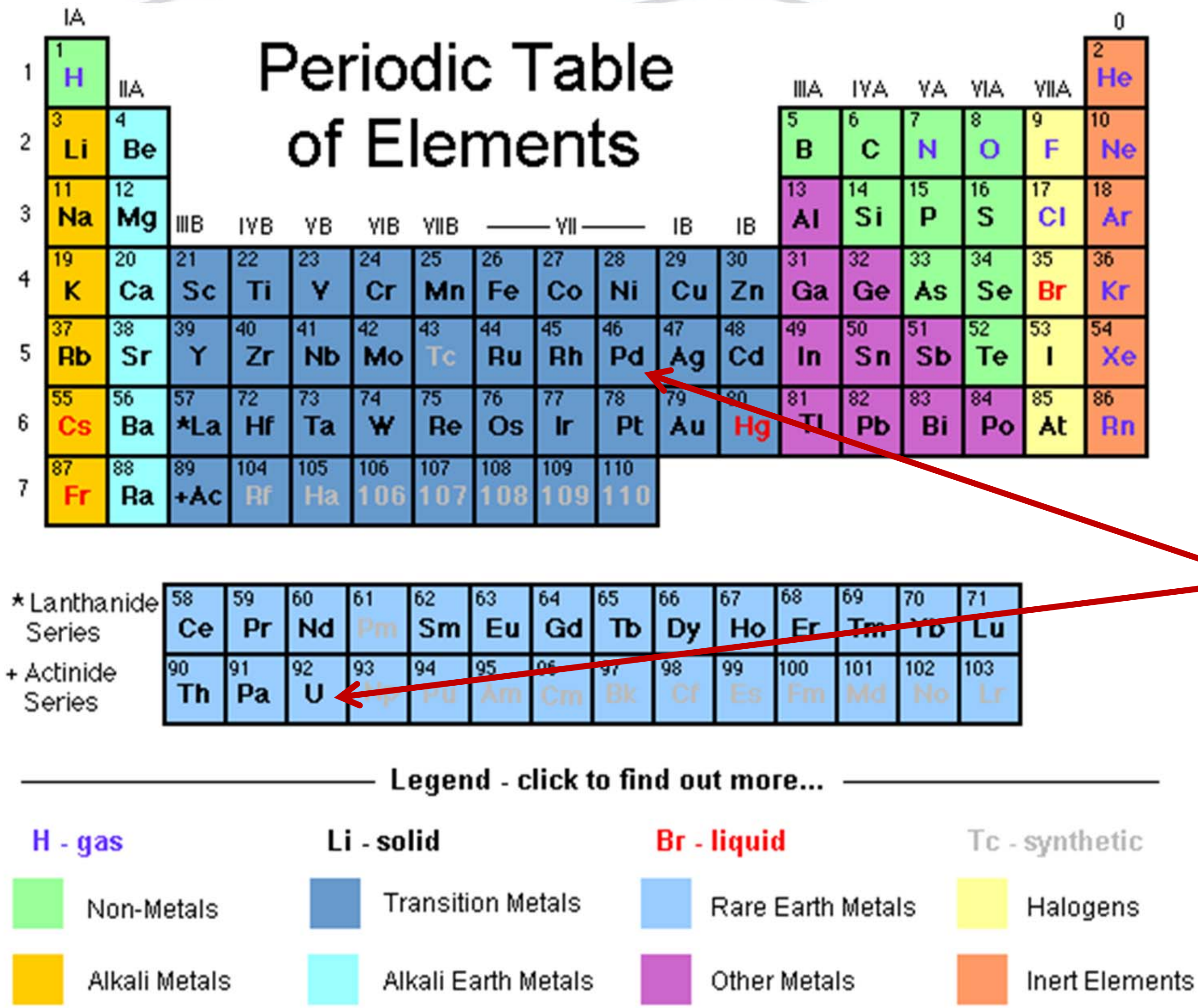
On 21 August 1945, An experiment by Los Alamos scientist Harry Daghlian went critical when he dropped a tungsten brick onto a sphere of Pu.

Nuclear Fission Products

What are fission products?



Fission Reaction from the Atomic Archive website



Does the fission of $_{92}$U result in a bunch of $_{46}$Pd?

Table of Elements from the ThinkQuest website

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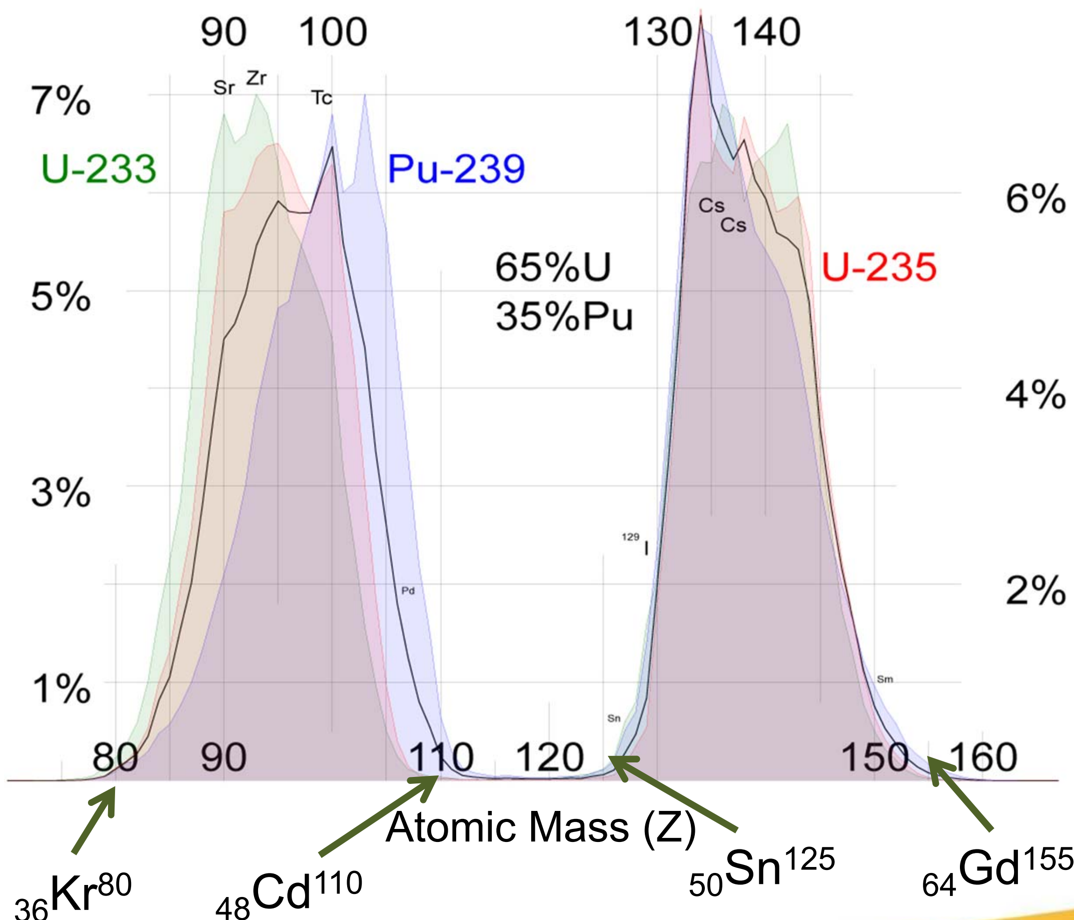
Fission Product Yield



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Actinide Production

- What are actinides?
- How do you create actinides

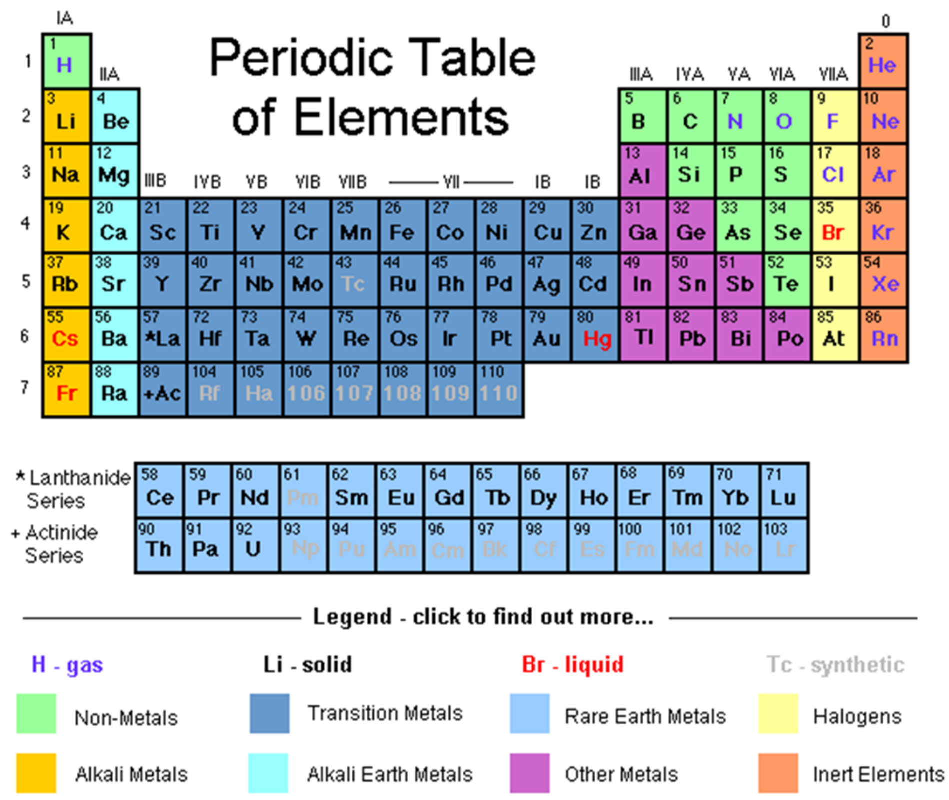
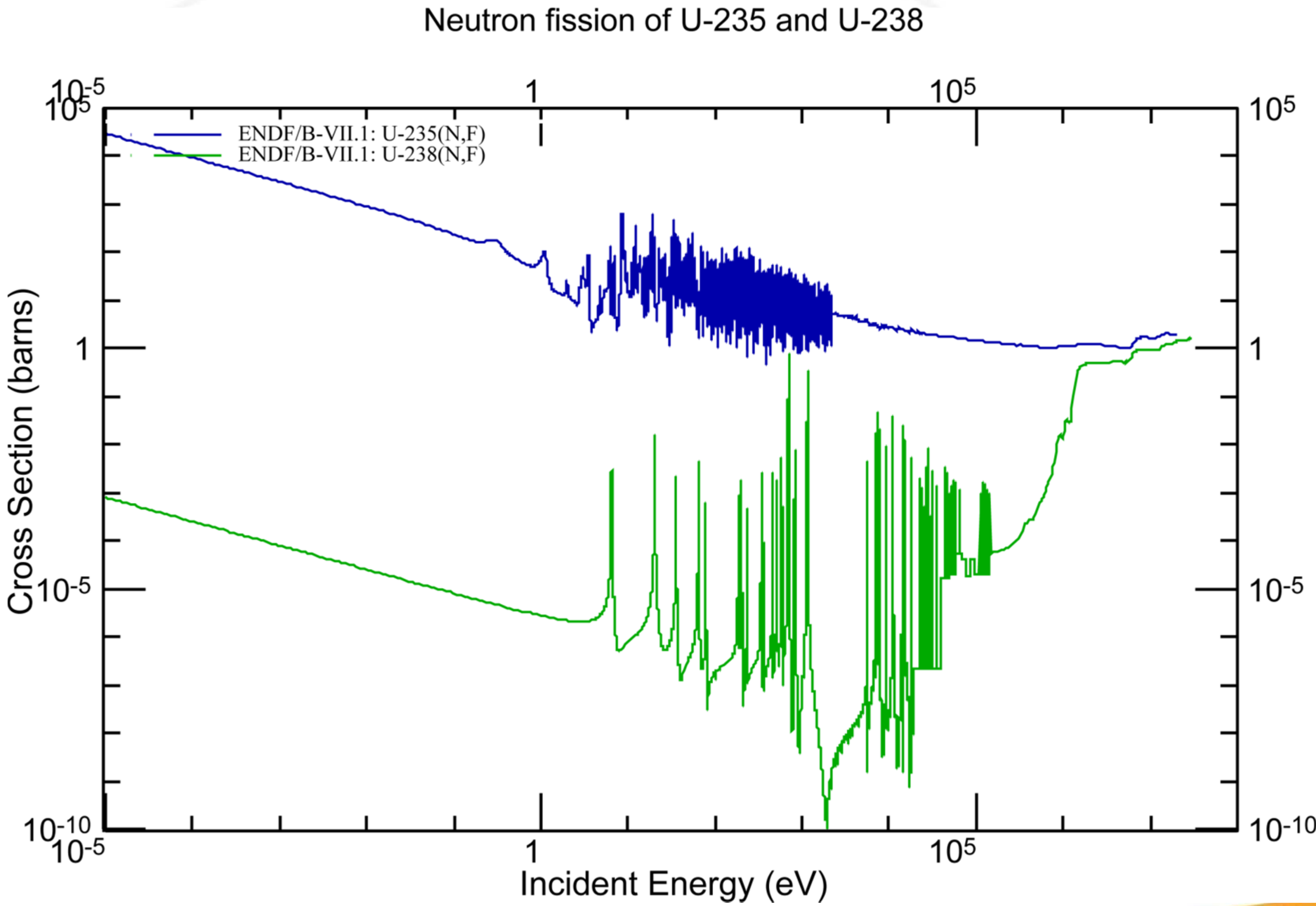


Table of Elements from the ThinkQuest website

Natural Uranium

- Natural Abundance of Uranium Nuclides
  - U-234 : 0.0055%
  - U-235 : 0.720%
  - U-238 : 99.2745%
- Note: The terms isotope and nuclide denote the same concept. The term nuclide is intended to focus on the nucleus and isotope the chemical properties.

Fission x-sections for U-235 vs U-238



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Going Critical with Uranium

- How do you create a fission chain reaction with Uranium?
  - 1. Use a really good moderator that doesn’t absorb many neutrons (heavy water)
  - 2. Use enriched uranium
    - Commercial Reactor (3-5% U-235)
    - Nuclear weapons grade uranium (>90% U-235)

Stability of Nuclides

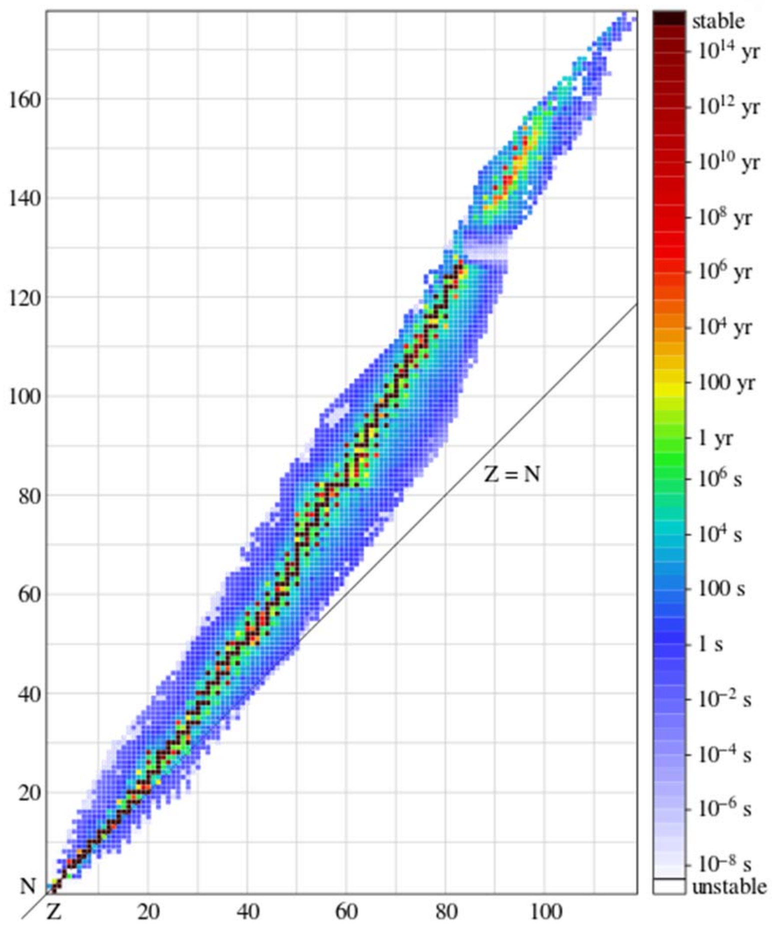

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Chart of the Nuclides

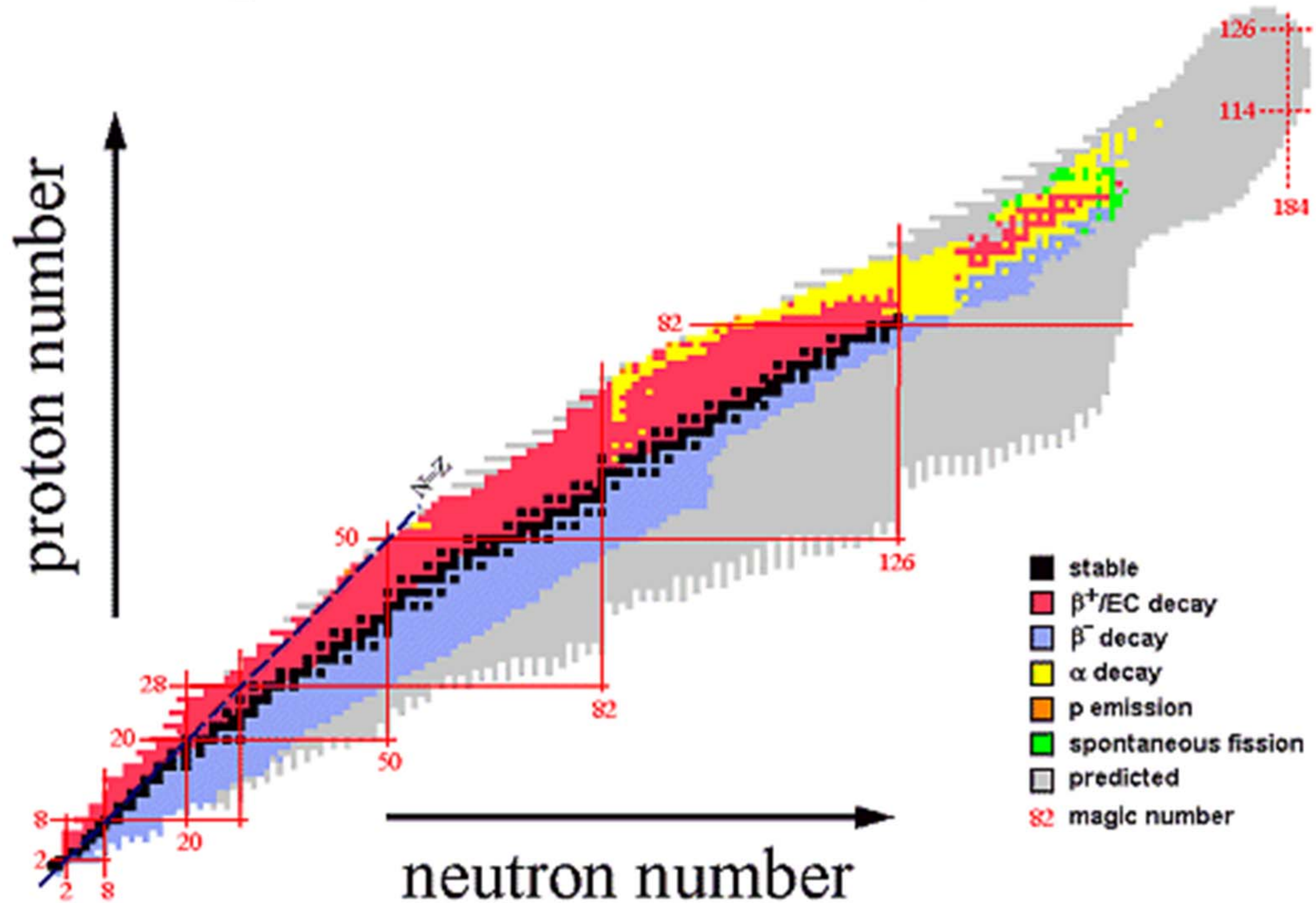

Chart of the nuclides from LUND University

Isotopes, Isobars and Isotones

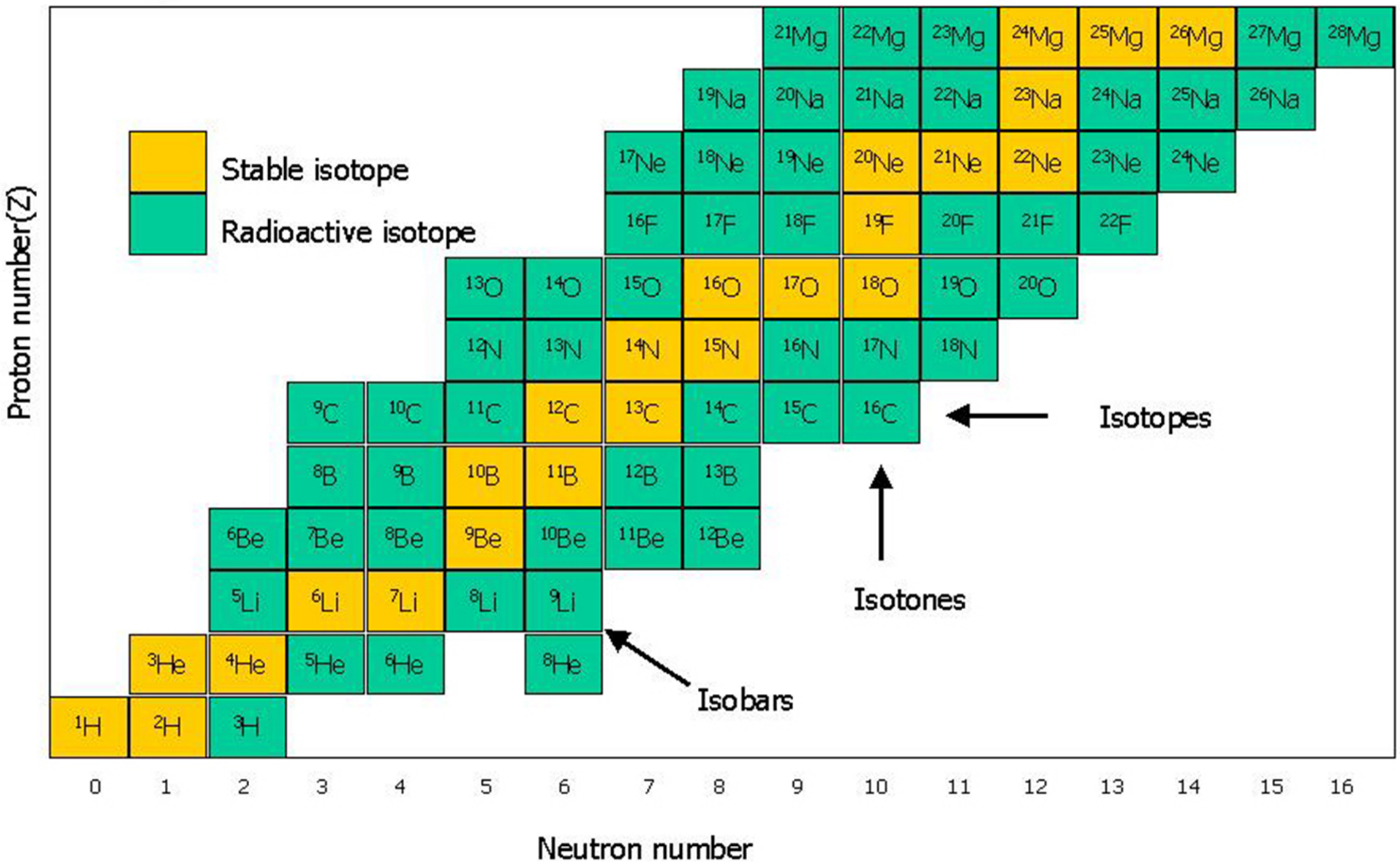

Chart of the Nuclides from University of Alaska

Up Close View of Nuclides Chart

Chart of the Nuclides from KAPL

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How Are Actinides Produced

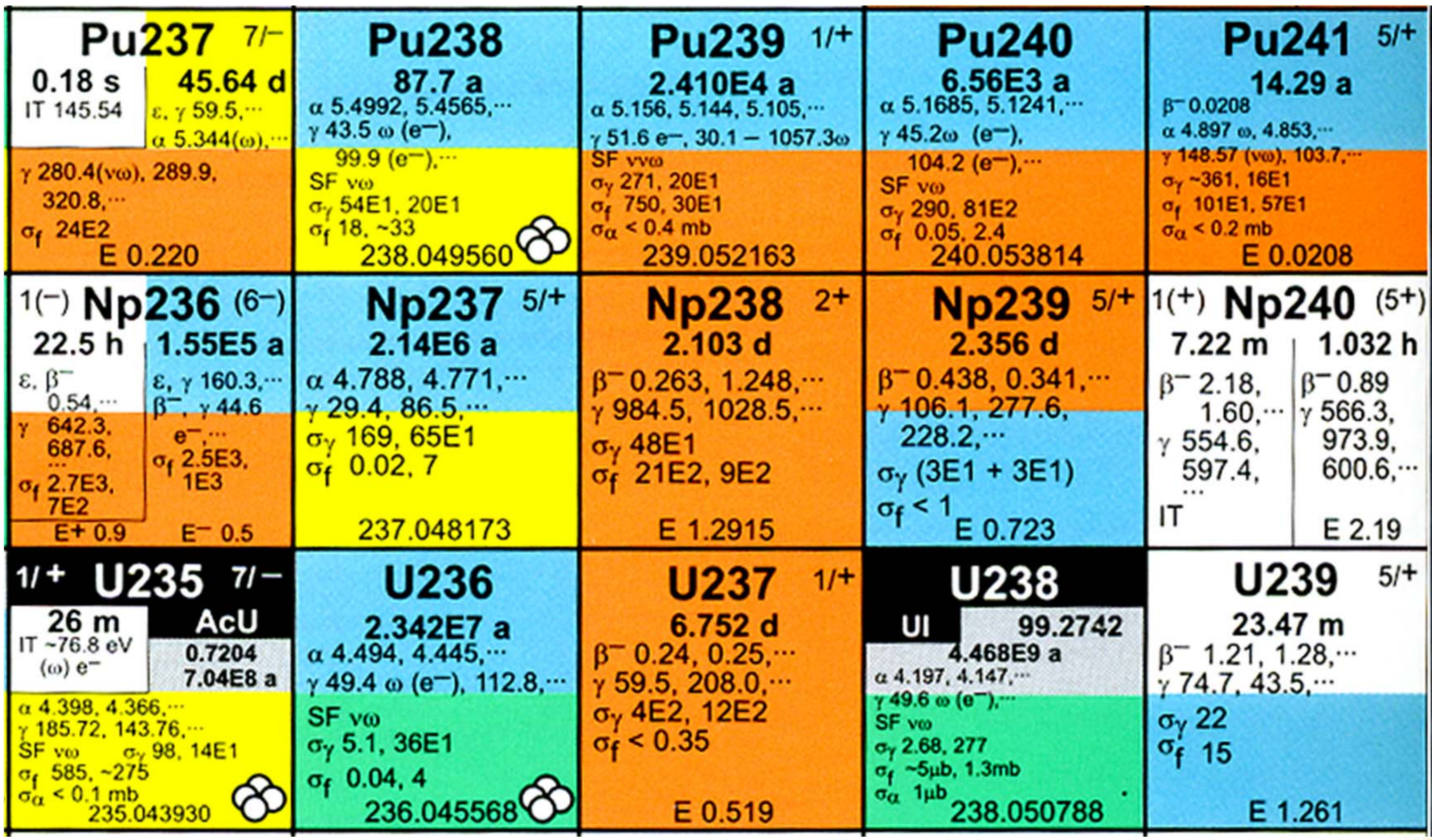

Chart of the Nuclides from KAPL

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Decay of Fission Products

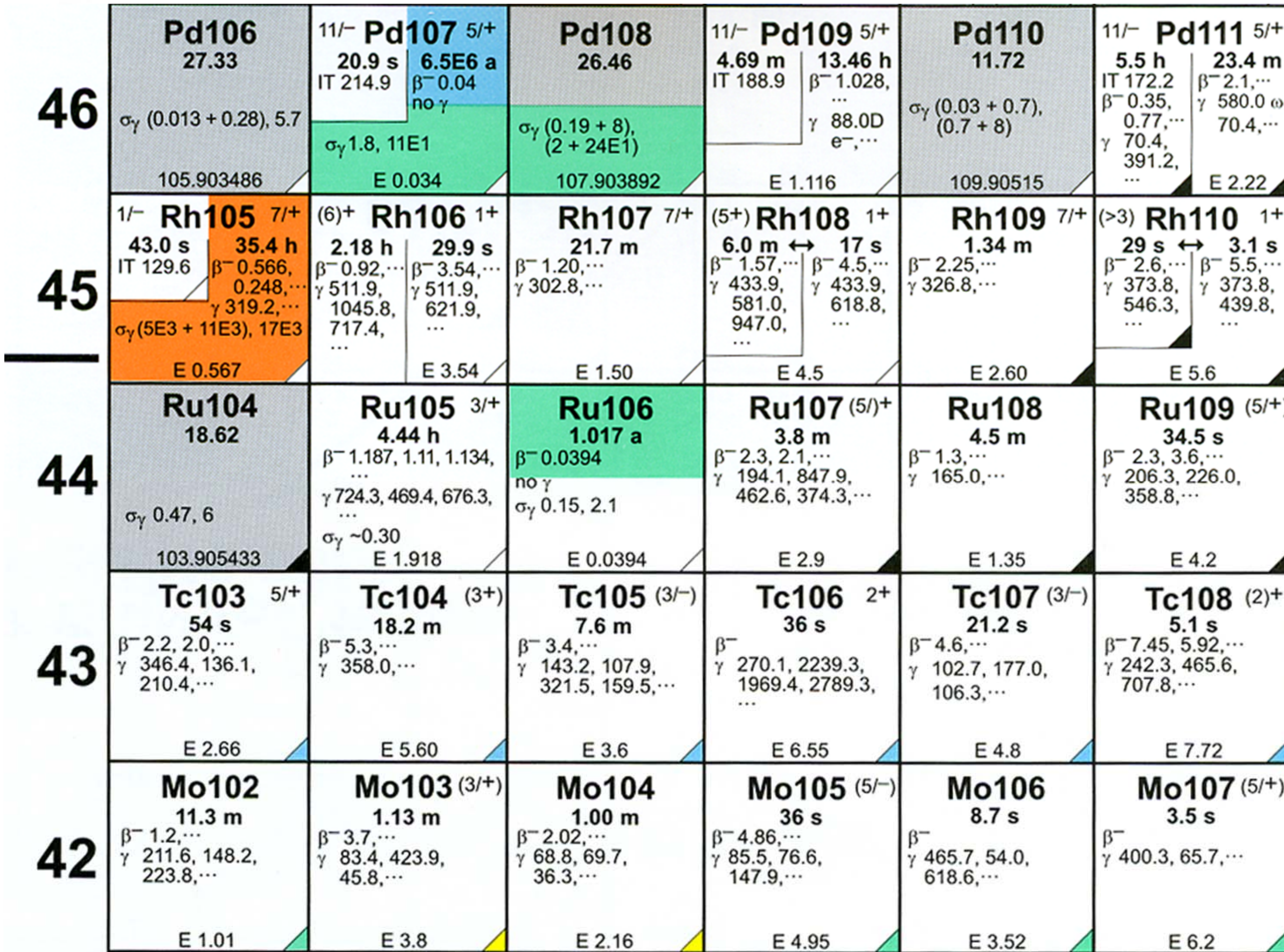

Chart of the Nuclides from KAPL

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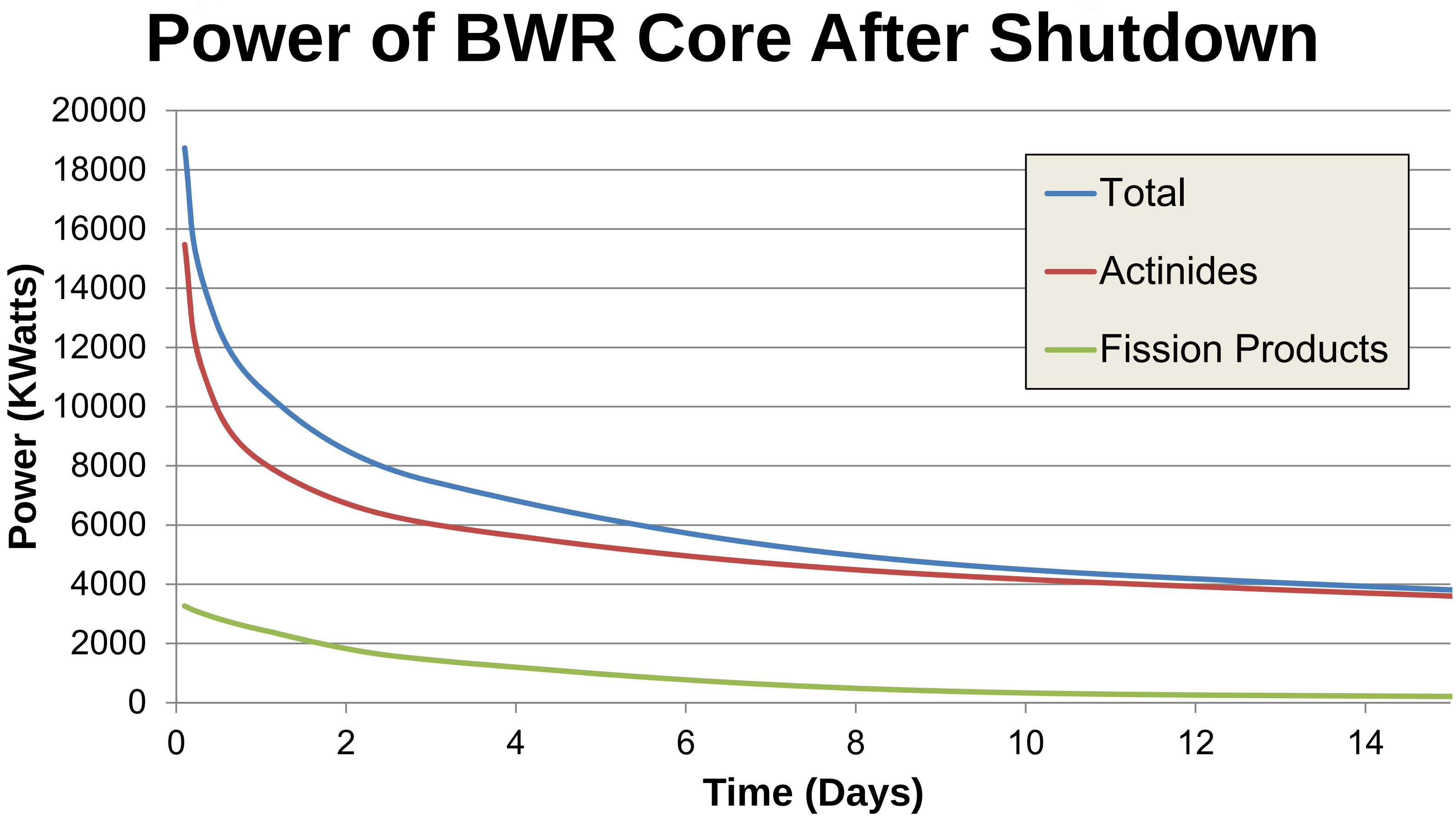
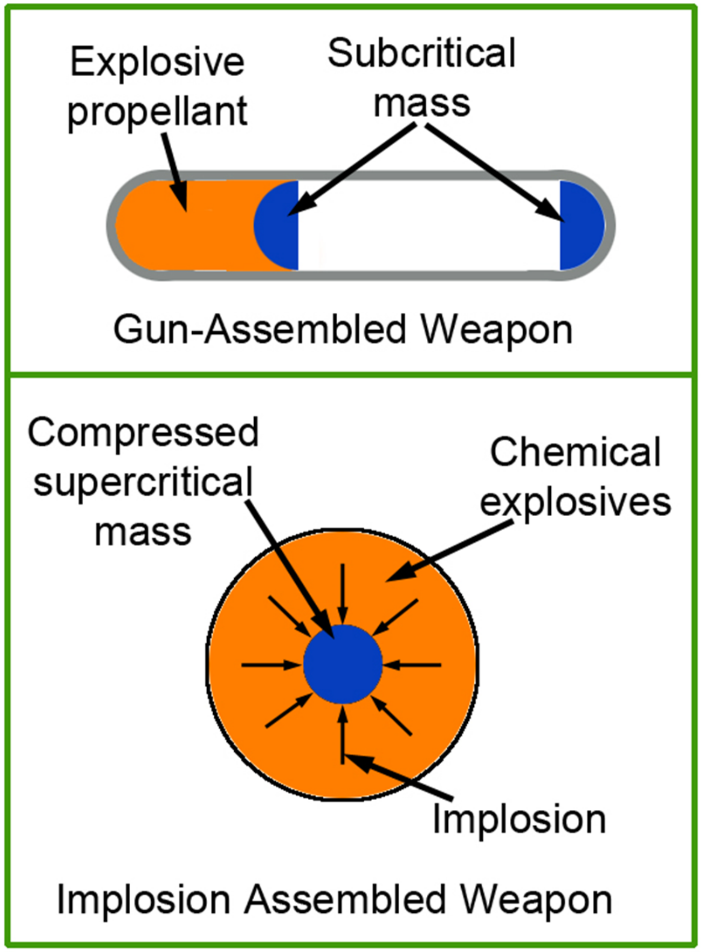
Decay Heat after Shutdown



Questions Before Break?

Pathways to a Nuclear Weapon

- To create a nuclear weapon you must first obtain weapons grade material.
  - Enriched uranium
  - Plutonium
- One of the most important methods to prevent a rogue state or group from creating a nuclear weapon is to prevent them from obtaining weapons grade material.



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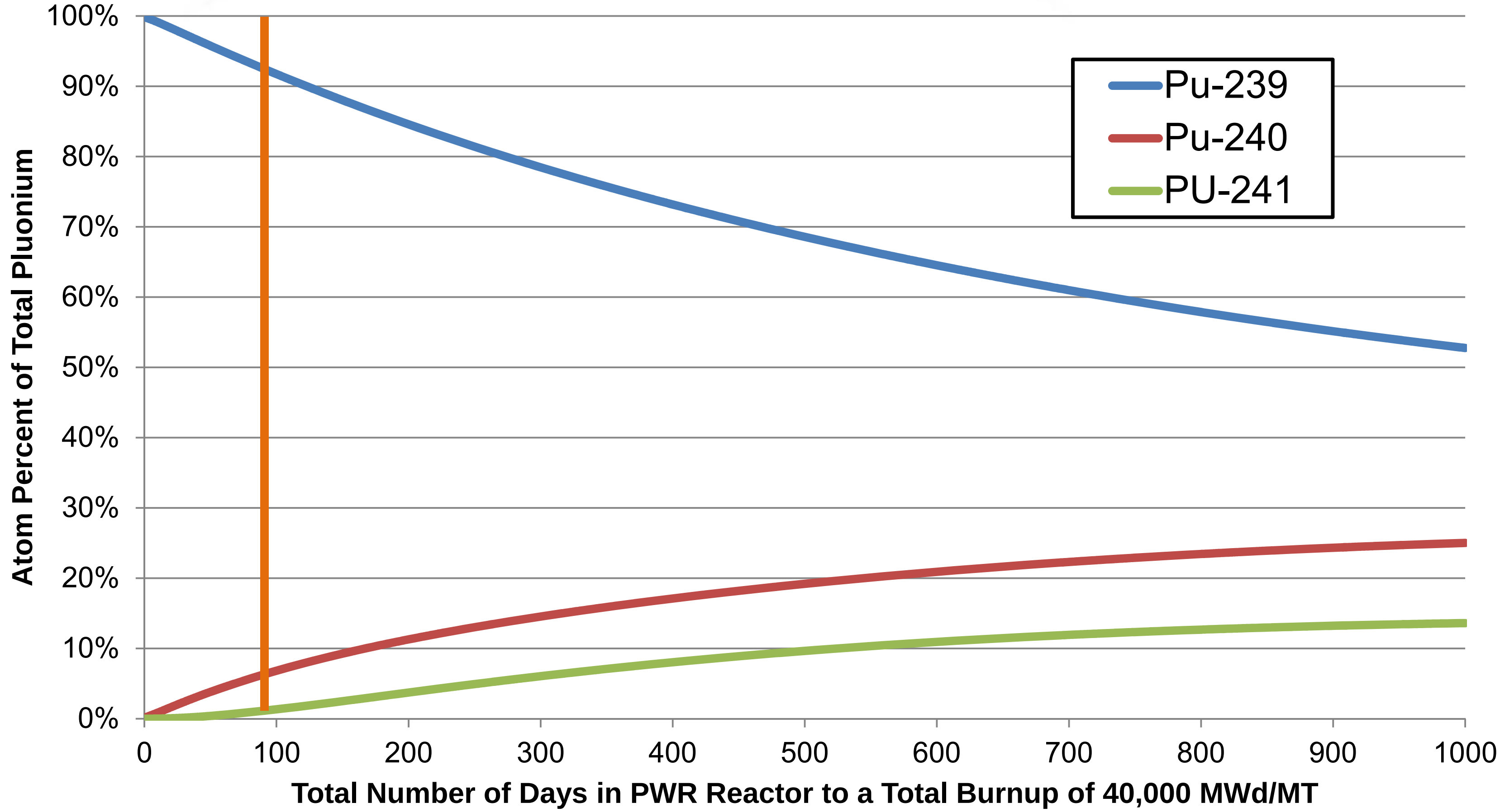
Plutonium Attribution

- The motivation may become higher for a state to maintain control of it’s plutonium stockpile if it knows it can be traced back to them.

Weapons Grade Plutonium



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Power vs Production Reactor

- Commercial Power Reactor
  - Purpose is to produce electric power for consumer use
  - Refueling occurs about every 18 months
  - Refueling requires a 20-45 day shutdown
- Production Reactor
  - Purpose is to produce weapons grade plutonium
  - Fuel is changed every few months
  - Some can refuel without shutting down

Reactor Depletion Codes

- ORIGEN 2
- SCALE 5 (ORIGEN-S, TRITON)
- MCNP w/ CINDER90
- Monteburns (MCNP and ORIGEN2)
- Attila
- WIMS
- Other specialized commercial codes

Inverse Modeling

Nuclide Monitor Measurements

Suspect Source Material

Burnup → Enrichment → Reactor Type → Cooling Time

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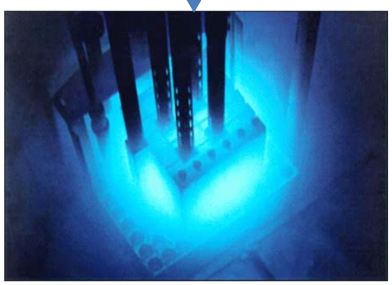
Spent Fuel Samples

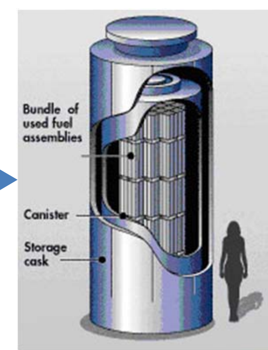
Fresh Fuel

?

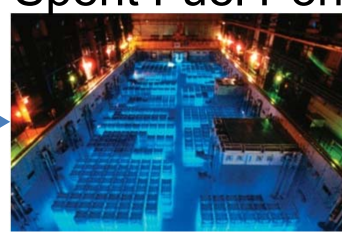


Reactor Core

Dry Storage Cask



Spent Fuel Pond



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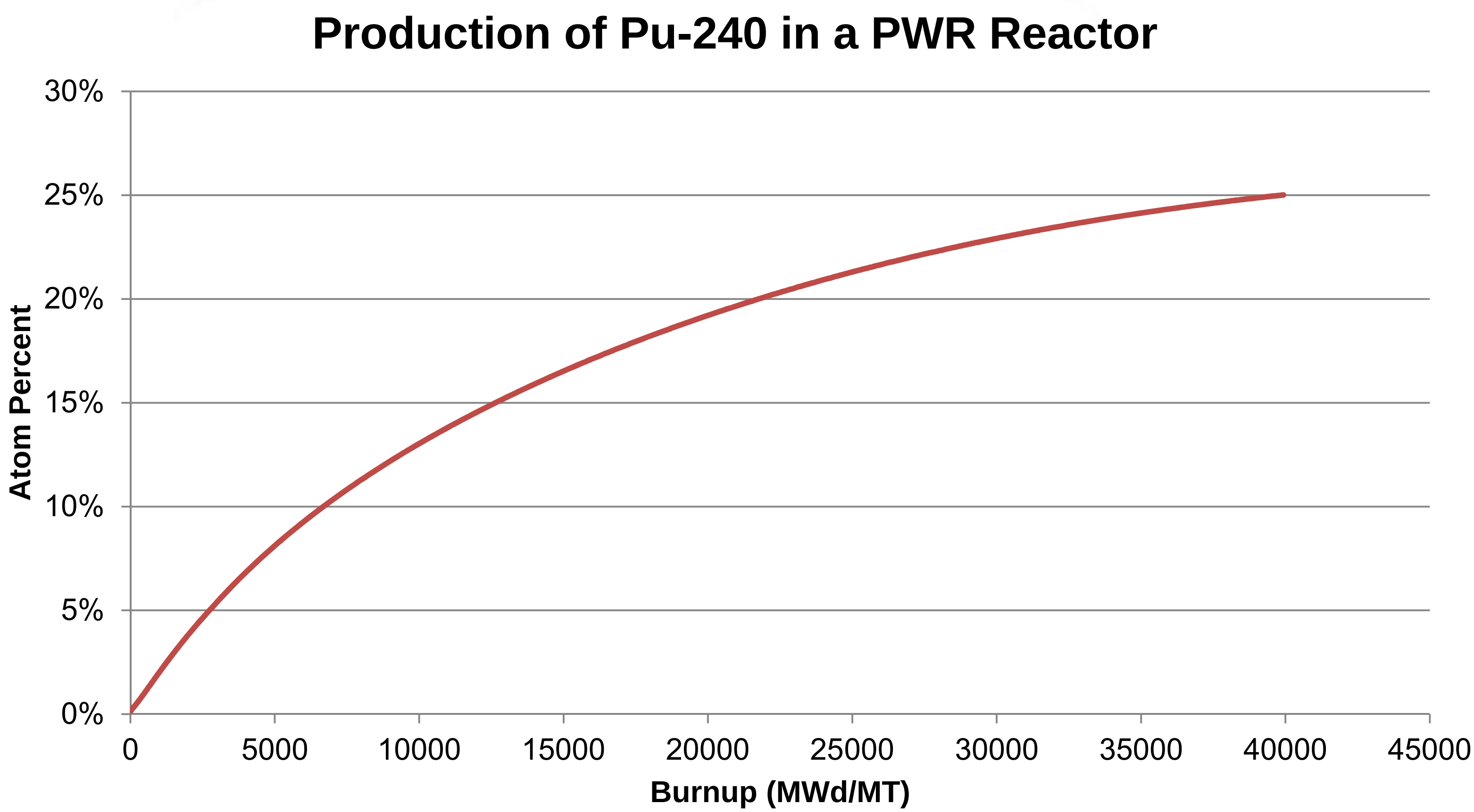
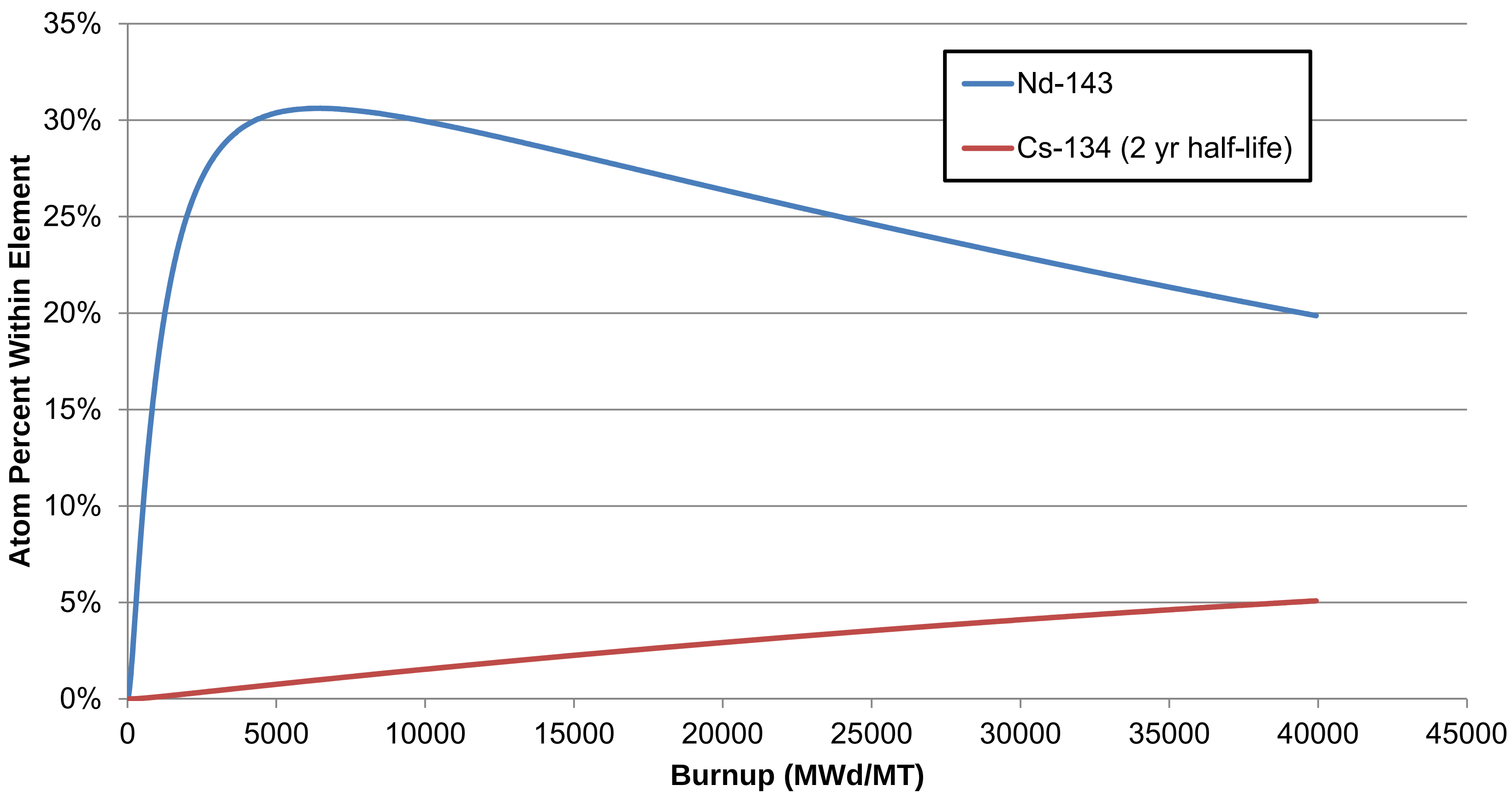
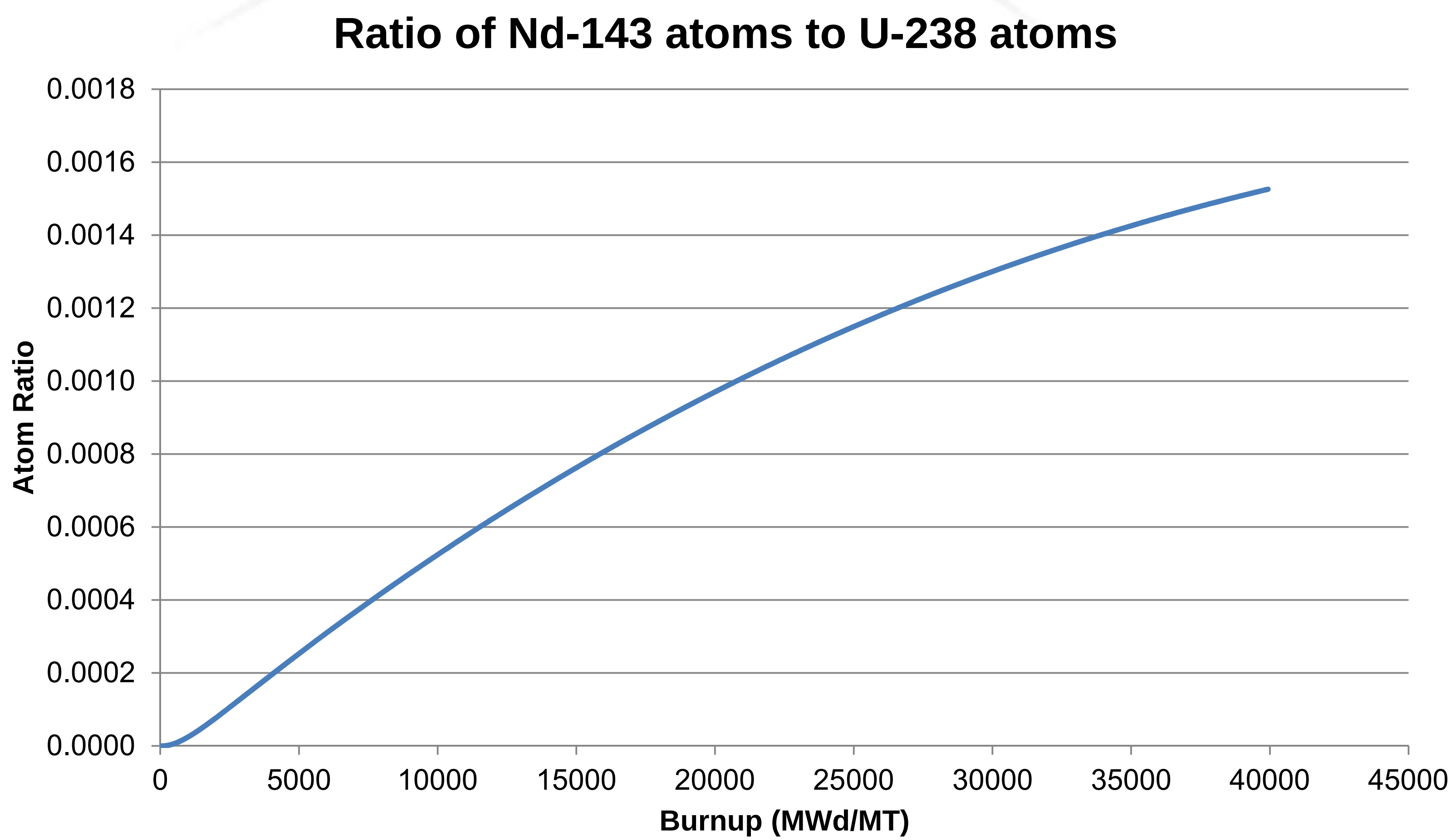
Nuclide Production as a Burnup Indicator



Not All Nuclides Are Appropriate



Ratios Are Your Friend



What is Burnup?

- A value that quantifies how much fission energy has been extracted from nuclear fuel.
- Most common units for burnup are:
  - MWd/MT (Megawatt days/ metric tonne)
  - kWd/kg (Kilowatt days / kilogram)
  - GWd/MT (Gigawatt days / metric tonne)

Burnup vs Number of Days in Reactor?

- Neutron flux is not flat across the reactor core.
- Fuel at different locations in the reactor experience different burn rates.
- Reactor operators may lower or raise the power level of a reactor.

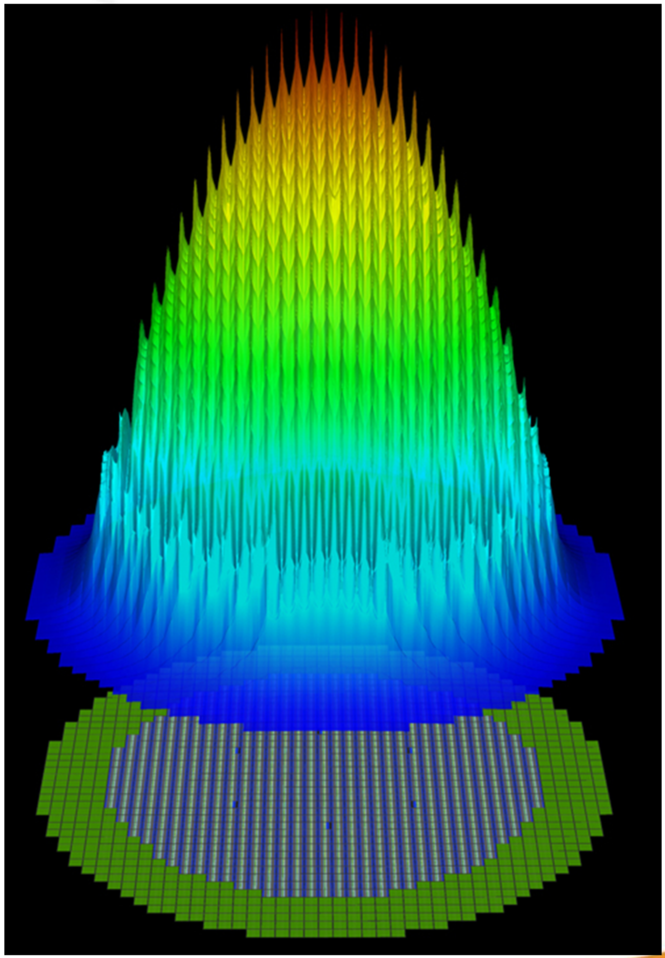


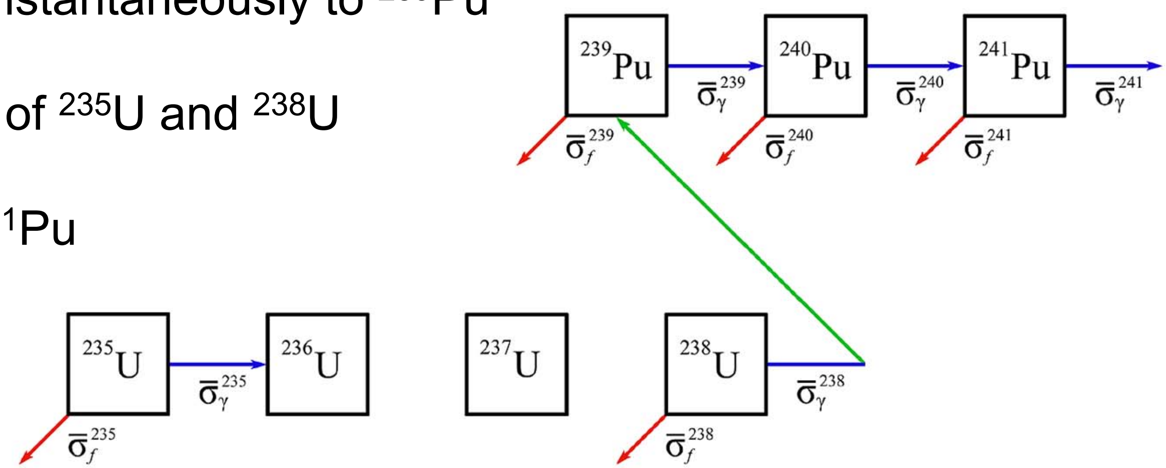
Figure from Argonne National Lab.

Initial Uranium Enrichment

The ratio of ^{235}U to U in the fuel before irradiation

Assumptions on fuel characteristics during irradiation:

- The only isotopes that fission are ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu
- ^{239}Np and ^{239}U decay instantaneously to ^{239}Pu
- There is no production of ^{235}U and ^{238}U
- Neglect the decay of ^{241}Pu



Burnup → **Enrichment** → **Reactor Type** → **Cooling Time**

Estimating the Initial Enrichment

$$H1 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U238}}{N_o^U}\right]\left(\left[\frac{N^{U235}}{N^{U238}}\right] + \left[\frac{N^{U236}}{N^{U238}}\right]\right)$$

$$H2 = -M_o^U BU(T) \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241}$$

$$H3 = N_a E_R \bar{\sigma}_f^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H4 = -N_a E_R \bar{\sigma}_f^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U234}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H5 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U239}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H6 = -N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H7 = -N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U234}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H8 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U240}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H9 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U239}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H10 = -N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241} \left[\frac{N^{U234}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H11 = N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H12 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_f^{Pu241} \left[\frac{N^{U241}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H13 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241} \left[\frac{N^{U240}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H14 = -N_a E_R \bar{\sigma}_a^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241} \left[\frac{N^{U239}}{N^{U238}}\right]\left[\frac{N^{U238}}{N_o^U}\right]$$

$$H15 = N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241} \left(1 - \left[\frac{N^{U238}}{N_o^U}\right]\right)$$

$$H16 = \bar{\sigma}_f^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} - \bar{\sigma}_a^{U238} \bar{\sigma}_a^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241}$$

$$H17 = \bar{\sigma}_\gamma^{U238} \bar{\sigma}_f^{Pu239} \bar{\sigma}_a^{Pu240} \bar{\sigma}_a^{Pu241} - \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_f^{Pu240} \bar{\sigma}_a^{Pu241}$$

$$H18 = \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241}$$

$$e_o = \frac{(H1 + H2 + \ldots .H15)}{H16 + H17 + H18}$$

Burnup → **Enrichment** → Reactor Type → Cooling Time

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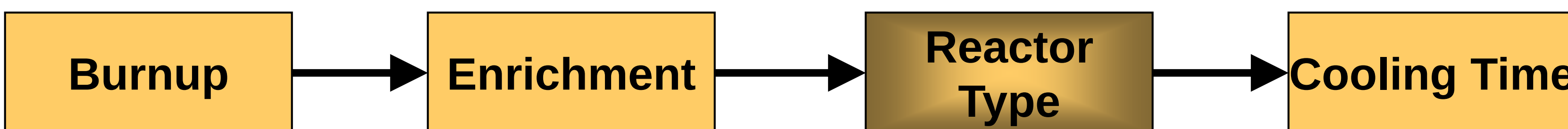
Reactor Type

Reactor Type:

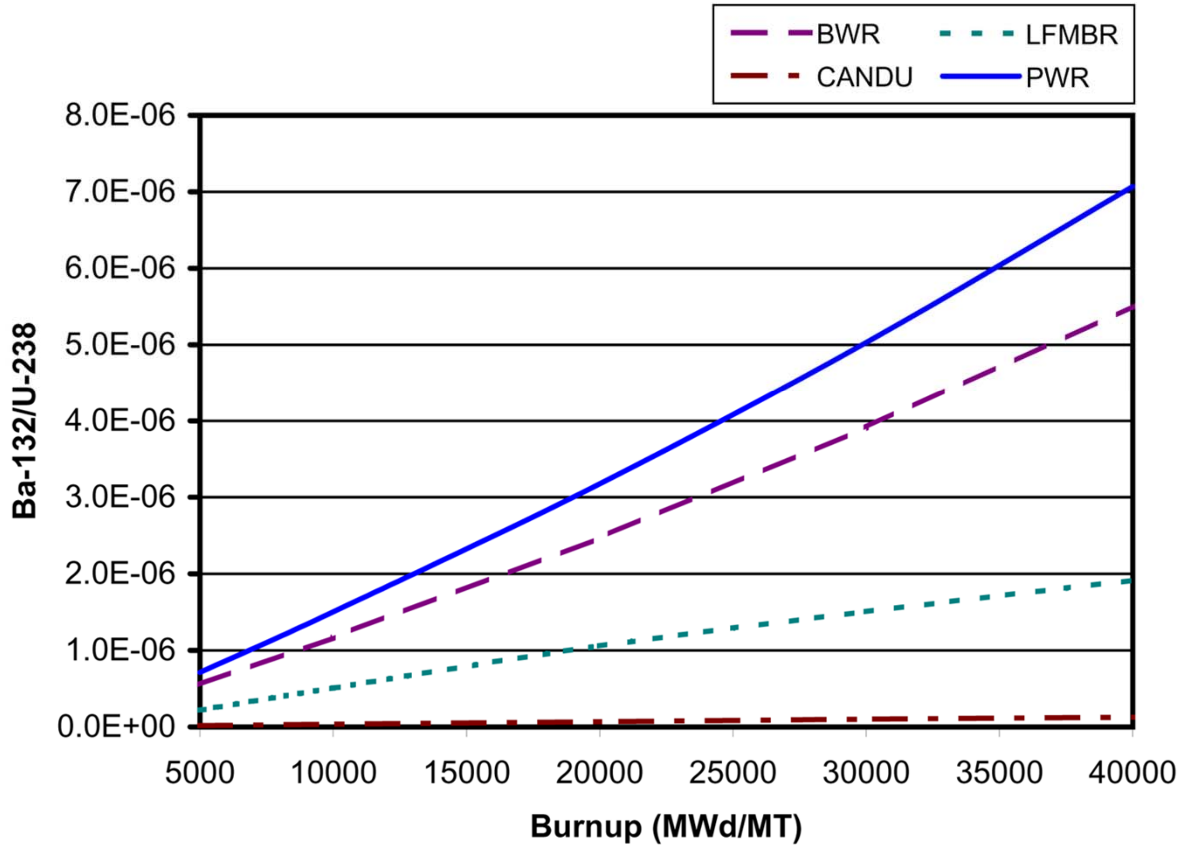
Nuclear reactors that share a common design concept
(e.g. PWR, BWR, CANDU, LMFBR)

Reactor Type Indicator Requirements:

- Fission yield and/or absorption rate changes for each reactor type
- Stable or long-lived nuclides



Reactor Type Indicator



Burnup → **Enrichment** → **Reactor Type** → **Cooling Time**

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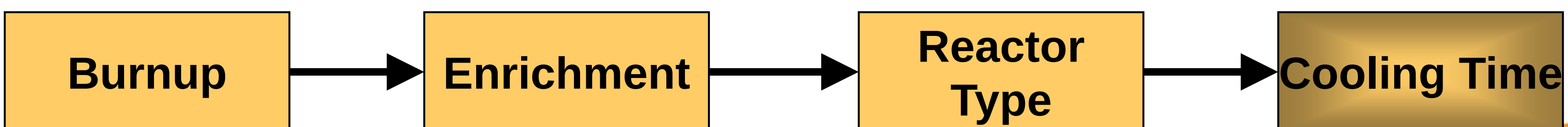
Fuel Age or Cooling Time

Cooling Time:

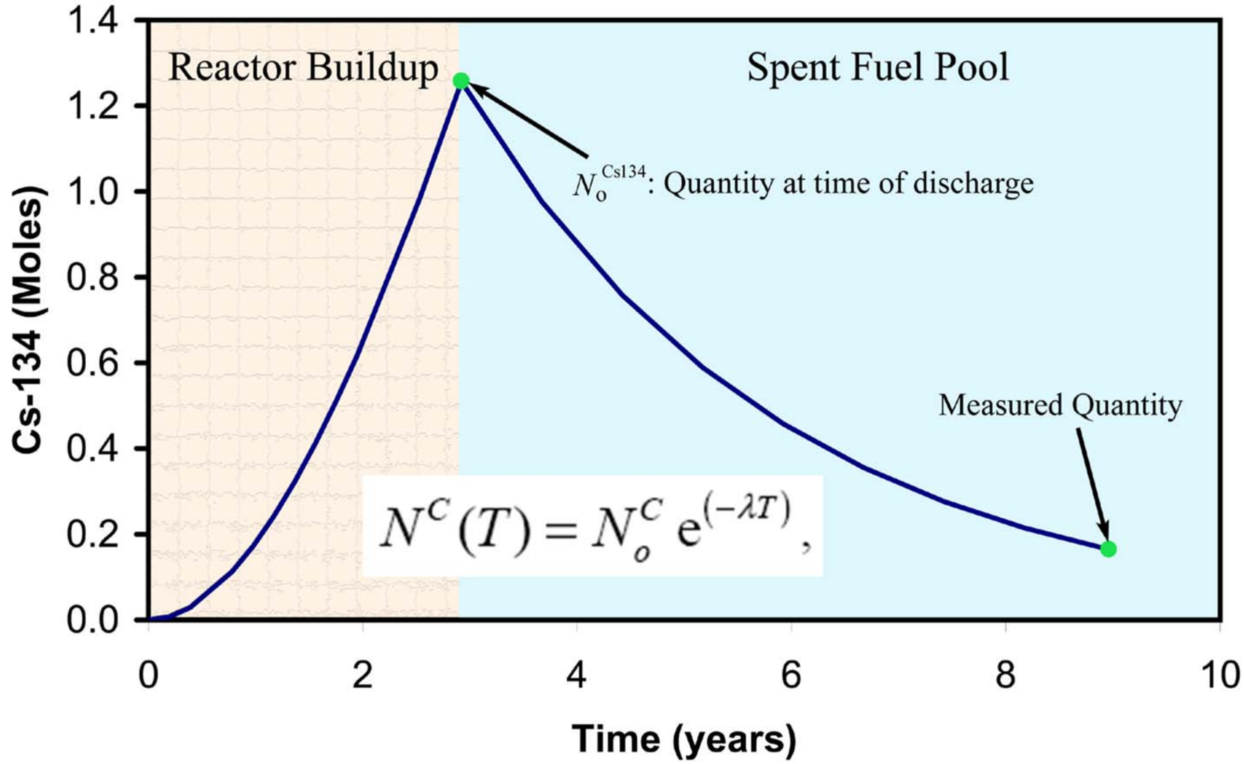
Cooling time is the period from the end of irradiation to the time of measurement

Cooling Time Monitor Requirements:

- Half-life is between 1-30 years
- At least 0.01 moles is produced per MT of fuel



Fuel Age or Cooling Time



Burnup → Enrichment → Reactor Type → **Cooling Time**

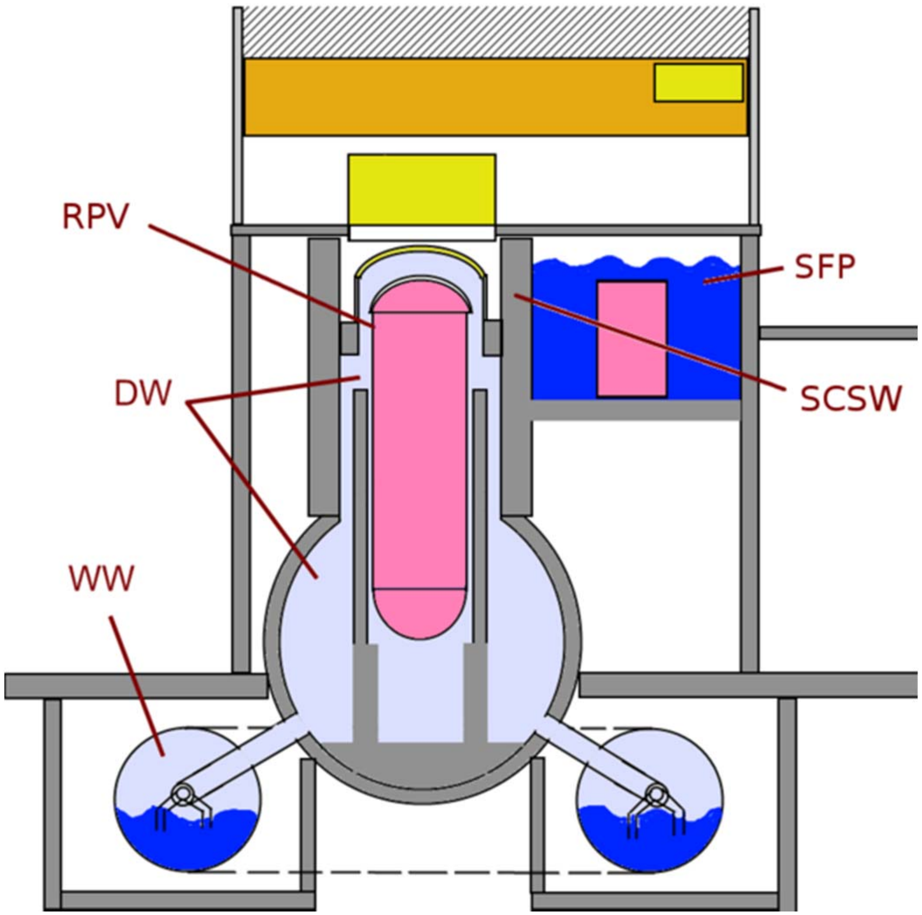
How Does it Help?

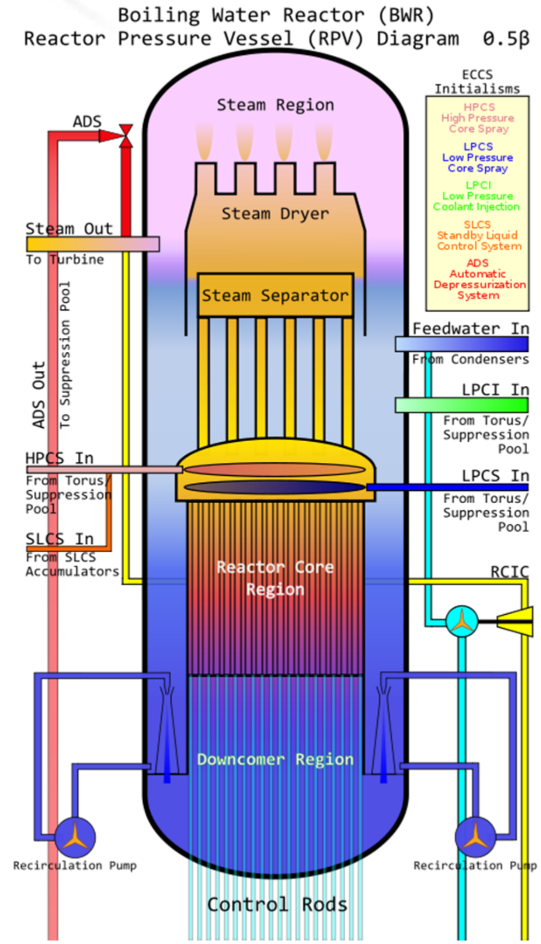
- Burnup
  - Quality and quantity of plutonium
  - Production vs commercial reactor
- Initial Enrichment
  - Natural vs light water reactor
  - Commercial reactors use between 3-5% enriched fuel
- Reactor Type
  - Countries tend to use only one or two reactor types
- Cooling Time
  - Determine how radioactive the fuel is
  - Estimate when was the fuel was discharged (fuel pond vs dry storage)

Fukushima Reactor Accident

- Fukushima Dai-ichi plant consists of 6 reactors.
- Three were operating and three were already shutdown before the earthquake.
- Earthquake struck at 2:46pm March 11 2011.
- Reactors were automatically shutdown, core still required power to cool the core from decay heat.
- Approximately 60 minutes later a 14-15 meter high tsunami destroyed the plant's backup generators and disabled the cooling systems.

Fukushima Reactor Building Layout





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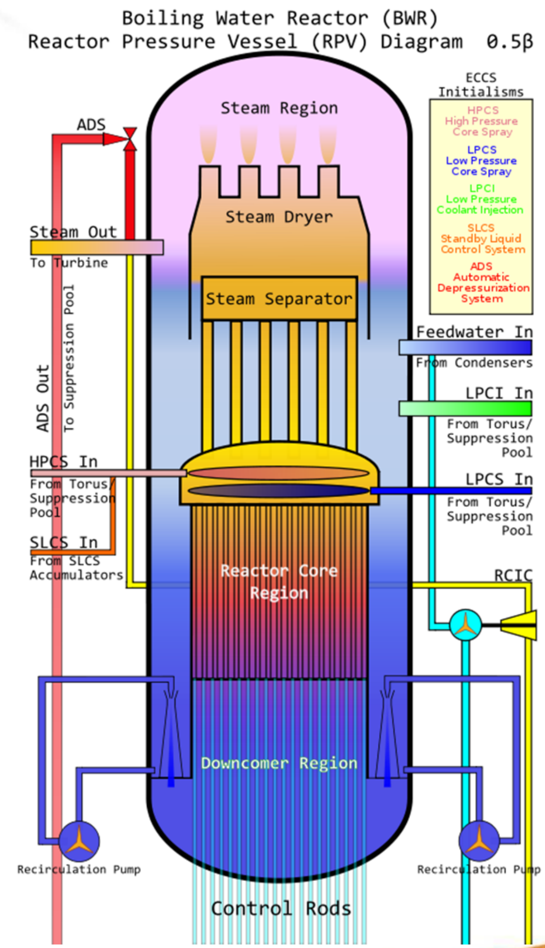
Pressure Relief

- After the cooling failed the main steam isolation valve closed (MSIV).
- Pressure built up in the pressure vessel as the water turned to steam.
- The steam needed to be vented from the pressure vessel to allow an injection of water.



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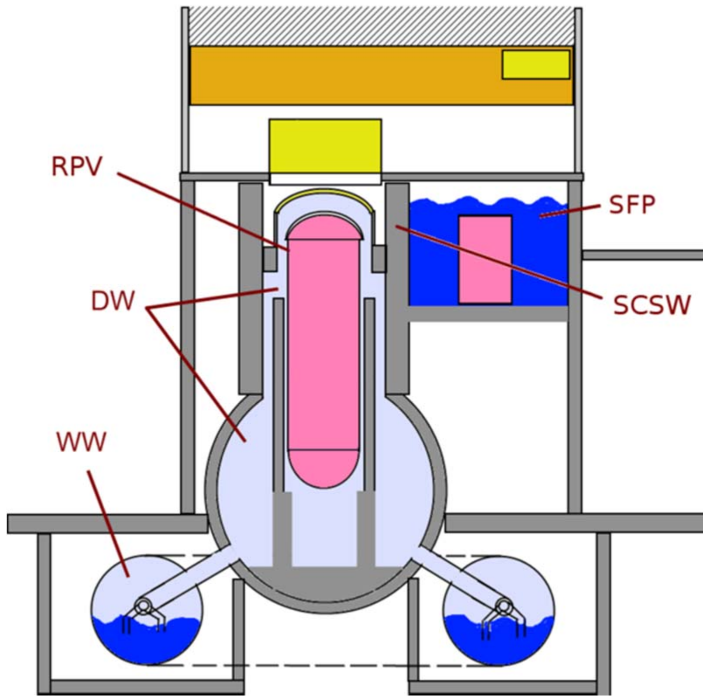
Fog of War

- No power to the reactor control room, no instruments were available to monitor the reactor cores.
- TEPCO and the Japanese government were trying to manage the crisis while in an almost complete blackout of information.
- There were no plans in place to address such a catastrophic failure.

Photograph from Japan Nuclear and Industrial Safety Agency

Critical Questions

- What was the water level within each of the reactor's pressure vessels?
- Has any nuclear fuel melted within the cores?
- What was the water level of the spent fuel ponds?
- Has the spent fuel ponds ignited?
- Where to use the small amount of battery power available?
- Where to use the fire truck pumps that were being deployed?



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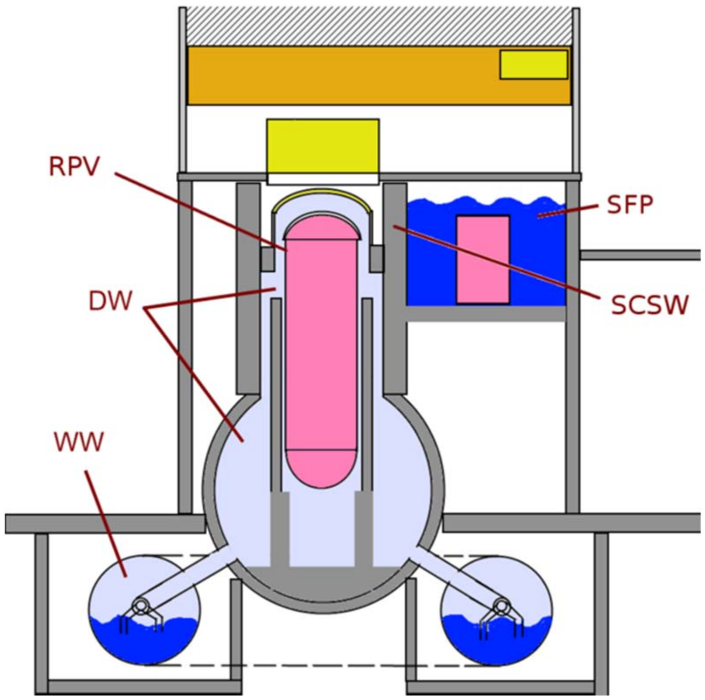
Steam Release From Fukushima

- What can be measured that might answer some of the questions being asked?
  - Steam was released that had some radioactive elements
- What radioactive debris would be expected in steam?
  - Water soluble elements
  - Fission products
  - Activated structural components

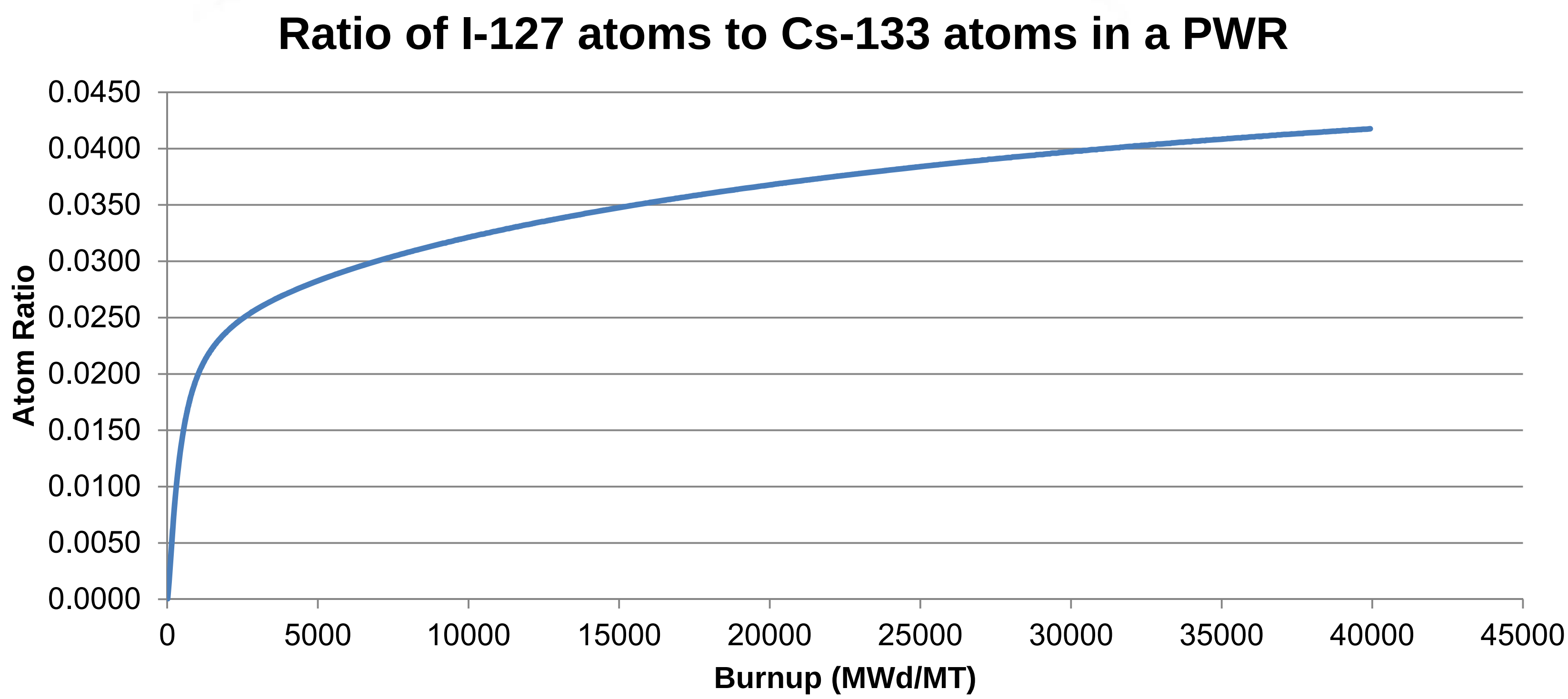


Iodine and Cesium

- The radioactive material released from Fukushima was mostly iodine and cesium.

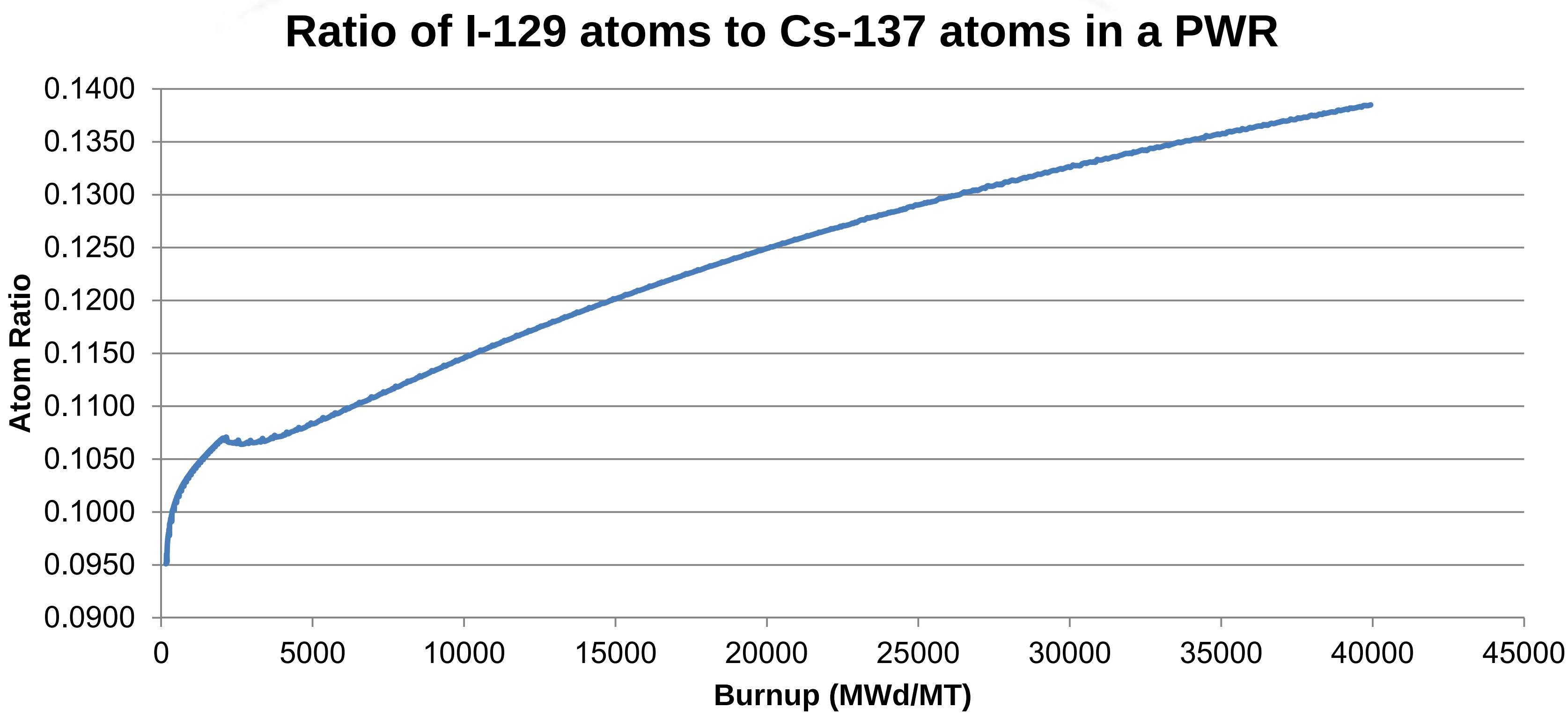
Nuclear Safety Commission (NSC) of Japan published the results of the total amount of radioactive materials released into the air during the accident at the Fukushima Daiichi Nuclear Power Station. “The total amounts released between 11 March and 5 April were 1.3×10^{17} Bq for iodine-131 and 1.1×10^{16} Bq for caesium-137, which is about 11% of Chernobyl emissions.”

Background Nuclides



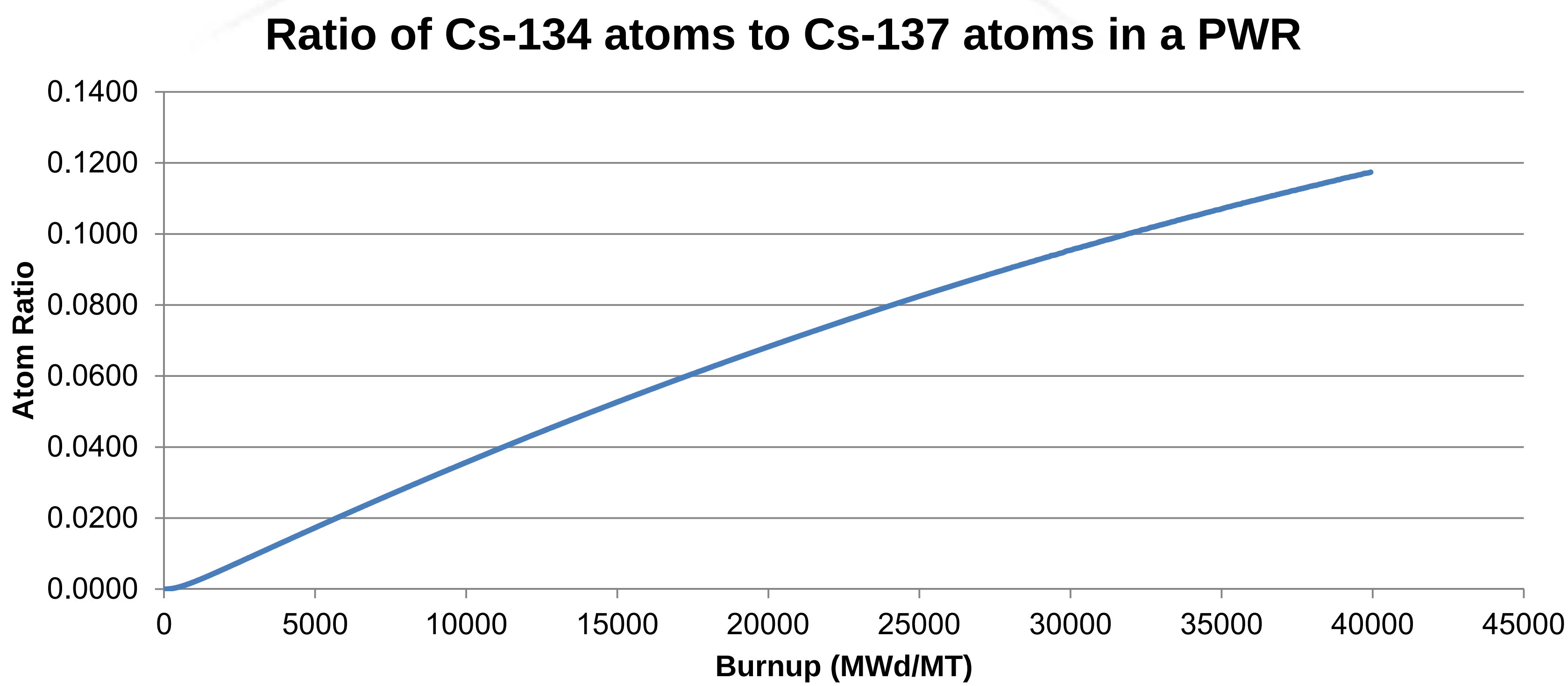
- Naturally occurring iodine and cesium is 100% I-127 and Cs-133.

Elemental Fractionation



- Elements may transport differently in the environment causing elemental fractionation.

Nuclide Ratios Within the Same Element

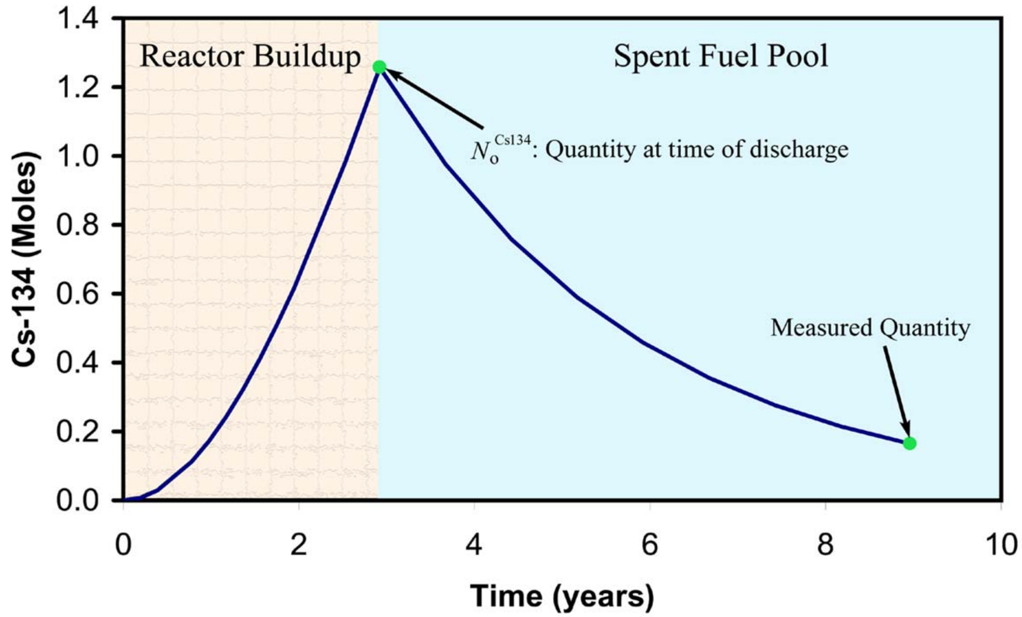


- Not a perfect burnup indicator due to half-lives.
  – Cs-134 has a 2 year half-life
  – Cs-137 has a 30 year half-life

Ground vs Air Sampling

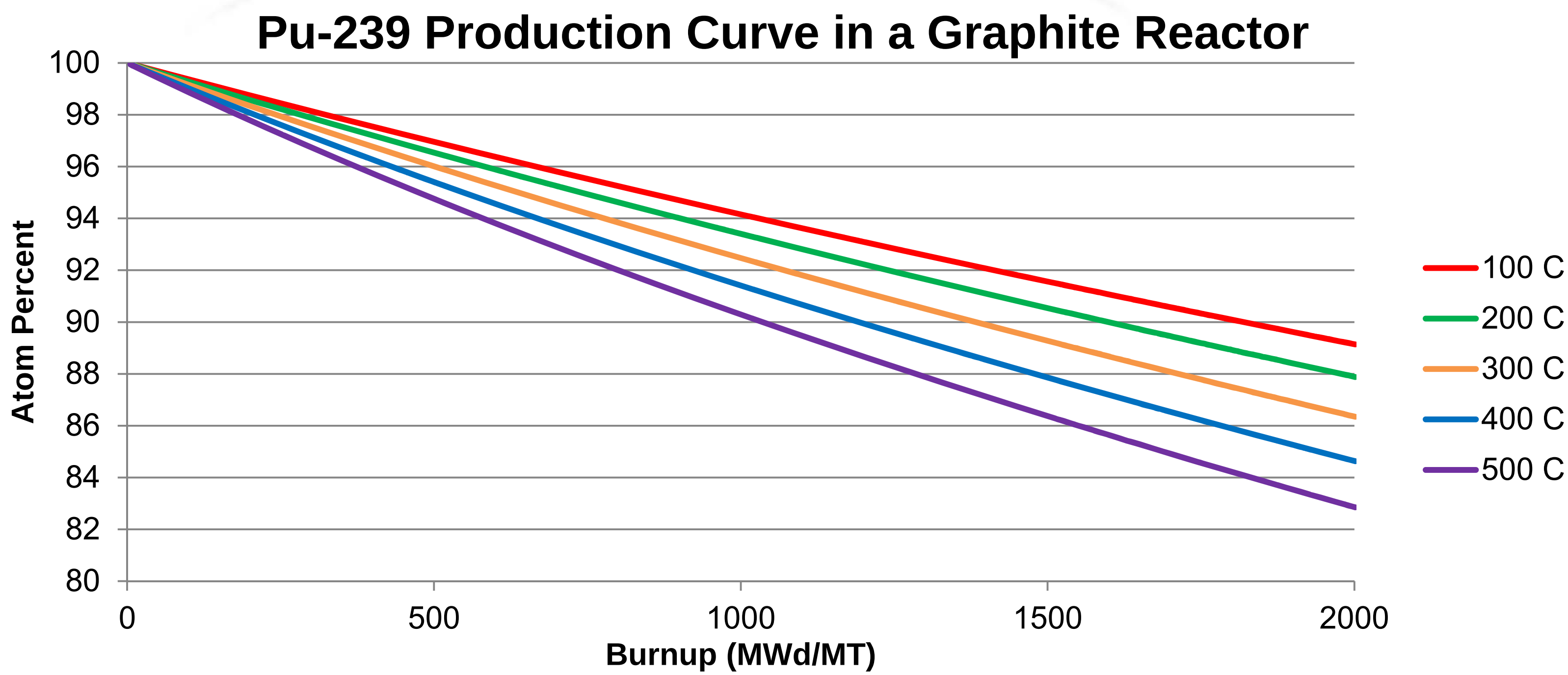
- Soil samples sent to a laboratory
  - Mass spectrometry
  - High-purity germanium detectors (HPGe)
- Onsite or In-Situ ground detection with NaI detectors
- Air Sampling
  - Real time with gamma detectors
  - Air filters collected for mass spectrometry
- Different sampling methods may be biased towards different nuclides

Spent Fuel Pond vs Reactor Core



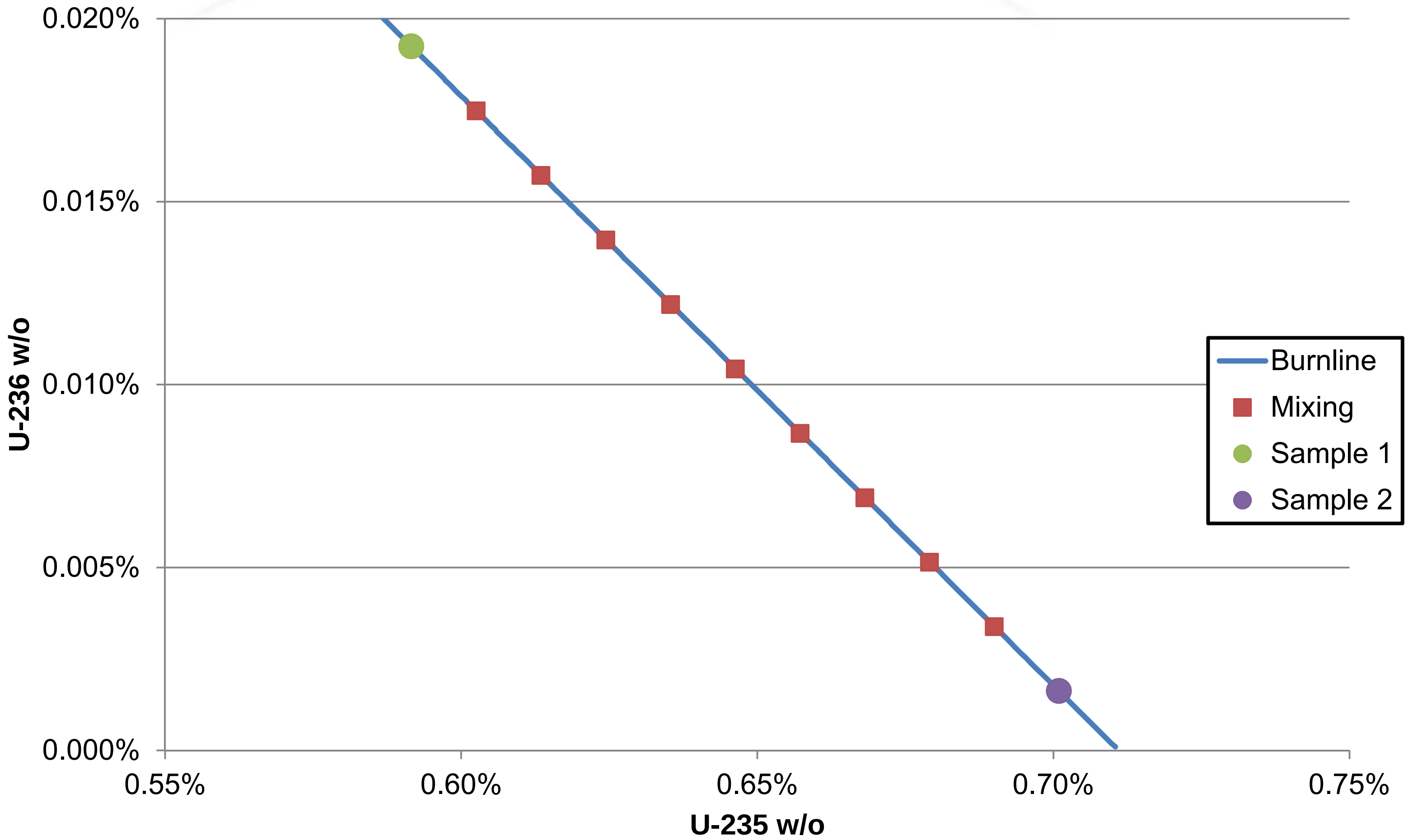
- Cs-136 has a 2 day half-life
- Ratio of Cs-137 atoms to Cs-136 would indicate if the radioactive material is coming from the core, the spent fuel pond, or a mixture.

Temperature Effects



- As the moderator changes temperature the neutron energy changes, which changes the capture and fission rates for a reactor.

Uranium Mixing Between Samples



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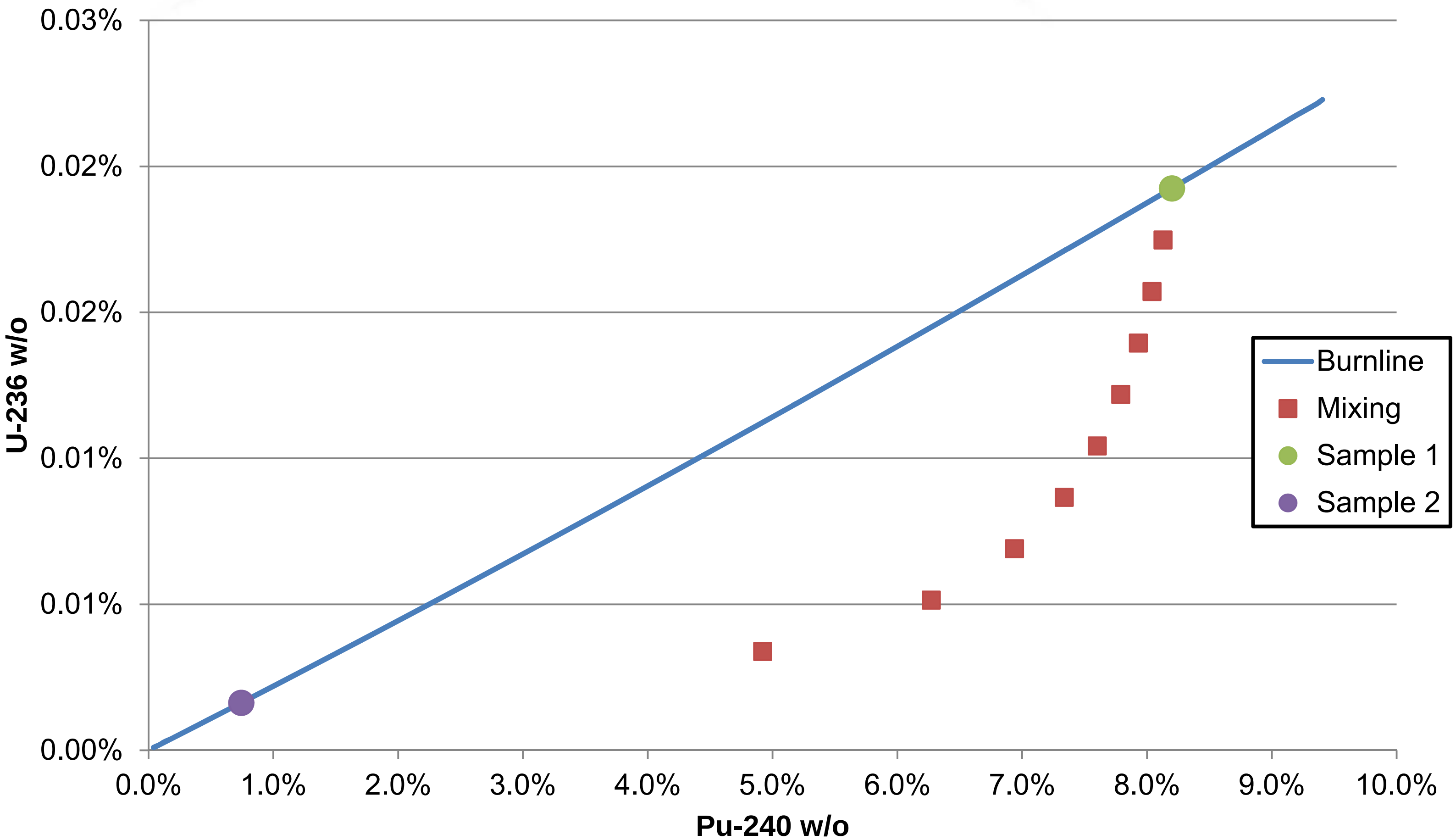
Plutonium Mixing Between Samples



Mixing of Plutonium Ingots

- To obtain the correct plutonium isotopic specifications for a nuclear weapon; a number of ingots may be mixed.
- The ingots may come from spent fuel or recycled plutonium from dismantled nuclear weapons.
- The Americium built up in the plutonium from Pu-241 decay will be removed, which resets the age clock.

Pinpointing the Source of a Pu Ingot

Would it be possible to pinpoint the position in a reactor where plutonium in a nuclear weapon came from?

Future Work in Reactor Forensics

- Relatively few real-world use cases
- Not a high-profile field of science
- High demand when events occur
- A lot of work still needs to be done