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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!

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# 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

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# 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

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# Nuclear Reactor Fuel

Fresh Fuel



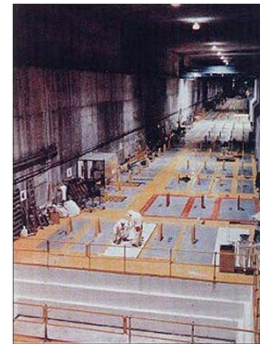
MOX Fuel



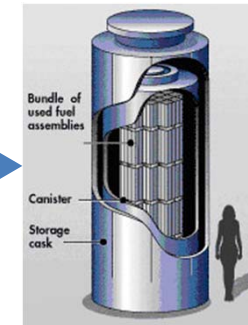
Plutonium



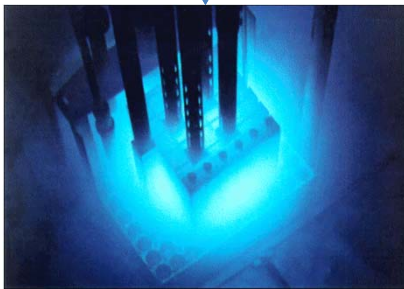
Reprocessing



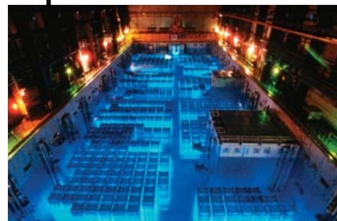
Dry Storage Cask



Reactor Core



Spent Fuel Pond



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# 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.

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# 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

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# 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.

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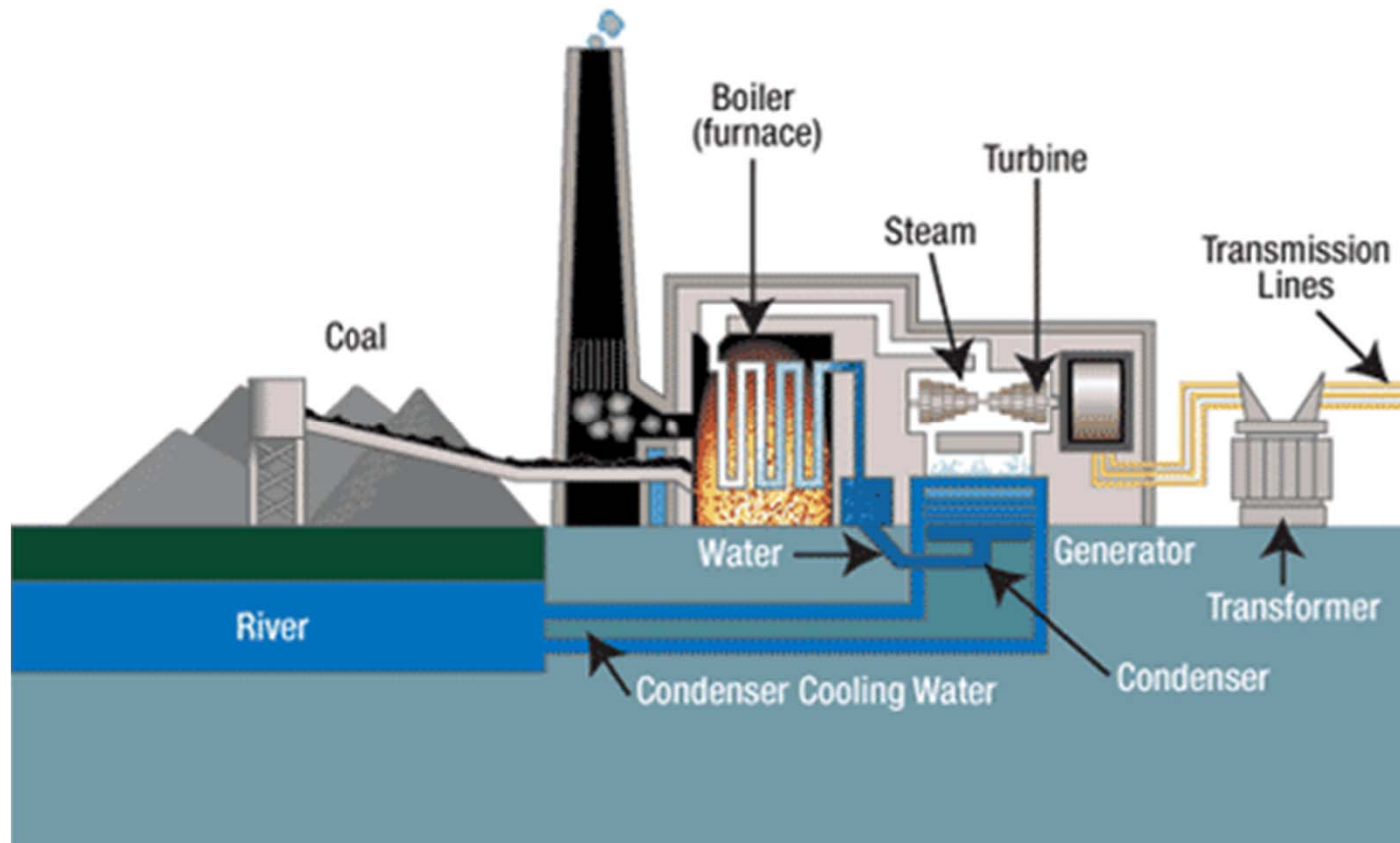


# Clip from Sum of All Fears

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# Electric Power Generation

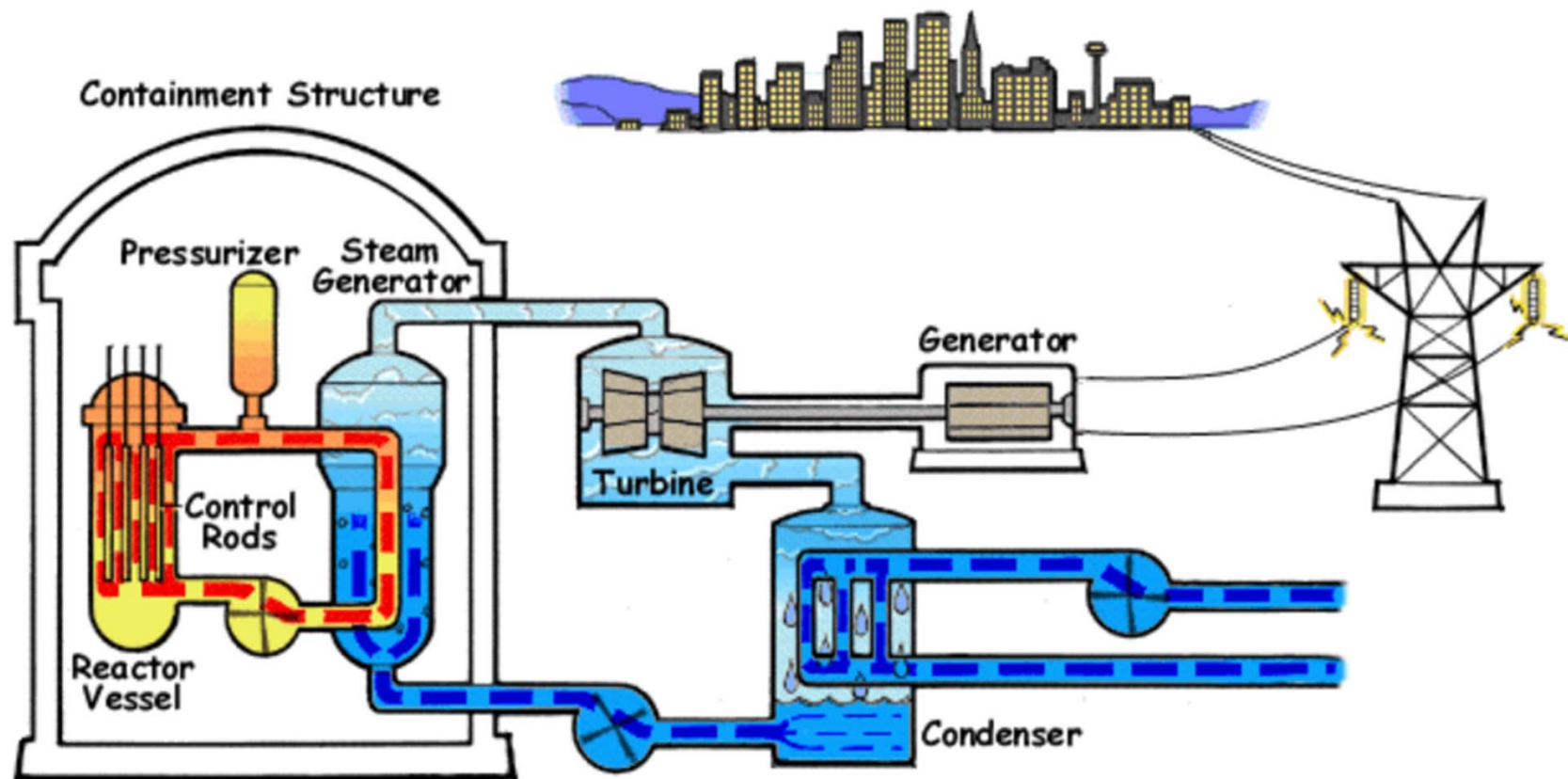


*Coal-fired power plant, diagram from Tennessee Valley Authority*

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# Pressurized Water Reactor (PWR)



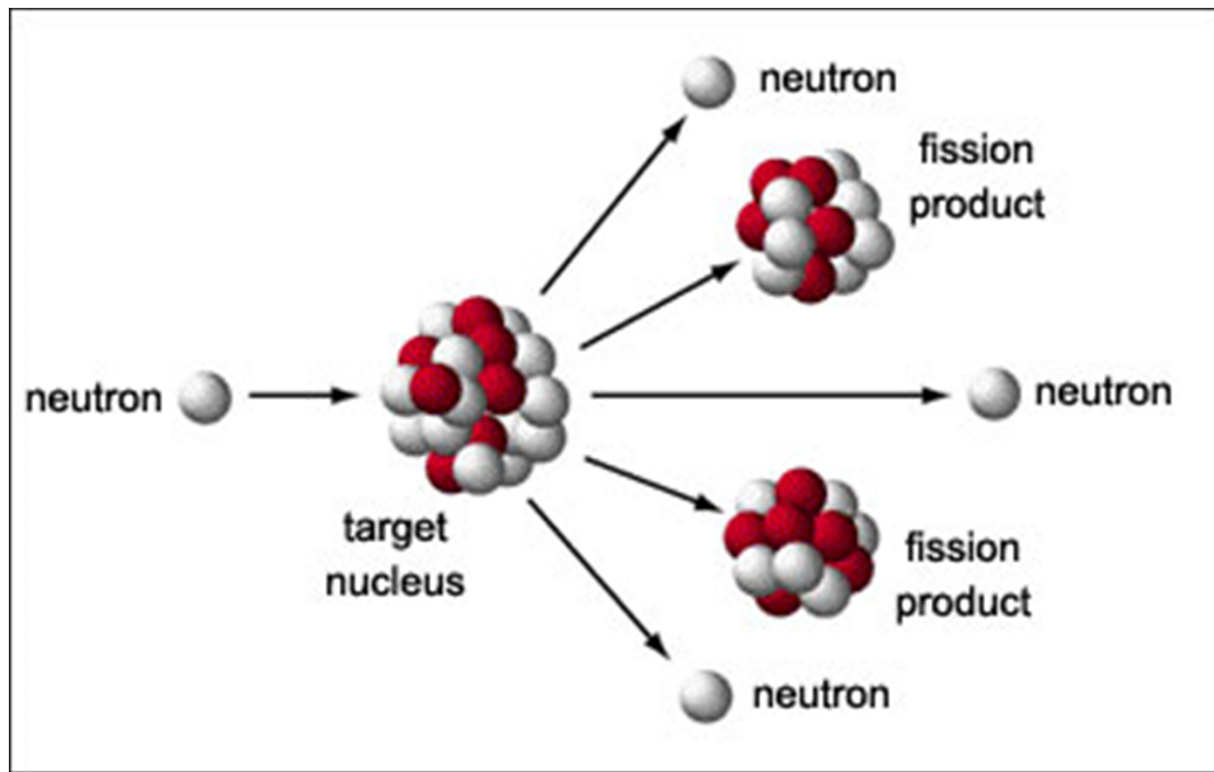
*Pressurized Water Reactor from the NRC website*

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# 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)
$\gamma$ (Gamma Rays)
Protons
Alpha
Beta
Neutrons (2n,3n,4n)
Fission products

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# 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.

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# Splitting a Coconut



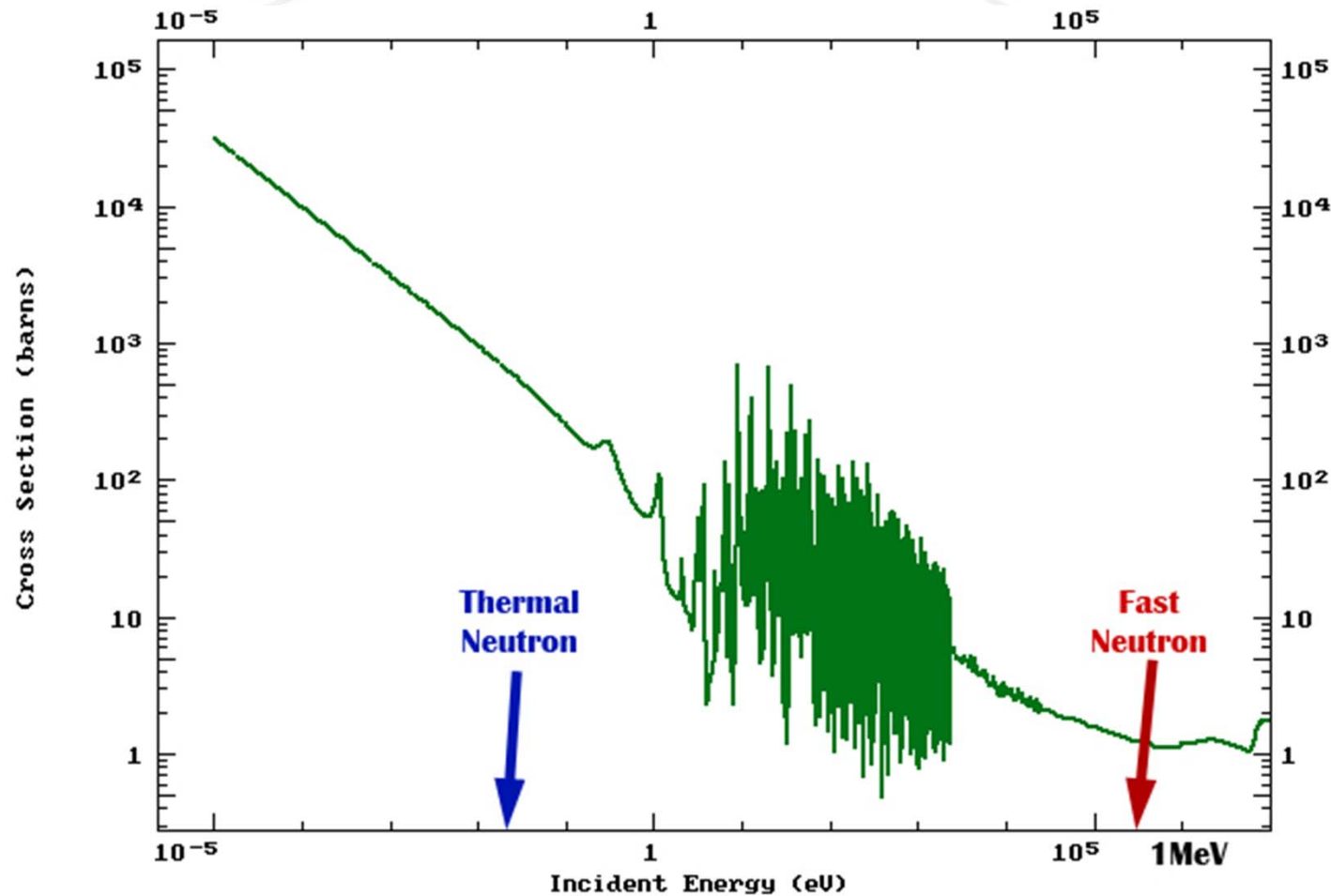
Increasing the velocity of the hammer striking the coconut has what kind of effect on the probability it will split?

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# U-235 Neutron Fission X-Section

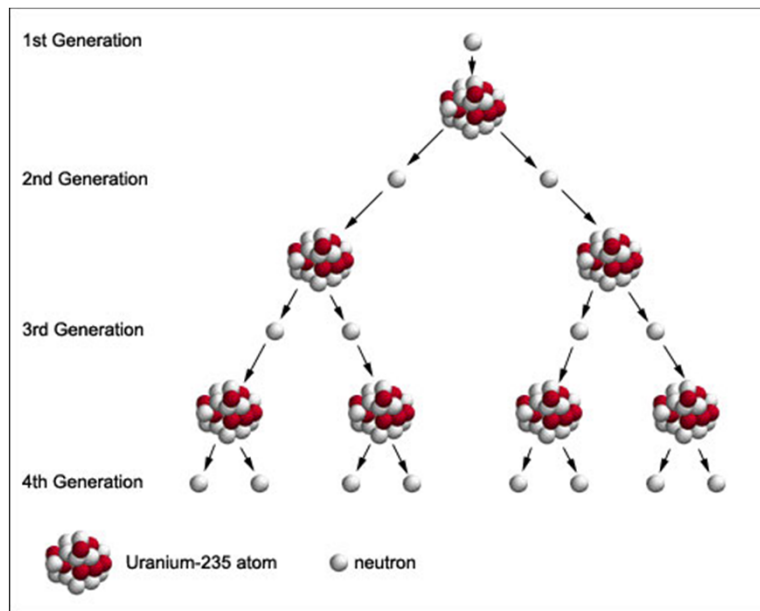


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# Energy Release From Each Fission



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**

Fission Reaction from the Atomic Archive website

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

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# How to Slow Down a Neutron

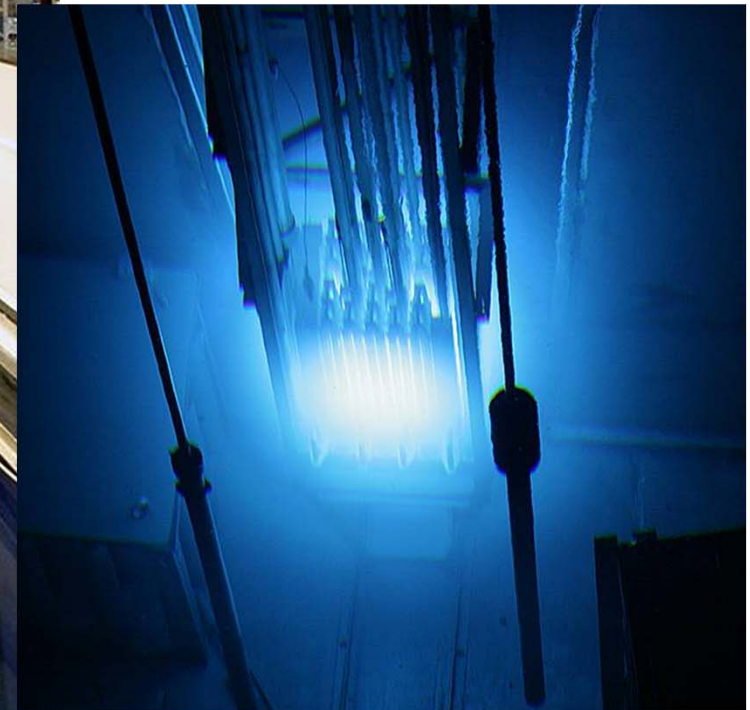
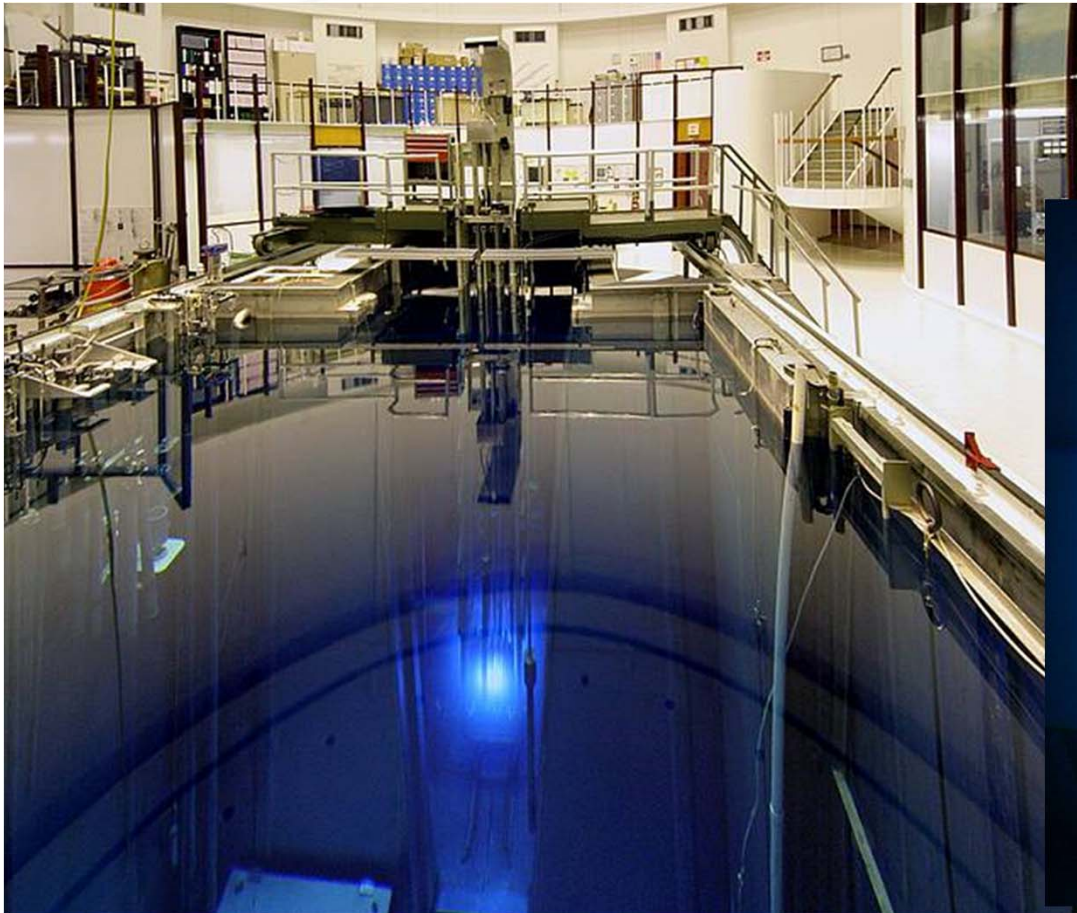
- Ideal neutron moderator
  - Low Z Material
  - High scattering x-section
  - Low absorption x-section
- Examples of neutron moderators
  - Water
  - Heavy water
  - Concrete
  - Graphite
  - Polyethylene

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# Texas A&M TRIGA Reactor

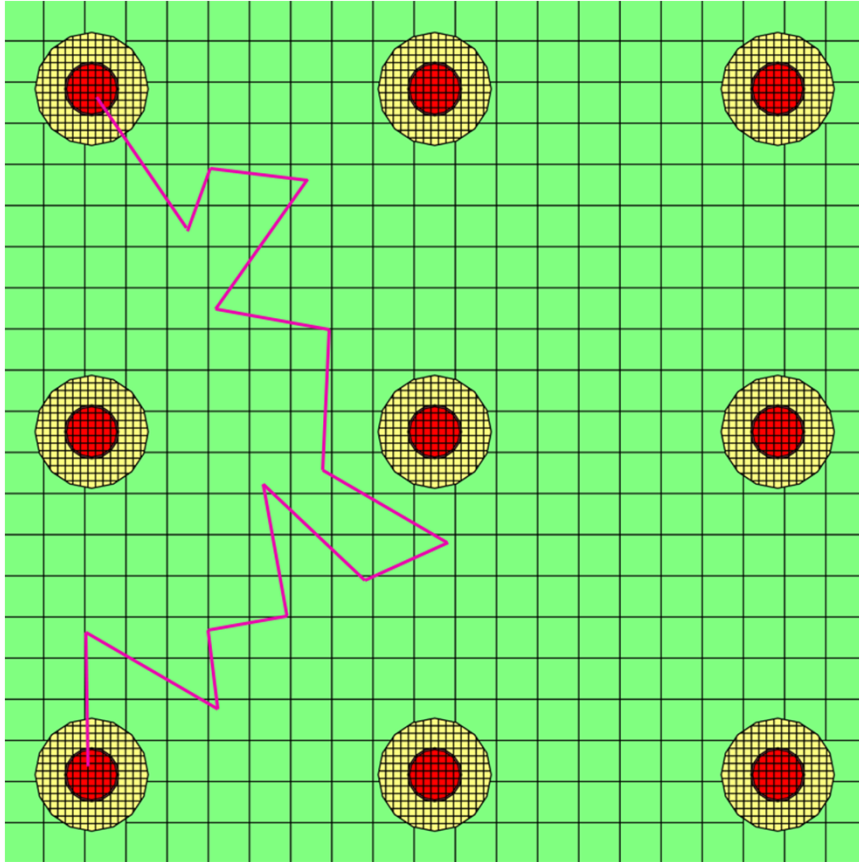


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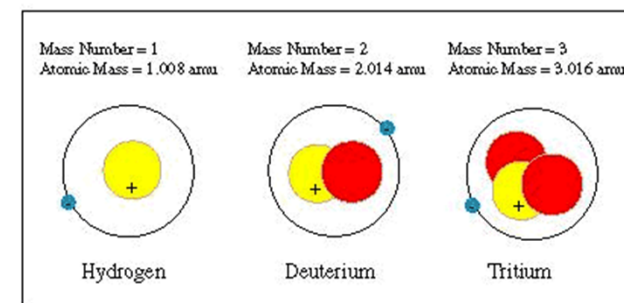
# Water as a Moderator and Coolant



On average water ( $\text{H}_2\text{O}$ ) will moderate a neutron after 16 collisions.

Heavy water ( $\text{D}_2\text{O}$ ) requires approximately 29 collisions to moderate a neutron.

Why is heavy water preferred?



*Hydrogen isotope diagram from NASA JPL*

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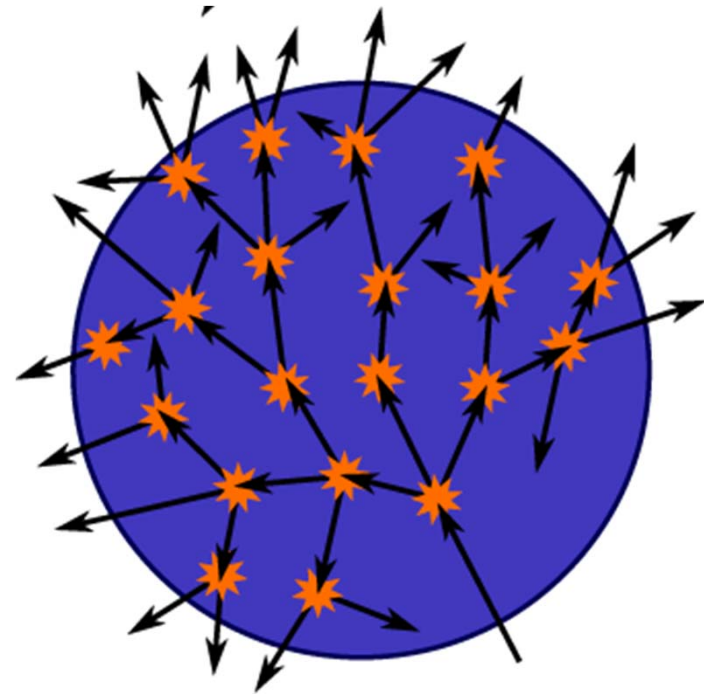


# Criticality

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

80 generations equate to  $6 \times 10^{23}$  (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

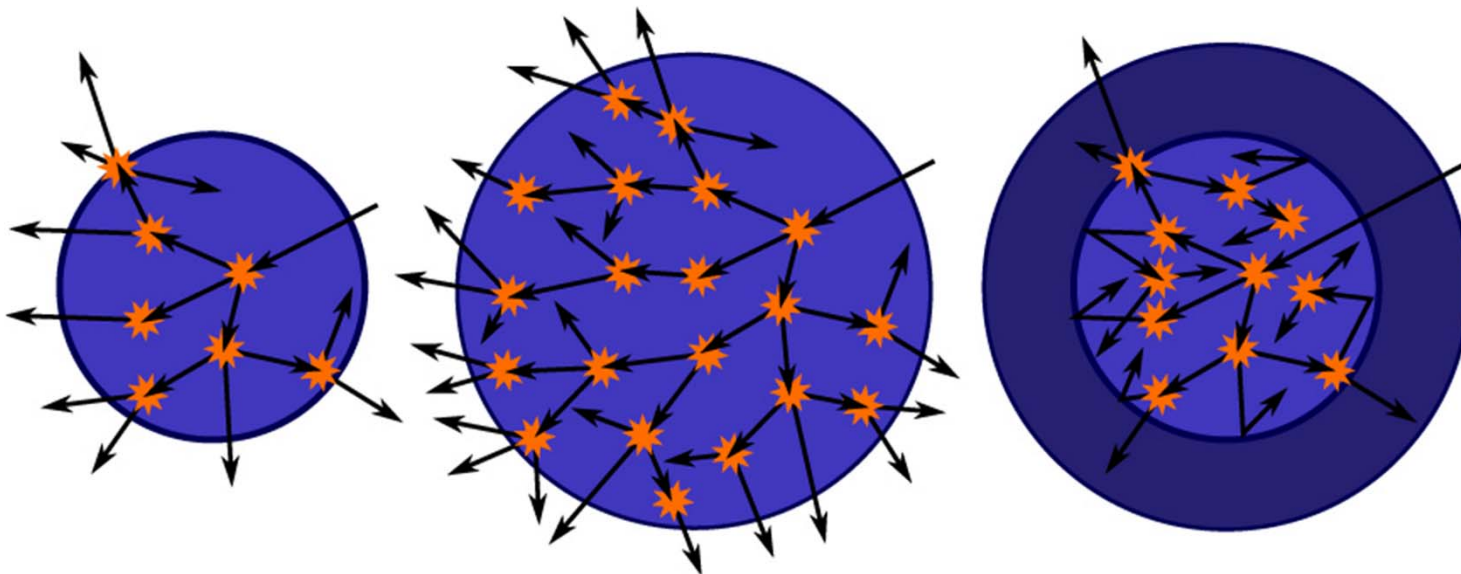


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# Critical Mass

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



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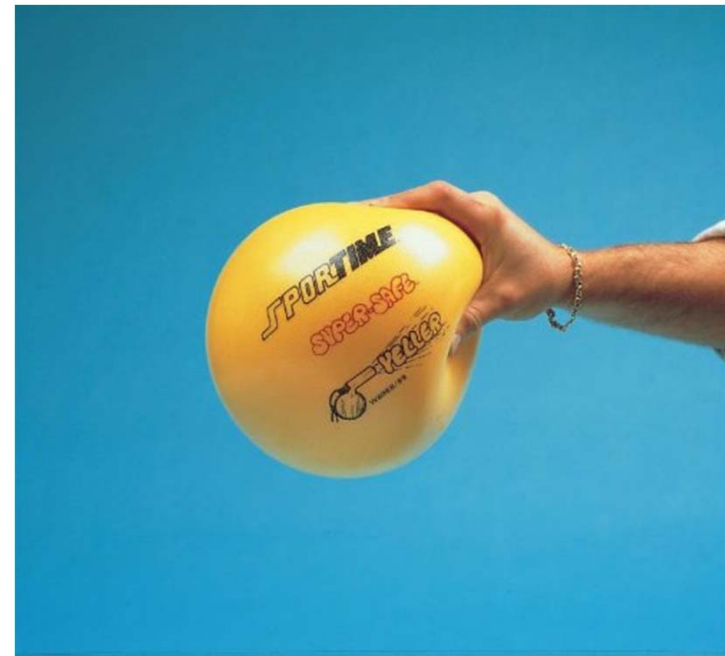


# Critical Mass of a Bare Sphere

Pu-239 Critical Mass:  
10kg at 9.9 cm



U-235 Critical Mass:  
52kg at 17 cm

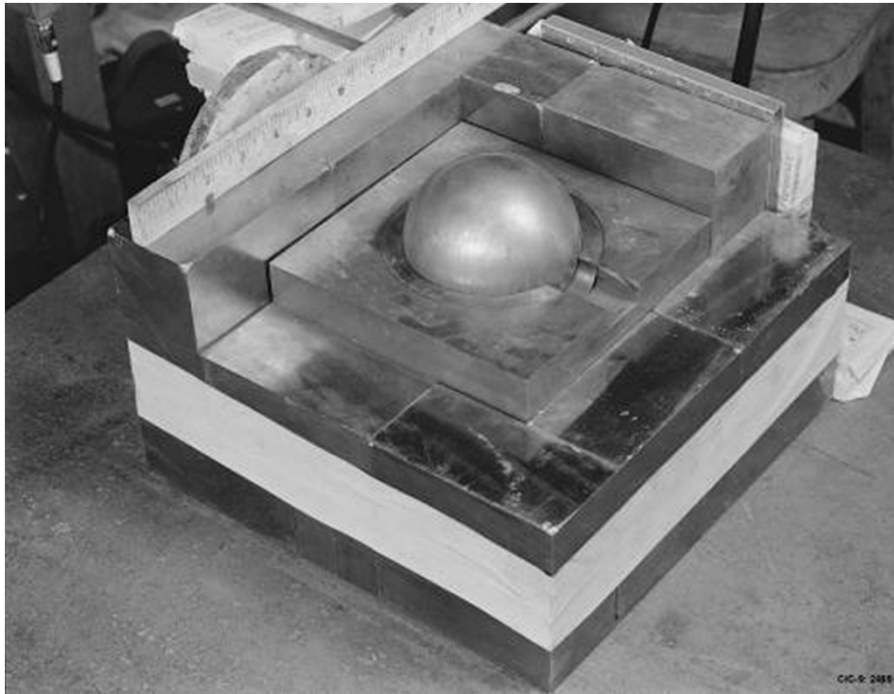


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# Criticality Accidents



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 Daghljan went critical when he dropped a tungsten brick onto a sphere of Pu.

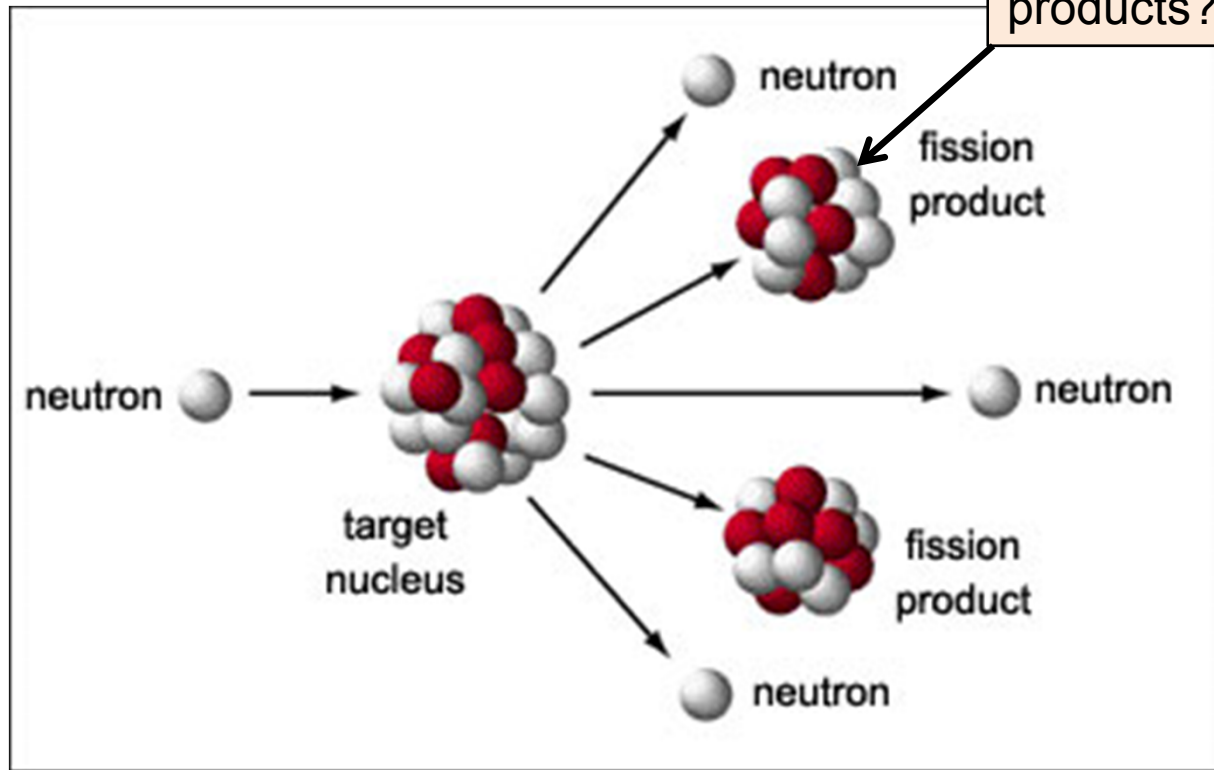
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# Nuclear Fission Products

What are fission products?



Fission Reaction from the Atomic Archive website

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# Periodic Table of Elements

# Periodic Table of Elements

1A	1	H	2	He
	3	Li	4	Be
	11	Na	12	Mg
	19	K	20	Ca
	37	Rb	38	Sr
	55	Cs	56	Ba
	87	Fr	88	Ra
	21	Sc	22	Ti
	23	V	24	Cr
	25	Mn	26	Fe
	27	Co	28	Ni
	29	Cu	30	Zn
	31	Ga	32	Ge
	33	As	34	Se
	35	Br	36	Kr
	41	Nb	42	Mo
	43	Tc	44	Ru
	45	Rh	46	Pd
	47	Ag	48	Cd
	49	In	50	Sn
	51	Sb	52	Te
	53	I	54	Xe
	57	*La	58	Ce
	59	Pr	60	Nd
	61	Pm	62	Sm
	63	Eu	64	Gd
	65	Tb	66	Dy
	67	Ho	68	Er
	69	Tm	70	Yb
	71	Lu	72	Hf
	73	Ta	74	W
	75	Re	76	Os
	77	Ir	78	Pt
	79	Au	80	Hg
	81	Tl	82	Pb
	83	Bi	84	Po
	85	At	86	Rn
	89	+Ac	90	Th
	91	Pa	92	U
	93	Np	94	Pu
	95	Am	96	Cm
	97	Bk	98	Cf
	99	Es	100	Fm
	101	Md	102	No
	103	Lr	104	Rf
	105	Ha	106	Db
	107	Bh	108	Hs
	109	Uu	110	Ds

\* Lanthanide Series

+ Actinide Series

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

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

Legend - click to find out more...

H - gas

Li - solid

Br - liquid

Tc - synthetic



Non-Metals



Transition Metals



Rare Earth Metals



Halogens



Alkali Metals



Alkali Earth Metals



Other Metals



Inert Elements

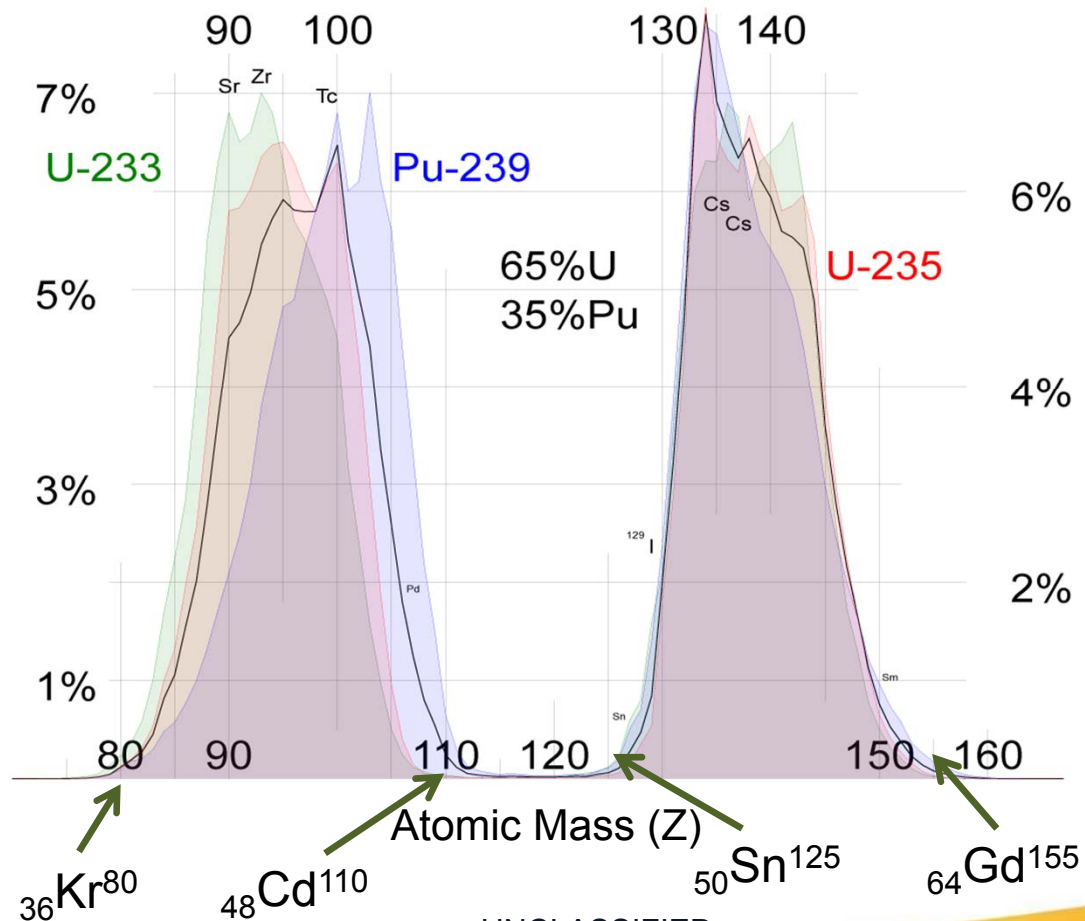
Table of Elements from the ThinkQuest website

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# Fission Product Yield



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# Actinide Production

- What are actinides?
- How do you create actinides

Periodic Table of Elements

1 H																	2 He		
3 Li	4 Be																	9 F	10 Ne
11 Na	12 Mg	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar											36 Kr	
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr		
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe		
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn		
87 Fr	88 Ra	89 Ac	104 Rf	105 Ha	106 Nh	107 Ds	108 Mt	109 Uu	110 Uub										

\* Lanthanide Series

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
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+ Actinide Series

90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr
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Legend - click to find out more...

H - gas	Li - solid	Br - liquid	Tc - synthetic
Non-Metals	Transition Metals	Rare Earth Metals	Halogens
Alkali Metals	Alkali Earth Metals	Other Metals	Inert Elements

Table of Elements from the ThinkQuest website

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# 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.

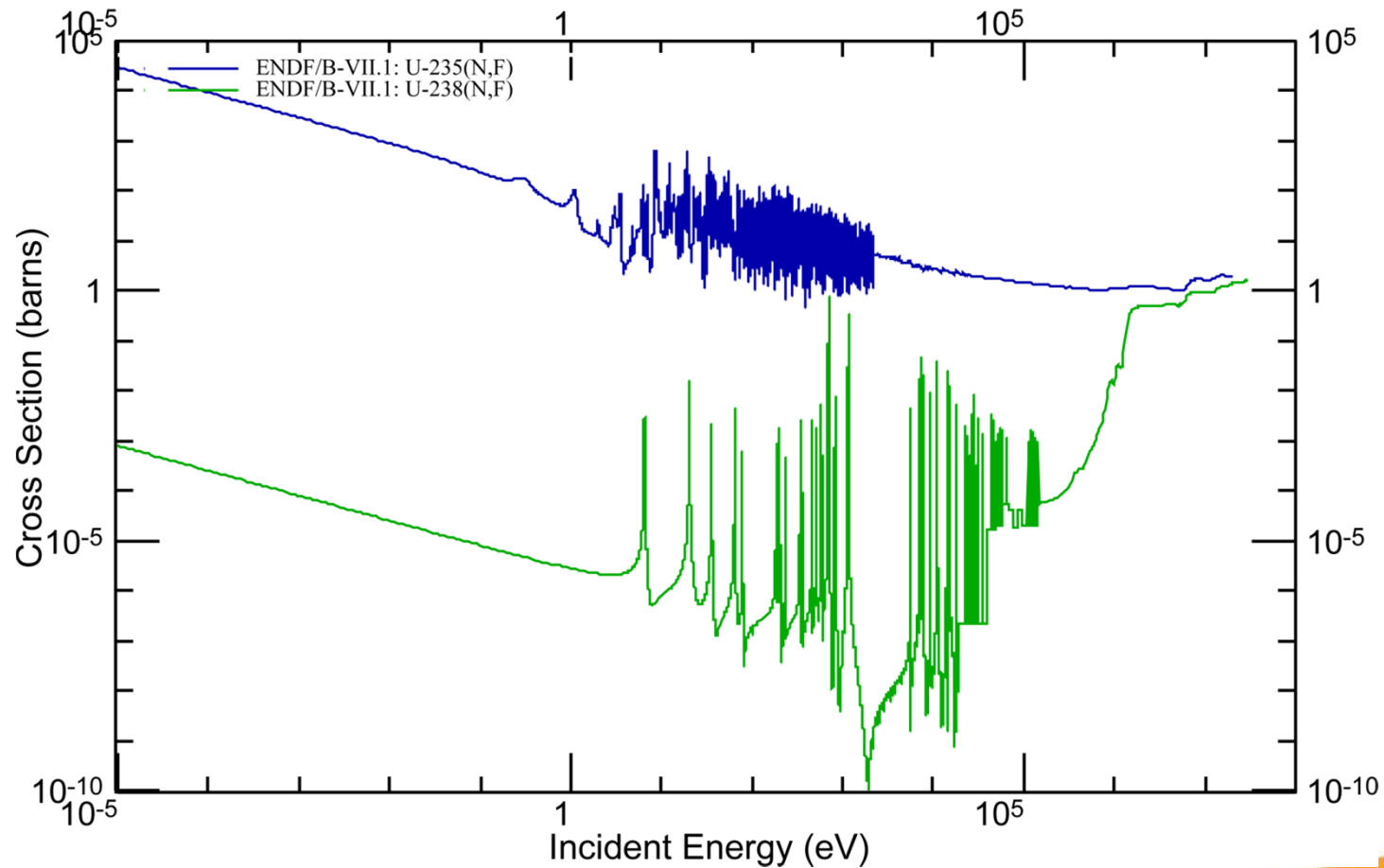
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# Fission x-sections for U-235 vs U-238

Neutron fission of U-235 and U-238



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# Going Critical with Uranium

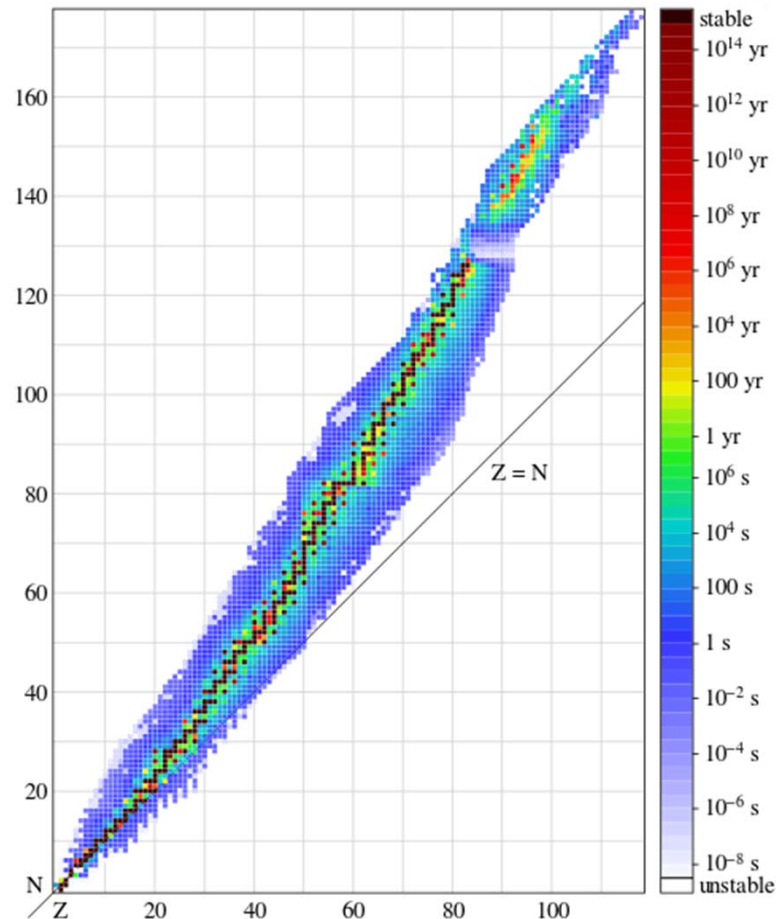
- How do you create a fission chain reaction with Uranium?
  - 1. Use a really good moderator that doesn't absorb many neutrons (heavy water)
  - 2. Use enriched uranium
    - Commercial Reactor (3-5% U-235)
    - Nuclear weapons grade uranium (>90% U-235)

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# Stability of Nuclides



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# Chart of the Nuclides

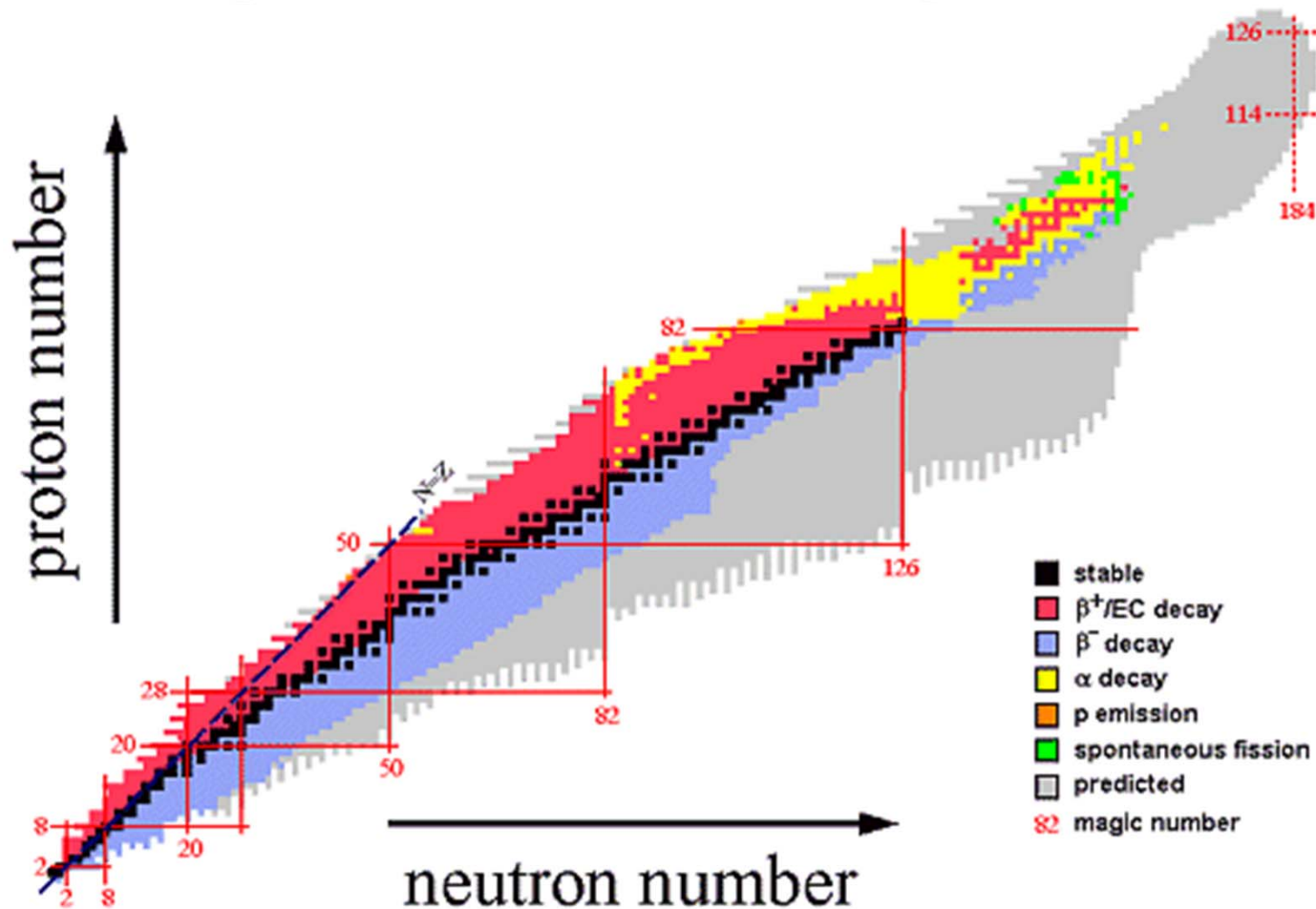


Chart of the nuclides from LUND University

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# Isotopes, Isobars and Isotones

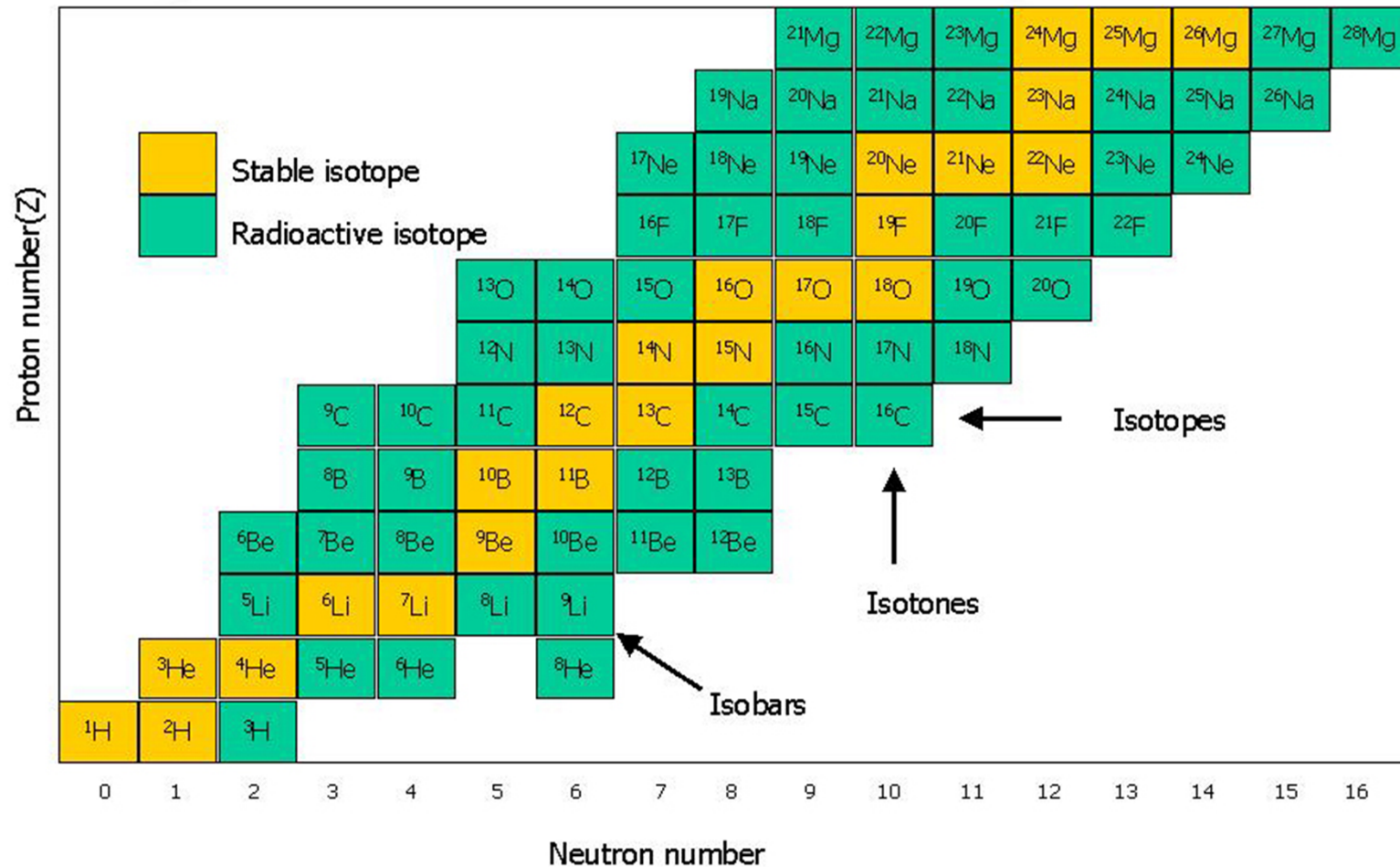


Chart of the Nuclides from University of Alaska

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# Up Close View of Nuclides Chart

97	Bk																	
	berkelium																	
96	Cm	Cm235	Cm236	Cm237	Cm238	Cm239	Cm240	Cm241	Cm242	Cm243	Cm244	Cm245	Cm246	Cm247	Cm248	Cm249	Cm250	Cm251
	curium																	
95	Am232	Am233	Am234	Am235	Am236	Am237	Am238	Am239	Am240	Am241	Am242	Am243	Am244	Am245	Am246	Am247	Am248	Am249
94	Pu231	Pu232	Pu233	Pu234	Pu235	Pu236	Pu237	Pu238	Pu239	Pu240	Pu241	Pu242	Pu243	Pu244	Pu245	Pu246	Pu247	Pu248
93	Np230	Np231	Np232	Np233	Np234	Np235	Np236	Np237	Np238	Np239	Np240	Np241	Np242	Np243	Np244	Np245	Np246	Np247
92	U229	U230	U231	U232	U233	U234	U235	U236	U237	U238	U239	U240	U241	U242	U243	U244	U245	U246
91	Pa228	Pa229	Pa230	Pa231	Pa232	Pa233	Pa234	Pa235	Pa236	Pa237	Pa238	Pa239	Pa240	Pa241	Pa242	Pa243	Pa244	Pa245
90	Th227	Th228	Th229	Th230	Th231	Th232	Th233	Th234	Th235	Th236	Th237	Th238	Th239	Th240	Th241	Th242	Th243	Th244
89	Ac226	Ac227	Ac228	Ac229	Ac230	Ac231	Ac232	Ac233	Ac234	Ac235	Ac236	Ac237	Ac238	Ac239	Ac240	Ac241	Ac242	Ac243

Chart of the Nuclides from KAPL

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# How Are Actinides Produced

<b>Pu237</b> 7/- <b>0.18 s</b> IT 145.54 <b>45.64 d</b> $\epsilon, \gamma$ 59.5,... $\alpha$ 5.344( $\omega$ ),... $\gamma$ 280.4( $\nu\omega$ ), 289.9, 320.8,... $\sigma_f$ 24E2 E 0.220	<b>Pu238</b> <b>87.7 a</b> $\alpha$ 5.4992, 5.4565,... $\gamma$ 43.5 $\omega$ ( $e^-$ ), 99.9 ( $e^-$ ),... SF $\nu\omega$ $\sigma_f$ 54E1, 20E1 $\sigma_f$ 18, ~33 238.049560	<b>Pu239</b> 1/+ <b>2.410E4 a</b> $\alpha$ 5.156, 5.144, 5.105,... $\gamma$ 51.6 $e^-$ , 30.1 – 1057.3 $\omega$ SF $\nu\omega$ $\sigma_f$ 271, 20E1 $\sigma_f$ 750, 30E1 $\sigma_\alpha$ < 0.4 mb 239.052163	<b>Pu240</b> <b>6.56E3 a</b> $\alpha$ 5.1685, 5.1241,... $\gamma$ 45.2 $\omega$ ( $e^-$ ), 104.2 ( $e^-$ ),... SF $\nu\omega$ $\sigma_f$ 290, 81E2 $\sigma_f$ 0.05, 2.4 240.053814	<b>Pu241</b> 5/+ <b>14.29 a</b> $\beta^-$ 0.0208 $\alpha$ 4.897 $\omega$ , 4.853,... $\gamma$ 148.57 ( $\nu\omega$ ), 103.7,... $\sigma_f$ ~361, 16E1 $\sigma_f$ 101E1, 57E1 $\sigma_\alpha$ < 0.2 mb E 0.0208
<b>1(-) Np236</b> (6-) <b>22.5 h</b> $\epsilon, \beta^-$ 0.54,... $\gamma$ 642.3, 687.6, ... $\sigma_f$ 2.7E3, 7E2 E+ 0.9	<b>Np237</b> 5/+ <b>2.14E6 a</b> $\alpha$ 4.788, 4.771,... $\gamma$ 29.4, 86.5,... $\sigma_f$ 169, 65E1 $\sigma_f$ 0.02, 7 237.048173	<b>Np238</b> 2+ <b>2.103 d</b> $\beta^-$ 0.263, 1.248,... $\gamma$ 984.5, 1028.5,... $\sigma_f$ 48E1 $\sigma_f$ 21E2, 9E2 E 1.2915	<b>Np239</b> 5/+ <b>2.356 d</b> $\beta^-$ 0.438, 0.341,... $\gamma$ 106.1, 277.6, 228.2,... $\sigma_f$ (3E1 + 3E1) $\sigma_f$ < 1 E 0.723	<b>1(+) Np240</b> (5+) <b>7.22 m</b>   <b>1.032 h</b> $\beta^-$ 2.18, 1.60,...   $\beta^-$ 0.89 $\gamma$ 554.6, 597.4,   $\gamma$ 566.3, ...   973.9, IT   600.6,... E 2.19
<b>1/+ U235</b> 7/- <b>26 m</b> IT ~76.8 eV ( $\omega$ ) $e^-$ <b>AcU</b> <b>0.7204</b> <b>7.04E8 a</b> $\alpha$ 4.398, 4.366,... $\gamma$ 185.72, 143.76,... SF $\nu\omega$ $\sigma_f$ 98, 14E1 $\sigma_f$ 585, ~275 $\sigma_\alpha$ < 0.1 mb 235.043930	<b>U236</b> <b>2.342E7 a</b> $\alpha$ 4.494, 4.445,... $\gamma$ 49.4 $\omega$ ( $e^-$ ), 112.8,... SF $\nu\omega$ $\sigma_f$ 5.1, 36E1 $\sigma_f$ 0.04, 4 236.045568	<b>U237</b> 1/+ <b>6.752 d</b> $\beta^-$ 0.24, 0.25,... $\gamma$ 59.5, 208.0,... $\sigma_f$ 4E2, 12E2 $\sigma_f$ < 0.35 E 0.519	<b>U238</b> <b>UI</b> <b>99.2742</b> <b>4.468E9 a</b> $\alpha$ 4.197, 4.147,... $\gamma$ 49.6 $\omega$ ( $e^-$ ),... SF $\nu\omega$ $\sigma_f$ 2.68, 277 $\sigma_f$ ~5 $\mu$ b, 1.3mb $\sigma_\alpha$ 1 $\mu$ b 238.050788	<b>U239</b> 5/+ <b>23.47 m</b> $\beta^-$ 1.21, 1.28,... $\gamma$ 74.7, 43.5,... $\sigma_f$ 22 $\sigma_f$ 15 E 1.261

Chart of the Nuclides from KAPL

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# Decay of Fission Products

46	<b>Pd106</b> 27.33 $\sigma_{\gamma}$ (0.013 + 0.28), 5.7 105.903486	<b>Pd107</b> 5/+ 20.9 s IT 214.9 $\beta^-$ 0.04 no $\gamma$ $\sigma_{\gamma}$ 1.8, 11E1 E 0.034	<b>Pd108</b> 26.46 $\sigma_{\gamma}$ (0.19 + 8), (2 + 24E1) 107.903892	<b>Pd109</b> 5/+ 4.69 m IT 188.9 $\beta^-$ 1.028, ... $\gamma$ 88.0D e-, ... E 1.116	<b>Pd110</b> 11.72 $\sigma_{\gamma}$ (0.03 + 0.7), (0.7 + 8) 109.90515	<b>Pd111</b> 5/+ 5.5 h 23.4 m IT 172.2 $\beta^-$ 0.35, ... $\gamma$ 580.0, ... $\gamma$ 70.4, 391.2, ... E 2.22
45	<b>Rh105</b> 7/+ 43.0 s IT 129.6 $\beta^-$ 0.566, ... $\gamma$ 319.2, ... $\sigma_{\gamma}$ (5E3 + 11E3), 17E3 E 0.567	<b>Rh106</b> 1+ 2.18 h 29.9 s $\beta^-$ 0.92, ... $\gamma$ 511.9, ... $\gamma$ 1045.8, 621.9, ... E 3.54	<b>Rh107</b> 7/+ 21.7 m $\beta^-$ 1.20, ... $\gamma$ 302.8, ... E 1.50	<b>Rh108</b> 1+ 6.0 m $\leftrightarrow$ 17 s $\beta^-$ 1.57, ... $\gamma$ 433.9, 581.0, 947.0, ... E 4.5	<b>Rh109</b> 7/+ 1.34 m $\beta^-$ 2.25, ... $\gamma$ 326.8, ... E 2.60	<b>Rh110</b> 1+ 29 s $\leftrightarrow$ 3.1 s $\beta^-$ 2.6, ... $\gamma$ 373.8, 546.3, ... E 5.6
44	<b>Ru104</b> 18.62 $\sigma_{\gamma}$ 0.47, 6 103.905433	<b>Ru105</b> 3/+ 4.44 h $\beta^-$ 1.187, 1.11, 1.134, ... $\gamma$ 724.3, 469.4, 676.3, ... $\sigma_{\gamma}$ ~0.30 E 1.918	<b>Ru106</b> 1.017 a $\beta^-$ 0.0394 no $\gamma$ $\sigma_{\gamma}$ 0.15, 2.1 E 0.0394	<b>Ru107</b> (5/-)+ 3.8 m $\beta^-$ 2.3, 2.1, ... $\gamma$ 194.1, 847.9, 462.6, 374.3, ... E 2.9	<b>Ru108</b> 4.5 m $\beta^-$ 1.3, ... $\gamma$ 165.0, ... E 1.35	<b>Ru109</b> (5/-)+ 34.5 s $\beta^-$ 2.3, 3.6, ... $\gamma$ 206.3, 226.0, 358.8, ... E 4.2
43	<b>Tc103</b> 5/+ 54 s $\beta^-$ 2.2, 2.0, ... $\gamma$ 346.4, 136.1, 210.4, ... E 2.66	<b>Tc104</b> (3+) 18.2 m $\beta^-$ 5.3, ... $\gamma$ 358.0, ... E 5.60	<b>Tc105</b> (3/-) 7.6 m $\beta^-$ 3.4, ... $\gamma$ 143.2, 107.9, 321.5, 159.5, ... E 3.6	<b>Tc106</b> 2+ 36 s $\beta^-$ 270.1, 2239.3, 1969.4, 2789.3, ... E 6.55	<b>Tc107</b> (3/-) 21.2 s $\beta^-$ 4.6, ... $\gamma$ 102.7, 177.0, 106.3, ... E 4.8	<b>Tc108</b> (2)+ 5.1 s $\beta^-$ 7.45, 5.92, ... $\gamma$ 242.3, 465.6, 707.8, ... E 7.72
42	<b>Mo102</b> 11.3 m $\beta^-$ 1.2, ... $\gamma$ 211.6, 148.2, 223.8, ... E 1.01	<b>Mo103</b> (3/+) 1.13 m $\beta^-$ 3.7, ... $\gamma$ 83.4, 423.9, 45.8, ... E 3.8	<b>Mo104</b> 1.00 m $\beta^-$ 2.02, ... $\gamma$ 68.8, 69.7, 36.3, ... E 2.16	<b>Mo105</b> (5/-) 36 s $\beta^-$ 4.86, ... $\gamma$ 85.5, 76.6, 147.9, ... E 4.95	<b>Mo106</b> 8.7 s $\beta^-$ 465.7, 54.0, 618.6, ... E 3.52	<b>Mo107</b> (5/-) 3.5 s $\beta^-$ 400.3, 65.7, ... E 6.2

Chart of the Nuclides from KAPL

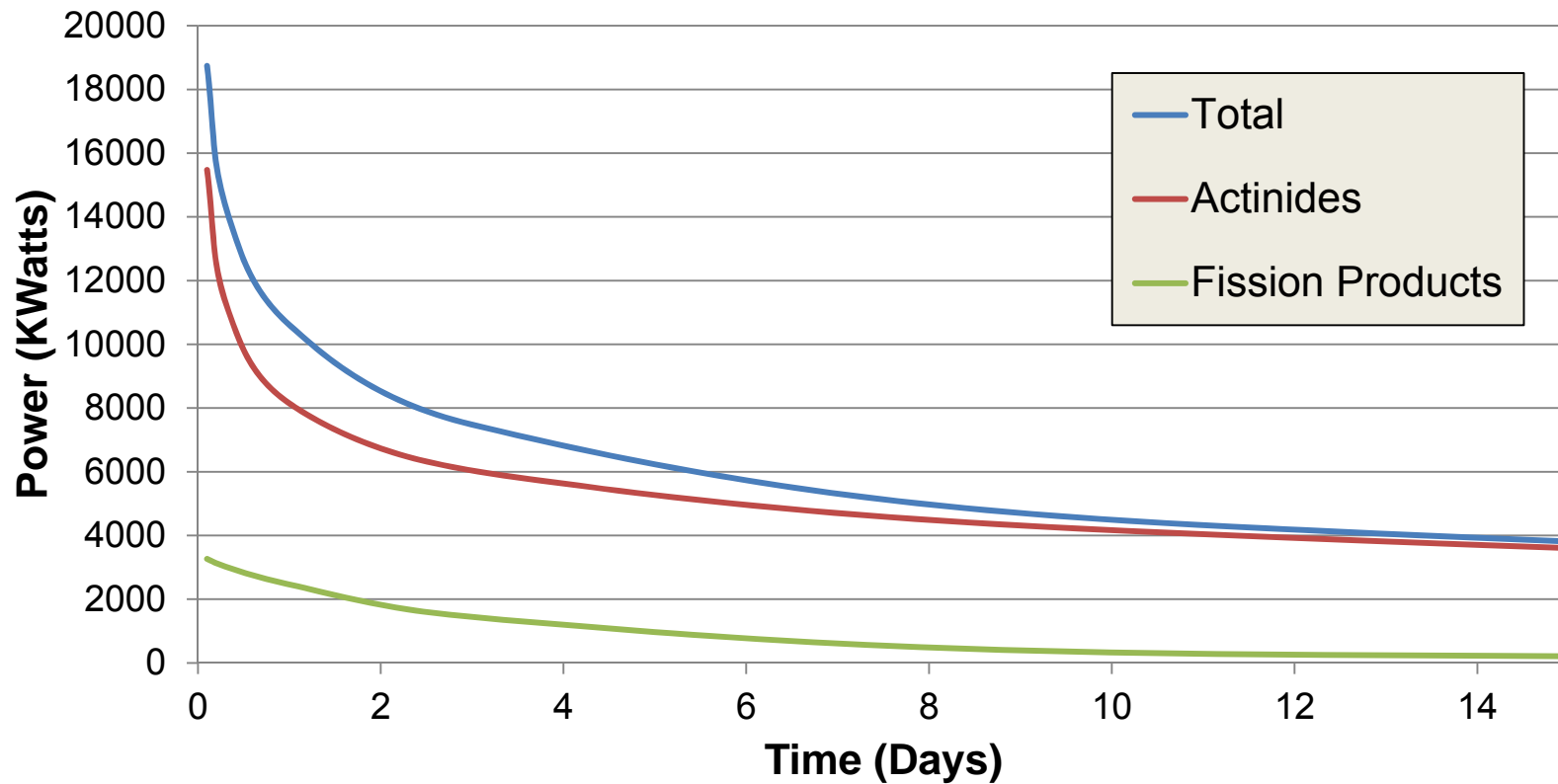
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# Decay Heat after Shutdown

## Power of BWR Core After Shutdown



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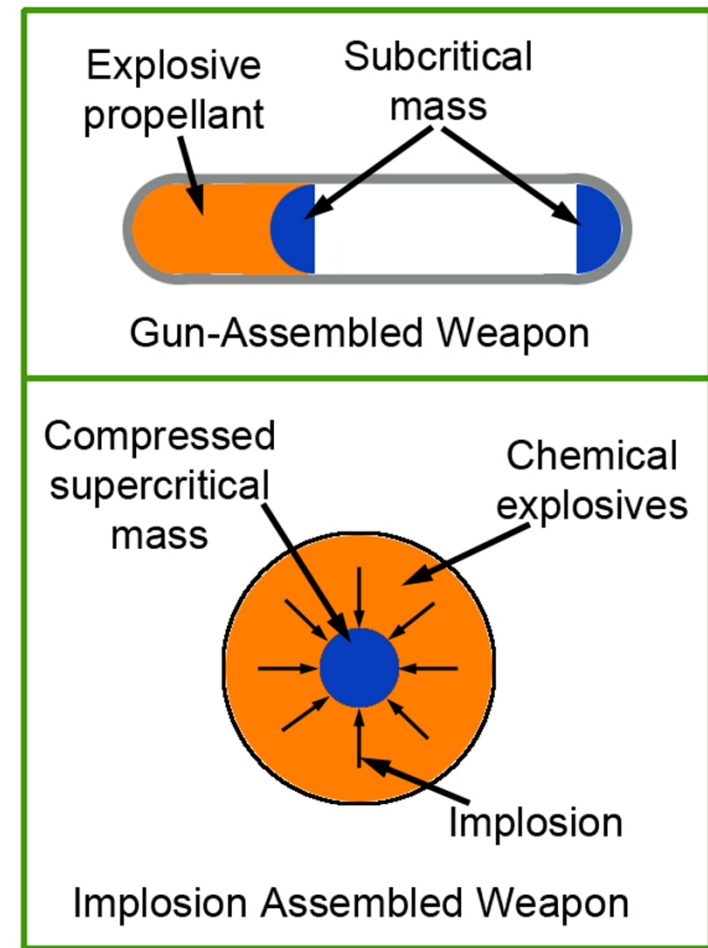
# Questions Before Break?

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# 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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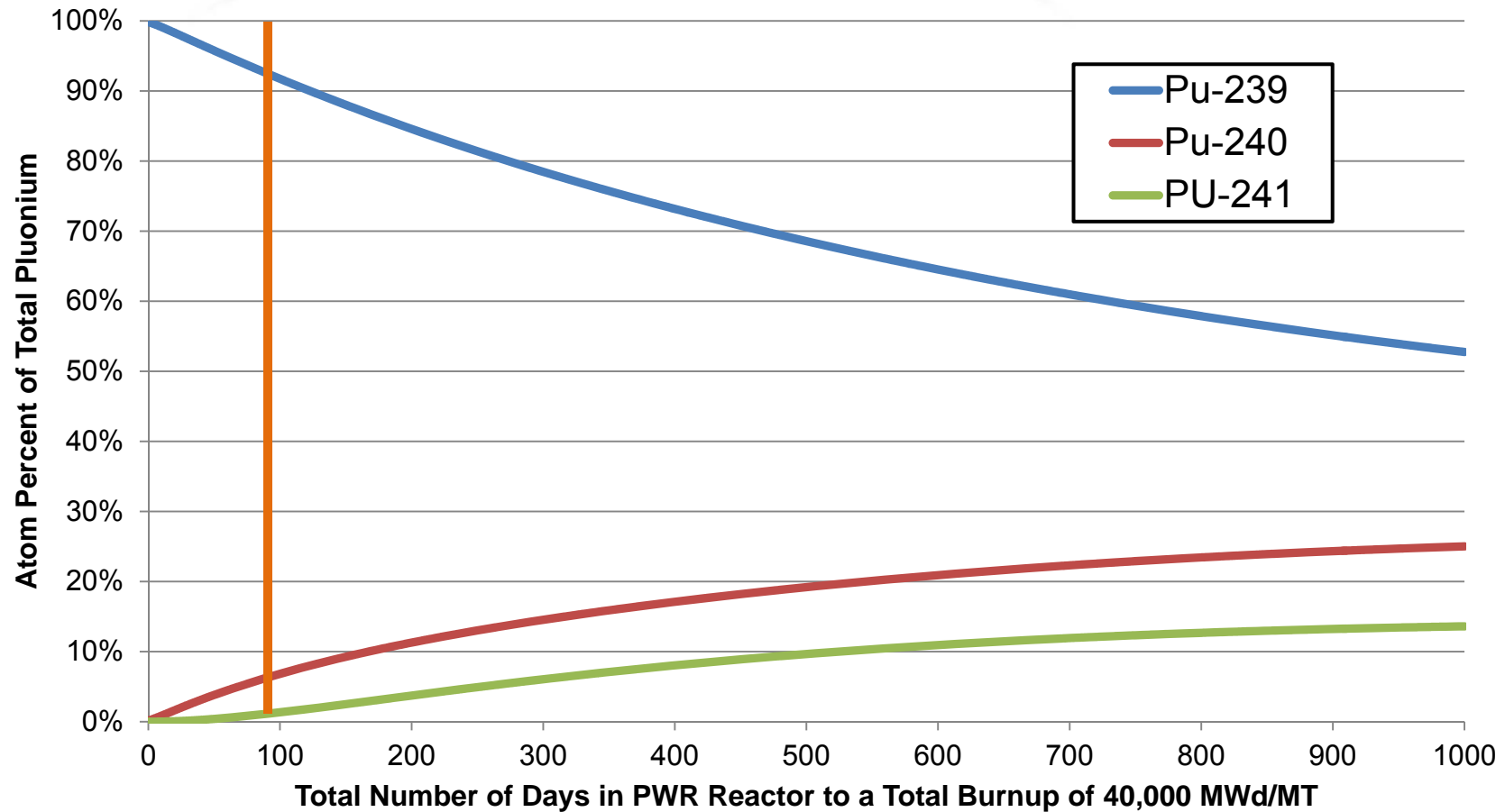
# Plutonium Attribution

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

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# 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

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# Reactor Depletion Codes

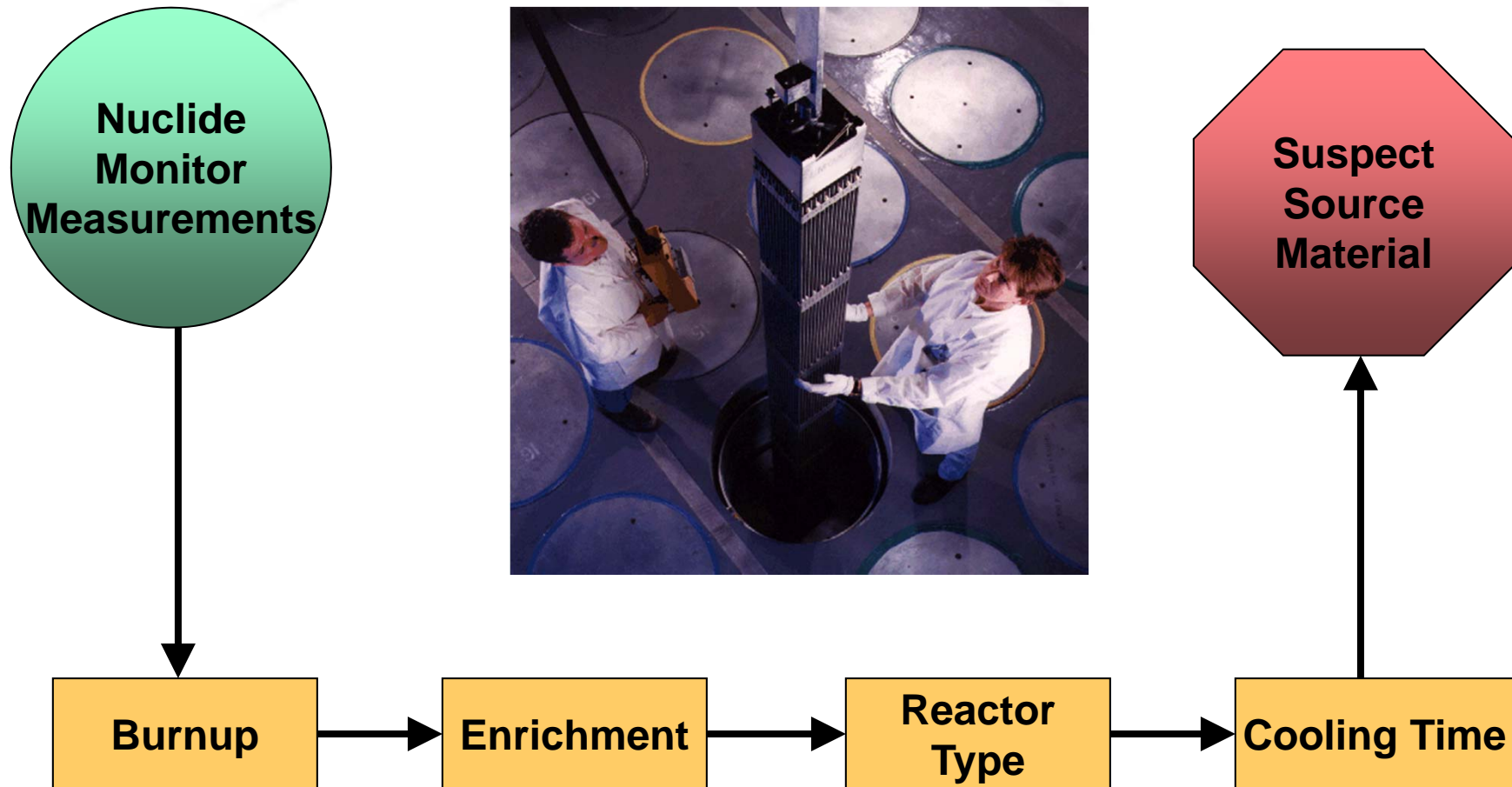
- ORIGEN 2
- SCALE 5 (ORIGEN-S, TRITON)
- MCNP w/ CINDER90
- Monteburns (MCNP and ORIGEN2)
- Attila
- WIMS
- Other specialized commercial codes

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# Inverse Modeling



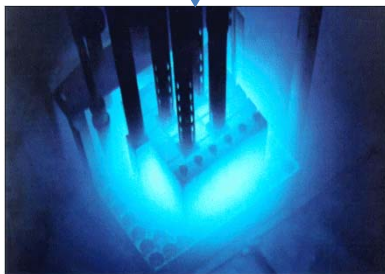
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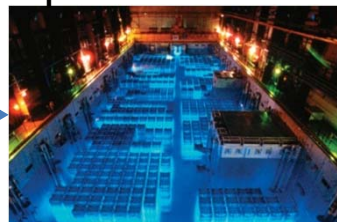
# Spent Fuel Samples

Fresh Fuel



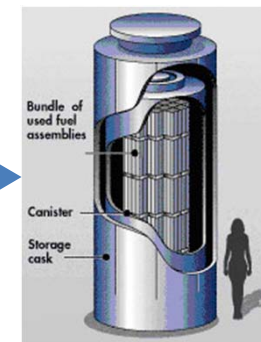
Reactor Core

Spent Fuel Pond



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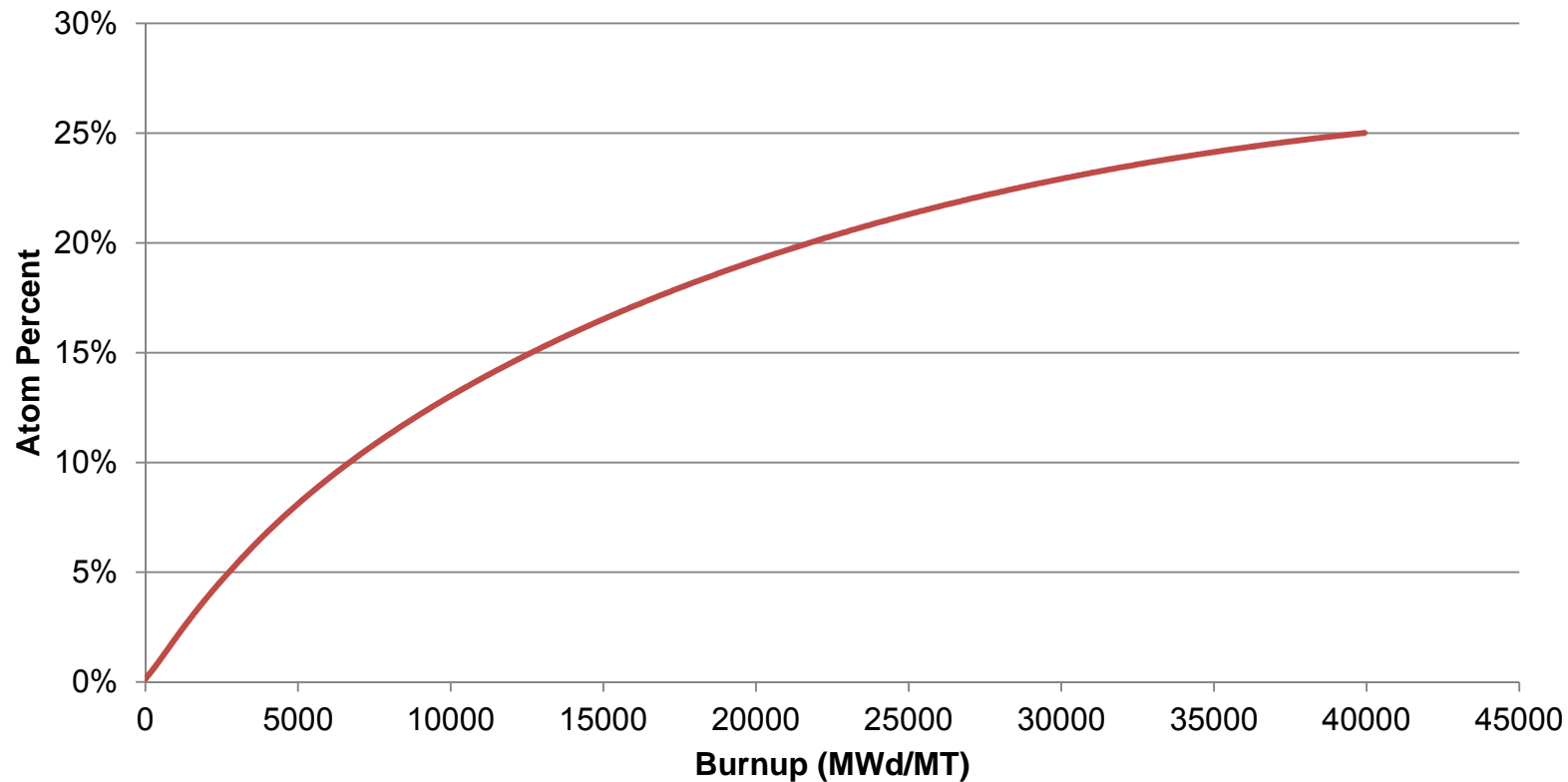
Dry Storage Cask





# Nuclide Production as a Burnup Indicator

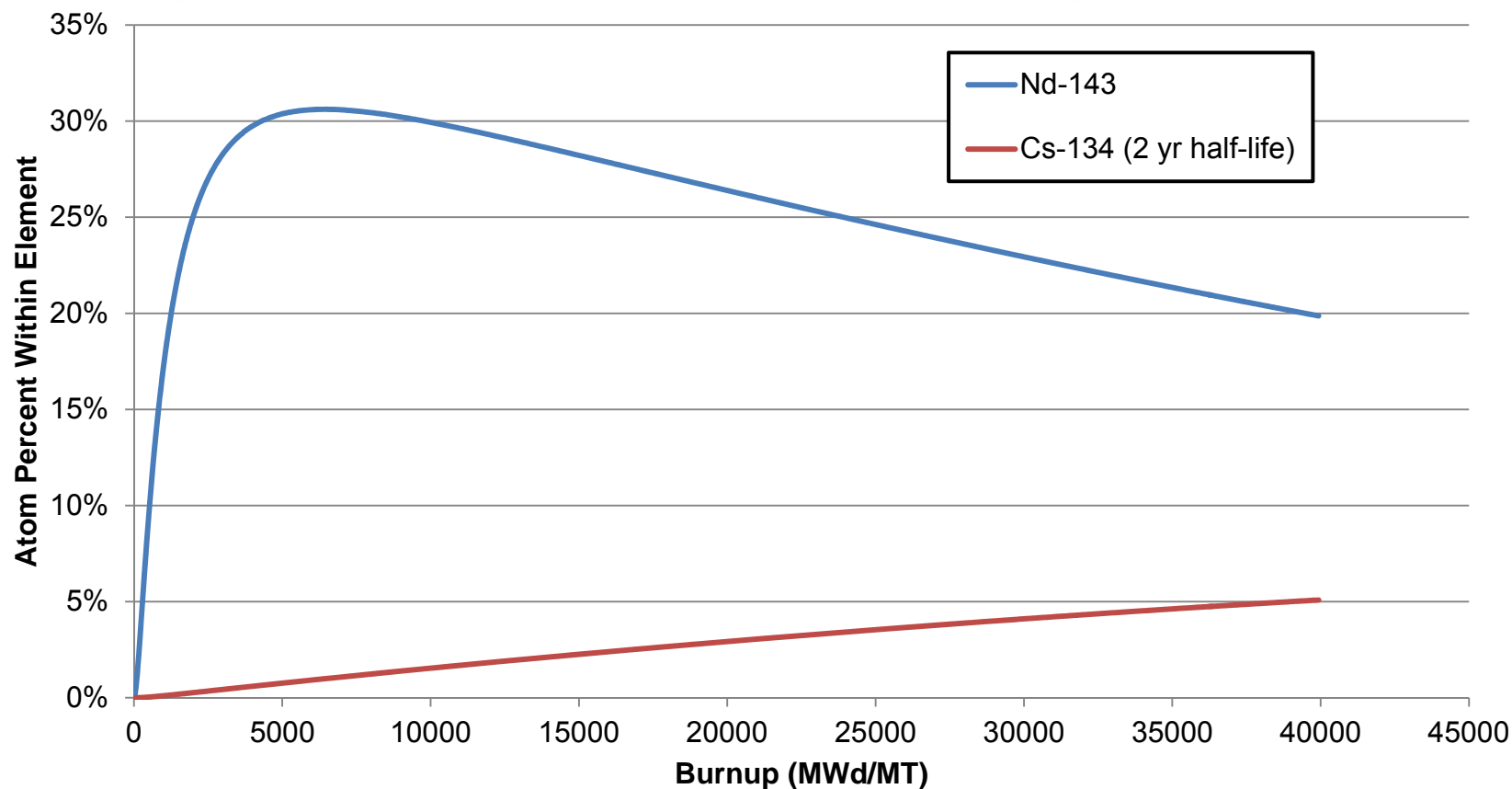
## Production of Pu-240 in a PWR Reactor



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# Not All Nuclides Are Appropriate

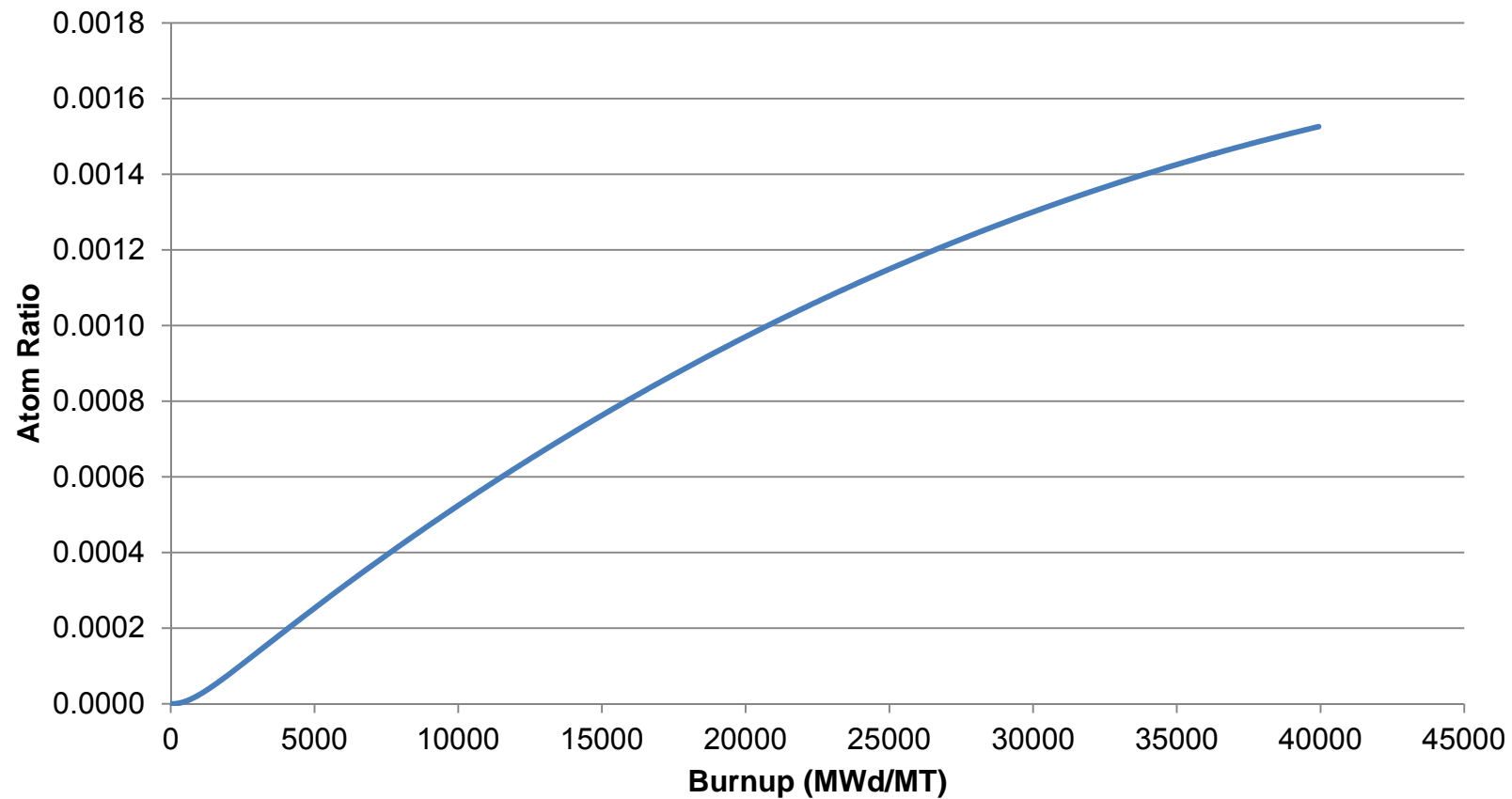


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# Ratios Are Your Friend

Ratio of Nd-143 atoms to U-238 atoms



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# 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)

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# 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.

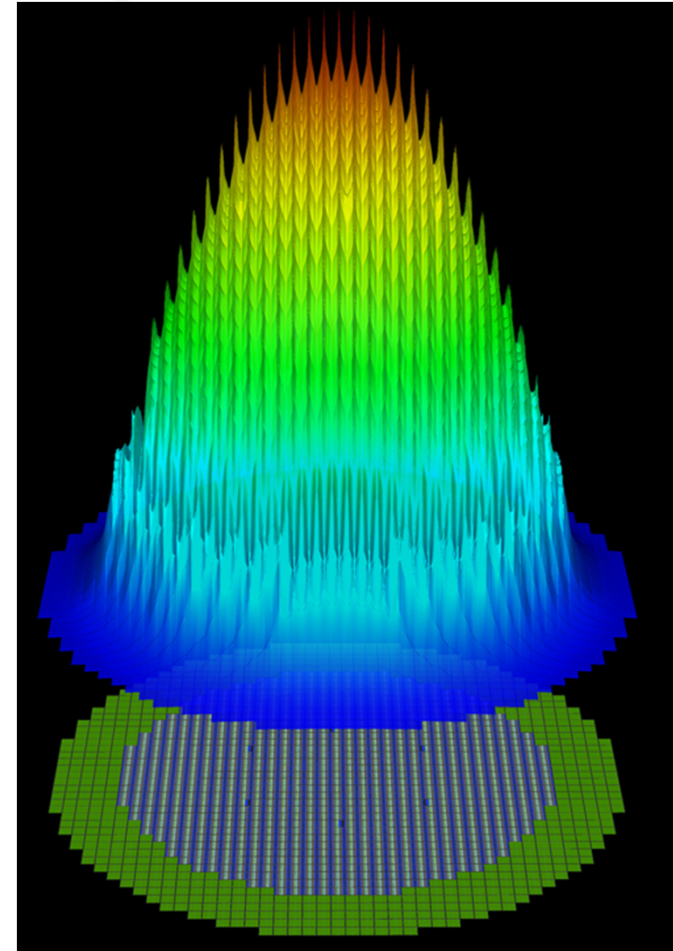


Figure from Argonne National Lab.

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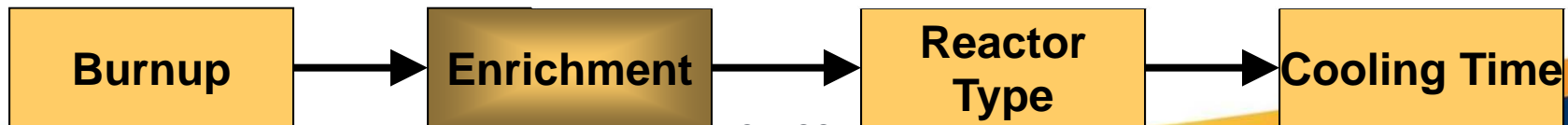
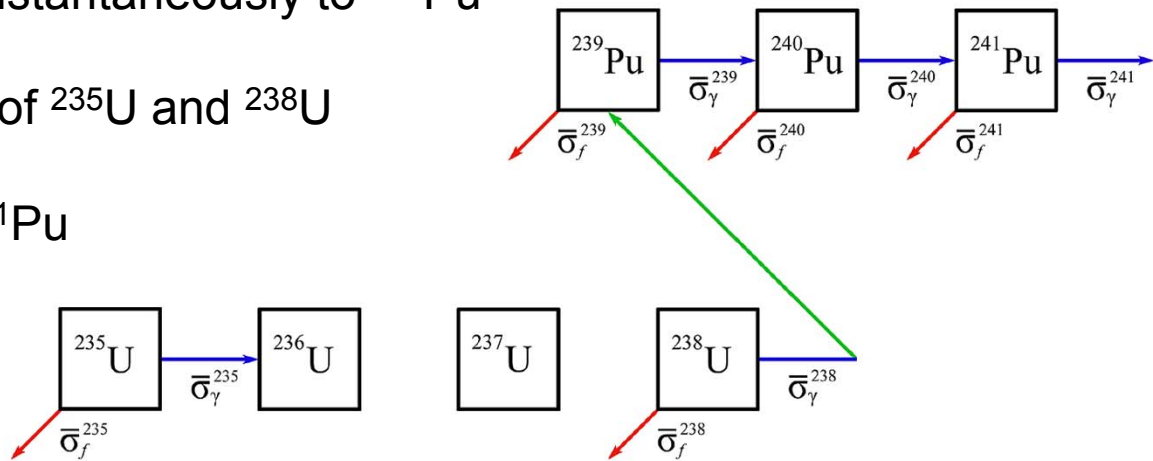


# Initial Uranium Enrichment

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

## Assumptions on fuel characteristics during irradiation:

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



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# Estimating the Initial Enrichment

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

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

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

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

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

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

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

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

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

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

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

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

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

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

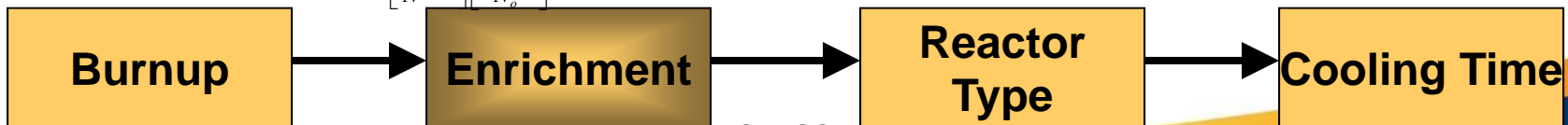
$$H15 = N_a E_R \bar{\sigma}_\gamma^{U238} \bar{\sigma}_\gamma^{Pu239} \bar{\sigma}_\gamma^{Pu240} \bar{\sigma}_f^{Pu241} \left( 1 - \left[ \frac{N^{U238}}{N_o^U} \right] \right)$$

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

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

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

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



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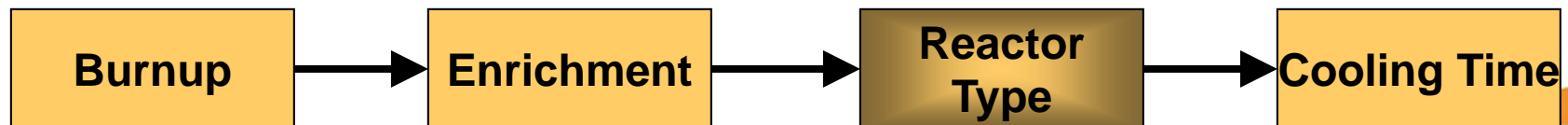
# Reactor Type

## 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

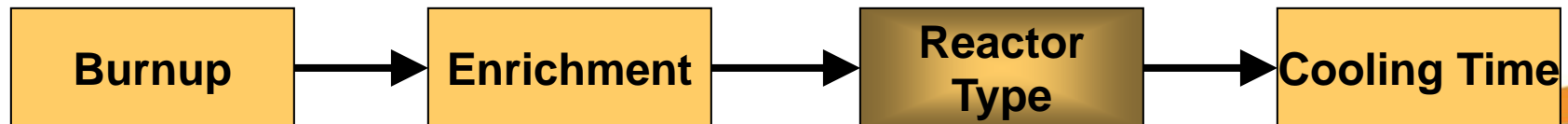
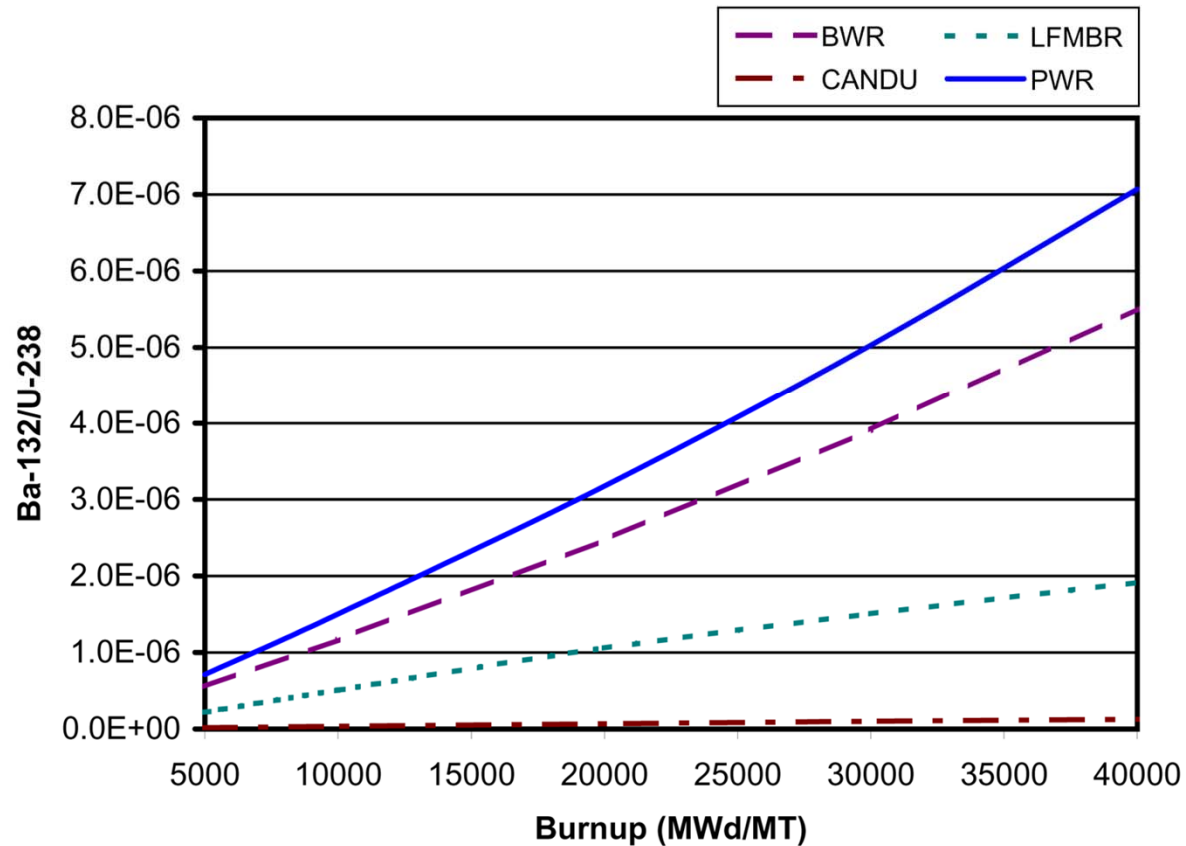


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# Reactor Type Indicator



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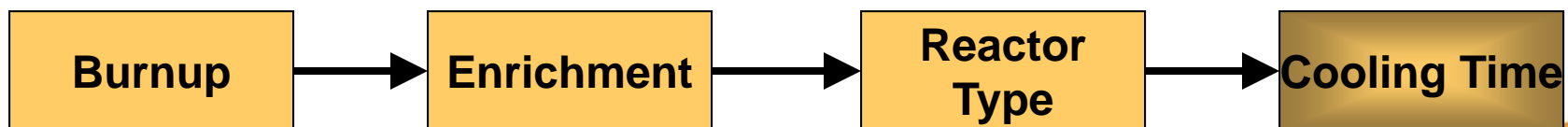
# Fuel Age or Cooling Time

## Cooling Time:

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

## Cooling Time Monitor Requirements:

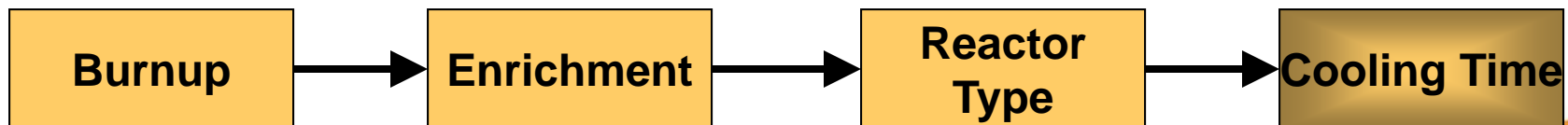
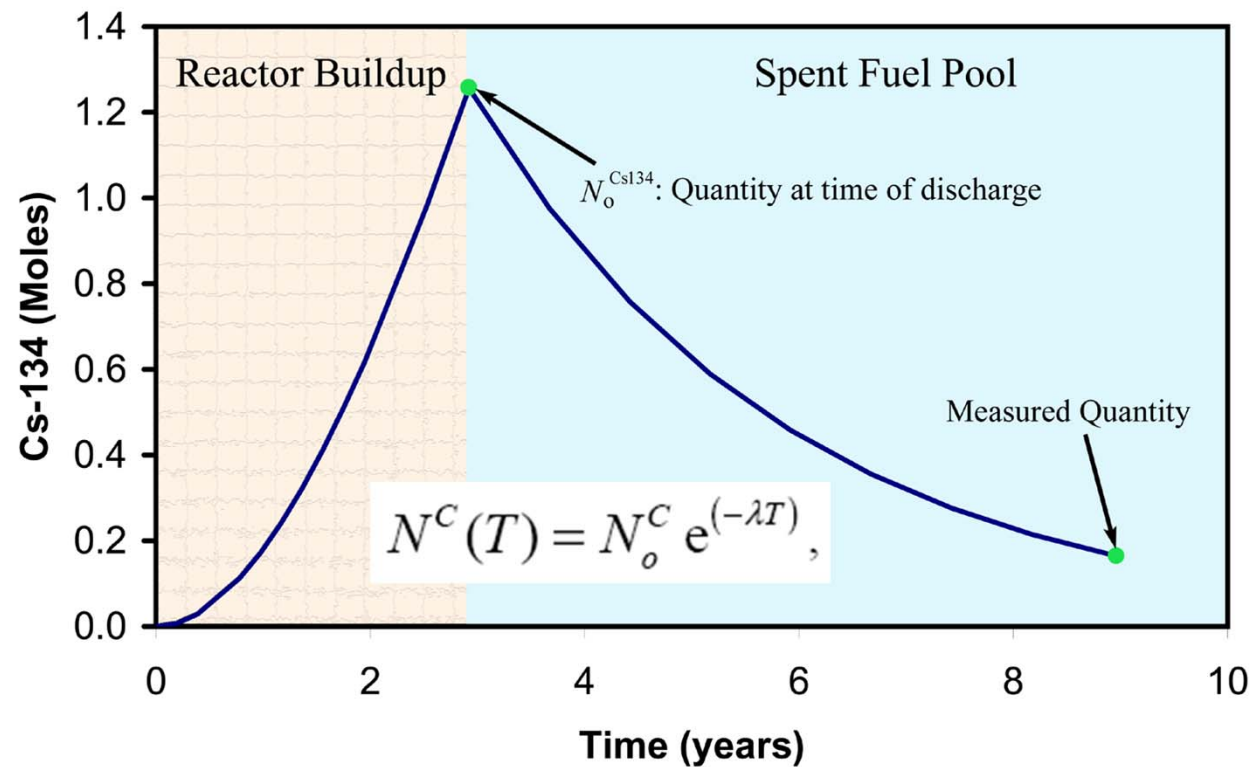
- Half-life is between 1-30 years
- At least 0.01 moles is produced per MT of fuel



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# Fuel Age or Cooling Time



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# 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)

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# Fukushima Reactor Accident

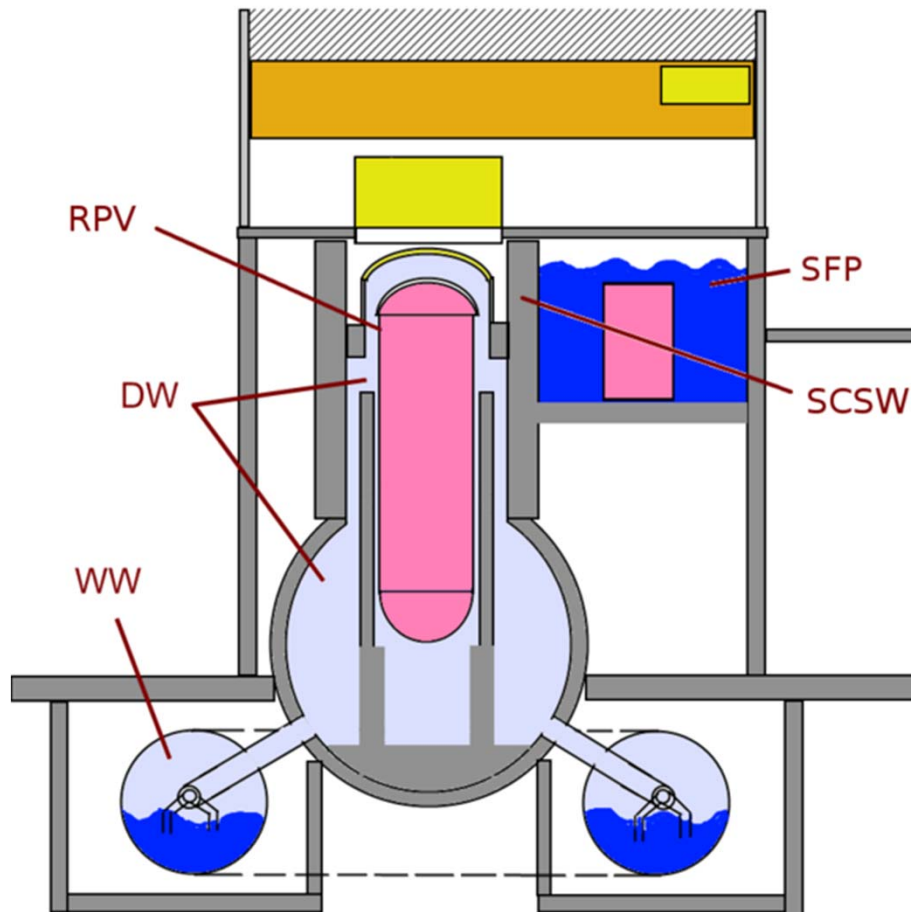
- Fukushima Dai-ichi plant consists of 6 reactors.
- Three were operating and three were already shutdown before the earthquake.
- Earthquake struck at 2:46pm March 11 2011.
- Reactors were automatically shutdown, core still required power to cool the core from decay heat.
- Approximately 60 minutes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

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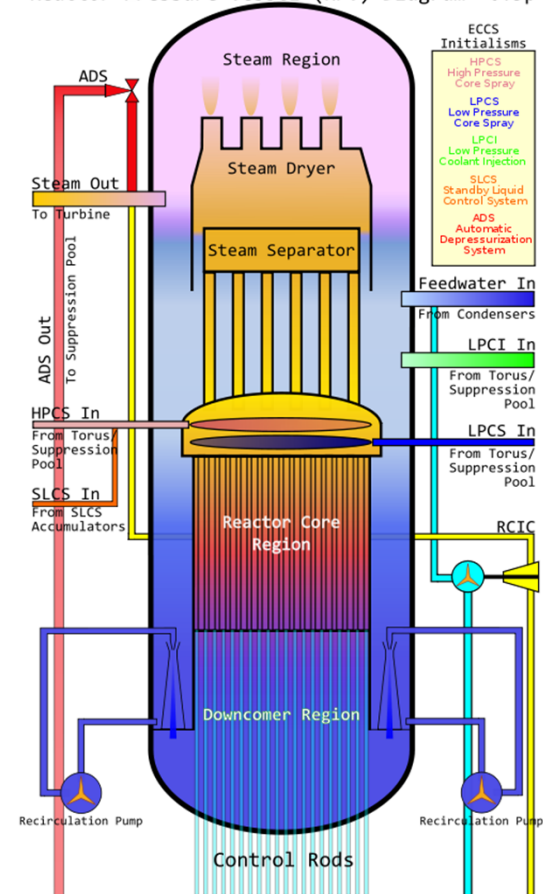




# Fukushima Reactor Building Layout



Boiling Water Reactor (BWR)  
Reactor Pressure Vessel (RPV) Diagram 0.5β



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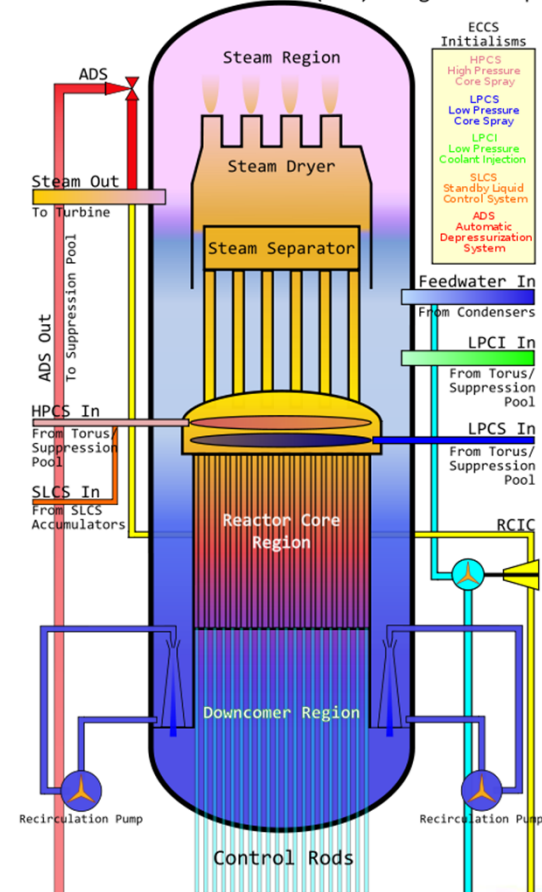




# Pressure Relief

- 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.

Boiling Water Reactor (BWR)  
Reactor Pressure Vessel (RPV) Diagram 0.5β



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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

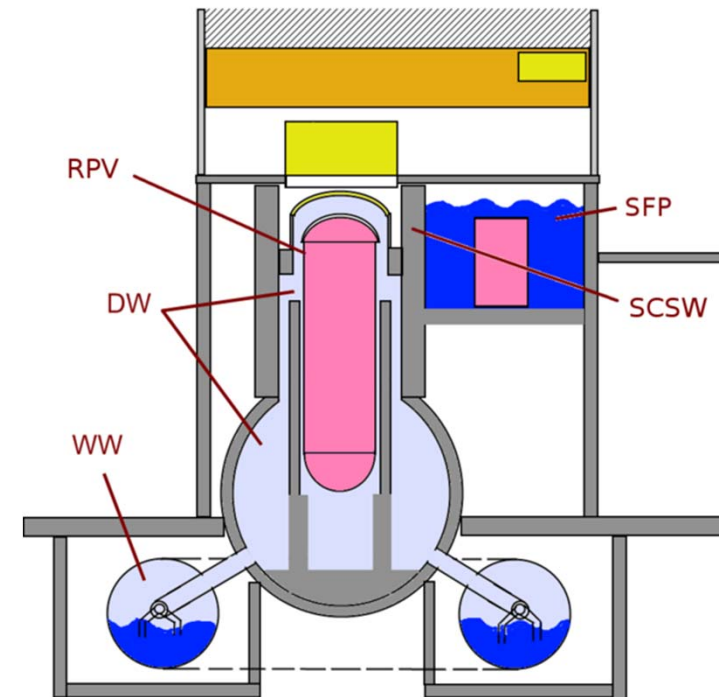
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# Critical Questions

- 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?



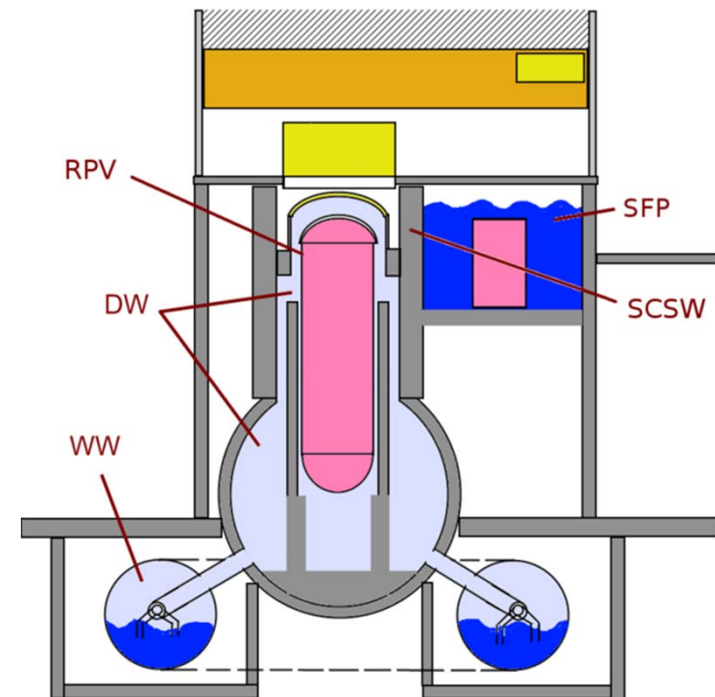
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# Steam Release From Fukushima

- 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



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# 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  $1.3 \times 10^{17}$  Bq for iodine-131 and  $1.1 \times 10^{16}$  Bq for caesium-137, which is about 11% of Chernobyl emissions.”

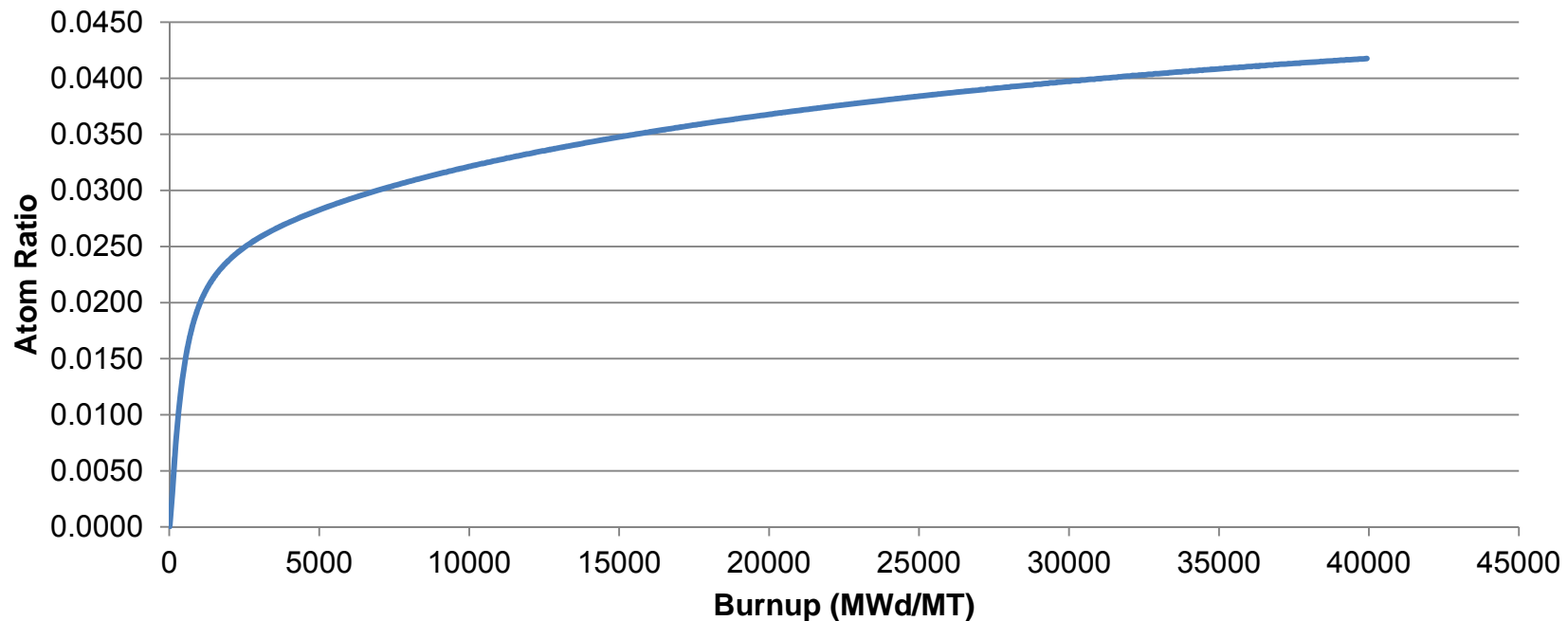
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# Background Nuclides

Ratio of I-127 atoms to Cs-133 atoms in a PWR



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

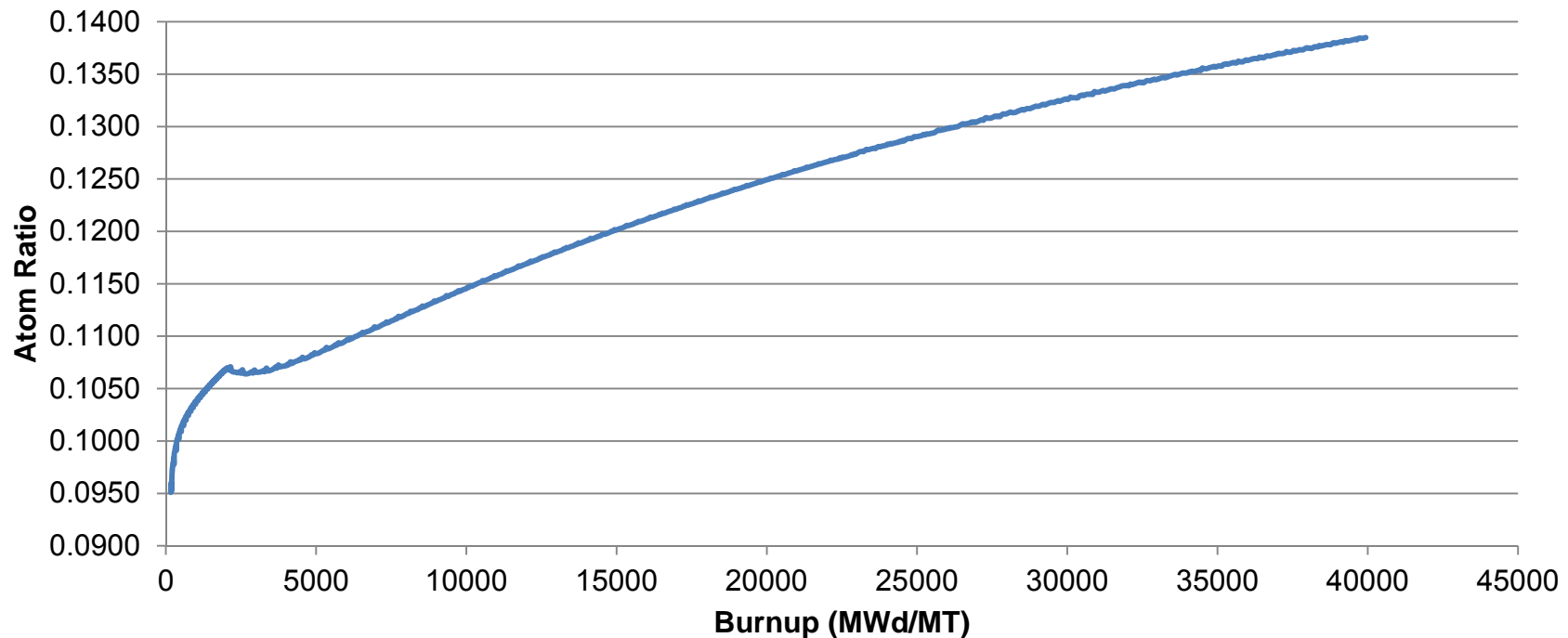
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# Elemental Fractionation

Ratio of I-129 atoms to Cs-137 atoms in a PWR



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

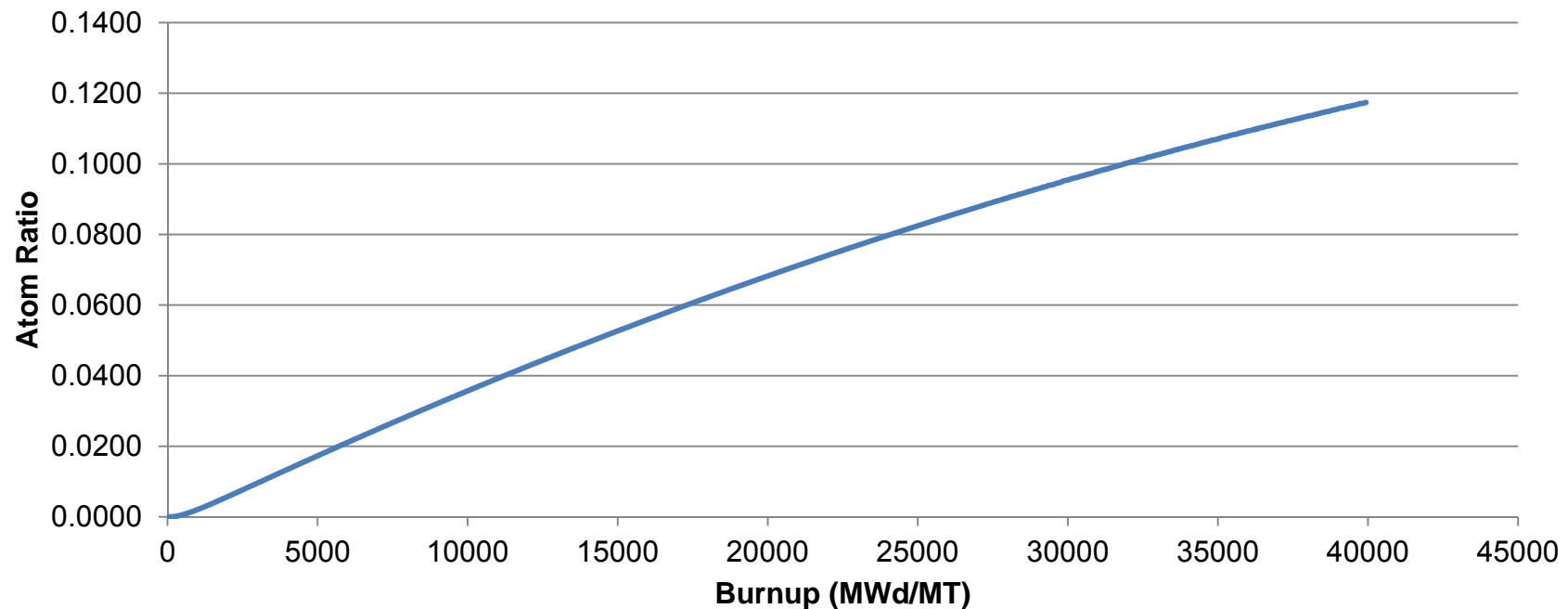
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# Nuclide Ratios Within the Same Element

Ratio of Cs-134 atoms to Cs-137 atoms in a PWR



- 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

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# Ground vs Air Sampling

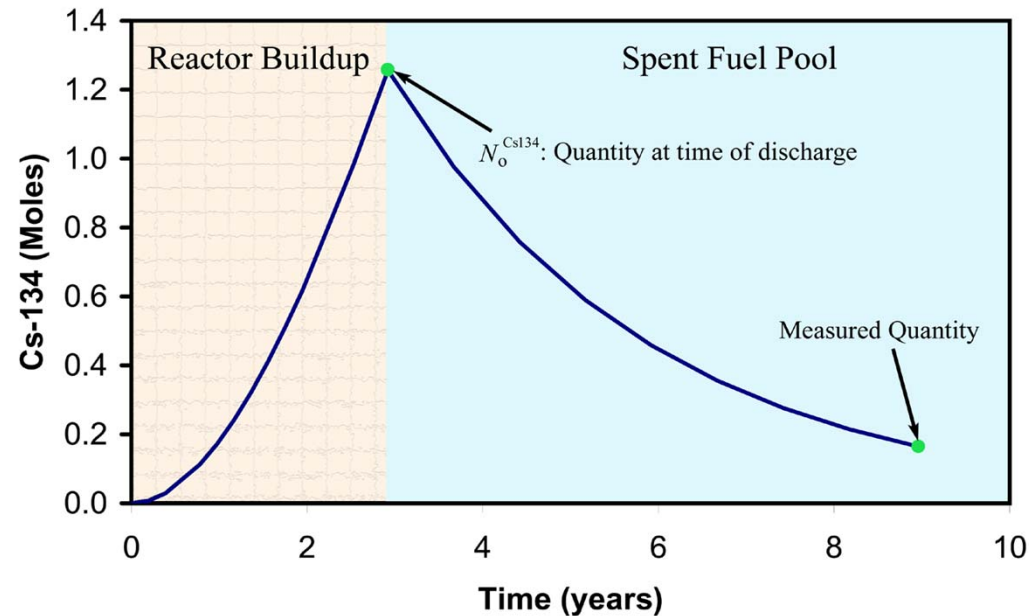
- Soil samples sent to a laboratory
  - Mass spectrometry
  - High-purity germanium detectors (HPGe)
- Onsite or In-Situ ground detection with NaI detectors
- Air Sampling
  - Real time with gamma detectors
  - Air filters collected for mass spectrometry
- Different sampling methods may be biased towards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

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# Spent Fuel Pond vs Reactor Core



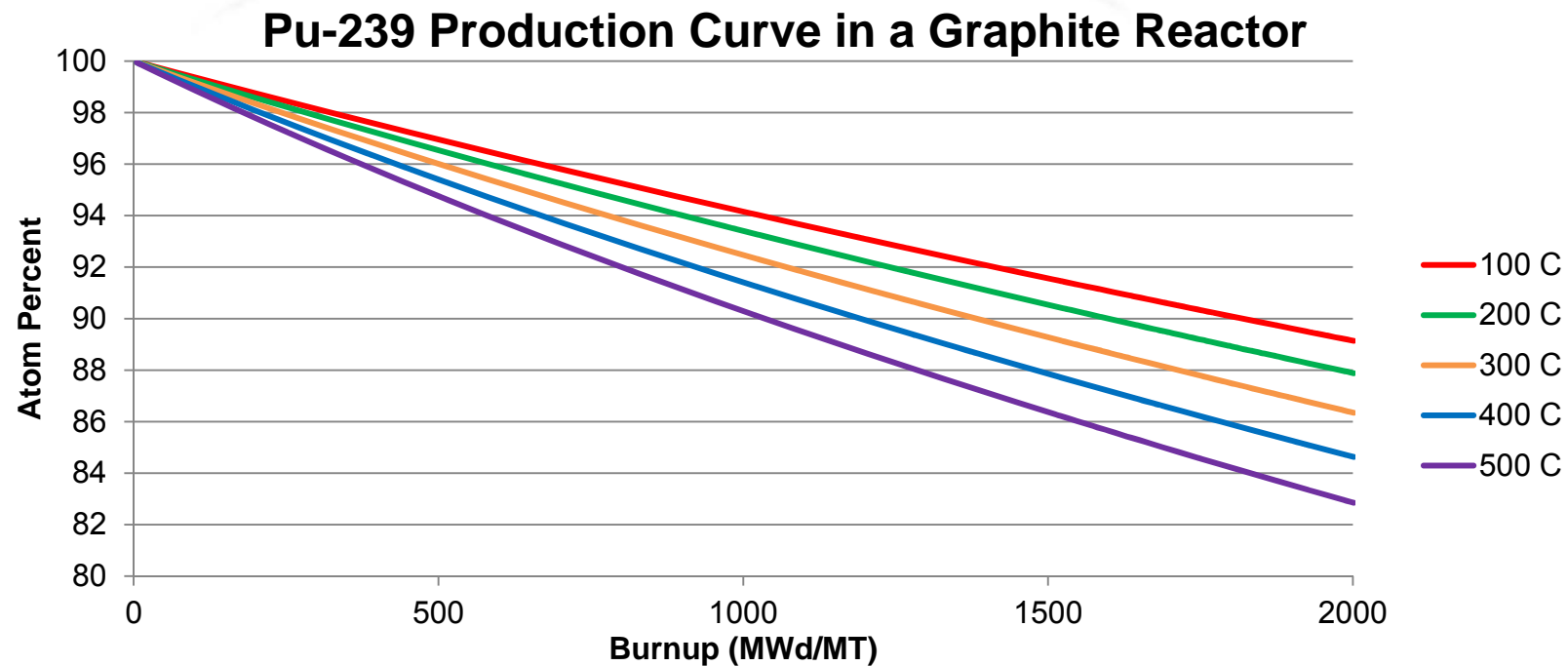
- Cs-136 has a 2 day half-life
- Ratio of Cs-137 atoms to Cs-136 would indicate if the radioactive material is coming from the core, the spent fuel pond, or a mixture.

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# Temperature Effects



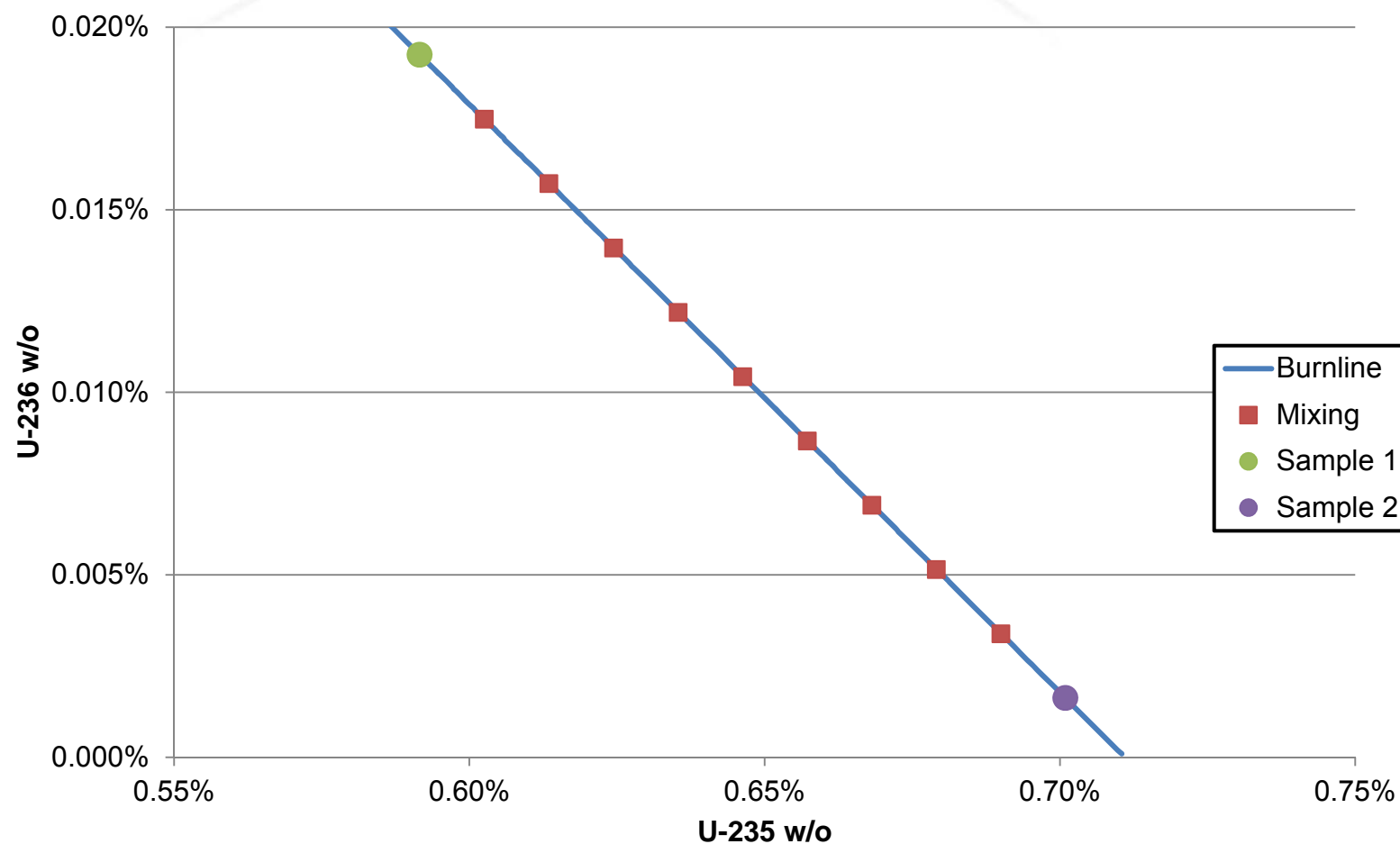
- As the moderator changes temperature the neutron energy changes, which changes the capture and fission rates for a reactor.

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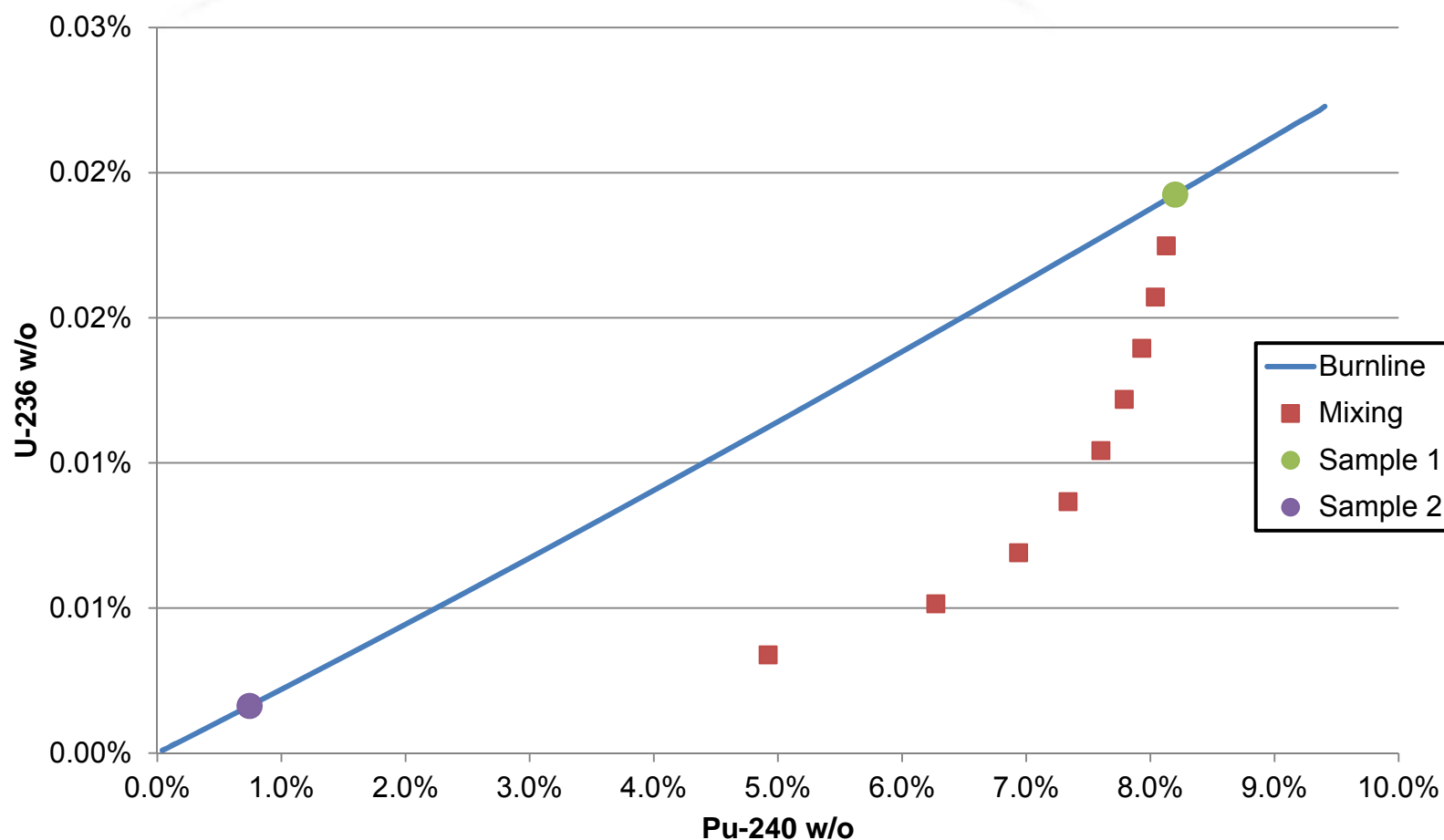
# Uranium Mixing Between Samples



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# Plutonium Mixing Between Samples



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# 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.

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# 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?

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# 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

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