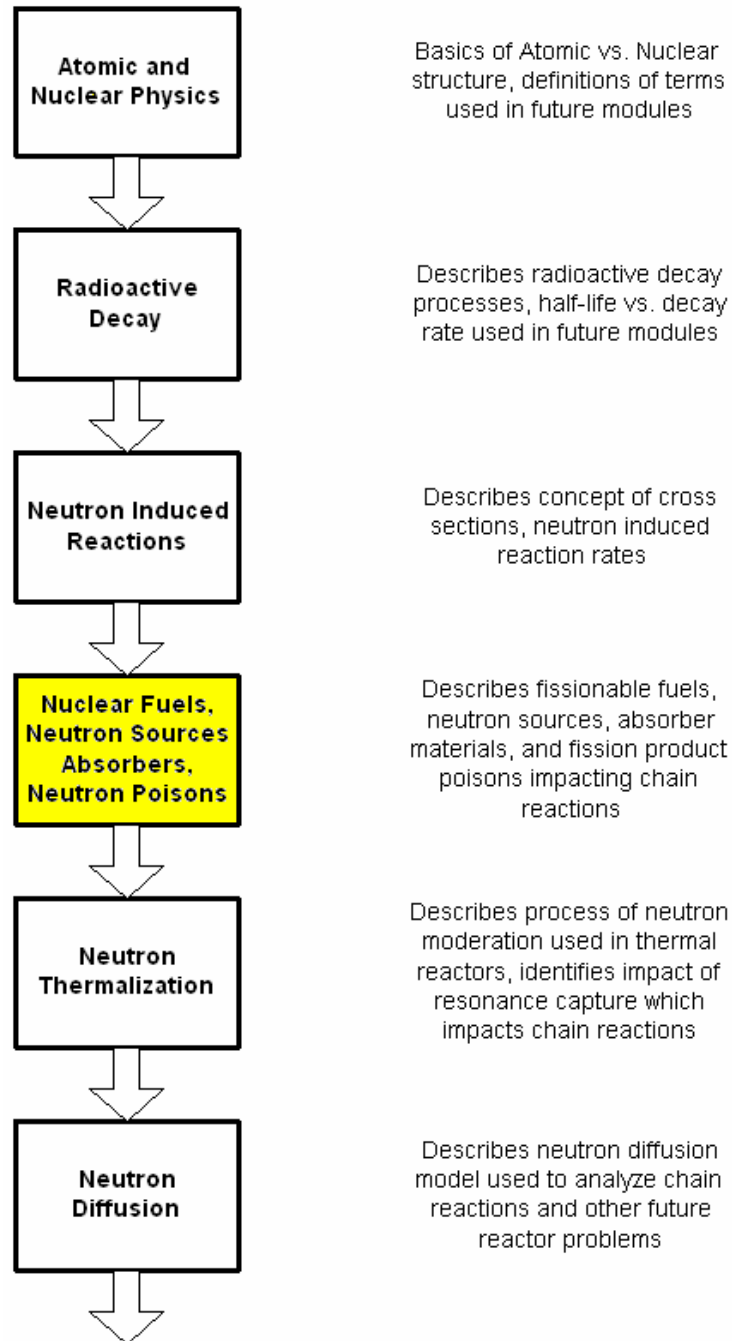


Fundamentals of Nuclear Engineering

Module 4: *Nuclear Fuels, Neutron Sources,
Neutron Absorbers, Neutron Poisons*

Dr. John H. Bickel



Objectives:

Identify key *isotopes, reactions, reaction rates* important to initiating and control of nuclear reactions, including:

1. Key *fissionable isotopes*
2. Key *neutron sources*
3. Key *neutron absorber* materials used for control
4. Key *neutron poisons* arising from fission product decay

Fissionable Isotopes

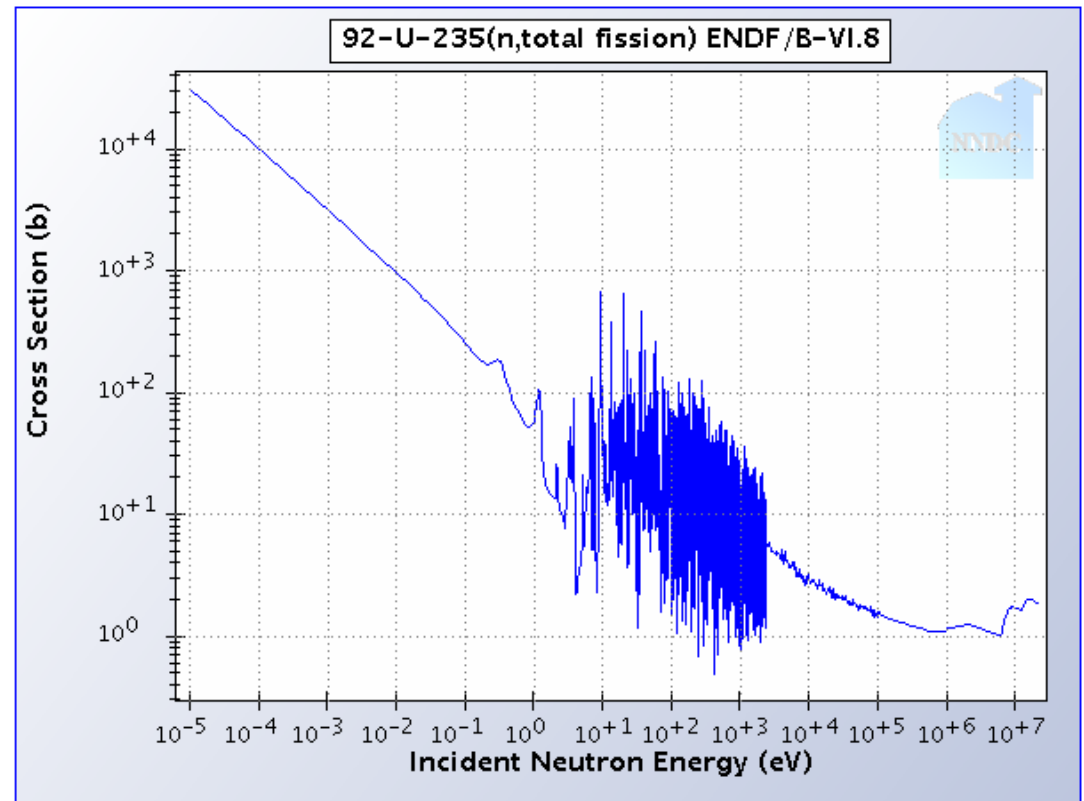
Isotopes Fissionable by Thermal Neutrons

- Many isotopes are capable of undergoing spontaneous fission and energy release
- For *controlled chain fission reaction* - interest is isotopes with: *long decay half-life: $t_{1/2}$, low spontaneous fission branching: α_f , and high thermal neutron fission rate: σ_{f-th}*
- Thermal averaged cross section σ_{f-th} is computed by averaging $\sigma_f(E)$ over thermal neutron energy distribution $\phi_{th}(E)$ – essentially a Maxwell-Boltzmann distribution

$$\sigma_{f-th} = \frac{\int_0^{\infty} \sigma_f(E) \phi_{th}(E) dE}{\int_0^{\infty} \phi_{th}(E) dE}$$

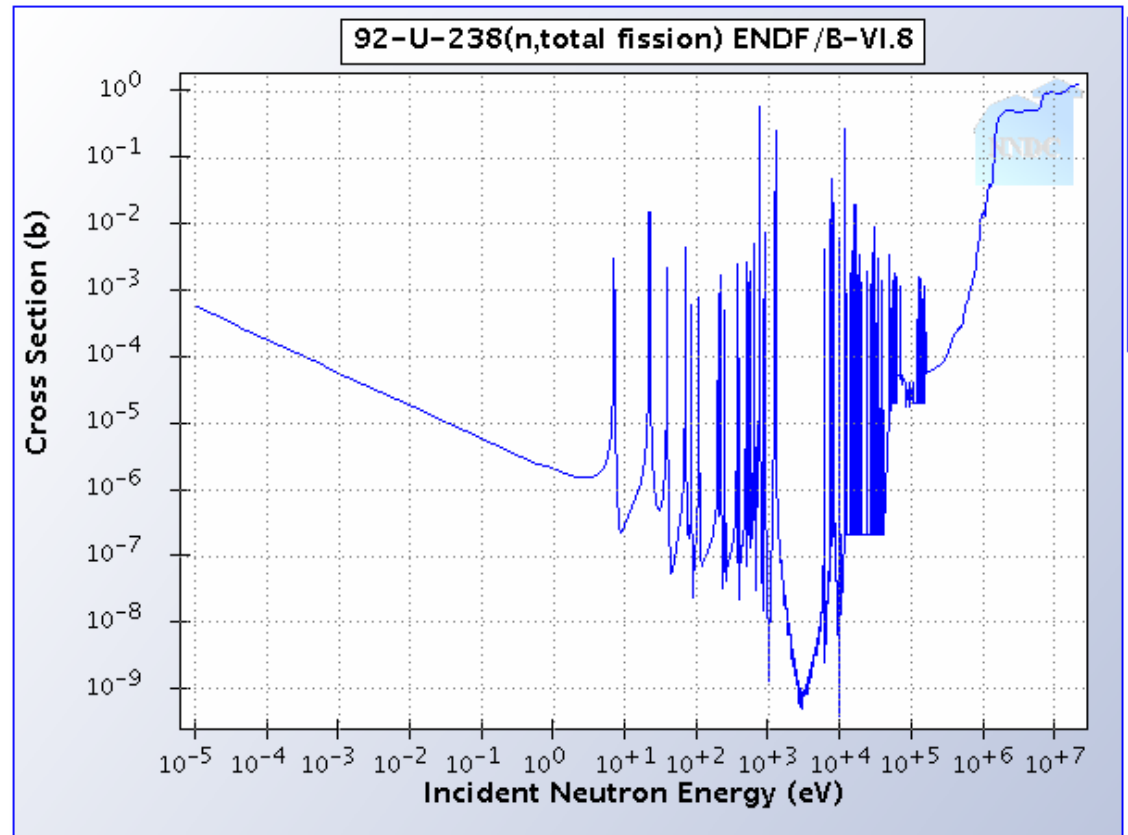
U^{235} Fission

- $t_{1/2} = 7.038 \times 10^8 \text{ yrs}$
- Spontaneous fission rate: $\alpha_{sf} = 7.0 \times 10^{-9}$
- $\sigma_{f-th} = 577 \text{ barns}$
- U^{235} yields ~ 2.43 neutrons/fission
- U^{235} naturally occurring
- Relative abundance 0.72%



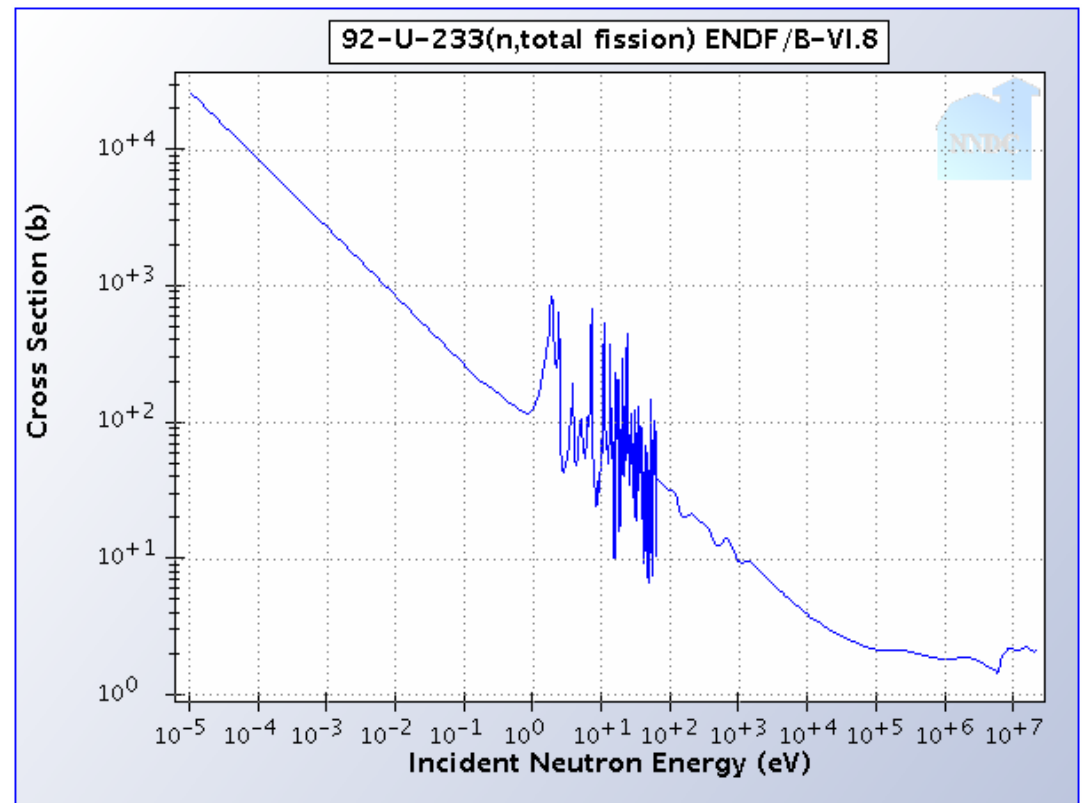
U^{238} Fission

- $t_{1/2} = 4.468 \times 10^9$ yrs
- Spontaneous fission rate: $\alpha_{sf} = 5.5 \times 10^{-5}$
- $\sigma_{f-th} \sim 10^{-5}$ barns
- Fast neutron fission rate is higher
- Overall U^{238} fission rate is small compared to U^{235}
- $\sigma_{f-f} \sim 0.5$ barns
- U^{238} naturally occurring
- Relative abundance 99.2745%



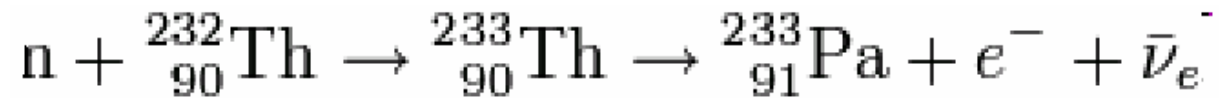
U^{233} Fission

- $t_{1/2} = 1.592 \times 10^5$ yrs
- Spontaneous fission rate: $\alpha_{sf} < 6.0 \times 10^{-11}$
- $\sigma_{f-th} = 527$ barns
- U^{233} yields ~ 2.48 neutrons/fission
- U^{233} is artificial isotope from Th^{232} neutron capture.



U²³³ Origins

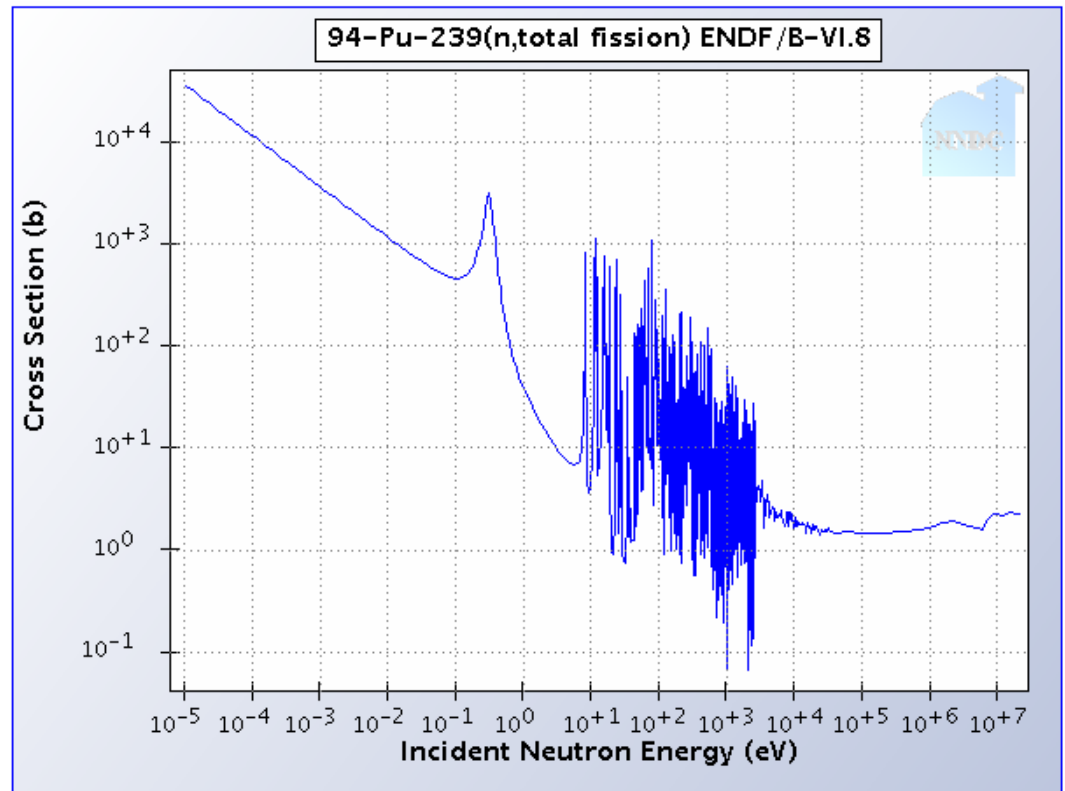
- *U²³³* is produced via following conversion chain from *Th²³²*:



- Recent interest in *U²³³* fission is due to non-proliferation, waste management considerations
- Thorium ore is 3x more plentiful than Uranium
- Large deposits exist in India, Canada, Norway

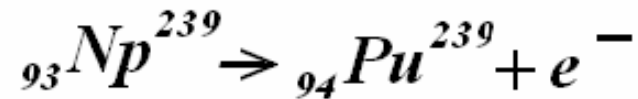
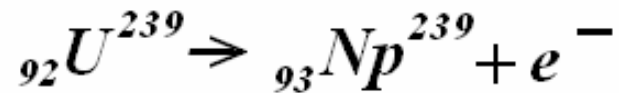
Pu^{239} Fission

- $t_{1/2} = 2.411 \times 10^4$ yrs
- Spontaneous fission rate: $\alpha_{sf} = 3.0 \times 10^{-10}$
- $\langle \sigma_f \rangle_{th} = 742$ barns
- Pu^{239} yields ~ 2.87 neutrons/fission
- Pu^{239} is produced via U^{238} neutron capture



*Pu*²³⁹ Origins

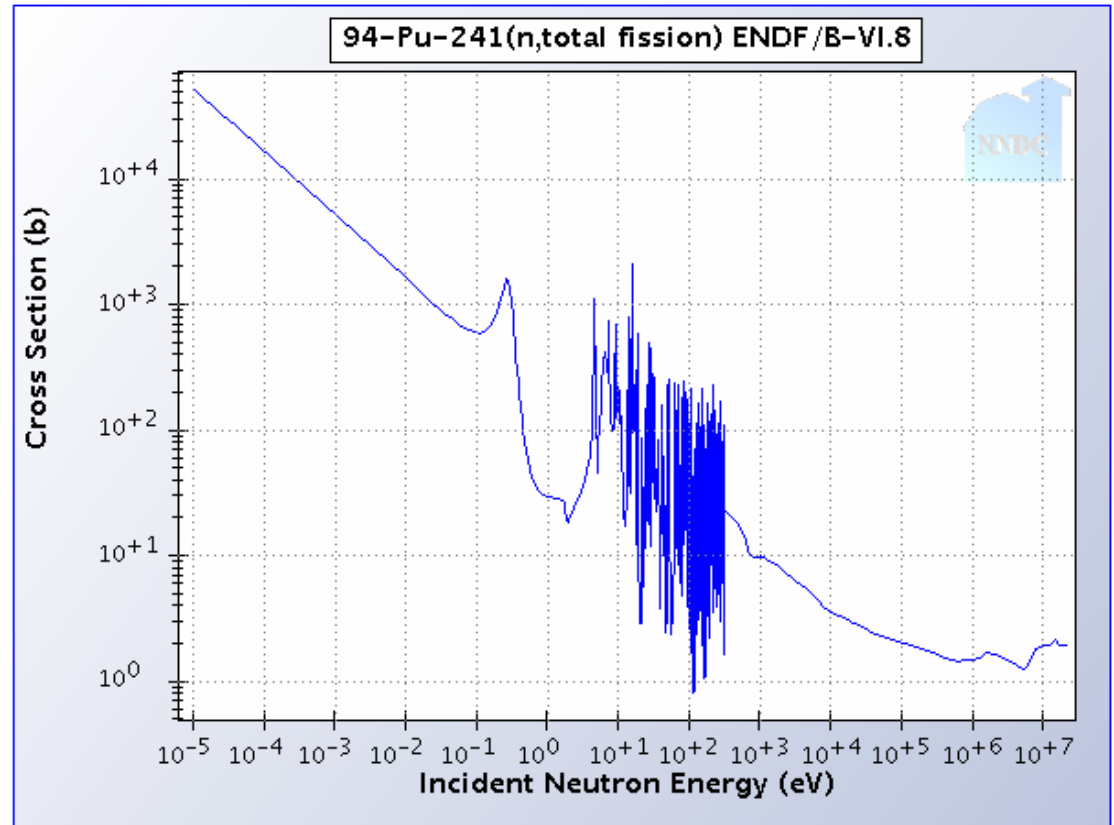
- *Pu*²³⁹ is produced via following conversion chain from *U*²³⁸:



- *Pu*²³⁹ is produced in any light water reactor using lightly enriched *U*²³⁵ Uranium
- Fission of *Pu*²³⁹ contributes significantly to power production at end of reactor fuel cycle as *U*²³⁵ is consumed

Pu^{241} Fission

- $t_{1/2} = 14.35$ yrs
- Very strong β -decay source
- Spontaneous fission rate: $\alpha_{sf} = 2.4 \times 10^{-14}$
- $\sigma_{f-th} = 1025$ barns
- Pu^{241} primarily produced via neutron capture



Three isotopes with desired properties:

- U^{235}
 - U^{233}
 - Pu^{239}
- All others found to: decay too quickly, have large spontaneous fission branching ratios, or too low a thermal neutron cross section.

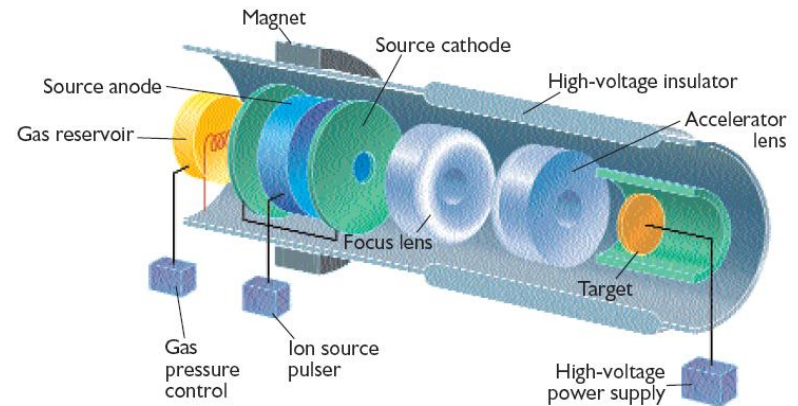
Neutron Sources

Neutrons can be generated by:

- (α, n) reaction from Radium α -particles hitting Beryllium and generating neutrons via: $Be^9(\alpha, n)C^{12}$ (Chadwick - 1932)
- Spontaneous fission neutrons –reliance on random U^{235} fission neutrons for chain reaction initiation is *not desirable*
- Cf^{252} spontaneous fission neutrons ($1\mu g Cf^{252} = 2.8 \cdot 10^6$ n/sec)
- Steady neutron source needed to initiate controlled chain fission reaction in fresh Uranium or Plutonium based fuel
- Radioactive sources via α -decay bombardment: *Ra-Be, Ra-B, Ra-F, Po-Be, Pu-Be*)
- Photo-neutron reactions: $Be^9(\gamma, n)Be^8$ with $E_\gamma > 1.6 MeV$,
 $H^2(\gamma, n)H^1$ with $E_\gamma > 2.23 MeV$
- Fusion reactions: $H^2 (H^2, n)H^3$ (pulsed portable n-generators)

Portable Neutron Generators

- Essentially miniature accelerator for producing fusion neutrons
- Used in laboratory and field survey applications
- $1.5 \cdot 10^8$ neutrons/sec at 14MeV
- 20 -250 kHz pulse rate
- Weight: ~ 25lbs
- Not used for reactor startup



Schematic design of a sealed-tube neutron generator with a Penning ion source.

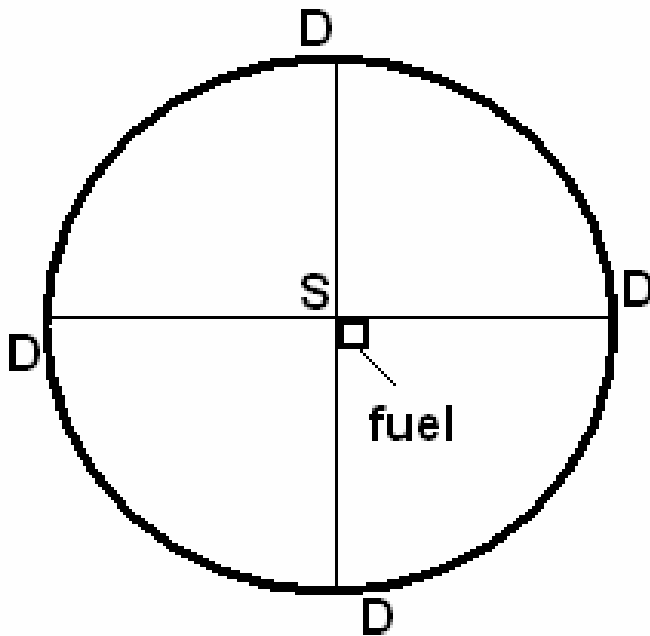
MP 320 Neutron Generator
Lightweight, portable neutron generator
for the most demanding field applications



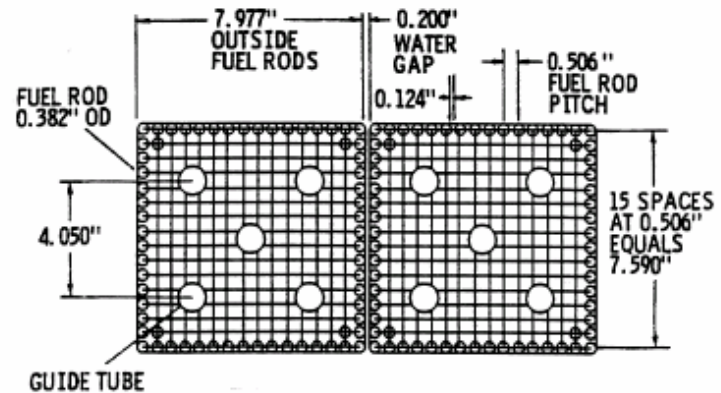
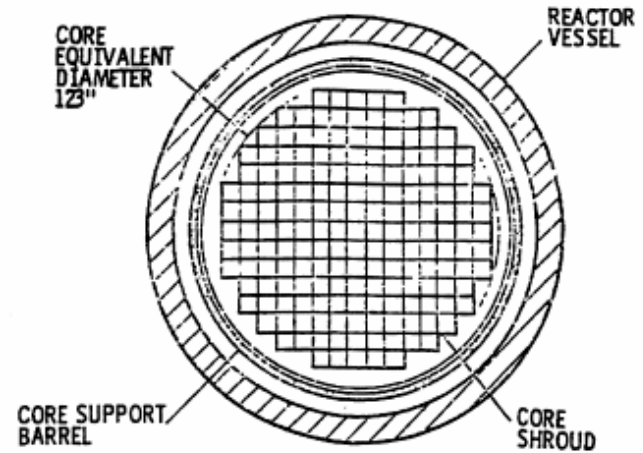
Possible (α, n) Sources for Reactor Startup

- 1 Cu $Ra^{226}-Be^9$ source capable of $1.0-1.5 \times 10^6$ neutrons/sec
- Disadvantage: $Ra^{226}-Be^9$ source has large γ source from Ra decay products.
- 1 Cu $Po^{210}-Be^9$ source capable of $\sim 2.8 \times 10^6$ neutrons/sec without excessive γ production.
- Disadvantage: Po^{210} scarcity.
- $Pu^{239}-Be^9$ source produces ~ 5 MeV neutrons in quantities of: 57.2 neutrons/ 10^6 α absorbed, without excess γ production
- Disadvantage: Pu^{239} is fissionable, has large σ_{f-th} .
- $Am^{241}-Be^9$ source produces ~ 5 MeV neutrons in quantities of: 71.5 neutrons/ 10^6 α absorbed but has small σ_{f-th}
- $Pu^{239}-Be^9$ and $Am^{241}-Be^9$ sources ($>10^6$ neutrons/sec) used for initial reactor startup with fresh fuel
- Source is placed in spare instrument slot in first fuel bundle inserted to reactor

Neutron Source for Initial Startup



S - Neutron Source
D - Neutron Detector



| | | | |
|--|---------------------|-----------------------|-----|
| SAR FIGURE NO. 4.1-2 | | | |
| AMENDMENT - 13 | | | |
| ARKANSAS NUCLEAR ONE UNIT 2 RUSSELLVILLE, ARKANSAS | | SCALE : NONE | |
| | | DRAWN : ENTERGY | |
| | | DESIGN : ENTERGY | |
| | | CAD NO : 00023793.qde | |
| REACTOR CORE CROSS SECTION | BASED ON DRAWING NO | SHEET | REV |
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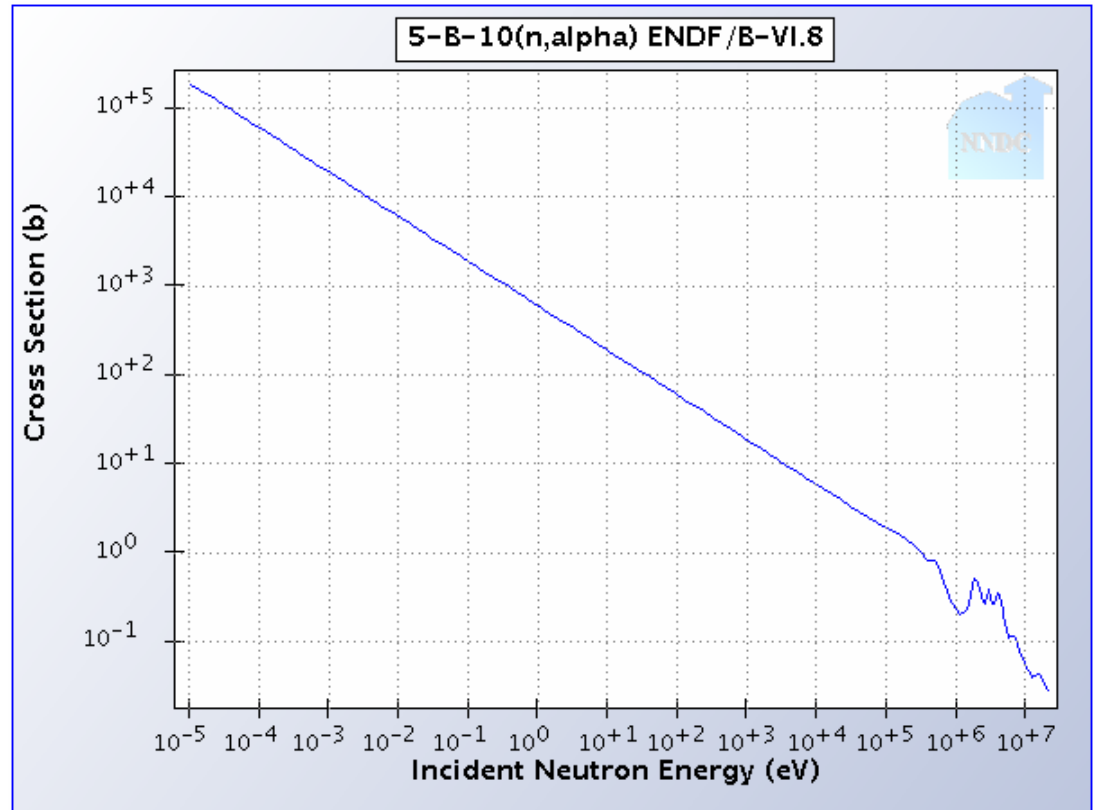
Neutron Absorbers

Neutron Absorbers Are Used to Control Nuclear Chain Reaction

- Key requirements for neutron absorber materials:
- Large neutron capture cross section σ_{c-th} for either (n, γ) or (n, α) type reactions
- Materials suitability for withstanding long term radiation exposure/damage and heat transfer

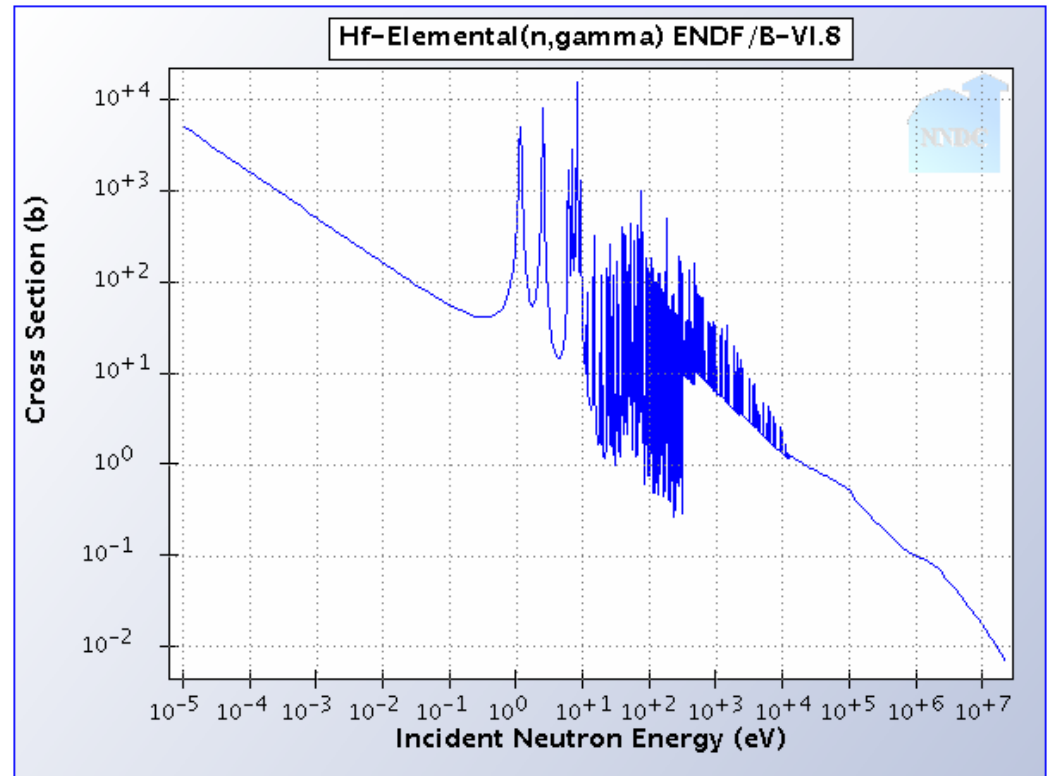
$B^{10}(n,\alpha)Li^7$ Reaction

- $\sigma_{c-th} = 3813 \text{ barns}$
- Typical material is Boron Carbide (B_4C)
- B_4C Melting point is: 2350°C (4262°F)
- Li^7 is non-radioactive
- Natural Boron is 19.9% B^{10} , 80.1% B^{11}
- B^{11} very weak absorber
 $\sigma_{c-th} < 0.05 \text{ barns}$
- Natural Boron (B^{10} , B^{11})
 $\sigma_{c-th} = 755 \text{ barns}$
- Boric Acid and Sodium Pentaborate solutions



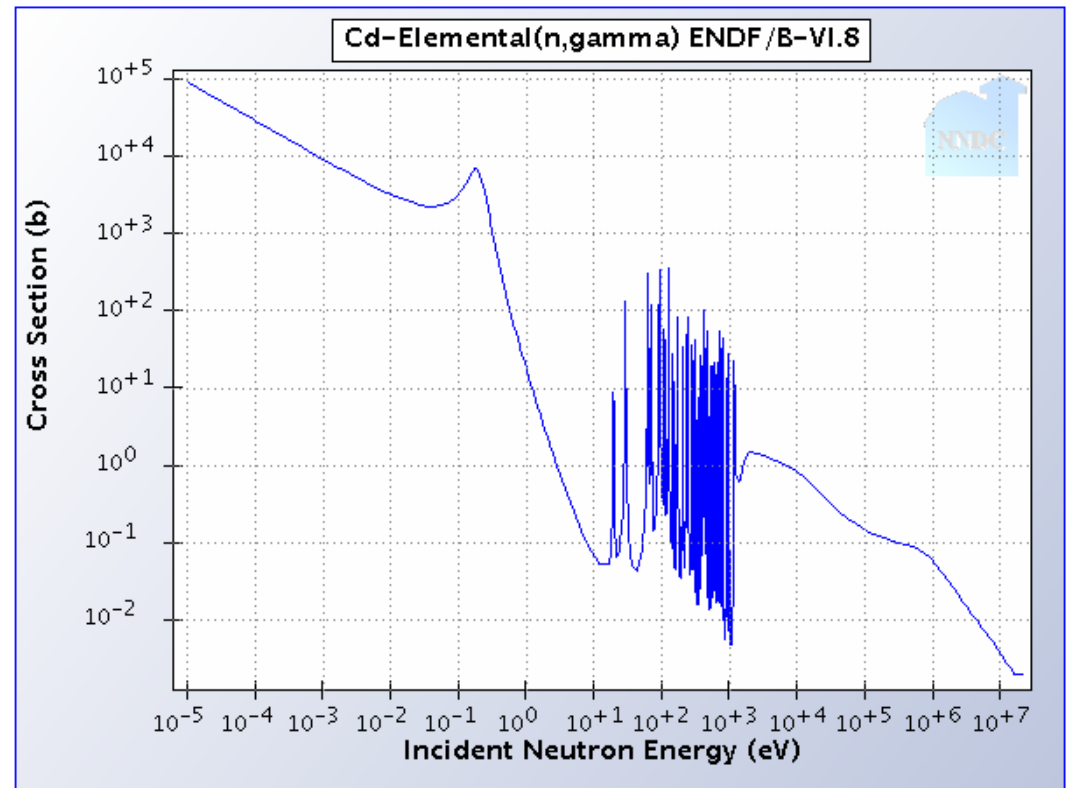
$Hf^x(n, \gamma)Hf^{x+1}$ Reactions

- $\sigma_{c-th} = 105 \text{ barns}$
- Elemental Hafnium is:
 - 35.100% Hf^{180}
 - 27.297% Hf^{178}
 - 18.606% Hf^{177}
 - 13.629% Hf^{179}
 - 5.206% Hf^{176}
 - 0.162% Hf^{174}
- Hafnium melting point is: 2233°C (4051°F)
- Commonly used in Naval Reactors



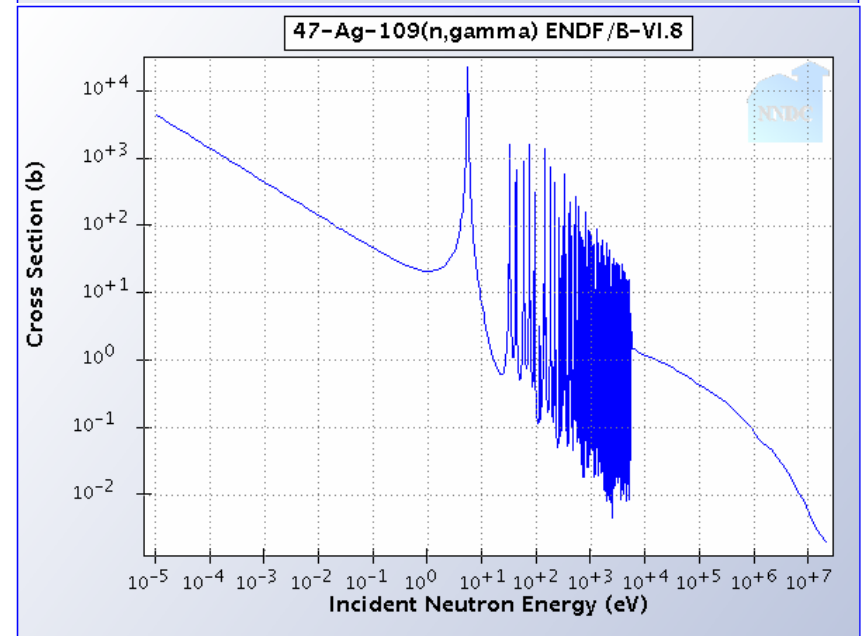
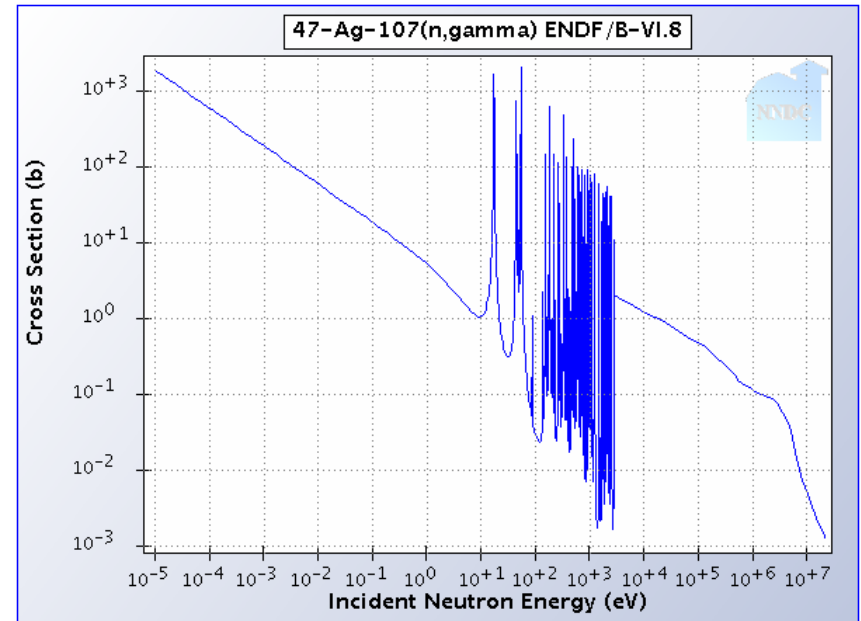
Cadmium (n, γ) Reactions

- $\sigma_{c-th} = 2450 \text{ barns}$
- Elemental Cadmium is:
 - 28.73% Cd^{114}
 - 24.13% Cd^{112}
 - 12.80% Cd^{111}
 - 12.49% Cd^{110}
 - 12.22% Cd^{113}
 - 7.49% Cd^{116}
 - 1.25% Cd^{106}
 - 0.89% Cd^{108}
- Melting Point is 321°C (609.9°F)
- Cadmium was used in first reactor (CP-1)



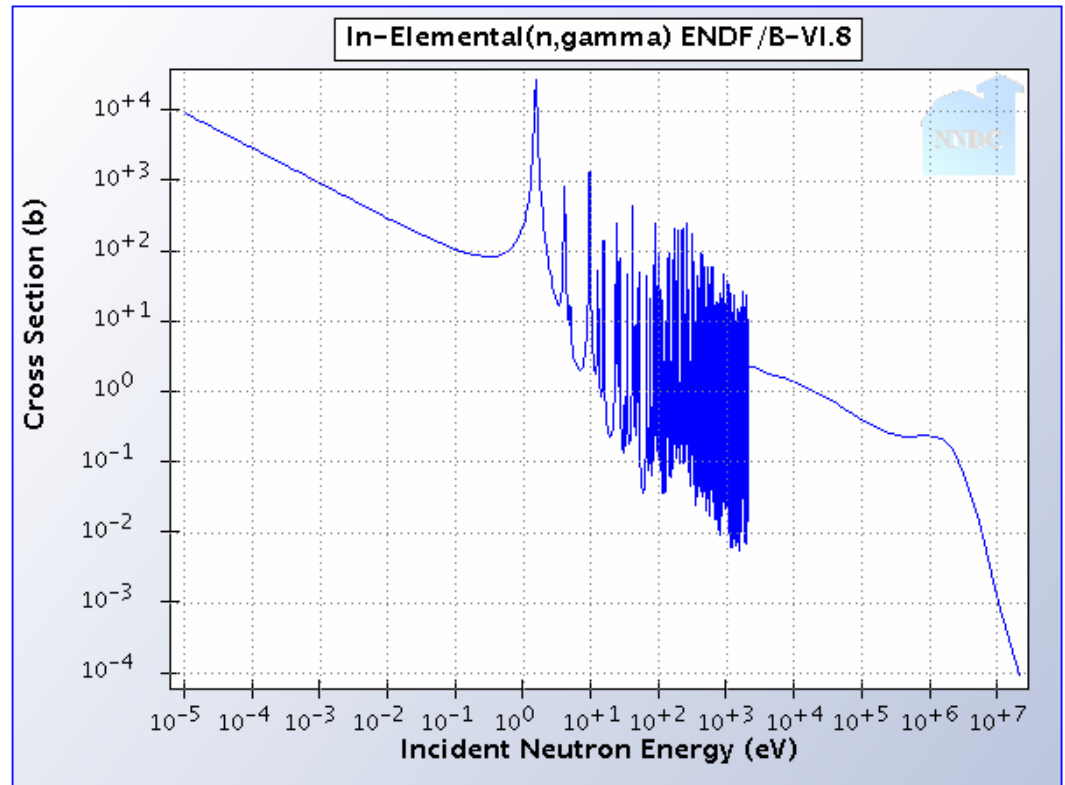
Silver (n, γ) Reactions

- Elemental Silver is:
 - 51.839% Ag^{107}
 - 48.161% Ag^{109}
- Natural Silver (Ag^{107}, Ag^{109})
 $\sigma_{c-th} = 63 \text{ barns}$
- *Capture in resonance region is very large*
- Melting Point is 961°C (1763°F)



Indium (n, γ) Reactions

- Elemental Indium is:
 - 95.7% In^{115}
 - 4.3% In^{113}
- $\sigma_{c-th} = 191 \text{ barns}$
- *Capture in resonance region is very large*
- Melting Point is 156.6°C (313.88°F)



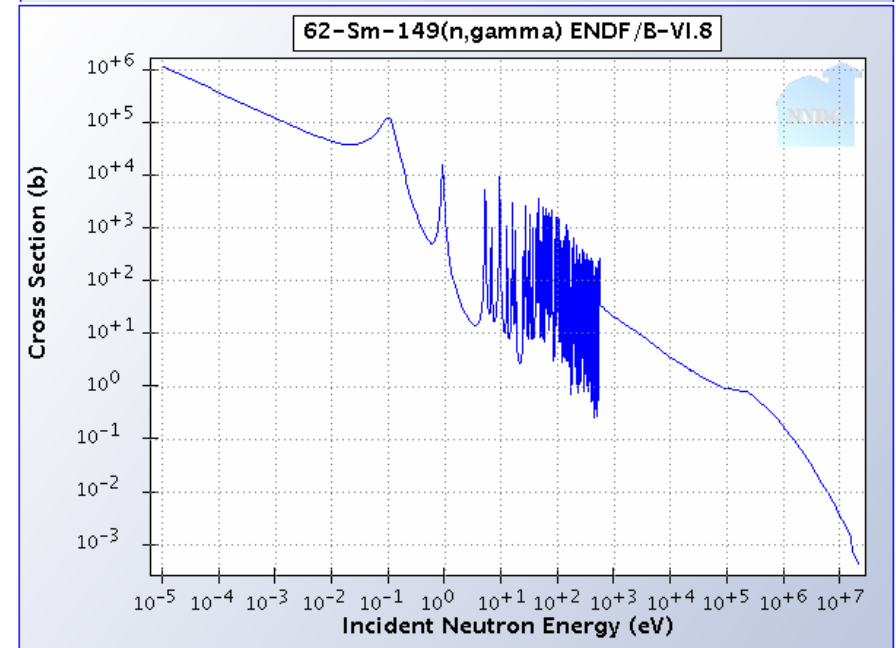
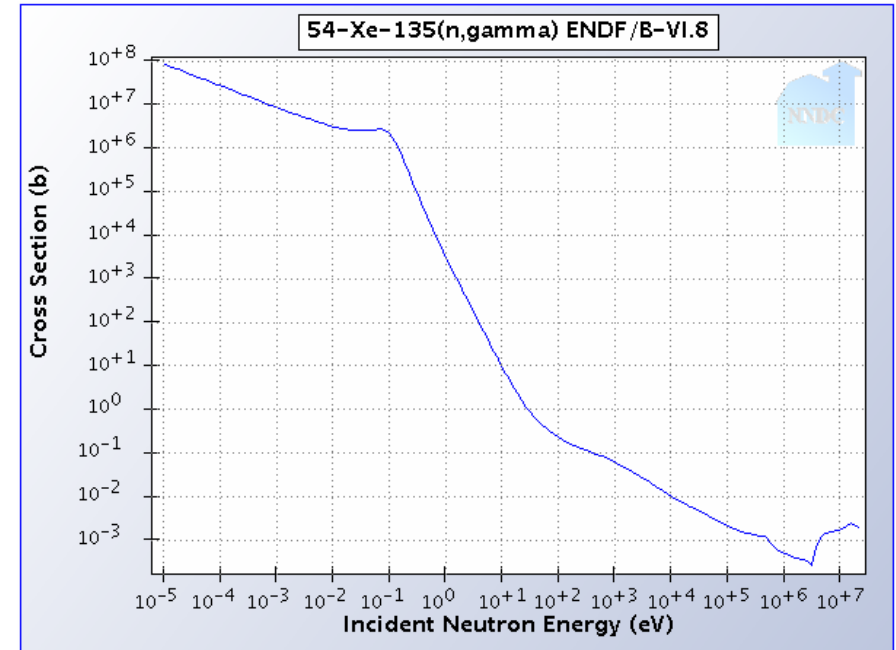
Silver-Indium-Cadmium Alloy

- Due to earlier unavailability of metallic Hafnium, an alloy of Silver-Indium-Cadmium was proposed as Civilian alternative
- Alloy mixture: 80% Silver, 15% Indium, 5% Cadmium
- $$\begin{aligned}\sigma_{c-th} &= 0.8 \sigma_{c-thAg} + 0.15 \sigma_{c-thIn} + 0.05 \sigma_{c-thCd} \\ &= 0.8(63 \text{ barns}) + 0.15(191 \text{ barns}) + 0.05(2450 \text{ barns}) \\ &= 201.55 \text{ barns}\end{aligned}$$
- Alloy mixture has similar nuclear absorption in thermal and resonance regions, but still has relatively low melting point

Fission Product Neutron Poisons

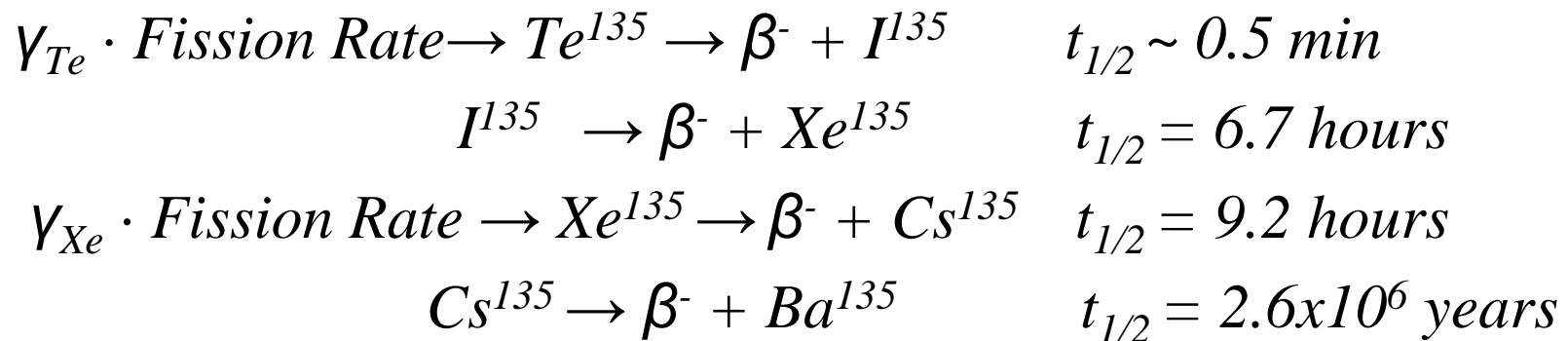
Certain Fission Products are Neutron Absorbers

- Majority of fission products have low neutron capture cross sections
- Two major exceptions:
- Xe^{135} $\sigma_{c-th} = 2.7 \times 10^6$ barns
- Sm^{149} $\sigma_{c-th} = 5.85 \times 10^4$ barns
- Because capture cross sections are large:
- Need to understand:
- Build-up, burn-up, decay physics of these fission products



Xe^{135} Poisoning

- Xe^{135} is direct U^{235} fission product (yield: $\gamma_{Xe} = 0.003$)
- Xe^{135} also produced via Te^{135} decay which is a fission product of U^{235} (yield: $\gamma_{Te} = 0.061$)
- Decay scheme is as follows:



- Thus: system of build-up caused by fission rate, decay, and possibly burn-up of Xe^{135} via neutron capture exists

Xe^{135} Poisoning in U^{235}

- Fission rate: $\varphi_{th} \Sigma_{f-th}$
- Direct production of I^{135} via fission: $Y_{Te} \varphi_{th} \Sigma_{f-th}$
- Elimination of I^{135} via neutron capture: negligible
- Elimination of I^{135} via decay: $-\lambda_I I(t)$ ($I(t)$ is I^{135} concentration)
- Direct production of Xe^{135} via fission: $Y_{Xe} \varphi_{th} \Sigma_{f-th}$
- Production of Xe^{135} via decay of I^{135} : $\lambda_I I(t)$
- Elimination of Xe^{135} via decay: $-\lambda_{Xe} Xe(t)$ ($Xe(t)$ is Xe^{135} conc.)
- Elimination of Xe^{135} via neutron capture is: $-\varphi_{th} \sigma_{c-th} Xe(t)$
- This yields following linear system of equations:

$$dI/dt = Y_{Te} \varphi_{th} \Sigma_{f-th} - \lambda_I I(t)$$

$$dXe/dt = Y_{Xe} \varphi_{th} \Sigma_{f-th} + \lambda_I I(t) - \varphi_{th} \sigma_{c-th} X(t) - \lambda_{Xe} Xe(t)$$

Xe^{135} Poisoning in U^{235}

- Under equilibrium conditions (constant φ_{th}):

$$dI/dt = 0 = Y_{Te} \varphi_{th} \Sigma_{f-th} - \lambda_I I(\infty) \quad \text{thus: } I(\infty) = Y_{Te} \varphi_{th} \Sigma_{f-th} / \lambda_I$$

$$dXe/dt = 0 = Y_{Xe} \varphi_{th} \Sigma_{f-th} + \lambda_I I(t) - \varphi_{th} \sigma_{c-th} Xe(\infty) - \lambda_{Xe} Xe(\infty)$$

- Thus: $Xe(\infty) = [Y_{Xe} \varphi_{th} \Sigma_{f-th} + \lambda_I I(\infty)] / [\varphi_{th} \sigma_{c-th} + \lambda_{Xe}]$
 $= \varphi_{th} \Sigma_{f-th} [Y_{Xe} + Y_{Te}] / [\varphi_{th} \sigma_{c-th} + \lambda_{Xe}]$

- If flux is constant, equilibrium Xe^{135} concentration reached

- When: $\lambda_{Xe} \ll \varphi_{th} \sigma_{c-th}$ - or: $\varphi_{th} \gg \lambda_{Xe} / \sigma_{c-th}$

- This is true when: $\varphi_{th} \gg (0.693/t_{1/2}) / \sigma_{c-th}$

$$\varphi_{th} \gg (0.693 / (6.7 \text{hrs} \cdot 3600 \text{sec/hr})) / (2.6 \cdot 10^6 \text{ barns} \cdot 10^{-24} \text{ cm}^2/\text{barn})$$

$$\varphi_{th} \gg 1.1 \cdot 10^{13} \text{ neutrons /sec. cm}^2$$

- *Commercial power reactors operate exactly in this range!* 31

Xe^{135} Poisoning in U^{235}

- Exact steady state buildup of Xe can be predicted from physics parameters *independent of neutron flux level*

- $Xe(\infty) = \Sigma_{f-th} [Y_{Xe} + Y_{Te}] / \sigma_{c-th}$

- Using numbers: $\Sigma_{f-th} = 48.7 \text{ cm}^{-1}$, $Y_{Xe} = 0.003$, $Y_{Te} = 0.061$,

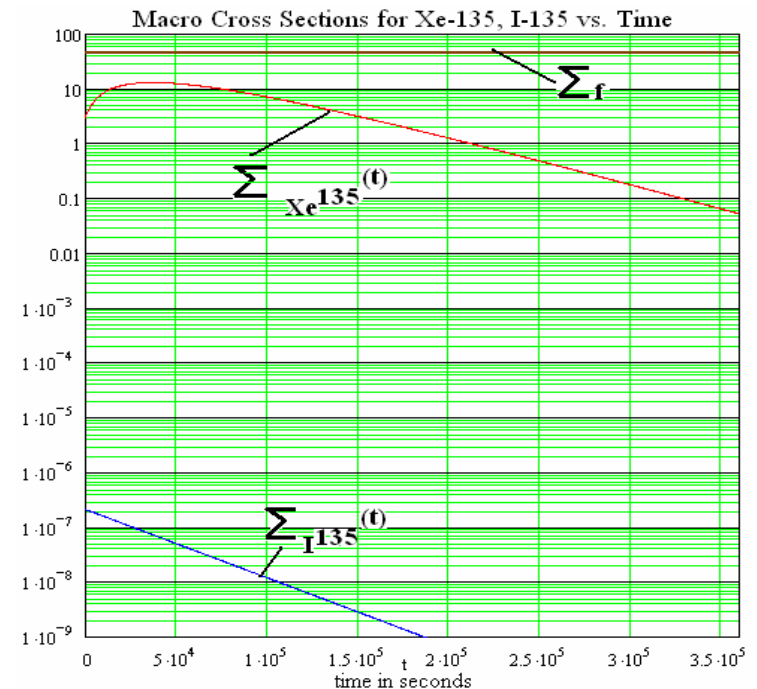
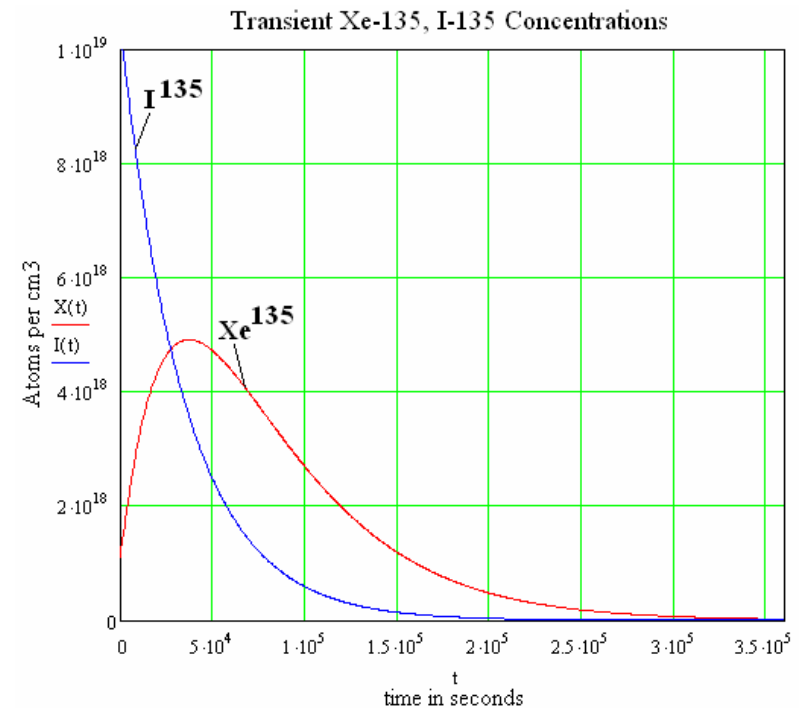
$$\sigma_{c-th} = 2.7 \times 10^6 \text{ barns} \times 10^{-24} \text{ cm}^2/\text{barn} = 2.6 \times 10^{-18} \text{ cm}^2$$

- Then: $Xe(\infty) = 1.1 \times 10^{18} \text{ atoms/cm}^3$

- What happens if after large flux level achieved, it suddenly disappears?

Xe^{135} Poisoning in U^{235}

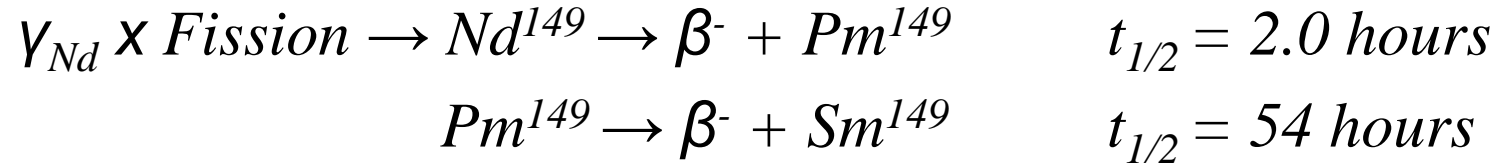
- Thermal flux: $\varphi_{th} \approx 10^{14}/cm^2 sec$
- $I(\infty) = 1 \times 10^{19} atoms/cm^3$
 $Xe(\infty) = 1.1 \times 10^{18} atoms/cm^3$
- Thermal flux drops to: $0/cm^2 sec$
- I^{135} production ceases and begins to decay away
- Xe^{135} production from fission ceases but production from I^{135} decay continues
- Removal of Xe^{135} by neutron capture ceases, but decay continues
- Xe^{135} reaches peak concentration of $4.4 \times 10^{18} atoms/cm^3$ at $\sim 11.6 hrs$
- Xe^{135} competes with fission



Sm^{149} Poisoning

- Sm^{149} is stable isotope produced via decay from Pm^{149} , which is from fission product: Nd^{149} (U^{235} yield: $\gamma_{Nd} = 0.011$)

- Decay scheme is as follows:



- Sm^{149} with $\sigma_{c-th} = 5.85 \times 10^4$ barns, can only be removed by *burning it up* with thermal neutrons
- System of equations governing Sm^{149} build-up/decay is:

$$dPm/dt = \gamma_{Nd} \varphi_{th} \Sigma_{f-th} - \lambda_{Pm} Pm(t)$$

$$dSm/dt = \lambda_{Pm} Pm(t) - \varphi_{th} \sigma_{c-th} Sm(t)$$

Sm^{149} Poisoning in U^{235}

- Under equilibrium conditions (constant φ_{th}):

$$dPm/dt = 0 = \gamma_{Nd} \varphi_{th} \Sigma_{f-th} - \lambda_{Pm} Pm(\infty)$$

- Thus: $Pm(\infty) = \gamma_{Nd} \varphi_{th} \Sigma_{f-th} / \lambda_{Pm}$

$$dSm/dt = 0 = \lambda_{Pm} P(\infty) - \varphi_{th} \sigma_{c-th} Sm(\infty)$$

- Thus: $Sm(\infty) = \lambda_{Pm} Pm(\infty) / \varphi_{th} \sigma_{c-th}$

- Substituting in for $P(\infty)$ yields:

$$Sm(\infty) = \gamma_{Nd} \Sigma_{f-th} / \sigma_{c-th}$$

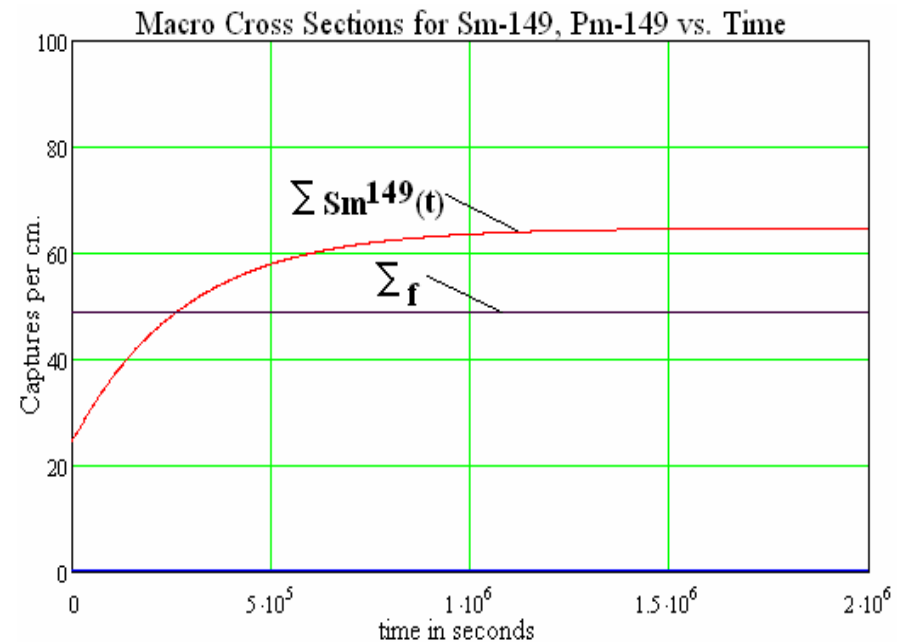
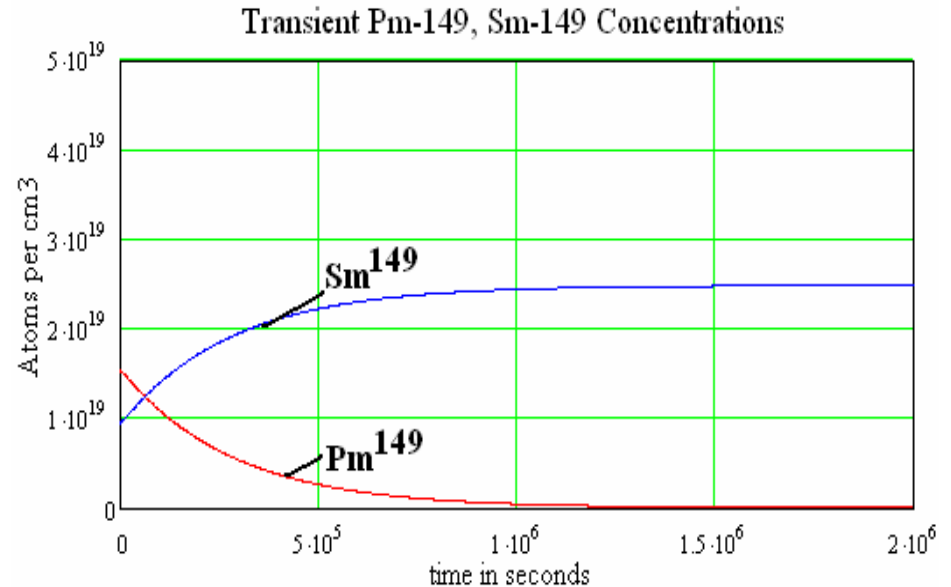
- Using: $\gamma_{Nd} = 0.011$, $\Sigma_{f-th} = 48.7 \text{ cm}^{-1}$,

$$\sigma_{c-th} = 5.85 \times 10^4 \text{ barns} \cdot 10^{-24} \text{ cm}^2/\text{barn} = 5.85 \times 10^{-20} \text{ cm}^2$$

- $Sm(\infty) = 9.4 \cdot 10^{18} \text{ atoms/cm}^3$

Sm^{149} Poisoning in U^{235}

- Thermal flux: $\varphi_{th} = 10^{14}/cm^2 sec$
- $Pm(\infty) = 5.5 \times 10^{19} atoms/cm^3$
 $Sm(\infty) = 9.4 \times 10^{18} atoms/cm^3$
- Thermal flux drops to: $0/cm^2 sec$
- Pm^{149} production ceases and begins to decay away
- Removal of Sm^{149} by neutron capture ceases, but production from Pm^{149} decay continues
- Sm^{149} eventually reaches value of $\sim 2.48 \times 10^{19} atoms/cm^3$
- Sm^{149} capture exceeds fission at $\sim 75 hours$



Reactor Design for Xe^{135} , Sm^{149}

- Obviously nuclear fuel design must consider:
 - All isotopes which capture neutrons: Xe^{135} , Sm^{149} , B^{10} , etc...
 - All isotopes present in fuel that fission: U^{235} , Pu^{239} , Pu^{241} , etc...
- During extended power operation equilibrium Xe^{135} , Sm^{149} capture becomes comparable to Σ_{f-th}
- Fuel design compensates by adjusting Uranium enrichment to increase Σ_{f-th} to cope with *equilibrium levels*
- Fuel design *does not* provide ability to override peak Xe^{135} condition (at 11.6 hours)
- If reactor trips and cannot immediately be restarted it will require waiting 18 - 24 hrs for Xe^{135} to decay

Summary

- Good fission fuels have:
 - Long $T_{1/2}$,
 - Isotope Availability,
 - High σ_f
- Isotopes meeting these requirements include:
 - U^{235}
 - U^{233}
 - Pu^{239}
- Good startup neutron sources:
 - Appreciable neutron production rates
 - Isotope Availability
 - Preferably not major γ -radiation source
- Neutron sources meeting these requirements include:
 - $Pu^{239}\text{-Be}^9$
 - $Am^{241}\text{-Be}^9$

Summary

- Good absorbers for chain reaction control should have:
 - large neutron capture cross section,
 - high melting point,
 - material availability.
- Some good absorbers include:
 - B^{10} Boron Carbide
 - Liquids B^{10} forms: *Boric Acid*, *Sodium Pentaborate*
 - *Halfnium*
 - *Silver-Indium-Cadmium Alloy*
- Most fission products are not major neutron absorbers.
- Fission products that are major neutron absorbers and impact operation:
 - Xe^{135}
 - Sm^{149}