Fundamentals of Nuclear Engineering

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

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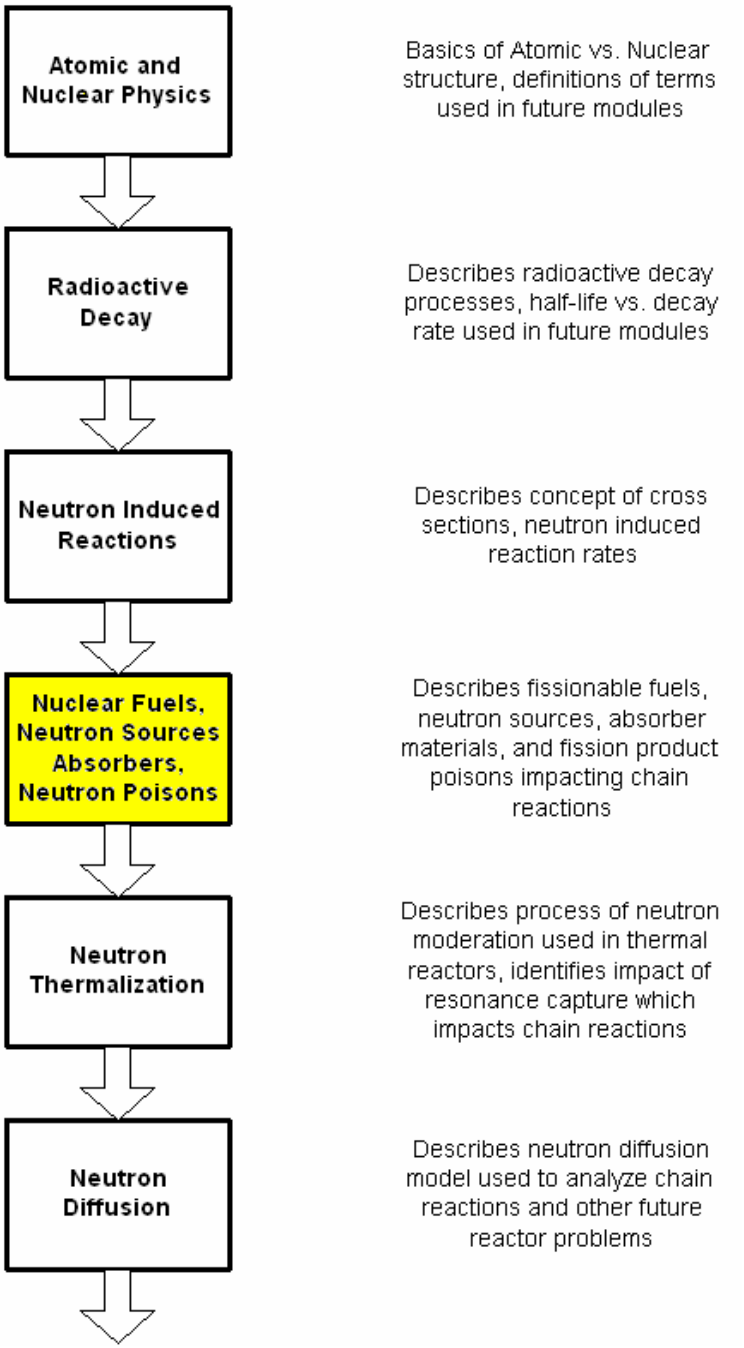

Atomic and Nuclear Physics
Basics of Atomic vs. Nuclear structure, definitions of terms used in future modules
Radioactive Decay
Describes radioactive decay processes, half-life vs. decay rate used in future modules
Neutron Induced Reactions
Describes concept of cross sections, neutron induced reaction rates
Nuclear Fuels, Neutron Sources Absorbers, Neutron Poisons
Describes fissionable fuels, neutron sources, absorber materials, and fission product poisons impacting chain reactions
Neutron Thermalization
Describes process of neutron moderation used in thermal reactors, identifies impact of resonance capture which impacts chain reactions
Neutron Diffusion
Describes neutron diffusion model used to analyze chain reactions and other future reactor problems

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

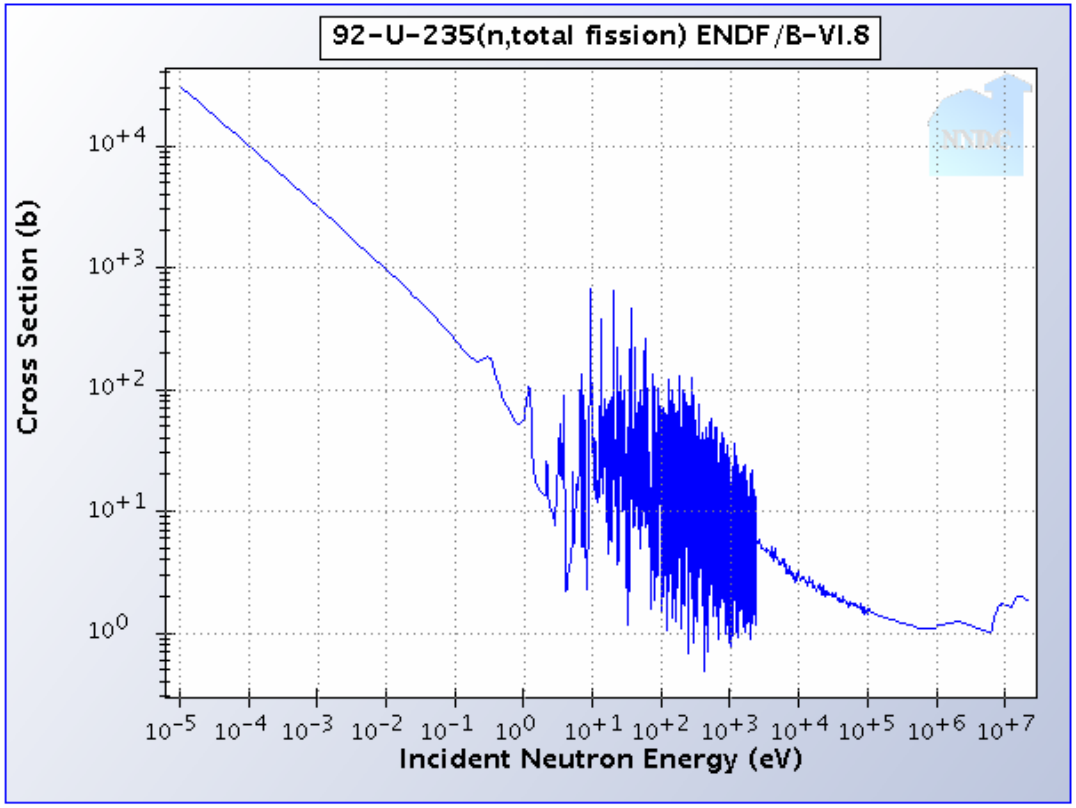
Isotopes Fissionable by Thermal Neutrons

- 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:* $\sigma_{f\text{-}th}$
- Thermal averaged cross section $\sigma_{f\text{-}th}$ is computed by averaging $\sigma_f(E)$ over thermal neutron energy distribution $\varphi_{th}(E)$ – essentially a Maxwell-Boltzmann distribution

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

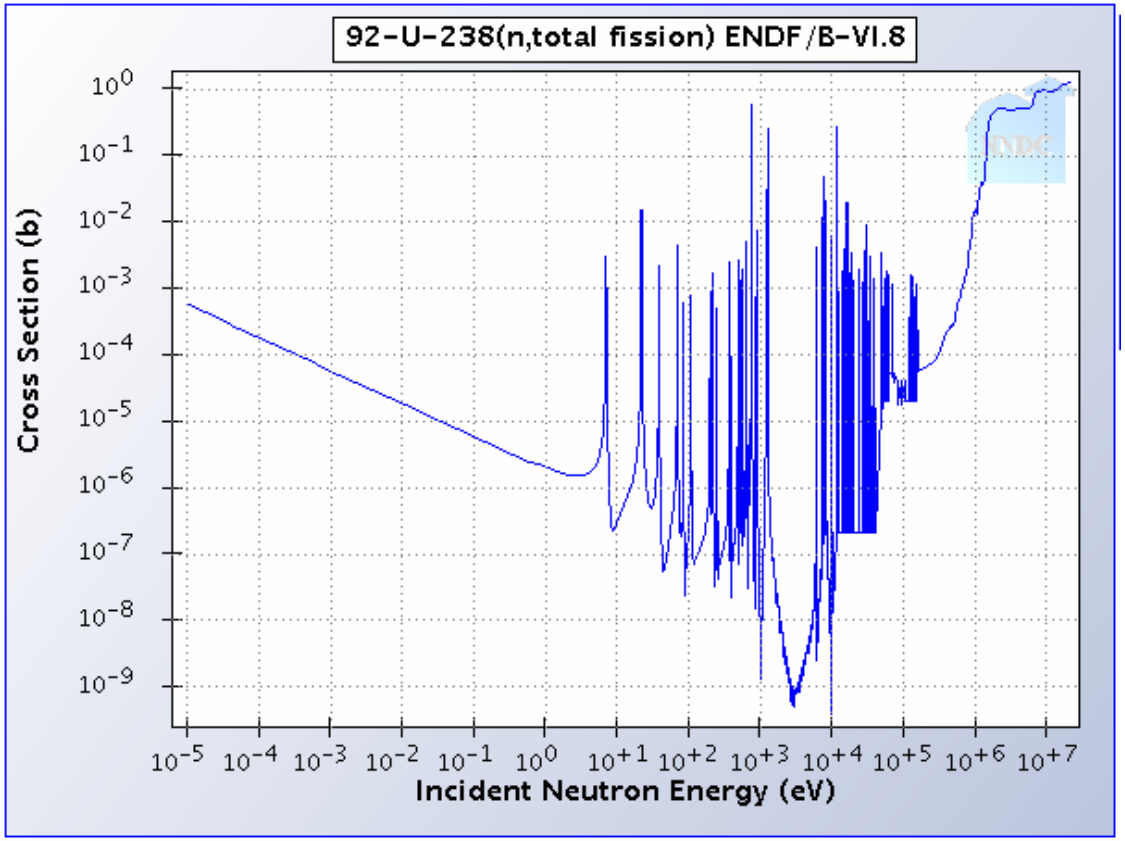
U^{235} Fission

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



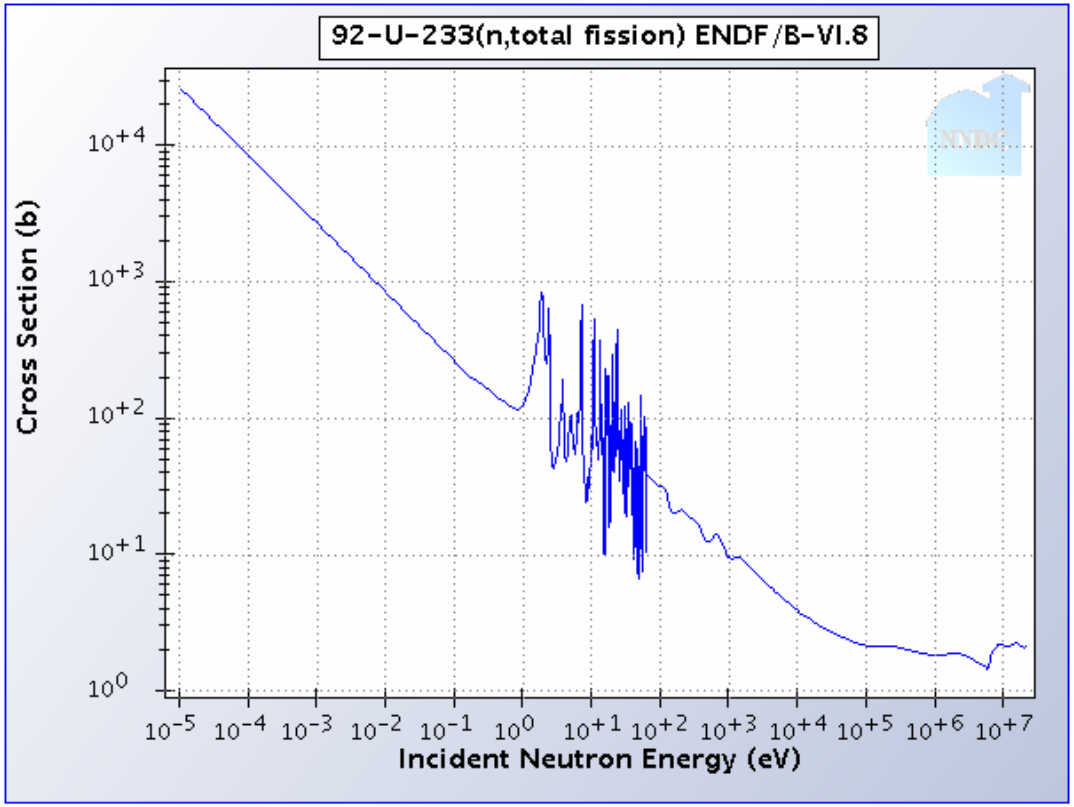
U^{238} Fission

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



U^{233} Fission

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



U^{233} Origins

- U^{233} is produced via following conversion chain from Th^{232}:

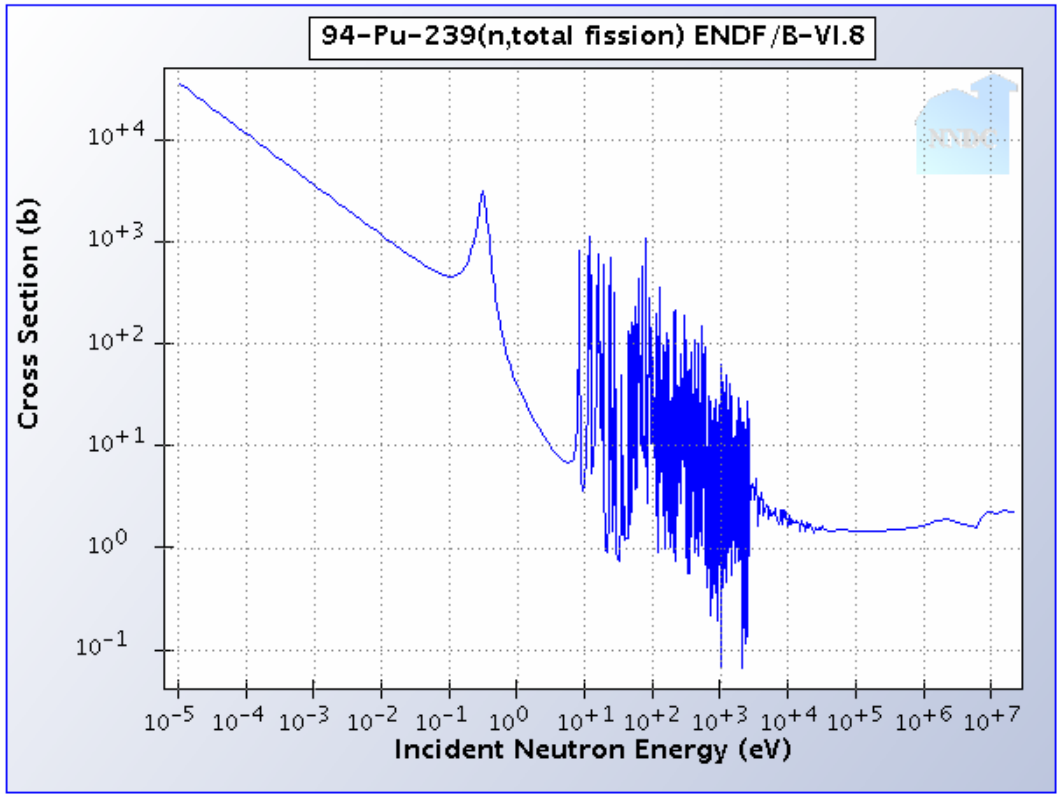
$$\mathrm{n} + {}^{232}_{90}\mathrm{Th} \rightarrow {}^{233}_{90}\mathrm{Th} \rightarrow {}^{233}_{91}\mathrm{Pa} + e^{-} + \bar{\nu}_e$$

$$ {}^{233}_{91}\mathrm{Pa} \rightarrow {}^{233}_{92}\mathrm{U} + e^{-} + \bar{\nu}_e$$

- Recent interest in U^{233} 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}:

$$_{92}U^{238} + n \rightarrow {}_{92}U^{239} + \gamma$$

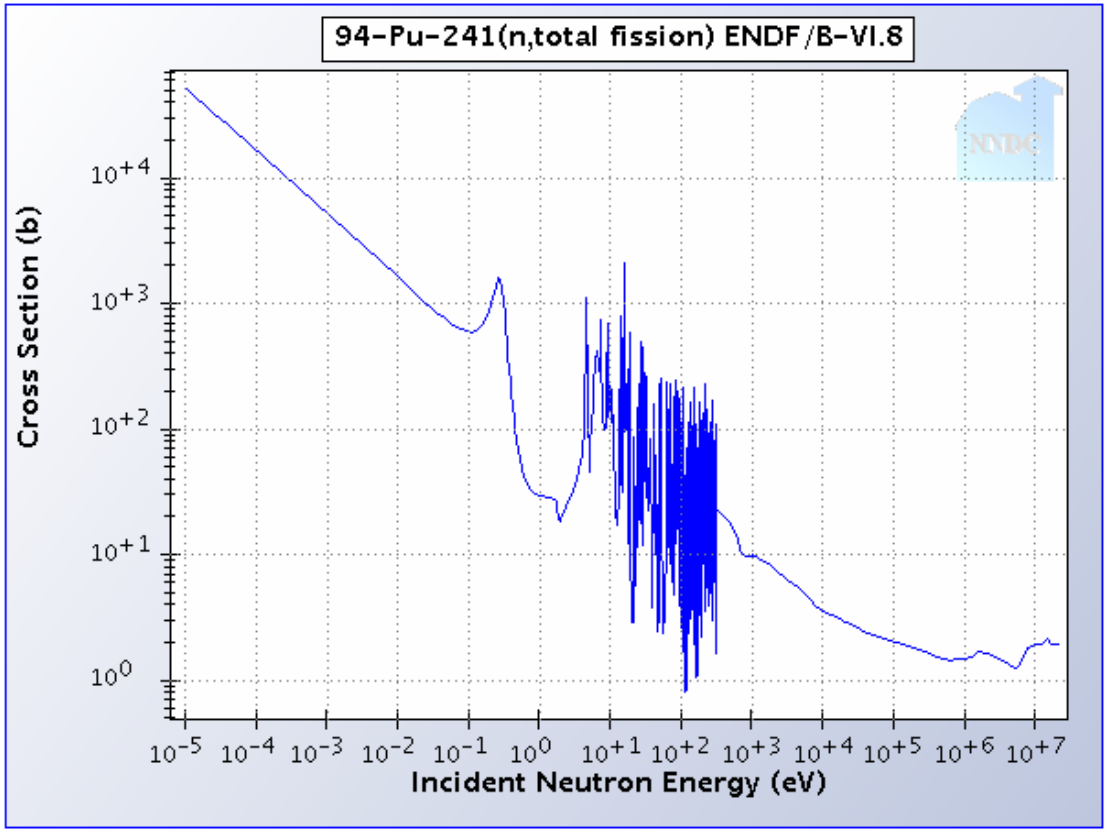
$$_{92}U^{239} \rightarrow {}_{93}Np^{239} + e^-$$

$$_{93}Np^{239} \rightarrow {}_{94}Pu^{239} + e^-$$

- 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: $\alpha_{sf} = 2.4 \times 10^{-14}$
- $\sigma_{f\text{-}th} = 1025\ barns$
- 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.

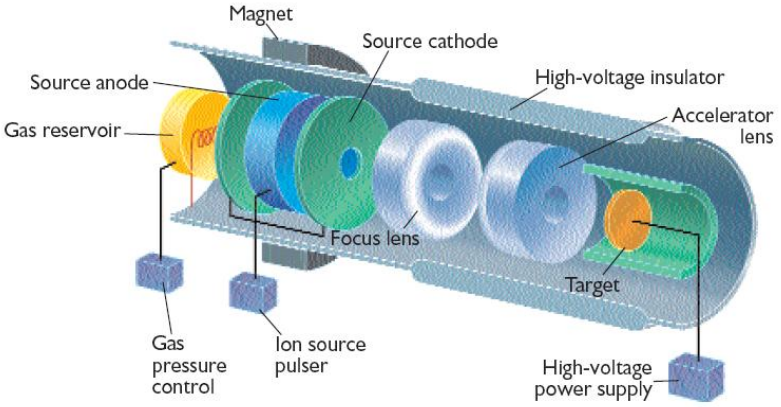
Neutron Sources

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 (1μg 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: $Ra\text{-}Be$, $Ra\text{-}B$, $Ra\text{-}F$, $Po\text{-}Be$, $Pu\text{-}Be$)
- Photo-neutron reactions: $Be^{9}(\gamma,n)Be^{8}$ with $E\gamma > 1.6MeV$, $H^{2}(\gamma,n)H^{1}$ with $E\gamma > 2.23MeV$
- 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

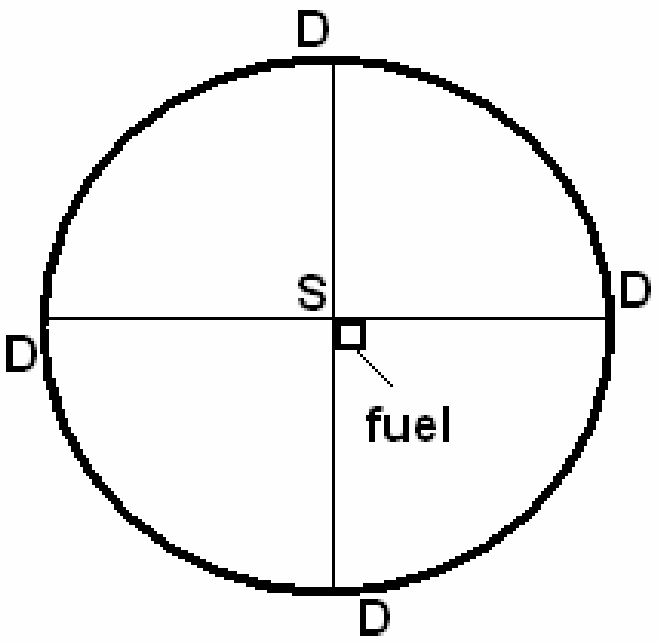


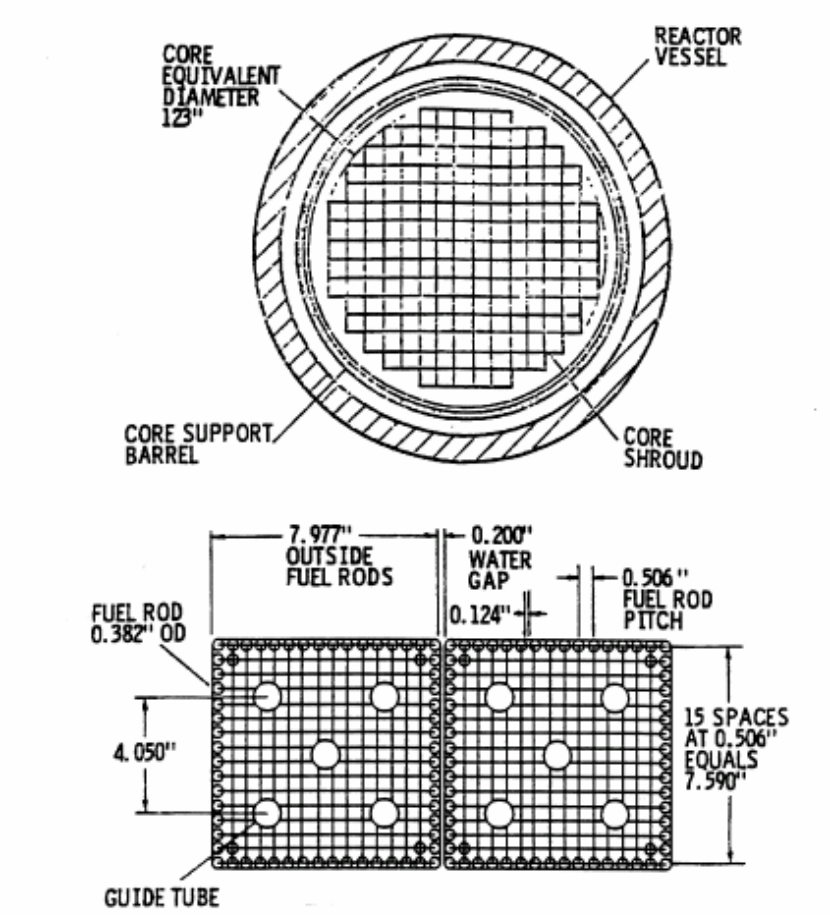
Schematic design of a sealed-tube neutron generator with a Penning ion source.

Possible (α,n) Sources for Reactor Startup

- 1 Cu Ra^{226}-Be^{9} source capable of 1.0-1.5 x 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 ~ 2.8 x 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 $\sigma_{f\text{-}th}$.
- Am^{241}-Be^{9} source produces ~5 MeV neutrons in quantities of: 71.5 neutrons/10^6 α absorbed but has small $\sigma_{f\text{-}th}$
- 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





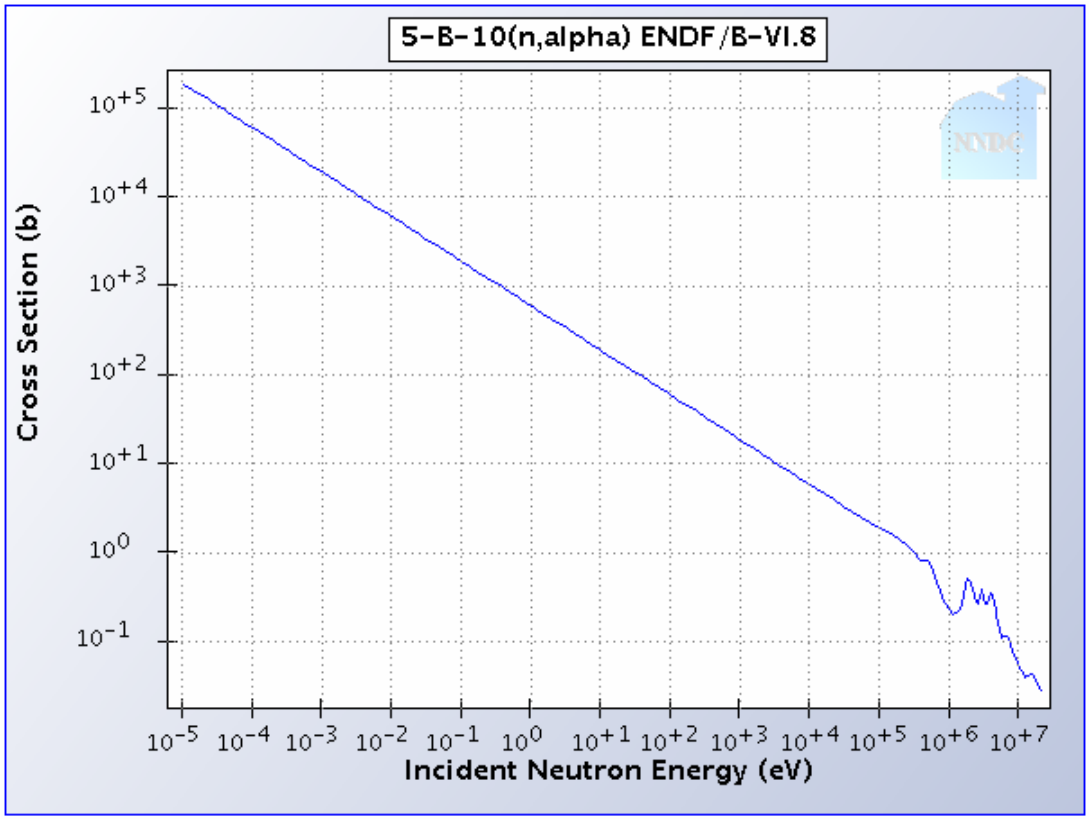
Neutron Absorbers

Neutron Absorbers Are Used to Control Nuclear Chain Reaction

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

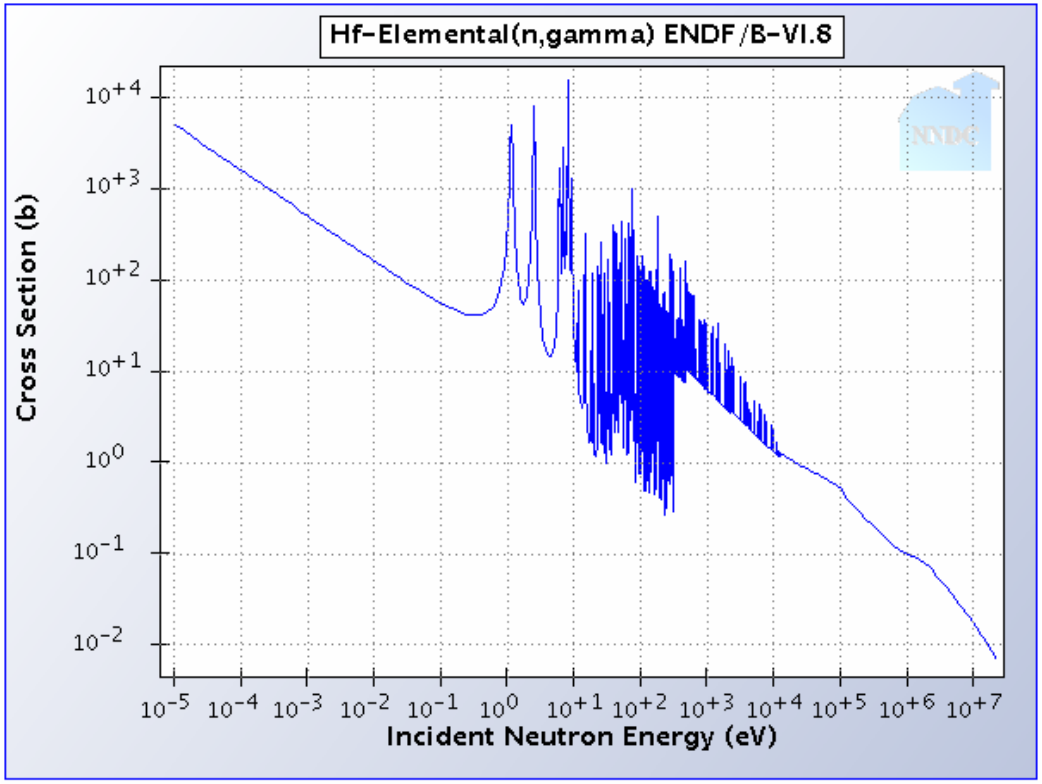
$B^{10}(n,\alpha)Li^{7}$ Reaction

- $\sigma_{c\text{-}th} = 3813$ *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* $\sigma_{c\text{-}th} < 0.05$ *barns*
- Natural Boron (B^{10}, B^{11}) $\sigma_{c\text{-}th} = 755$ *barns*
- Boric Acid and Sodium Pentaborate solutions



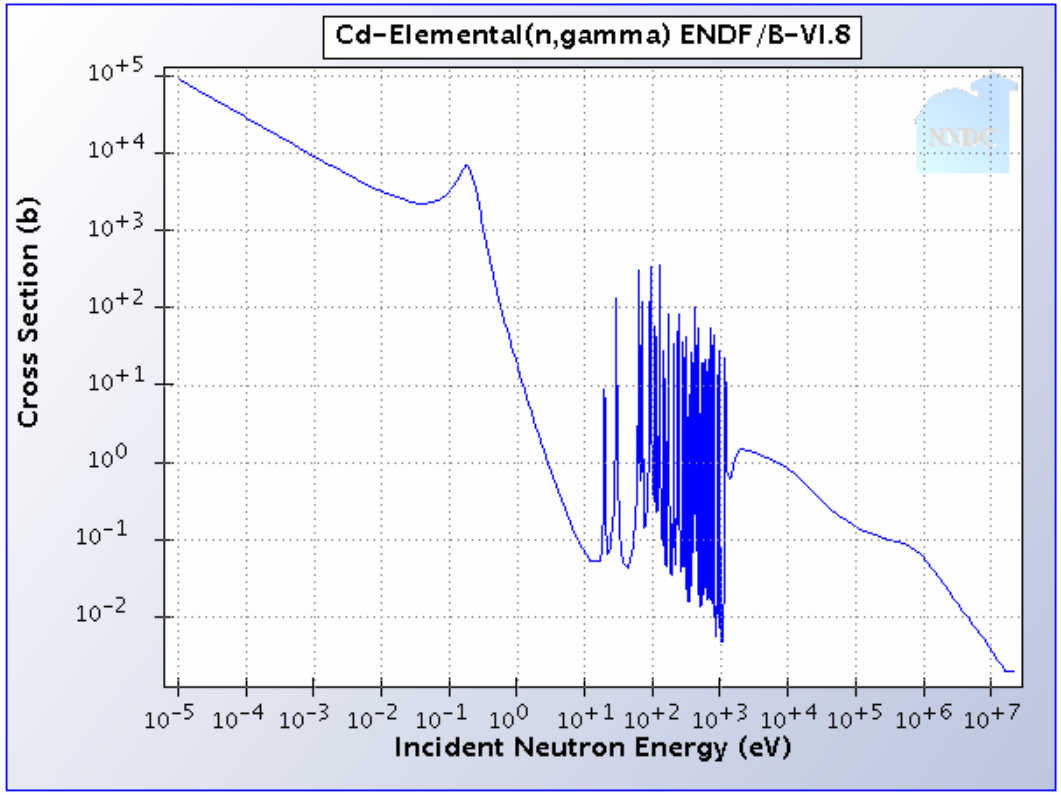
$Hf^{x}(n,\gamma)Hf^{x+1}$ Reactions

- $\sigma_{c\text{-}th}$ = *105 barns*
- Elemental Halfnium 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}
- Halfnium melting point is: 2233°C (4051°F)
- Commonly used in Naval Reactors



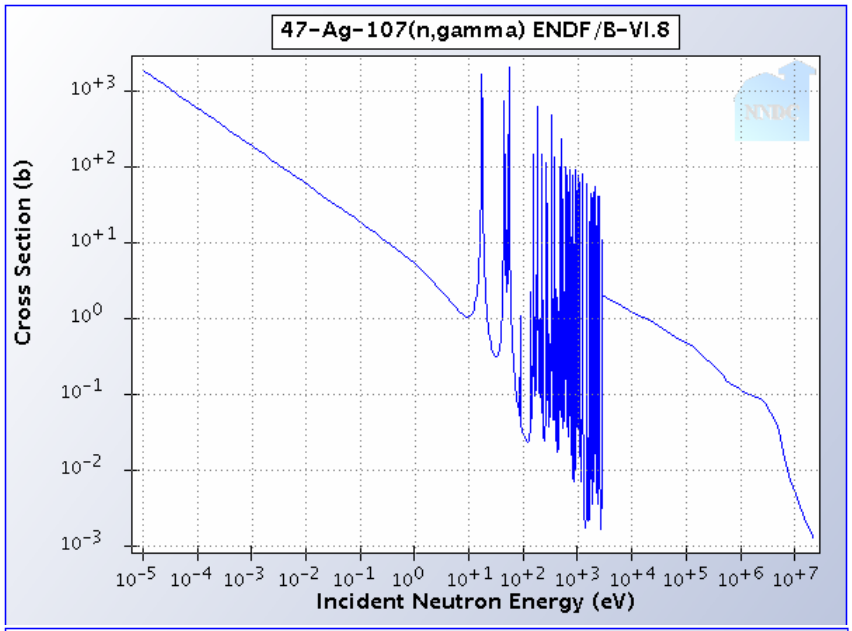
Cadmium (n,γ) Reactions

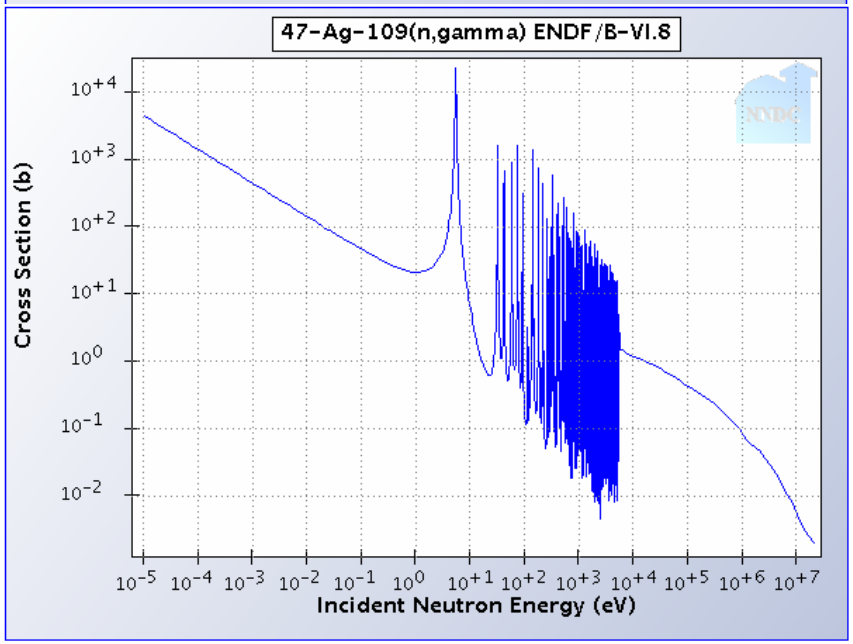
- $\sigma_{c\text{-}th}$ = *2450 barns*
- Elemental Cadmium is:
  - *28.73%* Cd^{114}
  - *24.13%* Cd^{112}
  - *12.80%* Cd^{111}
  - *12.49%* Cd^{110}
  - *12.22%* Cd^{113}
  - *7.49%* Cd^{116}
  - *1.25%* Cd^{106}
  - *0.89%* Cd^{108}
- Melting Point is 321°C (609.9°F)
- Cadmium was used in first reactor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



Silver (n,γ) Reactions

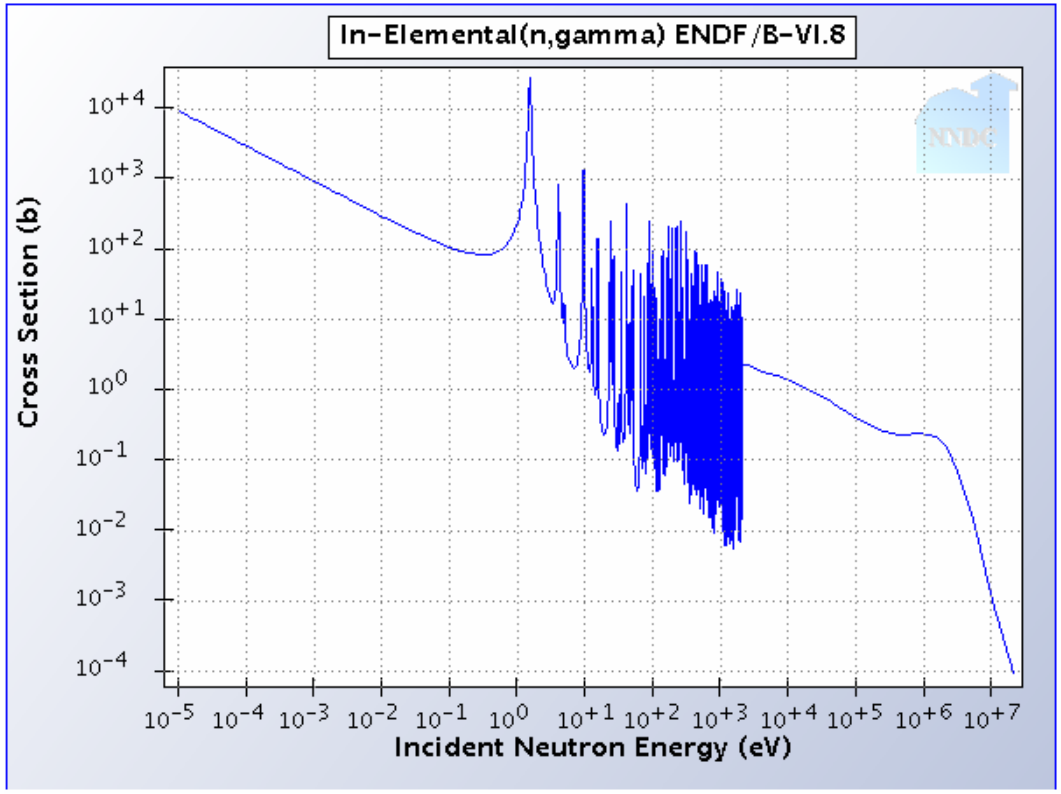
- Elemental Silver is:
  - *51.839%* Ag^{107}
  - *48.161%* Ag^{109}
- Natural Silver (Ag^{107}, Ag^{109})
  $\sigma_{c\text{-}th} = 63$ *barns*
- *Capture in resonance region is very large*
- Melting Point is 961°C (1763°F)





Indium (n,γ) Reactions

- Elemental Indium is:
  - *95.7%* In^{115}
  - *4.3%* In^{113}
- $\sigma_{c\text{-}th}$ *= 191 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 Halfnium, an alloy of Silver-Indium-Cadmium was proposed as Civilian alternative
- Alloy mixture: 80% Silver, 15% Indium, 5% Cadmium
- $\sigma_{c\text{-}th} = 0.8\,\sigma_{c\text{-}thAg} + 0.15\,\sigma_{c\text{-}thIn} + 0.05\,\sigma_{c\text{-}thCd}$

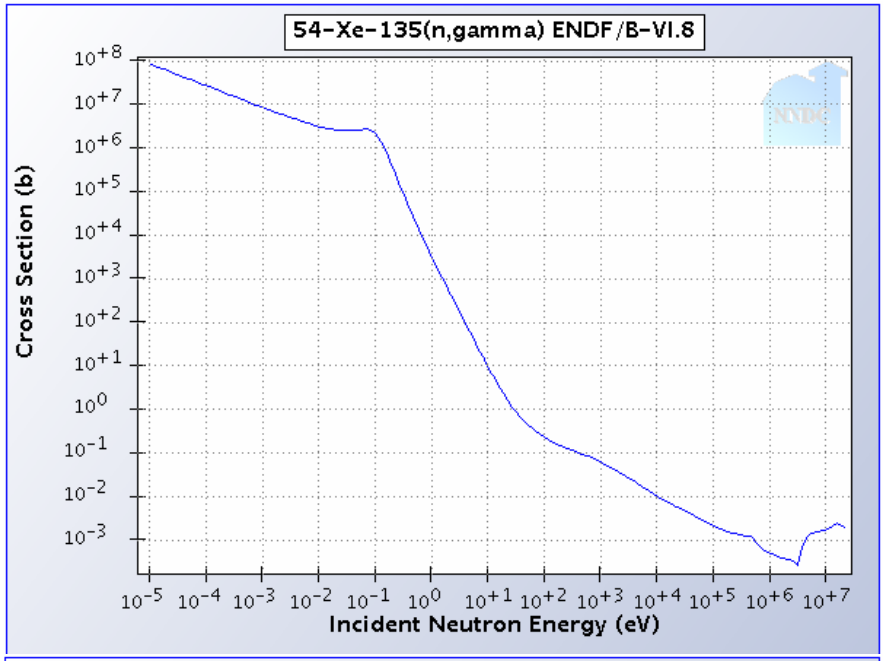
  $= 0.8(63 \text{ barns}) + 0.15(191 \text{ barns}) + 0.05(2450 \text{ barns})$

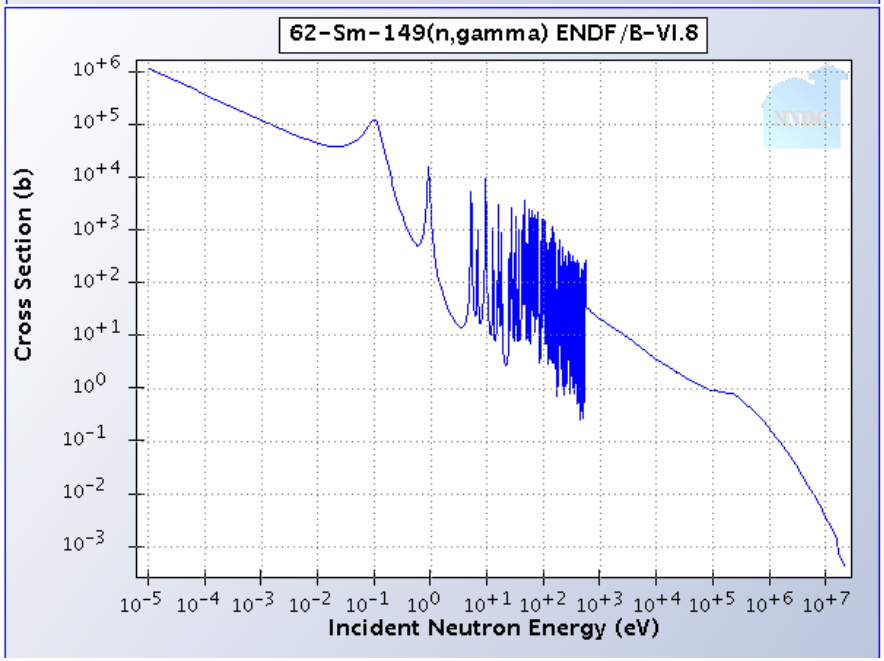
  $= 201.55 \text{ barns}$
- Alloy mixture has similar nuclear absorption in thermal and resonance regions, but still has relatively low melting point

Fission Product Neutron Poisons

Certain Fission Products are Neutron Absorbers

- Majority of fission products have low neutron capture cross sections
- Two major exceptions:
- $Xe^{135}\ \sigma_{c\text{-}th} = 2.7x10^{6}\ barns$
- $Sm^{149}\ \sigma_{c\text{-}th} = 5.85x10^{4}\ barns$
- 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:

$$\gamma_{Te} \cdot Fission\ Rate \rightarrow Te^{135} \rightarrow \beta^{-} + I^{135} \qquad t_{1/2} \sim 0.5\ min$$

$$I^{135} \rightarrow \beta^{-} + Xe^{135} \qquad t_{1/2} = 6.7\ hours$$

$$\gamma_{Xe} \cdot Fission\ Rate \rightarrow Xe^{135} \rightarrow \beta^{-} + Cs^{135} \qquad t_{1/2} = 9.2\ hours$$

$$Cs^{135} \rightarrow \beta^{-} + Ba^{135} \qquad t_{1/2} = 2.6x10^{6}\ years$$

- 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: $\varphi_{th}\Sigma_{f\text{-}th}$
- Direct production of I^{135} via fission: $\gamma_{Te}\,\varphi_{th}\,\Sigma_{f\text{-}th}$
- Elimination of I^{135} via neutron capture: <u>*negligible*</u>
- Elimination of I^{135} via decay: $-\lambda_I I(t)$ *($I(t)$ is I^{135} concentration)*
- Direct production of Xe^{135} via fission: $\gamma_{Xe}\,\varphi_{th}\,\Sigma_{f\text{-}th}$
- 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: $-\varphi_{th}\,\sigma_{c\text{-}th}\,Xe(t)$
- This yields following linear system of equations:

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

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

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

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

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

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

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

  $= \varphi_{th}\,\Sigma_{f\text{-}th}\,[\gamma_{Xe} + \gamma_{Te}] / [\varphi_{th}\,\sigma_{c\text{-}th} + \lambda_{Xe}]$

- If flux is constant, equilibrium Xe^{135} concentration reached
- When: $\lambda_{Xe} << \varphi_{th}\,\sigma_{c\text{-}th}$ - *or:* $\varphi_{th} >> \lambda_{Xe} / \sigma_{c\text{-}th}$
- This is true when: $\varphi_{th} >> (0.693/t_{1/2}) / \sigma_{c\text{-}th}$

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

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

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

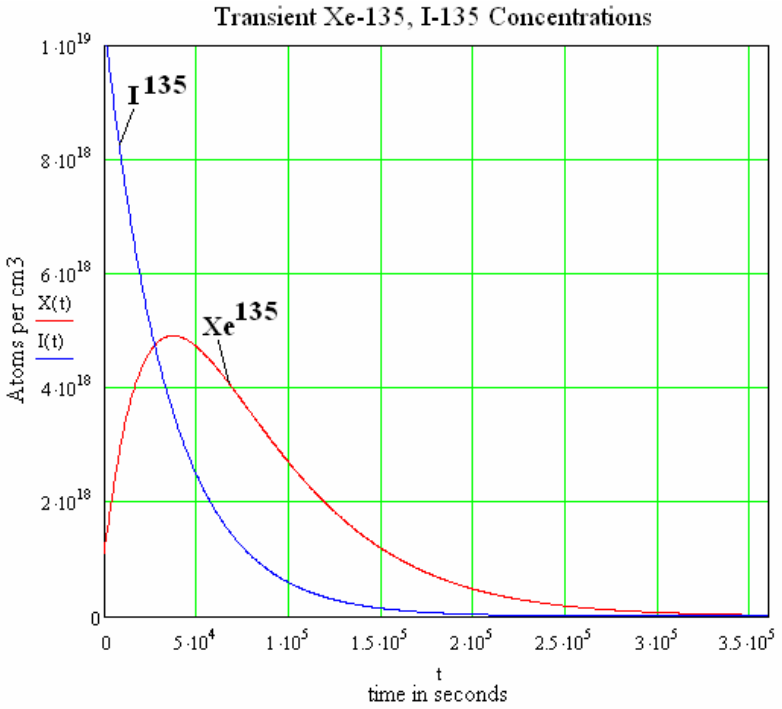
Xe^{135} Poisoning in U^{235}

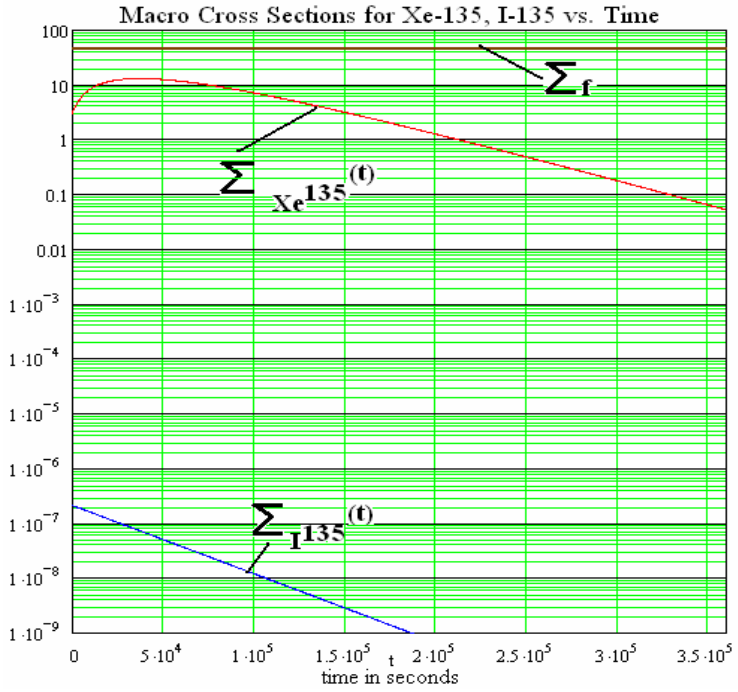
- Exact steady state buildup of Xe can be predicted from physics parameters *independent of neutron flux level*
- $Xe(\infty) = \Sigma_{f\text{-}th} [\gamma_{Xe} + \gamma_{Te}] / \sigma_{c\text{-}th}$
- Using numbers: $\Sigma_{f\text{-}th} = 48.7\ cm^{-1}$, $\gamma_{Xe} = 0.003$, $\gamma_{Te} = 0.061$,

  $\sigma_{c\text{-}th} = 2.7x10^{6}\ barns\ x\ 10^{-24} cm^2/barn = 2.6\ x10^{-18} cm^2$
- Then: $Xe(\infty) = 1.1\ x\ 10^{18}\ atoms/cm^3$
- What happens if after large flux level achieved, it suddenly disappears?

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

- Thermal flux: $\varphi_{th} \approx 10^{14}/cm^2\ sec$
- $I(\infty) = 1x10^{19}\ atoms/cm^3$
  $Xe(\infty) = 1.1x10^{18}\ atoms/cm^3$
- 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 $4.4x10^{18}\ atoms/cm^3$ at ~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:

$$\gamma_{Nd}\, x\, Fission \rightarrow Nd^{149} \rightarrow \beta^{-} + Pm^{149} \qquad t_{1/2} = 2.0\ hours$$

$$Pm^{149} \rightarrow \beta^{-} + Sm^{149} \qquad t_{1/2} = 54\ hours$$

- Sm^{149} with $\sigma_{c\text{-}th} = 5.85x10^{4}\ barns$, can only be removed by *burning it up* with thermal neutrons
- System of equations governing Sm^{149} build-up/decay is:

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

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

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

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

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

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

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

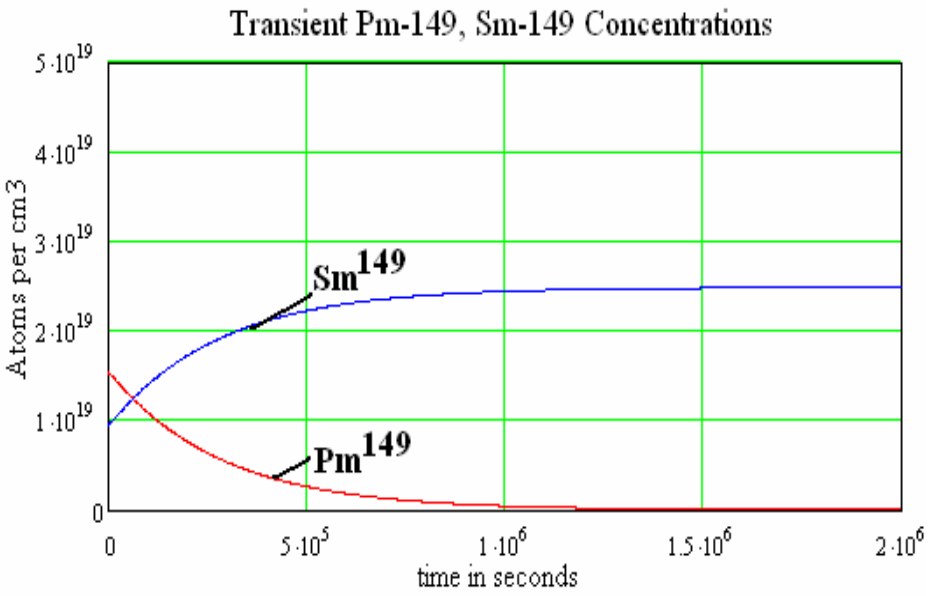
- Thus: $Sm(\infty) = \lambda_{Pm} Pm(\infty) / \varphi_{th}\,\sigma_{c\text{-}th}$
- Substituting in for $P(\infty)$ yields:

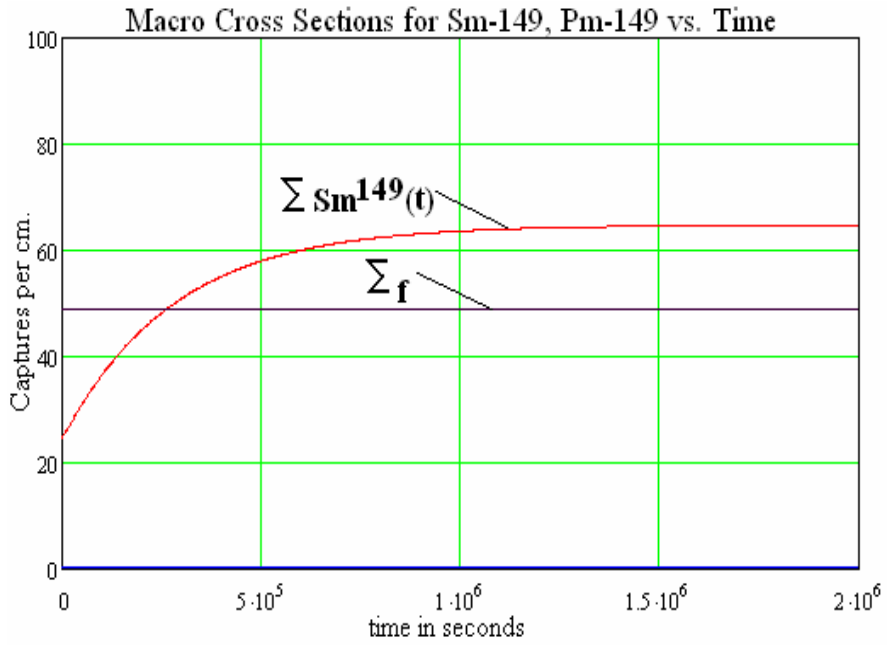
$$Sm(\infty) = \gamma_{Nd}\,\Sigma_{f\text{-}th} / \sigma_{c\text{-}th}$$

- Using: $\gamma_{Nd} = 0.011$, $\Sigma_{f\text{-}th} = 48.7 cm^{-1}$,
  $\sigma_{c\text{-}th} = 5.85 x10^{4} barns \cdot 10^{-24} cm^2/barn = 5.85\ x10^{-20} cm^2$
- $Sm(\infty) = 9.4 \cdot 10^{18}\ atoms/cm^3$

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

- Thermal flux: $\varphi_{th} = 10^{14}/cm^2\ sec$
- $Pm(\infty) = 5.5x10^{19}\ atoms/cm^3$
  $Sm(\infty) = 9.4x10^{18}\ atoms/cm^3$
- 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 $\sim 2.48x10^{19}\ atoms/cm^3$
- Sm^{149} ***capture exceeds fission at ~ 75hours***





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 $\Sigma_{f\text{-}th}$
- Fuel design compensates by adjusting Uranium enrichment to increase $\Sigma_{f\text{-}th}$ 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}-Be^{9}
  - Am^{241}-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}