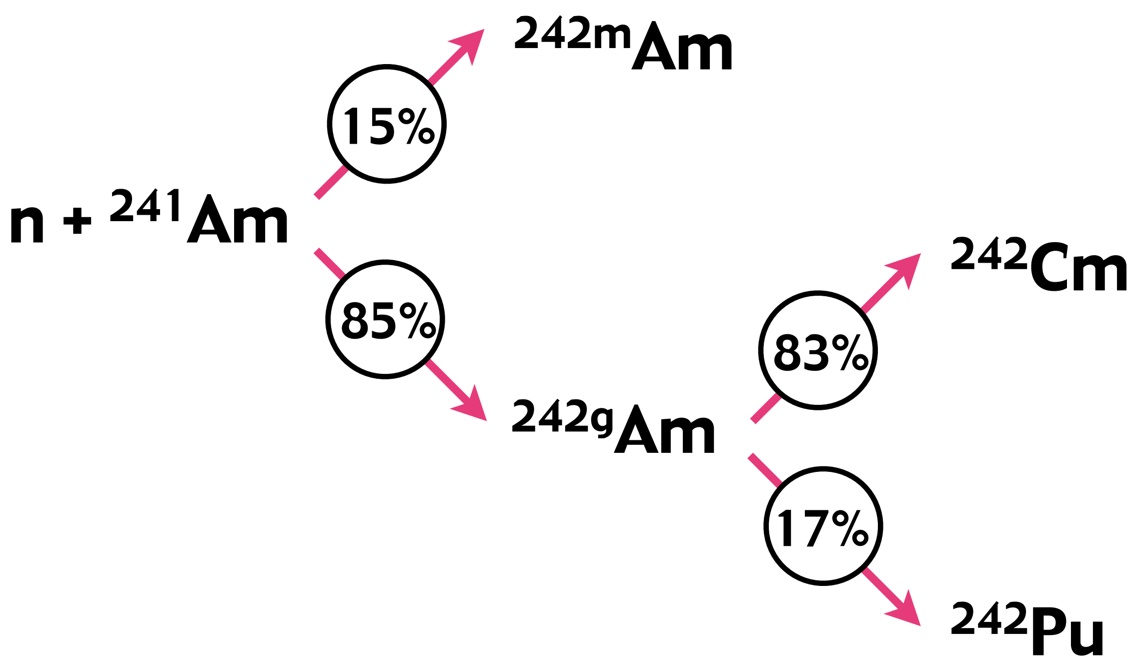
Minor actinide burning



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Intended learning outcomes

Fast spectrum Generation IV-reactors are capable of burning of minor actinides, thereby

Reduced time required to isolate residual waste to less than 1000 years

Reduce volume of geologic repository by factor 3-6

After this lecture you will be able to:

- **Evaluate the benefits of minor actinide burning on repository performance**
- **Assess the detrimental impact of minor actinides on safety parameters**
- **Design a fast reactor to accommodate minor actinides in the fuel**

Minor actinide production in LWRs

Np and Am are produced in LWRs as follows:

$$n + {}^{235}U \rightarrow {}^{236}U + \gamma$$

$$n + {}^{236}U \rightarrow {}^{237}U^{*} \rightarrow {}^{237}Np + \beta$$

$${}^{241}Pu \rightarrow {}^{241}Am + \beta$$

$$n + {}^{242}Pu \rightarrow {}^{243}Pu \rightarrow {}^{243}Am + \beta$$

Production rate of minor actinides

Elemets	kg/TWh$_e$	kg/TWh$_{th}$
TRU	32.3	11.0
Pu	28.8	9.8
MA	3.5	1.2
Am+Cm	1.8	0.6

Composition of spent PWR fuel after 50 GWd/ton burn-up and four years of cooling.

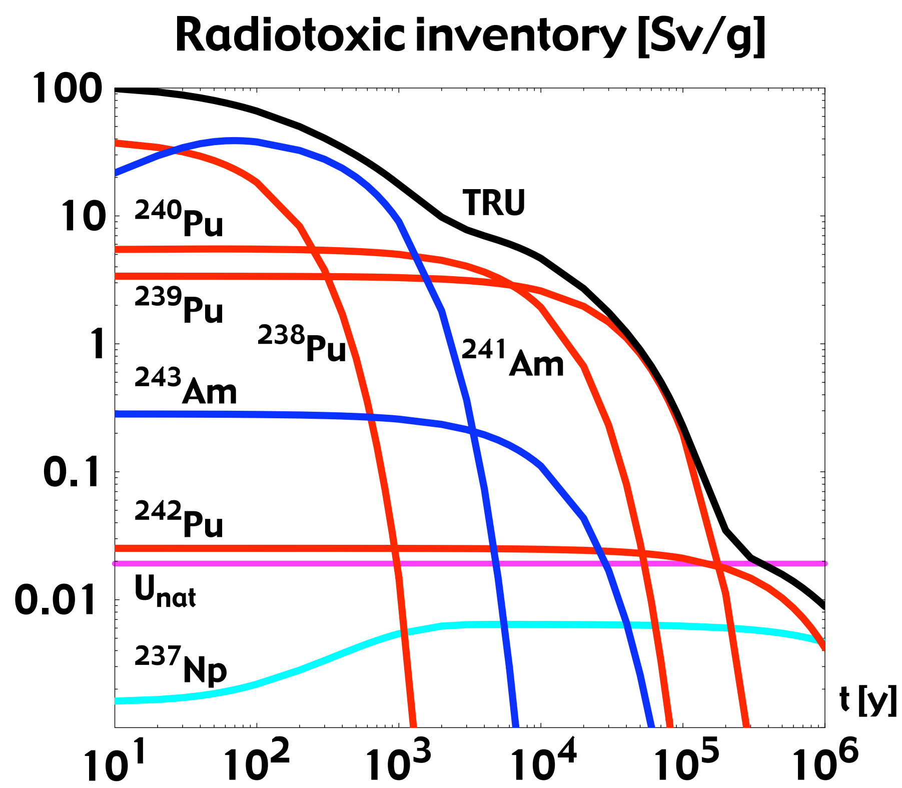
Nuclide	Fraction
Fission products	5,145 %
^{235}U	0.767%
^{236}U	0.552%
^{238}U	92.186%
^{237}Np	0.072%
^{238}Pu	0.042%
^{239}Pu	0.623%
^{240}Pu	0.286%
^{241}Pu	0.155%
^{242}Pu	0.095%
^{241}Am	0.038%
^{243}Am	0.028%
^{244}Cm	0.010%
^{245}Cm	0.001%

Radiotoxic inventory

Dose coefficients for ingestion

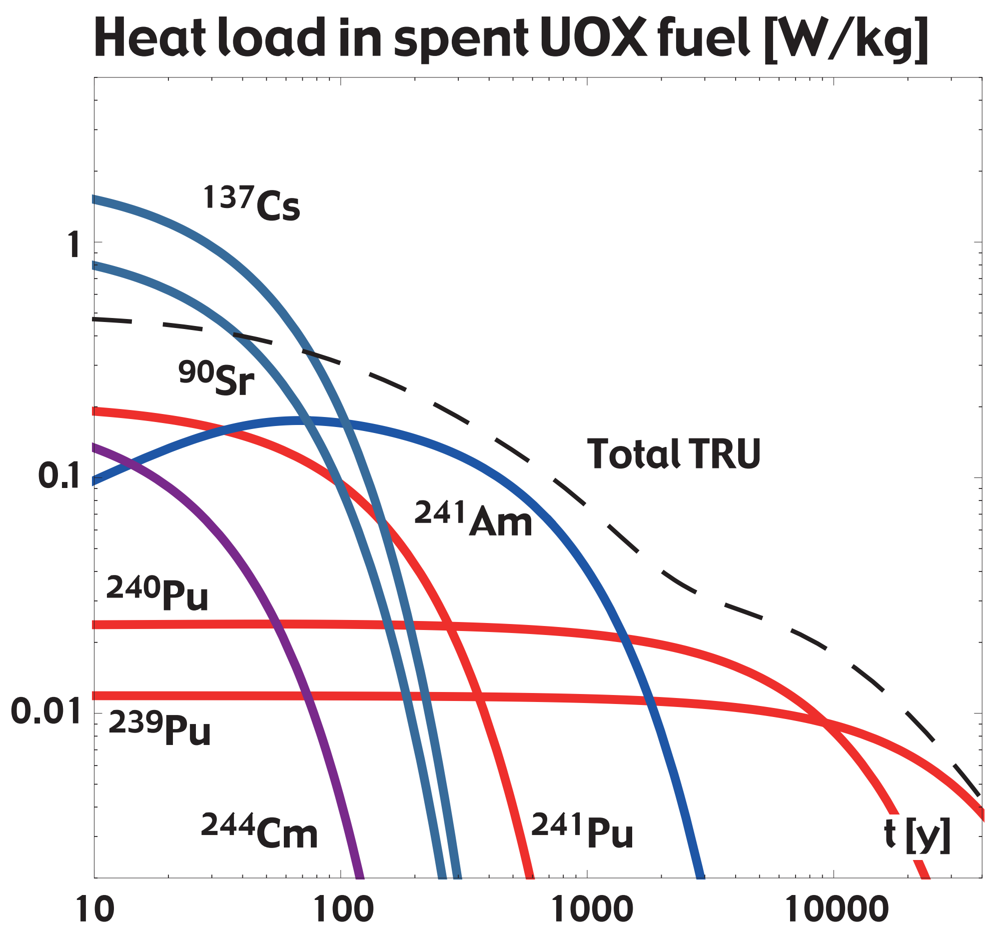
Nuclide	ε_{50} [nSV/Bq]
^{235}U	47
^{236}U	47
^{238}U	44
^{237}Np	110
^{238}Pu	230
^{239}Pu	250
^{240}Pu	250
^{241}Pu	5
^{242}Pu	240
^{241}Am	200
^{243}Am	200
^{244}Cm	120
^{245}Cm	210

Contribution of transuranium elements to radio-toxic inventory of spent PWR fuel after 50 GWd/ton burn-up and four years of cooling.



Time until decay of TRU reduces specific radio-toxicity to below that of uranium in nature ≈ 200 000 years

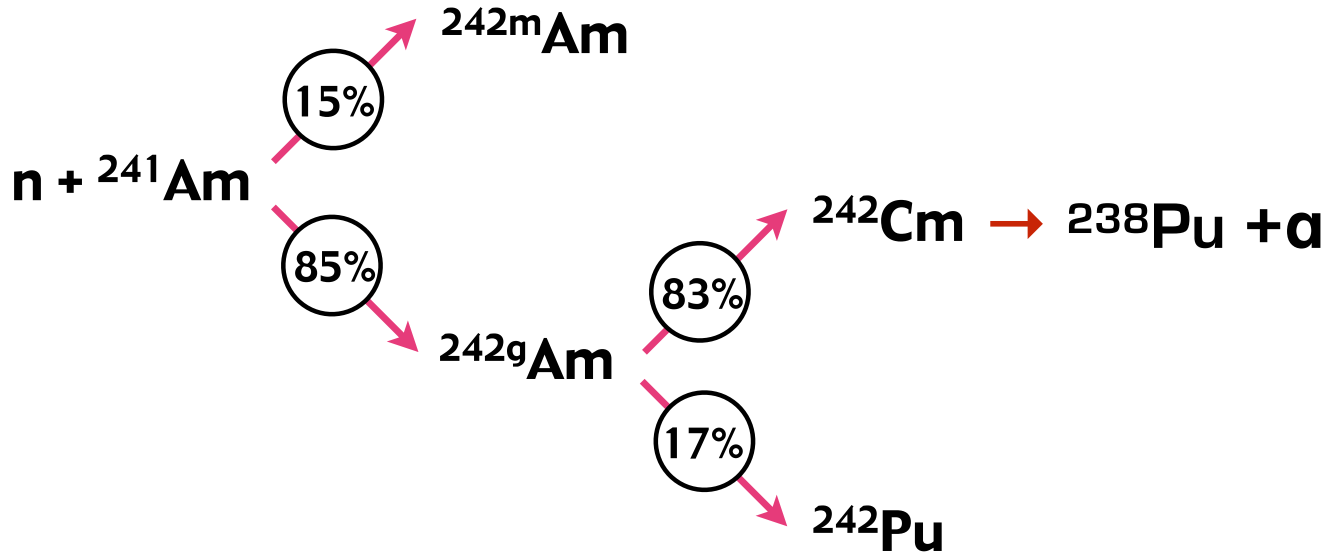
Heat production in repository



- Forced cooling of repository is conducted until decay of fission products permits closure.
- Heat from decay of ^{241}Am is dimensioning for the distance between spent fuel canisters
- Removal of ^{241}Pu and ^{241}Am from repository can reduce its specific volume by factor of 3-6

Transmutation of americium

- In Generation IV reactors with breeding ratio ≈ 1.0, the inventory of Pu is roughly constant.
- Americium is added to the fuel (or a burning blanket) for transmutation, mainly by neutron capture.
- Main transmutation products: ^{238}Pu, ^{242}Pu, ^{242m}Am, ^{244}Cm

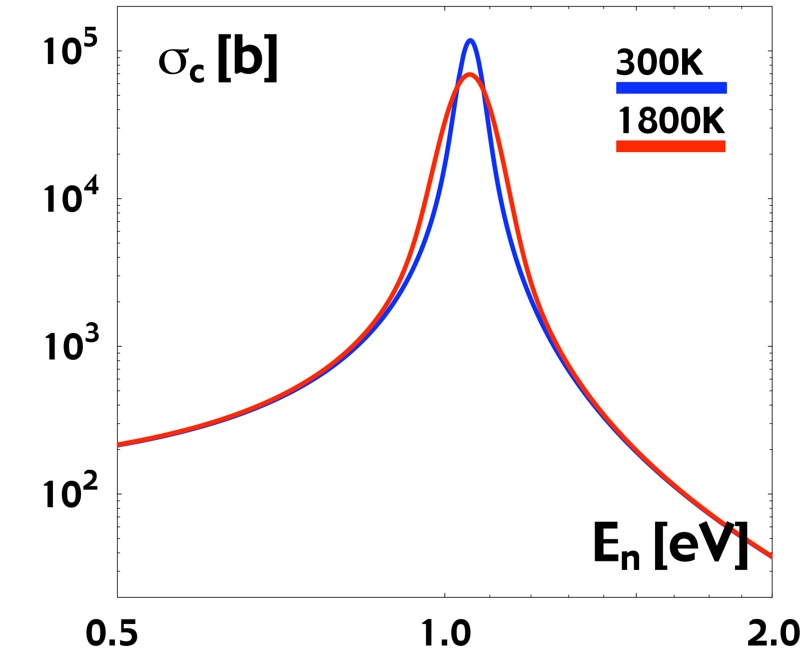


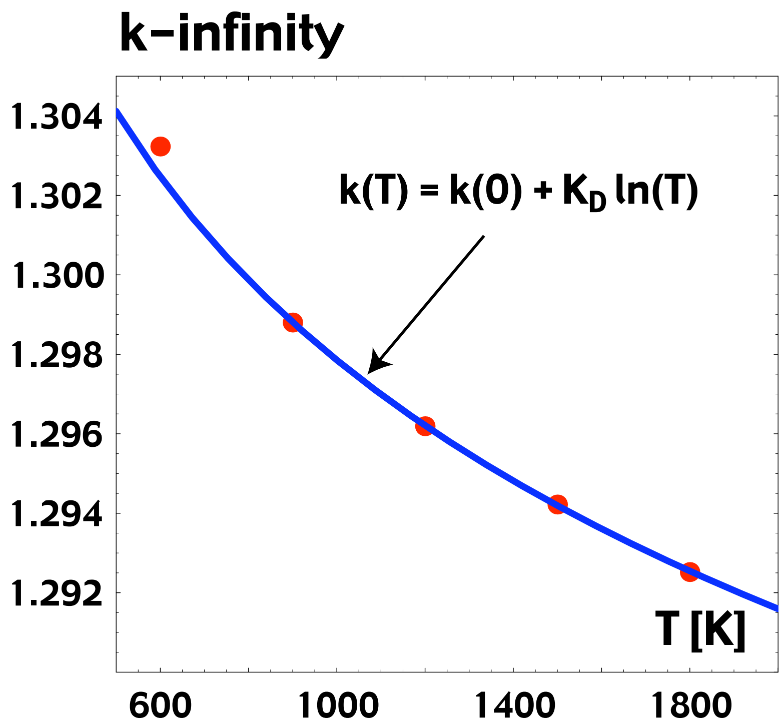
$$n + {}^{243}\mathrm{Am} \rightarrow {}^{244}\mathrm{Am} \rightarrow {}^{244}\mathrm{Cm} + \beta$$

Impact of americium on safety of fast reactors

- Introduction of americium will lead to
- Reduction of Doppler feedback
- Increase in coolant temperature coefficient
- Reduced delayed neutron fraction
- Degraded performance under transients

Doppler feedback





- When the fuel temperature increases, all nuclides vibrate with larger amplitude around their average crystal lattice positions.
- In the lab system, the cross section for resonance absorption is reduced at the peak and increases at the tails. Area under resonance peak is conserved.
- Neutrons under moderation first experience an increase in absorption cross section when approaching the higher energy tail. Fewer neutrons reach the energy of the resonance peak.
- Net effect: increase in spectrum averaged cross section for capture and reduction in reactivity.
- In fast reactors, the reactivity decreases logarithmically with temperature.

Doppler feedback (2)

The Doppler coefficient α_D is the change of reactivity with temperature due to broadening of absorption cross section.

$$\alpha_D \equiv \frac{d\rho}{dT} = \frac{1}{k^2}\frac{dk}{dT}$$

In a fast spectrum, reactivity decreases logarithmically with temperature.

$$\rho(T) = \rho(0) + K_D \ln(T)$$

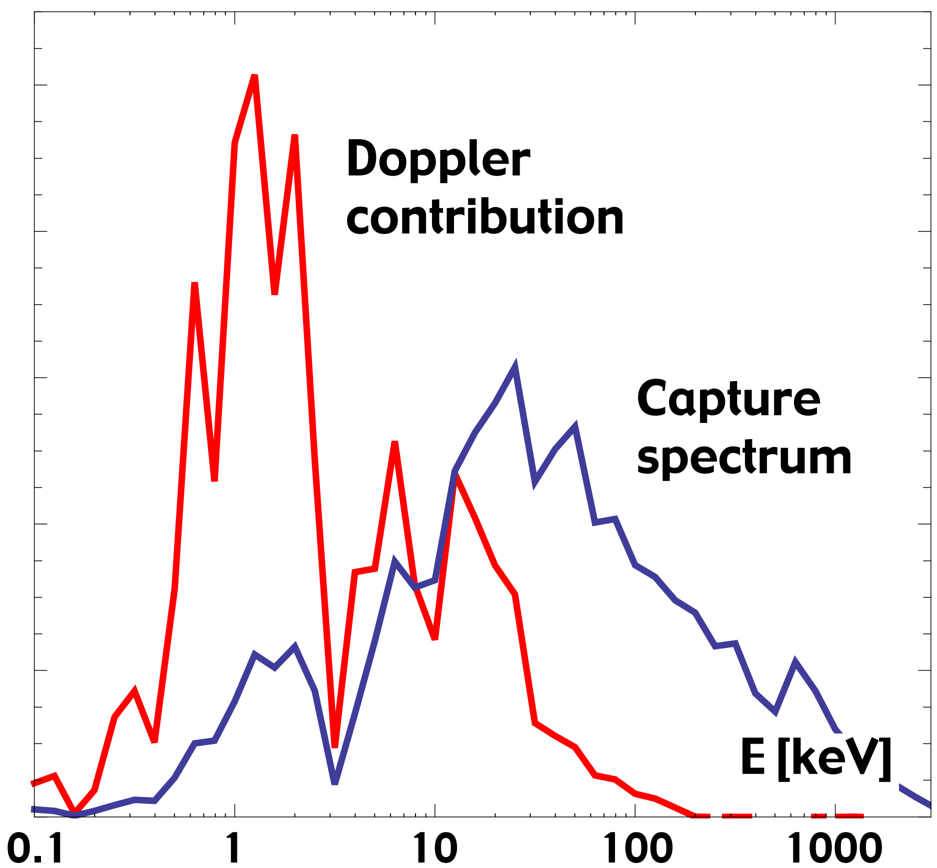
The constant of proportionality K_D is called ”The Doppler constant”

$$K_D = T\frac{d\rho}{dT}$$

The Doppler coefficient is obtained by dividing the Doppler constant with T

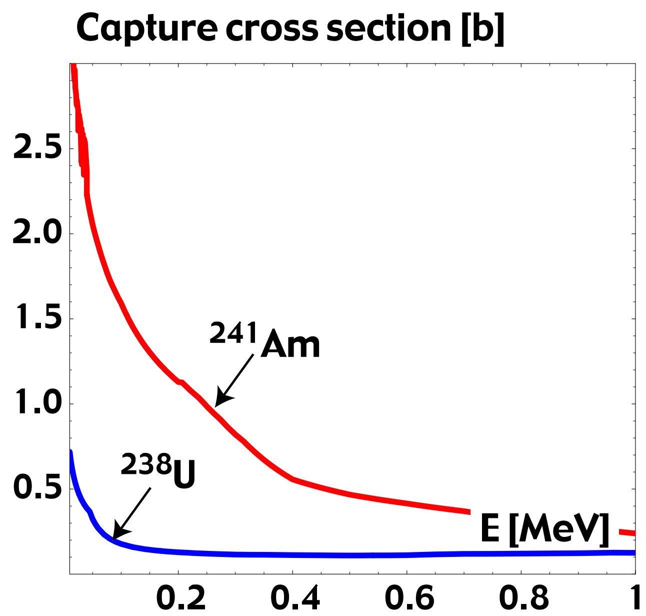
$$\alpha_D = \frac{K_D}{T}$$

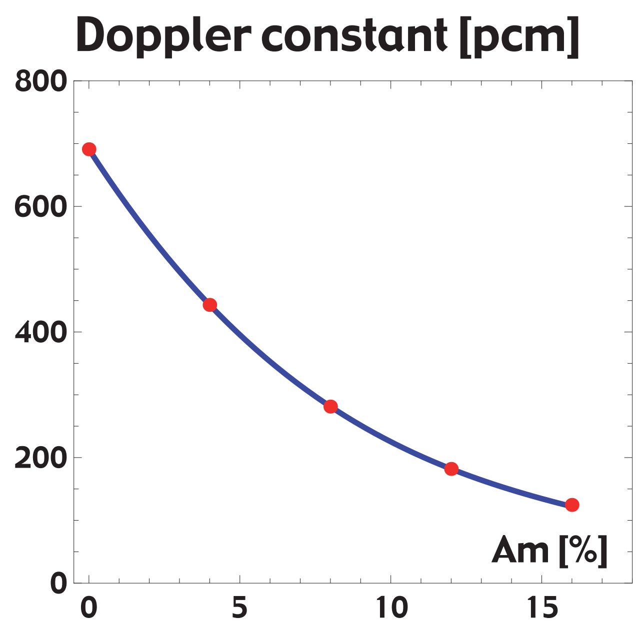
Doppler feedback (3)



- Doppler feedback mainly derives from neutron captures occurring below 100 keV
- The lower the energy of the resonance where capture occurs, the more efficient is the Doppler feedback
- In an SFR with standard MOX fuel, 65% of the Doppler feedback derives from neutron captures below 3 keV, constituting only 15% of all captures!
- Typical order of magnitude: K_D = −500 pcm
- Spectrum dependence is significant!

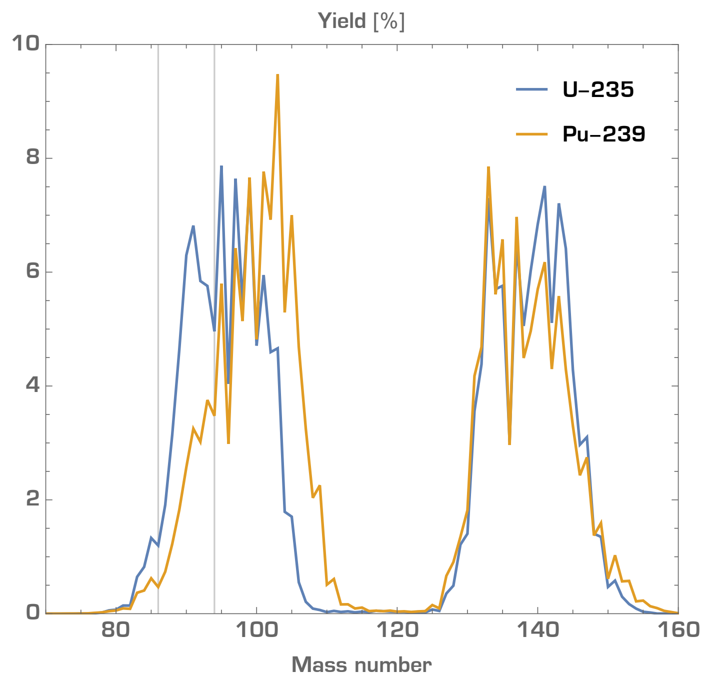
Doppler feedback (4)





- Cross section for neutron capture in ^{241}Am order of magnitude higher than for ^{238}U
- Even with modest concentration of Am in the fuel, fewer neutrons are slowed down to energy region (E_n < 30 keV) where Doppler broadening is of relevance.
- Doppler constant is reduced by factor three when Am concentration is ≈ 10% (even if ^{238}U concentration is > 70%!)

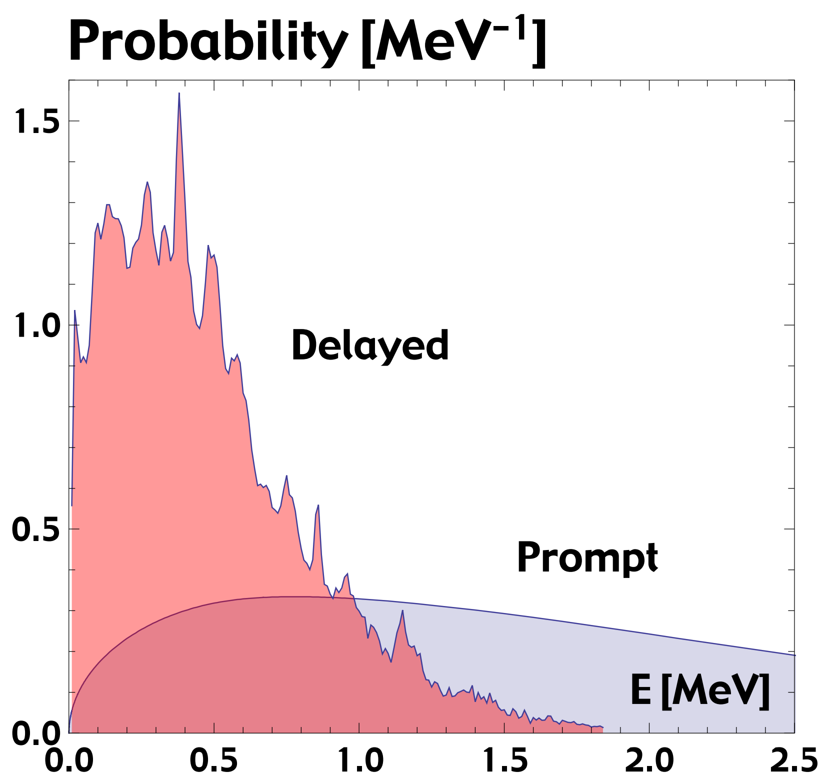
Delayed neutrons



Nuclide	ν_{tot}	ν_d / ν_{tot}
^{238}U	2.53	1.89%
^{239}Pu	3.02	0.22%
^{241}Am	3.37	0.13%
^{244}Cm	3.42	0.13%

- Delayed neutrons are emitted by unstable fission products having a half-life of up to 55 s.
- Main contributors: Kr, Br, I, Rb, Cs
- Increasing mass of mother nuclide shifts mass of the lighter fission product (Why?)
- Yield of Kr and Br is reduced.
- Delayed neutron yield in fission of Am and Cm is extremely low!

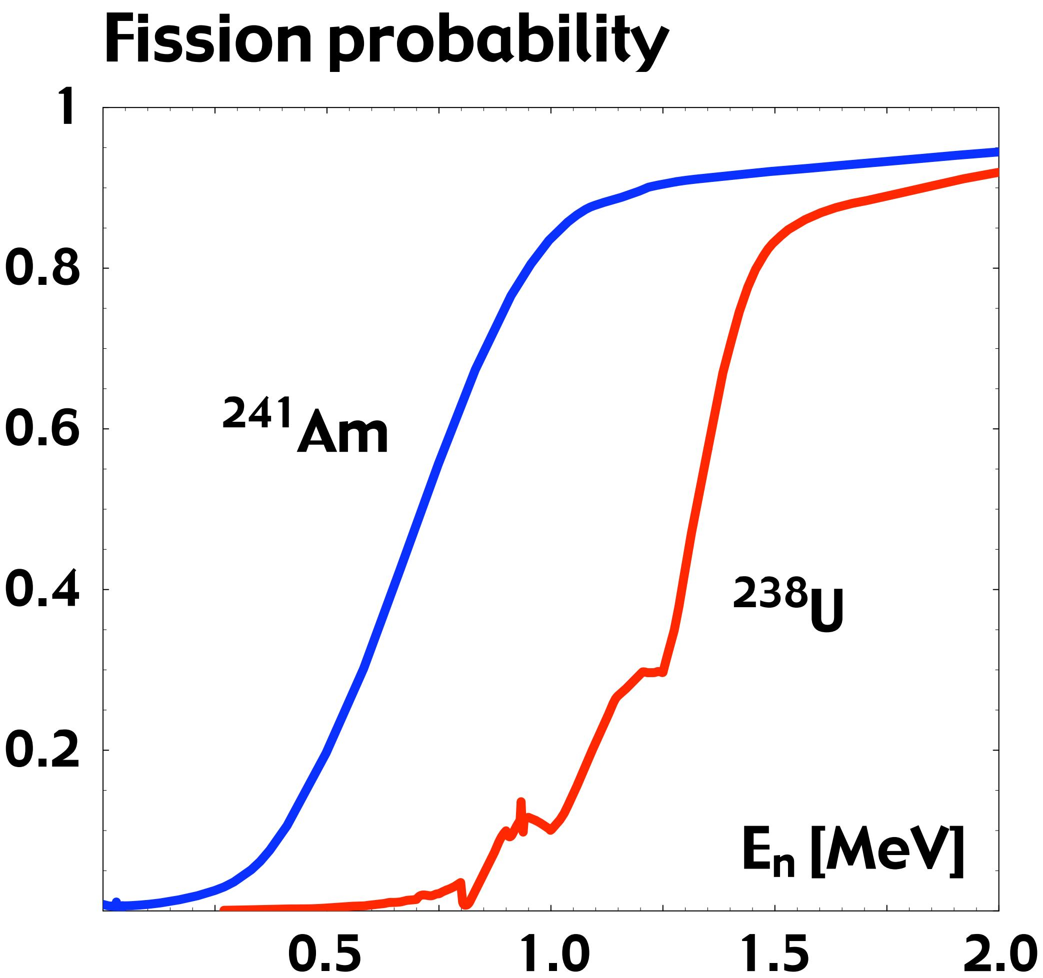
Effective delayed neutron fraction



Fuel	β_{eff}/β
$(U_{0.8},Pu_{0.2})O_2$	390/460
$(U_{0.7},Pu_{0.2},Am_{0.1})O_2$	330/430
$(U_{0.6},Pu_{0.2},Am_{0.2})O_2$	270/390
$(U_{0.5},Pu_{0.2},Am_{0.3})O_2$	220/350

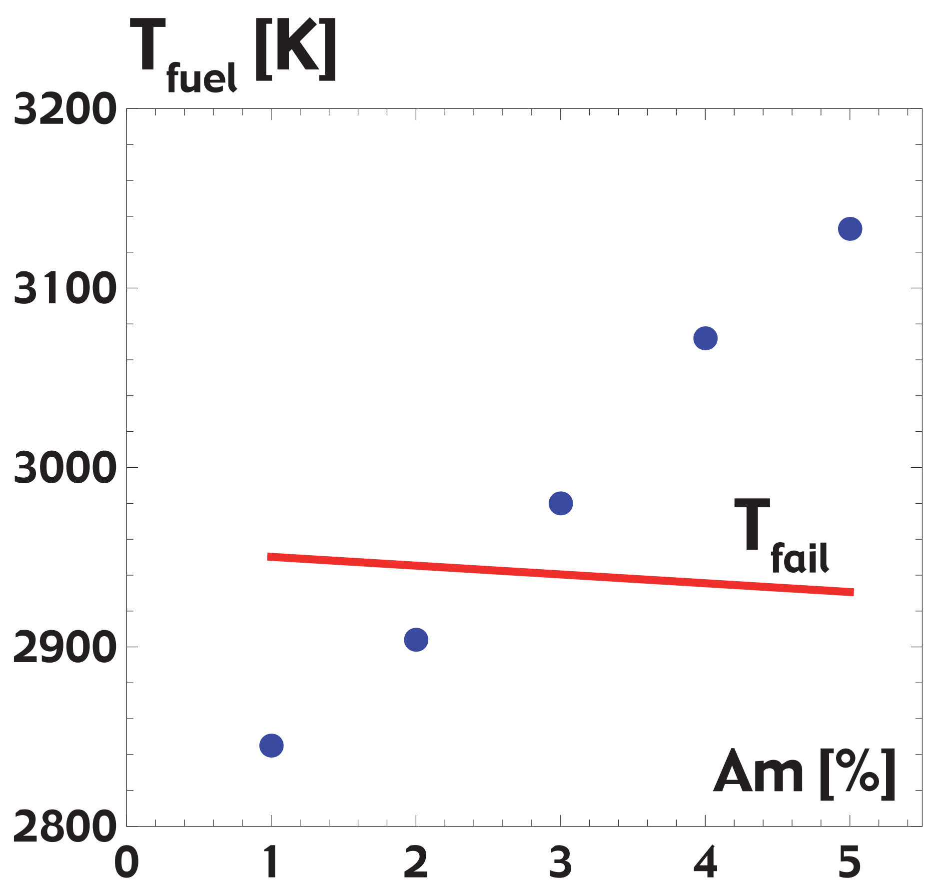
- Effective delayed neutron fraction β_{eff} is the fraction of fissions induced by delayed neutrons.
- Delayed neutron spectrum is softer than prompt neutron spectrum
- Am captures delayed neutrons with a higher probability than prompt neutrons.
- β_{eff} is smaller than β in the presence of Am.
- β_{eff} drops faster than β as function of Am concentration.

Coolant temperature feedback



- When the coolant heats up, the neutron spectrum hardens
- Capture cross section of ^{241}Am drops faster than of ^{238}U (see slide 11)
- Threshold for increase in fission probability is situated at lower neutron energy
- Spectrum component of coolant temperature coefficient is more positive, and increases with concentration of Am

Impact on transient behaviour



Peak fuel temperature during UTOP

- The combined deterioration of Doppler feedback, effective delayed neutron fraction and coolant temperature coefficient leads to poorer performance during transients.
- UTOP = Unprotected Transient Over-Power (e.g. due to control rod withdrawal without SCRAM)
- If a core with 1% Am in the fuel is designed to respect fuel melting with 100 K margin during a UTOP, it will fail if Am concentration is raised to 3%.

Home assignment 2

- Use Serpent to calculate the Doppler constant and coolant temperature coefficient in a fast reactor with rod diameter 10.0 mm, fuel cladding thickness of 0.5 mm a d fuel column height 1.0 m. Do the calculation for Am concentrations of 2, 5 and 8%, and a constant Pu fraction of 15%. That means: **U/Pu/Am = 1 - 0.15 - C(Am) / 0.15 / C(Am)**.
- The Doppler constant can be calculated as: $K_D = \dfrac{\rho(T_1) - \rho(T_2)}{\ln(T_2/T_1)}$, changing cross section library only.
- The coolant temperature coefficient is calculated by changing its density in the active core region only.
- Compare the ratio of fuel Doppler coefficient to the coolant temperature feedback. Discuss how the safety parameters depend on Am concentration.

Group No	Sodium	Lead	Helium
Oxide	1	5	9
Nitride	2	6	10
Carbide	3	7	11
Metal alloy	4	8	