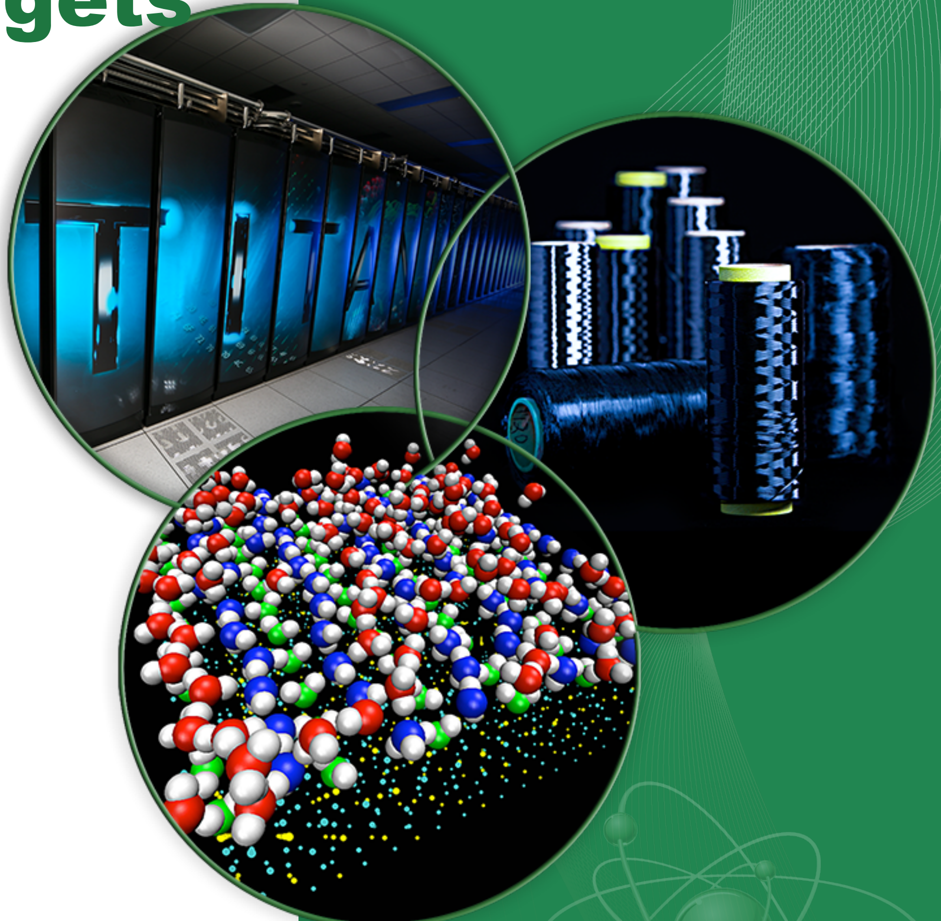
Nuclear Data Needs for ^{238}Pu Production Targets in HFIR



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Nuclear Data Needs and Capabilities for Applications
Lawrence Berkley National Laboratory
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Background

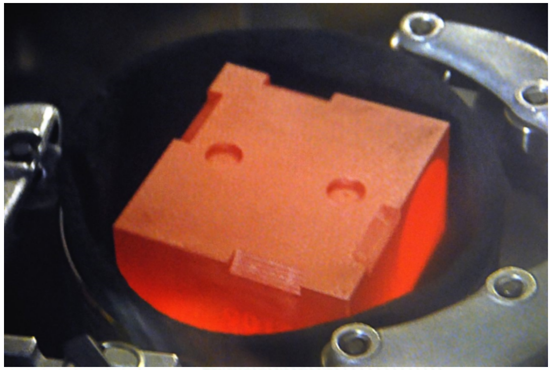
- Historically U.S. used ^{238}Pu in RTG[(1)] and RHU[(2)]
- U.S. ^{238}Pu production ended 1988 with the shutdown of SRS[(3)] reactors
- Since 1993, 16.5 kg ^{238}Pu purchased from Russia (ended 2009)
- Domestic supply necessary (DOE request in 2009)
- DOE goal is to produce 1.5 to 2 kilograms of ^{238}Pu/year within the DOE complex (at HFIR and ATR) by 2018

Curiosity's RTG is fueled by 4.8 kg (11 lb) of ^{238}Pu dioxide supplied by the U.S. DOE

125 watts of electrical power from about 2,000 watts of thermal power at the start of the mission

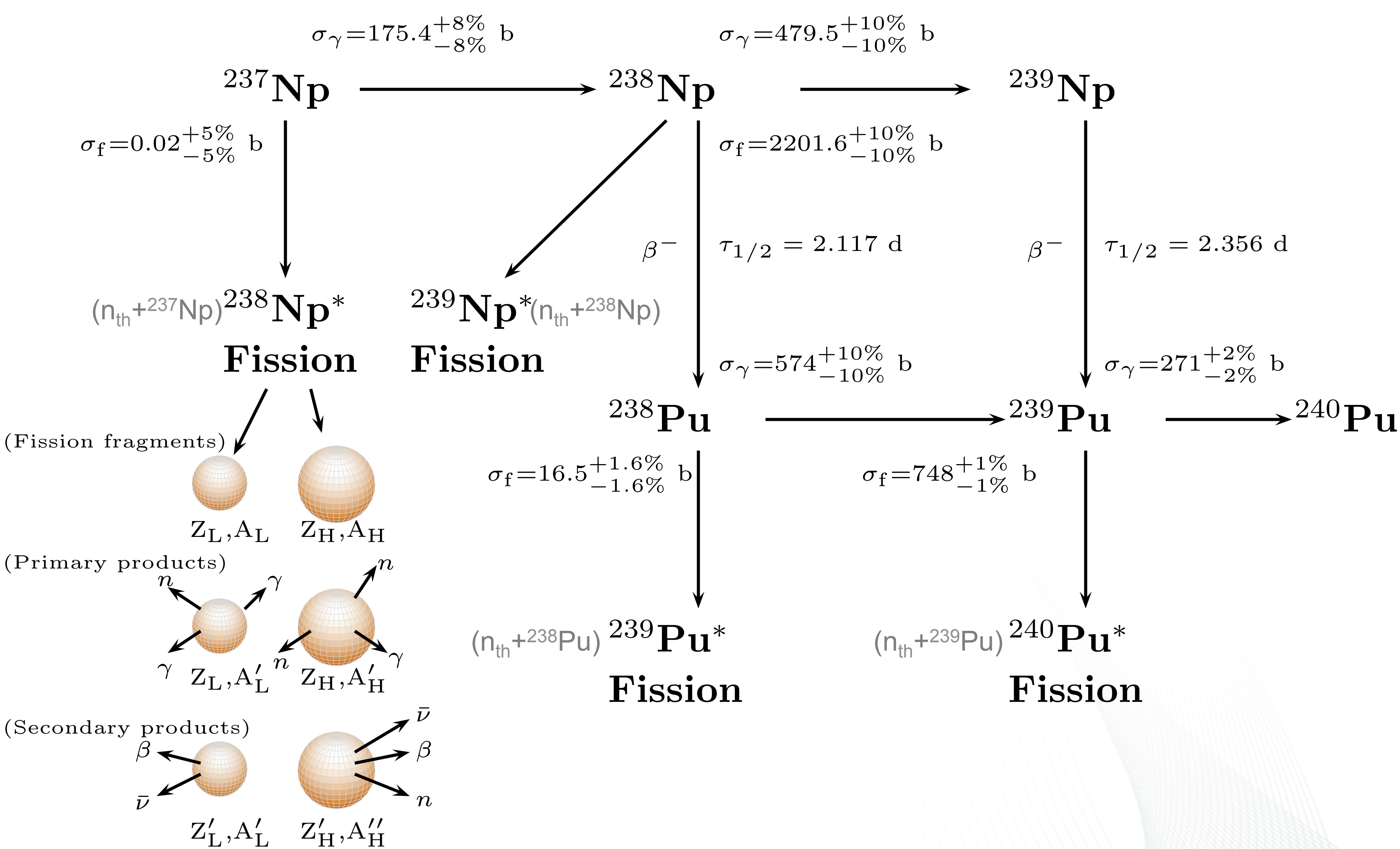
^{238}Pu oxide pellet

Radioisotope within a graphite shell goes into the generator

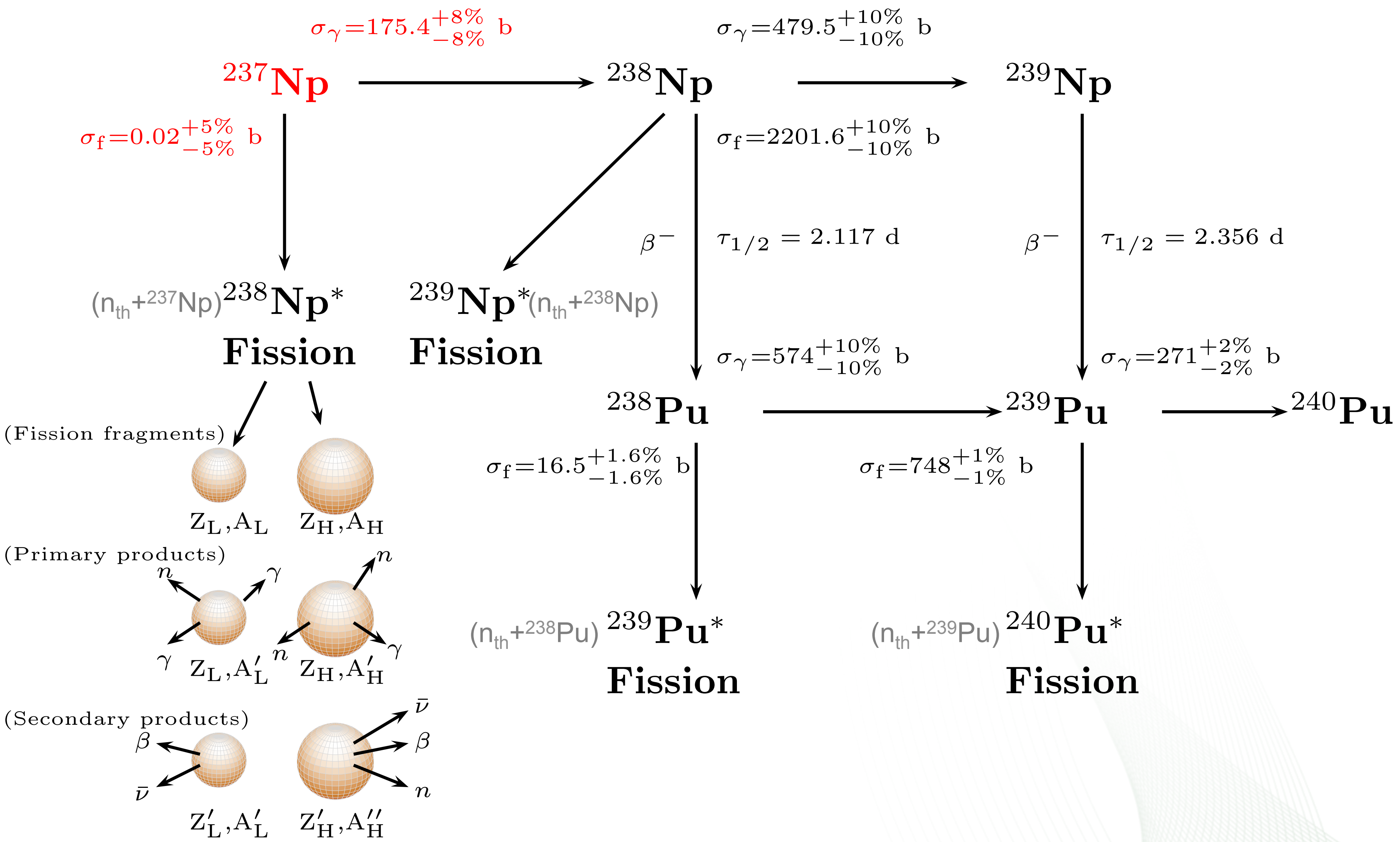


(1) Radioisotope Thermoelectric Generators to generate electrical power
(2) Radioisotope Heater Units to produce heat for electronics and environmental control for deep space missions
(3) In the late '50s Savannah River Site began generating, collecting ^{237}Np targets that were then irradiated with neutrons to produce ^{238}Pu (Fuller et al. WM2013 Conference, Phoenix, Arizona)

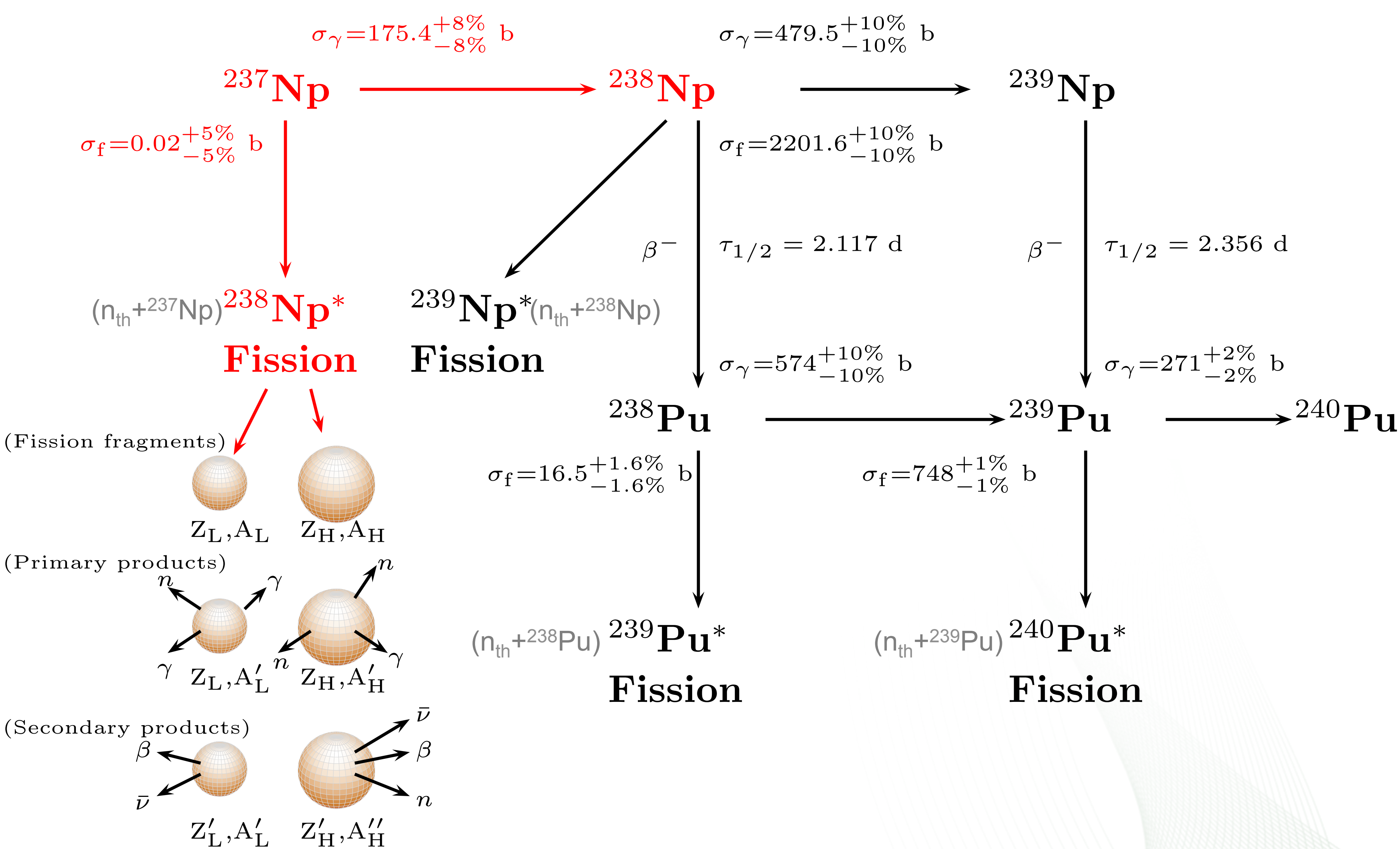
^{238}Pu Production Diagram



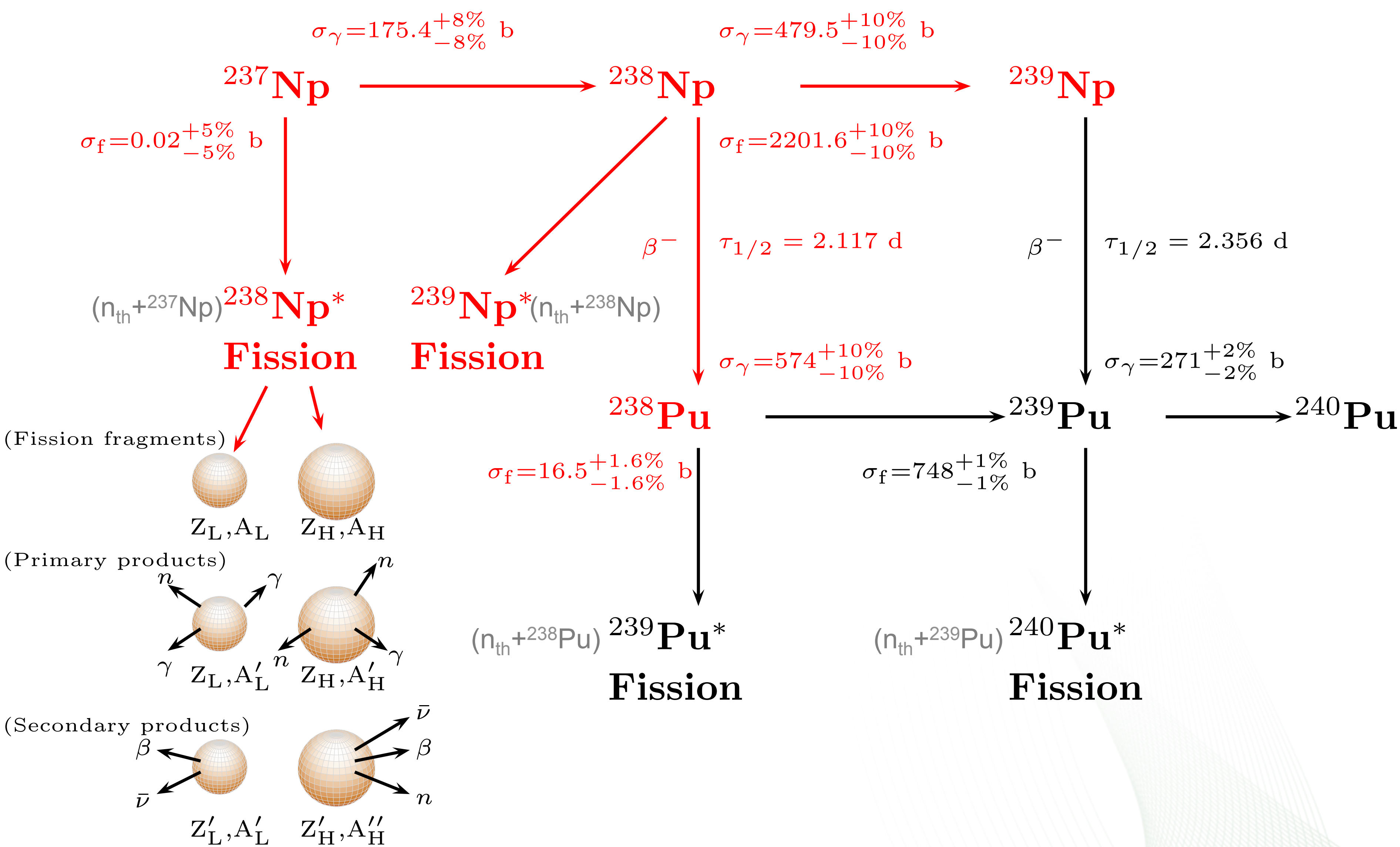
^{238}Pu Production Diagram



^{238}Pu Production Diagram



^{238}Pu Production Diagram



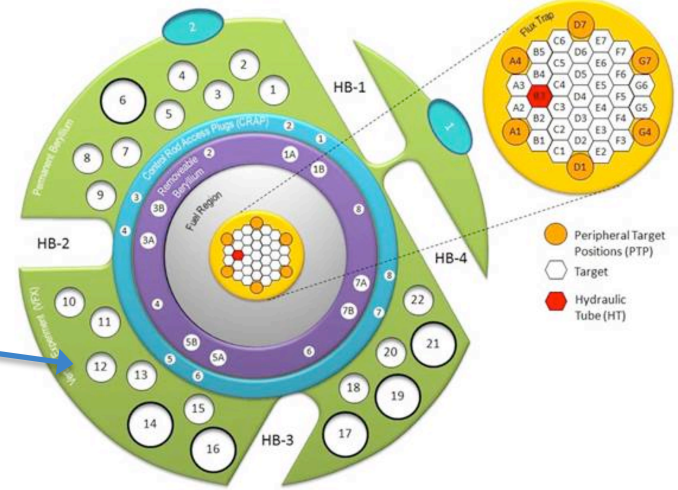
^{238}Pu Production in HFIR

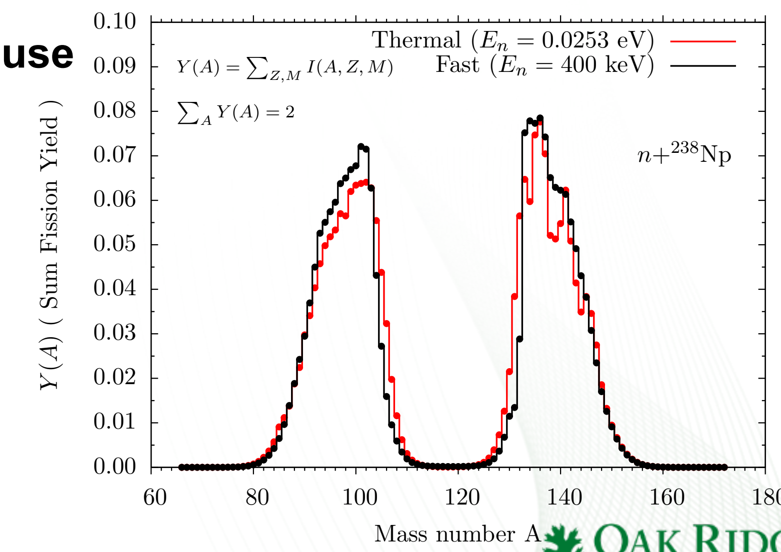
- $^{237}NpO_2$–Al targets irradiated in HFIR reflector VXF locations
- Analyses required for both production (cross section) and safety (fission product)
- Thermal limits(*) primarily from the fission of ^{238}Np and ^{239}Pu isotopes (current target loading is limited to 20% of $^{237}NpO_2$)
- Radiochemical measurements of fission products in irradiated 1-cycle targets found large discrepancies between predictions and measurements (> 30%) in fission products suggesting possible overestimate of fission rates
- However, validation of calculated fission rates used in the safety analysis is difficult due to uncertainties in nuclear data

(*) Heat generation also due to gammas from capture, prompt- and delayed-gammas, beta and alpha decay.

Nuclear Data – Fission Product Yields

- **Large discrepancies between predicted and measured individual FPs is likely due to poor fission yield data for ^{238}Np used in calculations**
  - **ENDF/B-VII.1 does not contain fission product yields for ^{238}Np thermal fission – only fast fission (not appropriate for thermal VXF sites)**
  - **Use of ^{238}Np fast fission yields for targets can result in 10 to 30% errors**
  - **Yields have very large uncertainties (10-40%) for measured fission products, e.g., ^{137}Cs (±30%), ^{148}Nd (±28%), ^{144}Ce (±55%)**
  - **These uncertainties make it very difficult to use individual FP measurements to validate ^{238}Np fission rates**





Comparison of ^{238}Np thermal and fast fission sum yields (data from JEFF 3.1)

Nuclear Data – Thermal Cross Sections

- There are also large uncertainties in the cross section data for ^{238}Pu production from ^{238}Np (+/-10%)

Data Library	^{237}Np		^{238}Np		^{238}Pu	
	(n,f)	(n,γ)	(n,f)	(n, γ)	(n,f)	(n, γ)
ENDF/B-VII.1	0.0203±5(%)	175.4±8(%)	2201.6±10(%)	479.5±10(%)	17.7±1.6(%)	412.8±10(%)
ENDF/B-VII.0	0.0203	161.7	2071.0	450.3	17.0	560.9
JEFF-3.0/A	0.0180	181.1	2028.8	203.0	17.2	546.62

- Changes from ENDF/B-VII.0 → -VII.1
  - 237Np(n_{th},γ) +8%
  - 238Pu(n_{th},γ) -26%
  - 238Np(n_{th},γ) +6%
- ^{238}Np(n_{th},f)
  - Spencer et al. (1969) σ = 2070 +/- 30 b
  - Abramovich et al.(1995) σ = 2110 +/- 74 b
  - Danon et al. (1996) σ = 2641 +/- 58 b
  - Furutaka et al. (2008) σ = 2201 +/- 34 b
- Better overall agreement between calculated and measured actinides (^{238}Pu and ^{239}Pu) observed when using ENDF/B-VII.1 cross sections

Decay data and cumulative FPY

Black dots : ratio of cumulative FPYs obtained by independent FPY and decay data in ENDF/B-VII.1 to cumulative FPYs in ENDF/B-VII.1.

Although deviations are small, ratios should be one!!

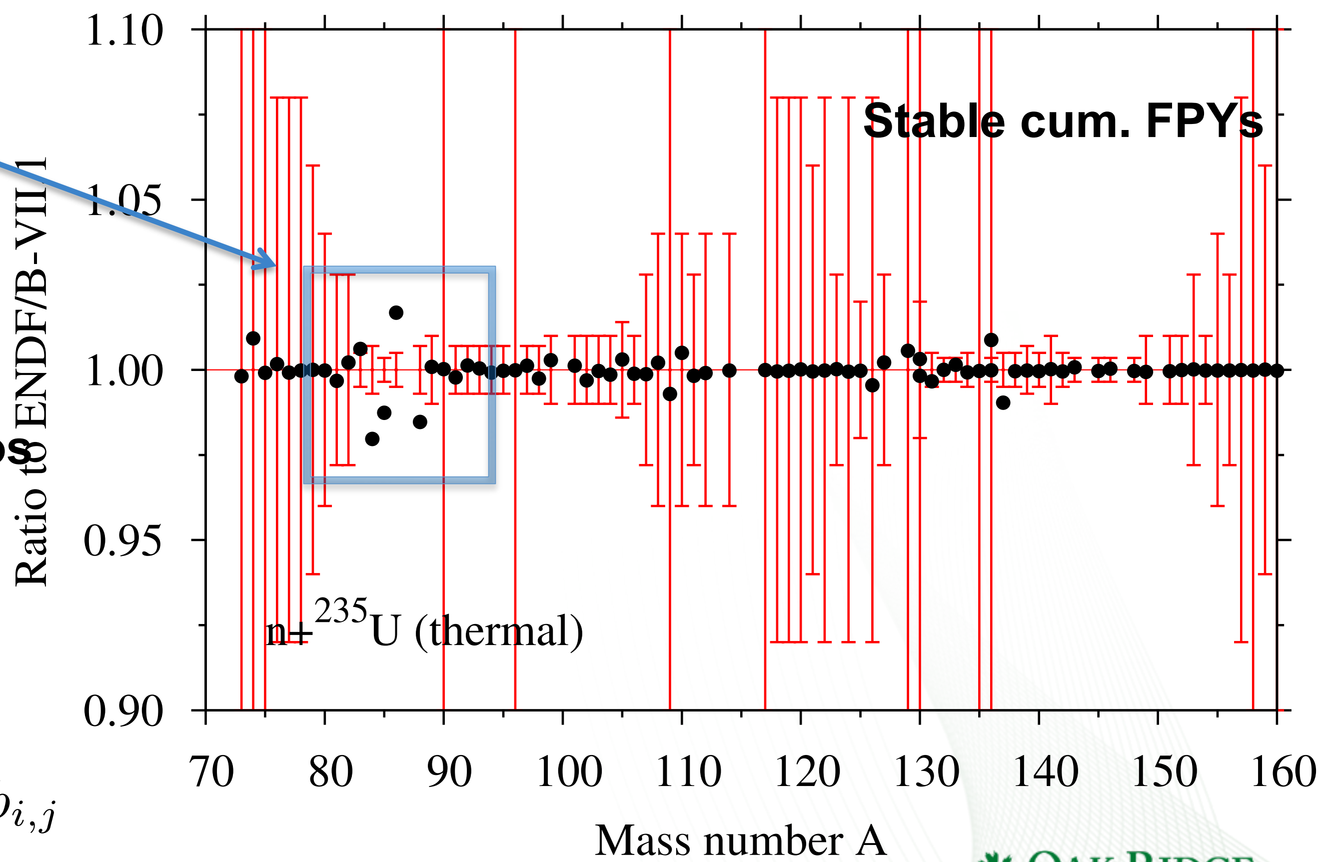
In red uncertainties (%) of cumulative yields in ENDF/B-VII.1

Biggest deviations

Cumulative yields

Independent FPY

Branching ratios



$$C_i(\mathbf{I}) = I_i + \sum_{j \in \mathbf{k}^i} C_j(\mathbf{I})\ b_{i,j}$$

Summary

- **Improvement of ^{238}Np nuclear data evaluations (cross sections and fission product yields) are needed for ^{238}Pu production**
- **Cross sections and fission product uncertainties and correlations**: **^{238}Np thermal cross section has 10% uncertainty, fission products very high uncertainties**
- **Fission product yields: large discrepancies (>30%) between predicted and measured**
- **Develop improved and reliable fission yield models for ^{238}Np**