Attribution Work on Americium (LA-UR-03-4354)

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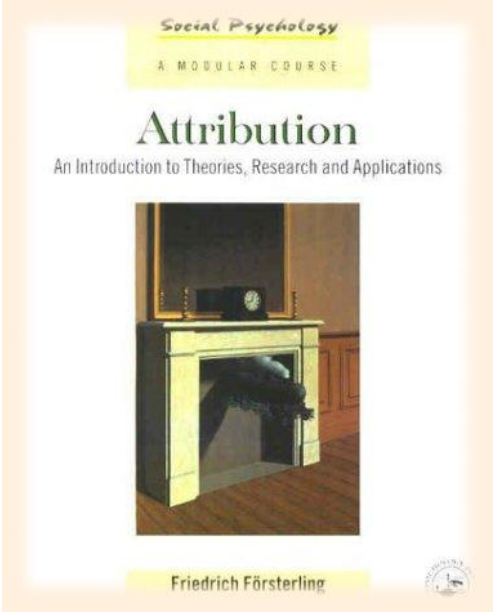

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Introduction

^{241}Am to ^{240}Am

- Americium is an impurity in plutonium.
- Americium isotopes can play a valuable role in attribution of a domestic nuclear explosion due to a terrorist or rogue state.
- We have initiated an effort to evaluate americium cross sections.
- In 2003 we have evaluated ^{241}Am$(n, 2n)$, fission, and capture cross sections, using nuclear reaction theory and experimental data.
- In 2004 The capture cross section of ^{241}Am was updated, and evaluations for some other isotopes were completed.

New LDRD Project at LANL

- New americium project was approved as a Laboratory-Directed Research & Development project (from FY05 to FY07, $>$ \$1.6M / year, between T, LANSCE, C, X, and D), work also supported by NA22.
- The goal of this project is to develop a new suite of accurate neutron reaction cross sections on americium isotopes.
  - Fabrication of americium targets (^{241}Am at LANL, ^{242m}Am at LLNL)
  - Experimental effort
    — neutron capture measurements with DANCE detector at LANSCE
  - Evaluation
    — development of GNASH, and modeling the neutron interaction with americium isotopes. (esp. important for cases where few measurements exist.)
  - Benchmark testing
    — NJOY processing, and MCNP simulations against TA18 critical assembly measurements.
  - Uncertainty quantification work.

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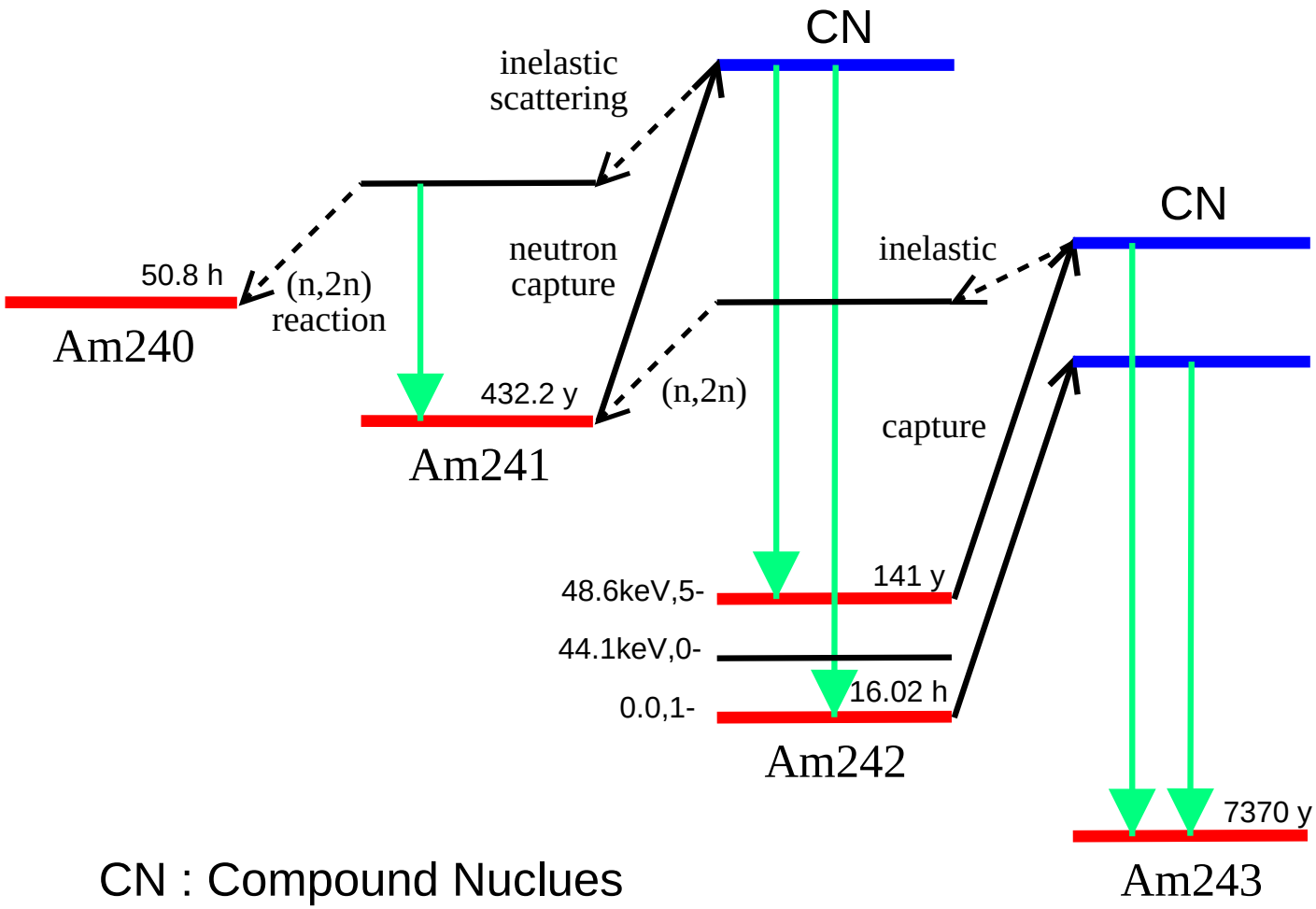
Nuclear Reaction Chain on Am Isotopes



CN : Compound Nuclues

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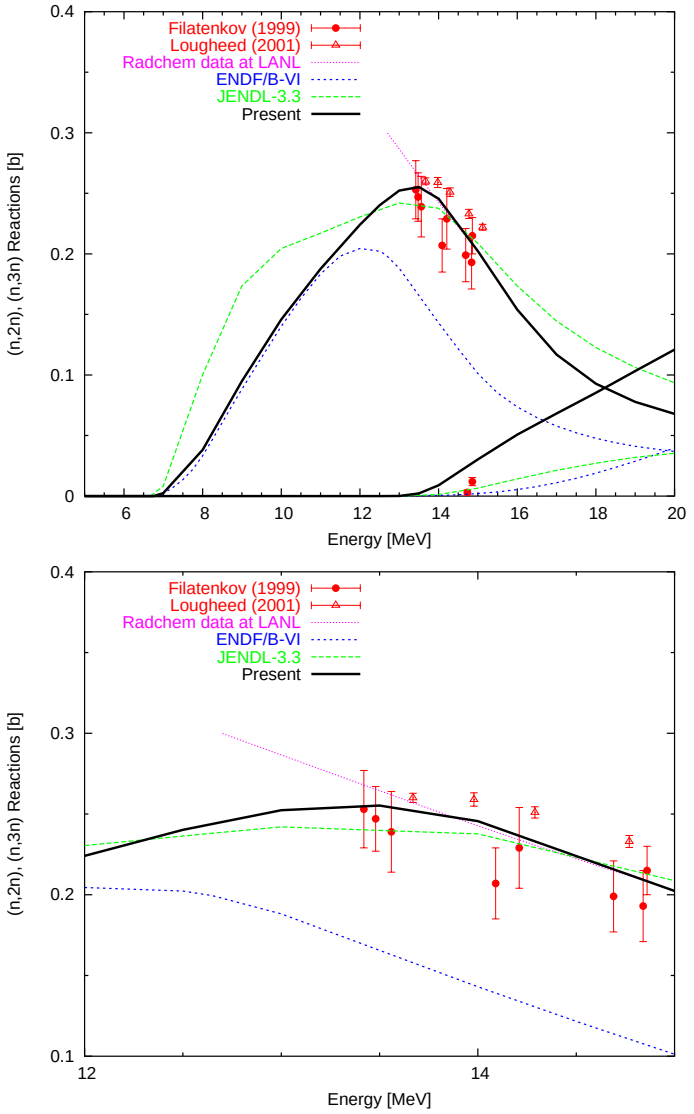
(n,2n) Reaction Cross Section - Am-241

Based on measurements at LANL and LLNL

- R.W. Lougheed, et al. (2001)
- Rad-Chem data at LANL (energy dependence near 14 MeV)
- A.A. Filatenkov, et al. (1999)

GNASH calculations

- Fission barriers were determined by adjusting to the experimental $(n, 2n)$ and fission cross sections.
- The KALMAN code was used for the adjustment.
- Further theory work is needed to better predict the cross section below 12 MeV where no measurements exist.



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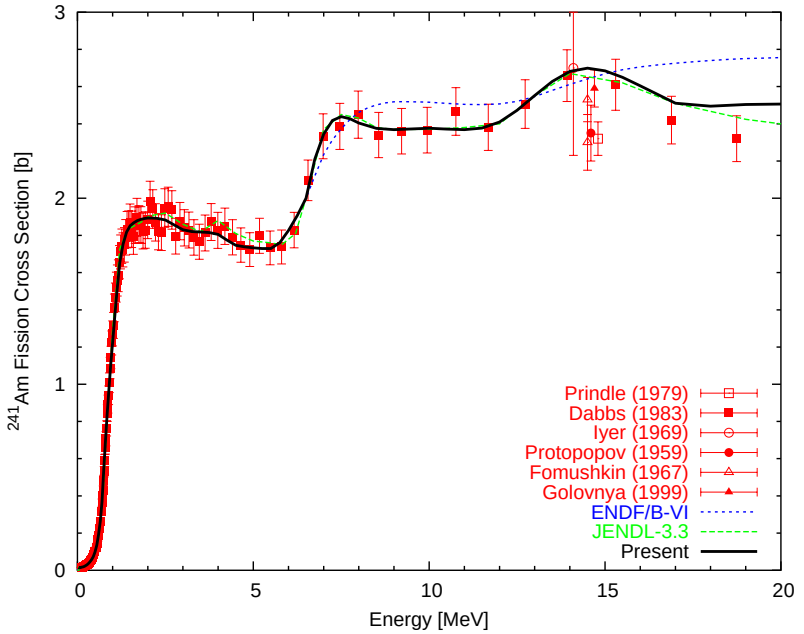
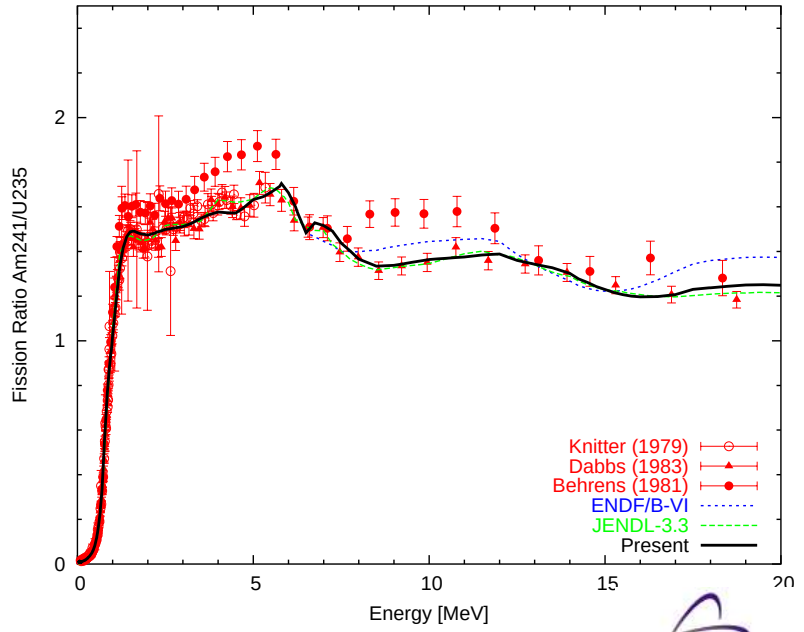
Fission Cross Section - Am-241

Statistical Analysis of Measurements

- The experimental data included were absolute measurements of $^{241}\mathrm{Am}(n,f)$ and $^{241}\mathrm{Am}(n,f)$ / $^{235}\mathrm{U}(n,f)$ ratio data
- New evaluation for $^{235}\mathrm{U}$ done by LANL, T16, was used as a standard.
- The LESQ fitting was made with a generalized least-squares fitting code
  - The fission cross section data are often high quality, and this procedure allows us to try to resolve discrepancies in measured values to determine the best value.
  - Based on the Bayesian statistics.





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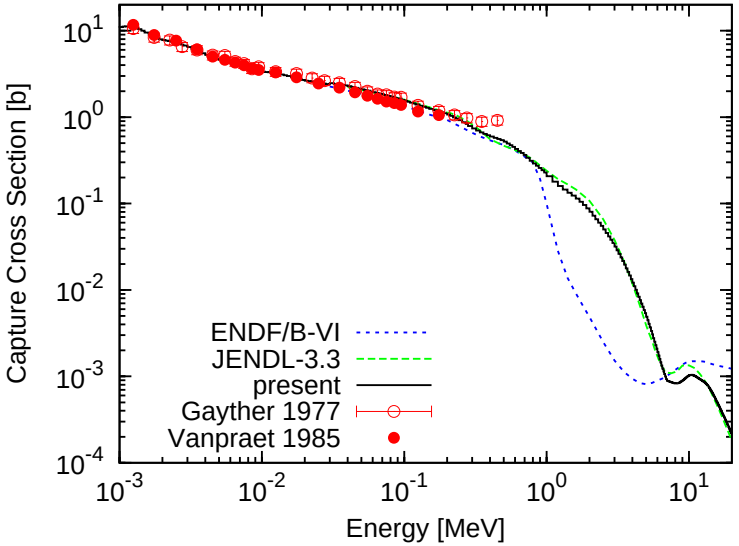
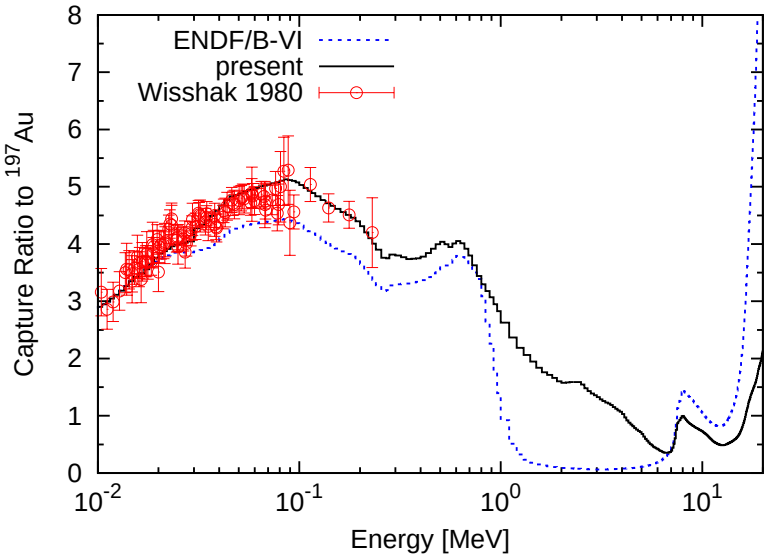
Capture Cross Section - Am-241

Statistical HF calculation

- ENDF/B-VI data were evaluated so as to reproduce Vanpraet data.
- However, the evaluated capture data are inconsistent with ^{241}Am / ^{197}Au ratio measurement by Wisshak and Käppelar.
- New data were evaluated to reproduce both Gayther *et al.* and Wisshak and Käppelar, simultaneously.
- The direct/semidirect capture process was included at higher energies.
- The new evaluation is about 15% higher than ENDF/B-VI.





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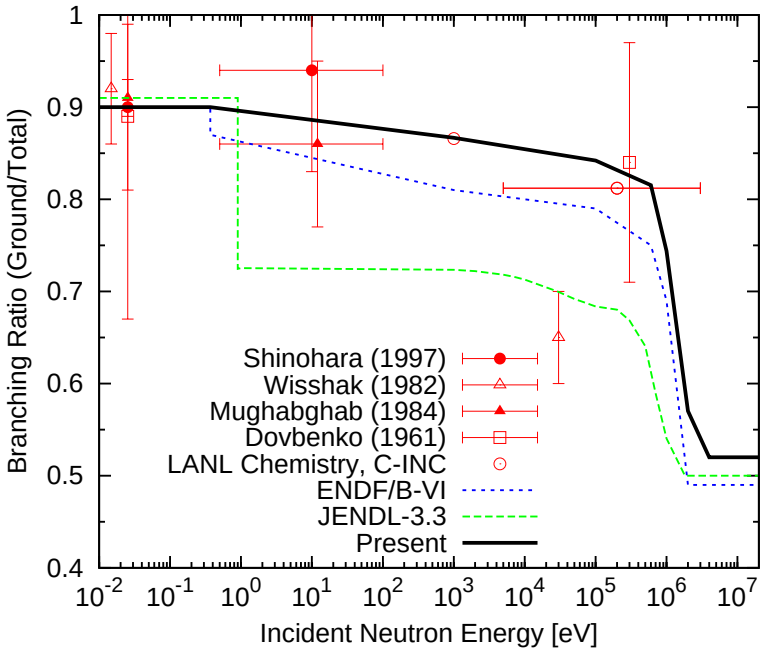
^{241}Am Capture (Isomeric Ratio)

Experimental database

- Thermal and epi-thermal regions
  - Shinohara, et al. (1997)
  - Wisshak, et al. (1982)
  - Mughabghab et al. (1984)
- High energy region
  - LANL data — ^{242m}Am/^{242g}Am production ratio
  - Averaged over Maxwell spectrum at kT =1 keV (C-INC)
  - Averaged over prompt fission neutron spectrum (C-INC)



The IR values were calculated with GNASH, and renormalized to the experimental data above. The higher IR is also consistent with the fast spectrum averaged value of Dovbenko (1961), and recent Post-Irradiation analyses at JNC (Ohki, et al.)

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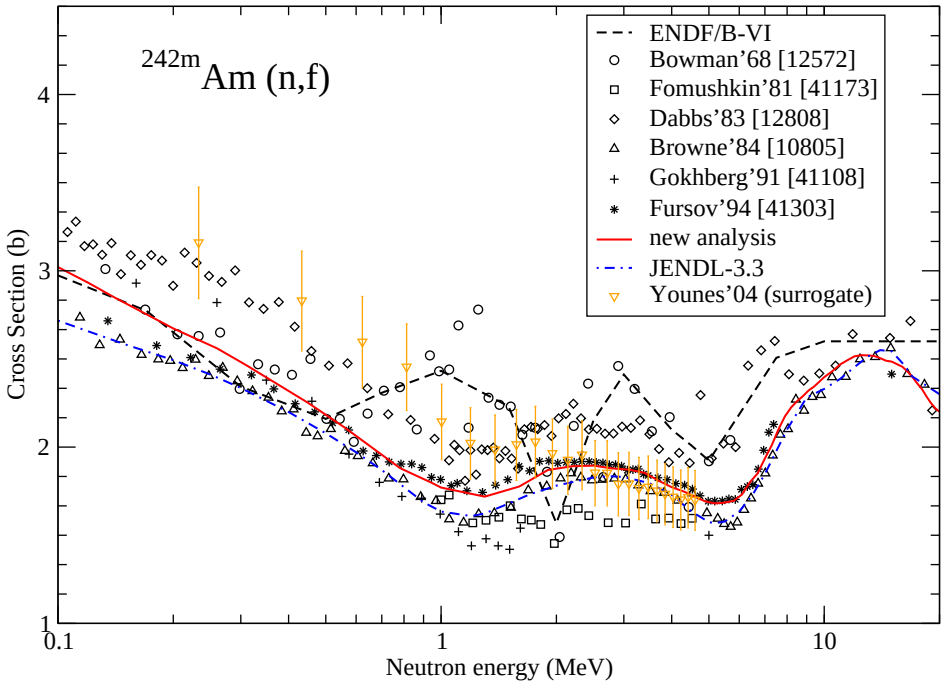
Fission Cross Section - Am-242m

Statistical Analysis

- The experimental data includ were the ratio to $^{235}\mathrm{U}(n,f)$ a $^{239}\mathrm{Pu}(n,f)$.
- New evaluations for $^{235}\mathrm{U}$ and ^{239} done by LANL, T16, were used.
- New fission data of Younes *et al.* we also included, which were obtained w the "surrogate technique."



The new evaluation basically follows the experimental data of Browne *et al.* and Fursov *et al.* The surrogate data also support our evaluation in the MeV energy range, however, the data seem to be higher at low energies.

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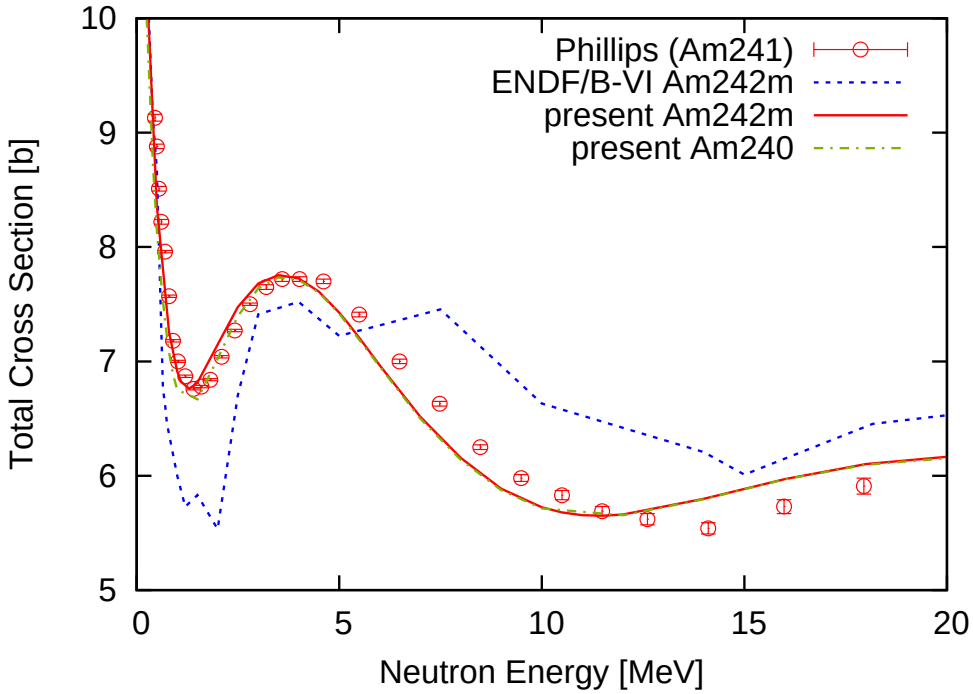
Total Cross Sections - Am-242m and Am-240

Coupled-Channels Calculation

- The coupled-channels optical potential obtained for ^{241}Am was used for those two nuclei.
- The coupling schemes used were :
  - 5^--6^--7^- for ^{242m}Am
  - 3^--4^--5^- for ^{240}Am



Since no experimental data exist for ^{240}Am and ^{242m}Am, data for ^{241}Am are shown for convenience. Because we assumed the same optical potential with an isospin correction term, the difference between ^{240}Am and ^{242m}Am is very small.

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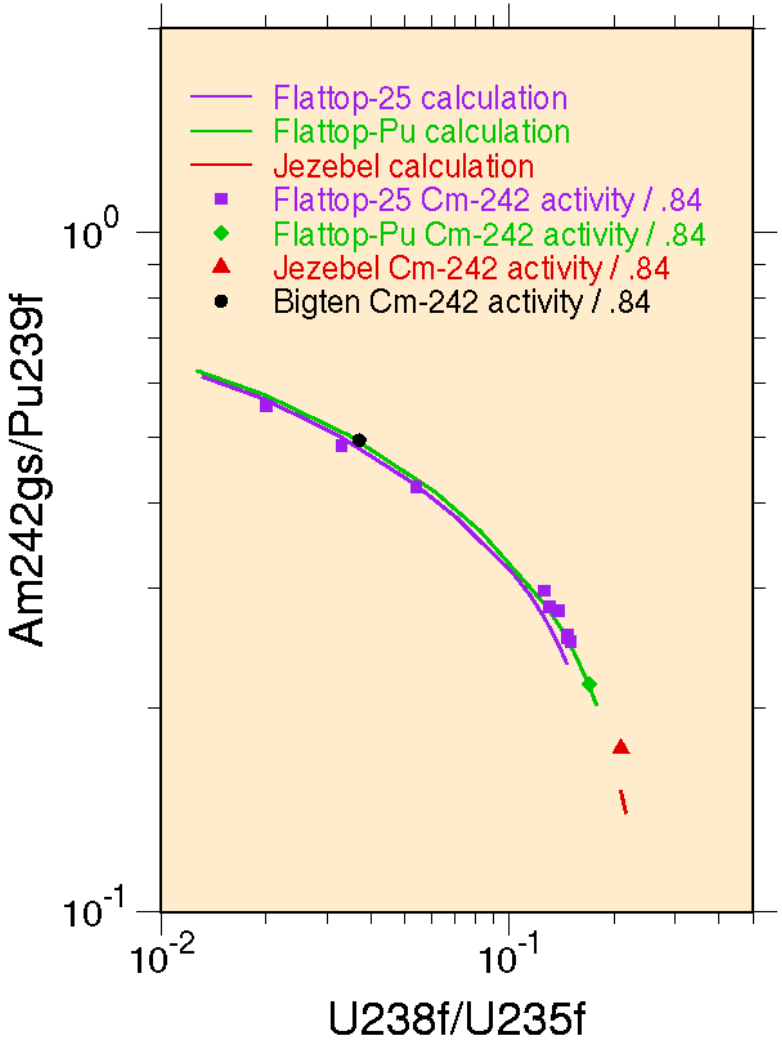
Data Testing for Integral Validation

- The evaluated americium cross sections are tested against TA18 (LACEF) critical assembly.
- A small americium sample is placed in different critical assemblies (Flattop, Jezebel, etc), and a reaction rate is measured as a function of neutron spectrum hardness.
- The MCNP simulations are carried out for the TA18 experiments to validate and provide feedback on the americium cross sections.



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Concluding Remarks

- We have initiated a new LDRD — americium ΔA project.
- The nuclear data for americium isotopes were upgraded. The new evaluation took advantage of recent measurements and advances in calculational modeling methods. The statistical Hauser-Feshbach theory was widely used for our evaluations.
- New measurements of ^{241,242m}Am capture will be performed at LANSCE with the DANCE detector.
- We also generated an evaluated nuclear data for ^{240}Am. We will refine this in the future.
- The evaluated americium cross sections are tested against TA18 (LACEF) critical assemblies.

Smoke Detector

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