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DRIVING THE FUTURE OF NUCLEAR TECHNOLOGY

Criticality Characterization of Pu-Fe Systems

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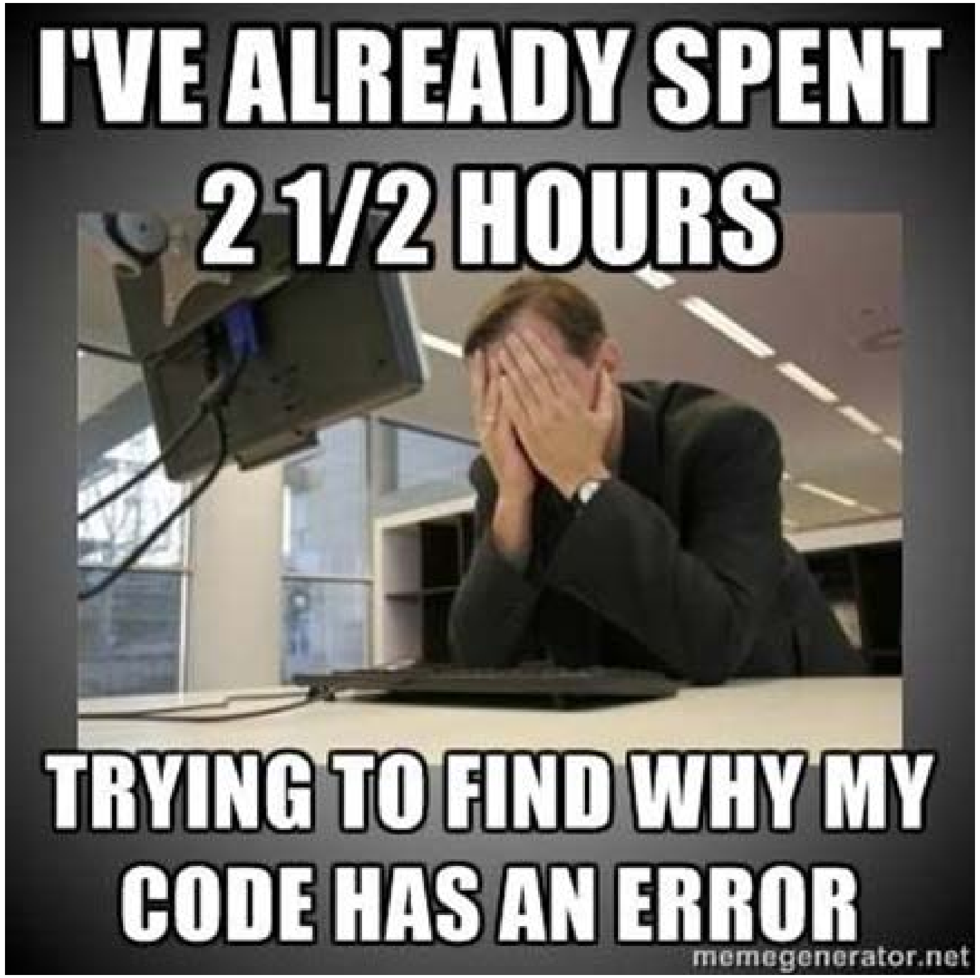


Outline

- Background
- Approach
- Results
  - Fe Sensitivity
  - Applicable Benchmarks
- Conclusion/Applicability

Background

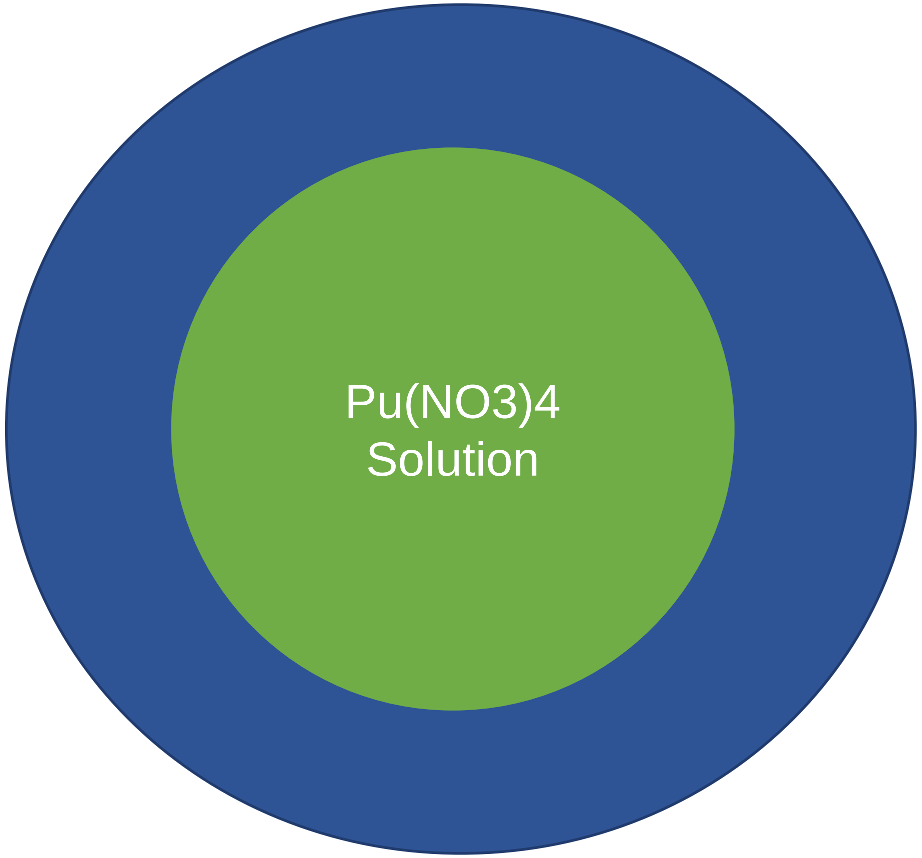
- Criticality safety for waste processing where iron is relied upon to maintain subcriticality
  - Are there any benchmarks that support this application?
    - ANSI/ANS 8.24-2017: “When no benchmark data are available, establishment of a calculational margin and upper subcritical limit (USL) is not possible within the requirements of this standard.”
    - ANSI/ANS 8.24-2017: “When limited benchmarks are available, other experimental data (e.g., cross-section differential data) may be used in the development of the appropriate bias uncertainty or margin of subcriticality. Sensitivity analysis may be used to determine the importance of specific nuclides to the system of interest.
  - What are the differences between the benchmarks and application?
    - ANSI/ANS 8.24-2017: “The determination of bias uncertainty should contain allowances for uncertainties in benchmark physical properties and measurement techniques; uncertainties due to limitations in the geometric, material, or neutronic representations (e.g., cross sections) used in a calculational model; and statistical and convergence uncertainties.”

Approach

- Selected an application
  - Plutonium solution maintained subcritical with iron bearing solids in suspension
- Conduct sensitivity analysis
  - How much iron is in the benchmarks in the ICSBEP?
  - How many benchmarks have iron in suspension?
  - How much iron can be added to the system before the system is subcritical?
  - Are there benchmarks that correlate with a subcritical system?

Approach
Model Definition and Assumptions

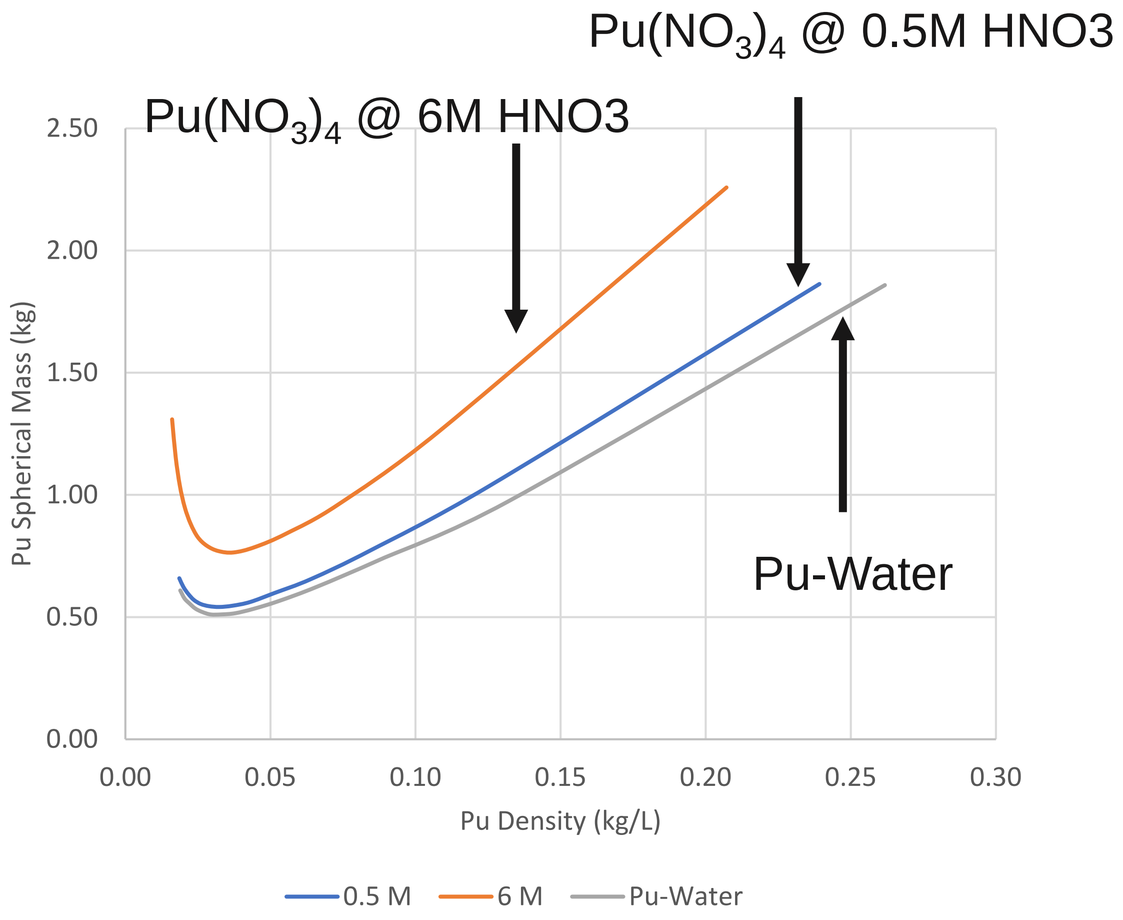
- Sphere of $Pu(NO_3)_4$
  - ^{239}Pu
- ARH-600 Method
  - Pu Density
  - HNO_3 molarity range 0.5 M – 6.0 M
- 30 cm of water reflection
- Calculated critical mass and radius



Approach

Overview of Parametric Studies

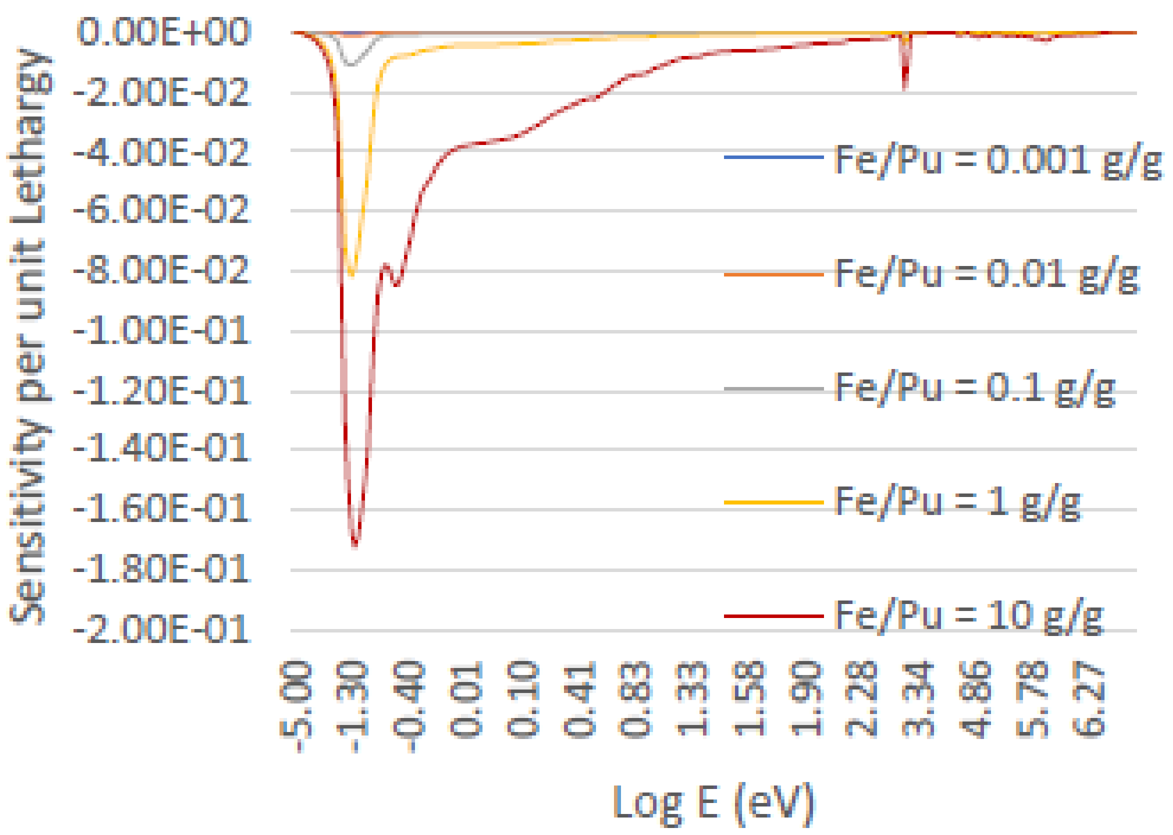
- Selected critical mass and radius
  - H/Pu = 1100
  - $MHNO_3$ = 6.0 M
- Added iron
  - Fe/Pu (g/g) range 0.001 – 10
- Critical radius recalculated



Approach

Sensitivity Studies

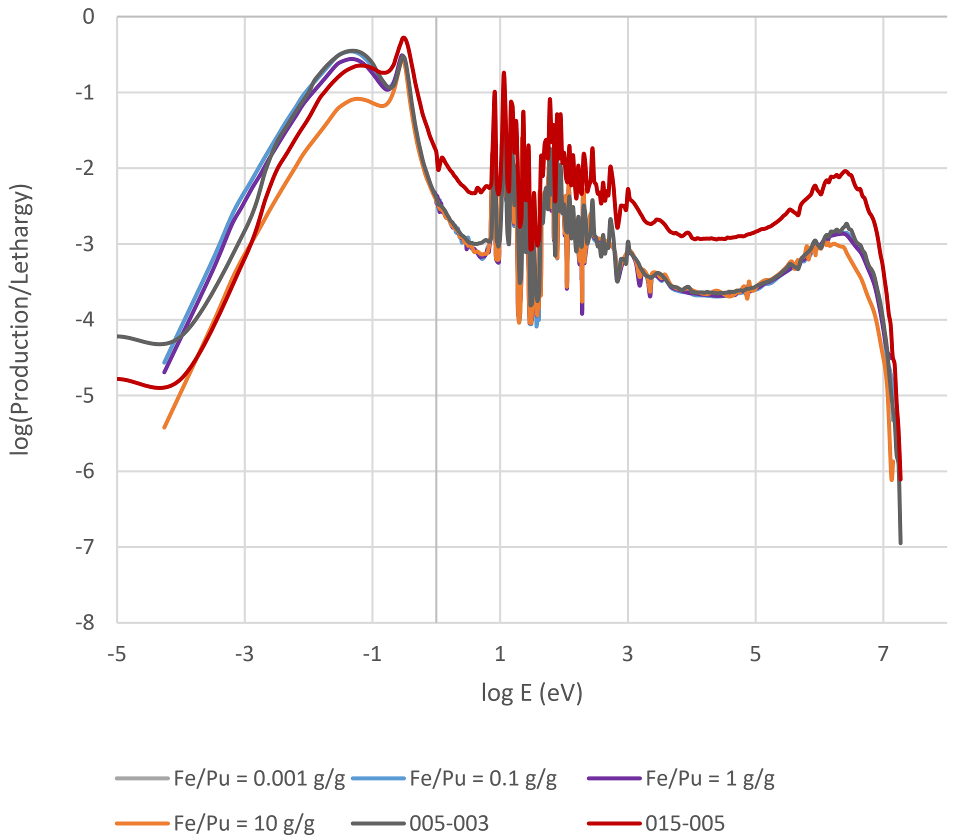
- Performed to determine the sensitivity of k-eff to the concentration of iron in the system
  - TSUNMAI-1D was used for this.
  - SDF files were generated for concentrations of iron ranging from 0.001 g/g to 10 g/g
- Sensitivity profiles from applications were used to compare to PU-SOL-THERM-XXX ICSBEP benchmarks to determine applicability



Approach

Neutron production evaluation

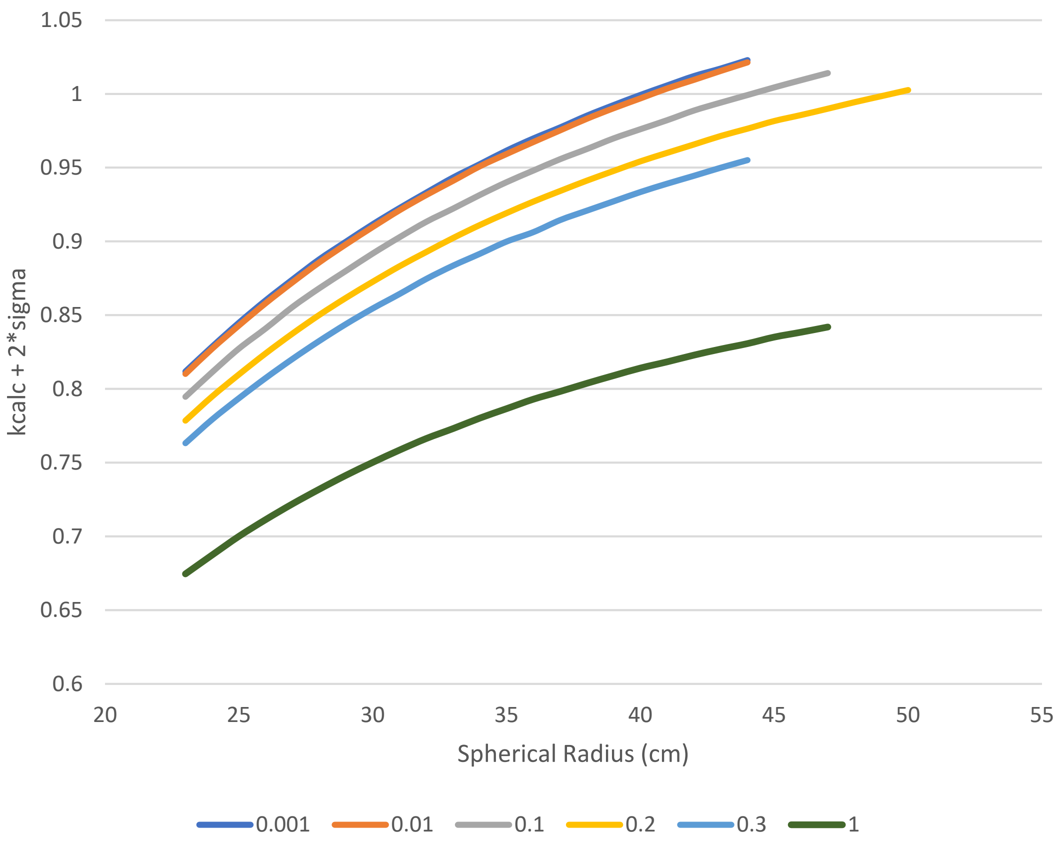
- Applicability of benchmarks:
  - Evaluated the neutron production of application cases with iron range of 0.001 g/g to 10 g/g
  - Compared the application cases to neutron production generated for PU-SOL-THERM-XXX benchmarks from ICSBEP



Results

Fe Sensitivity Effects on Subcriticality

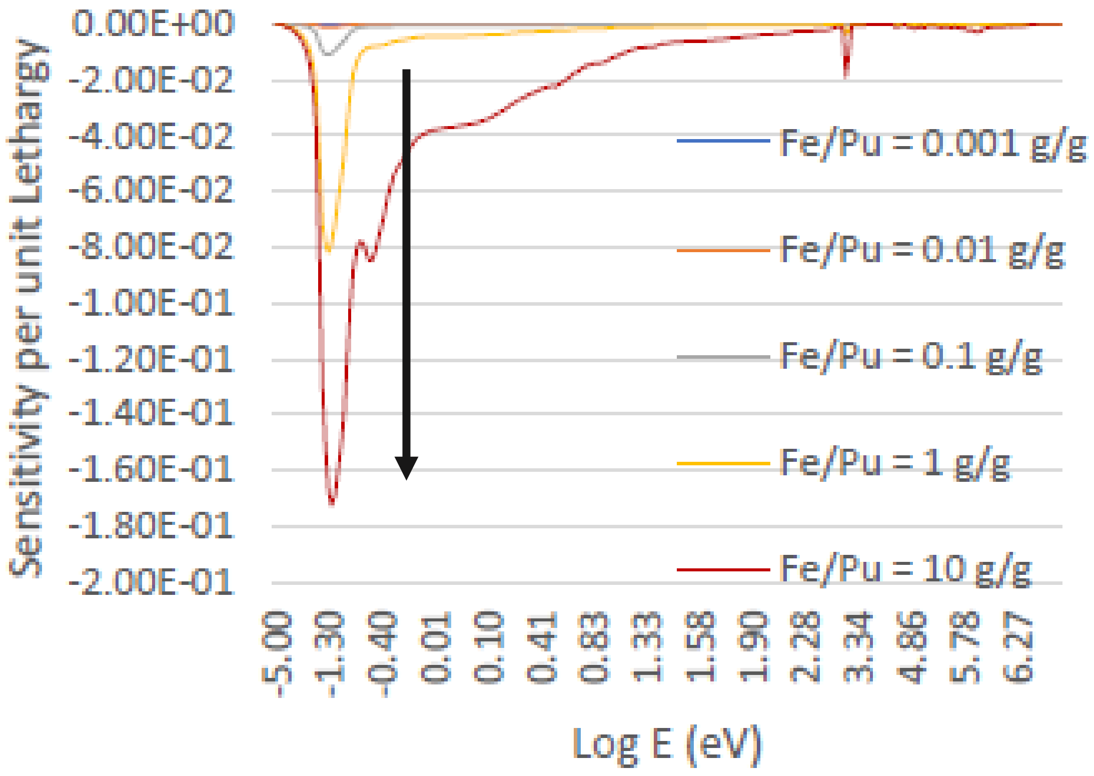
- Criticality could not be achieved for a $Pu(NO_3)_4$ system with a Fe/Pu > 0.2 g/g
- 6M HNO_3 and H/Pu = 1100



Results

Fe Sensitivity Effects on Subcriticality of Application Cases

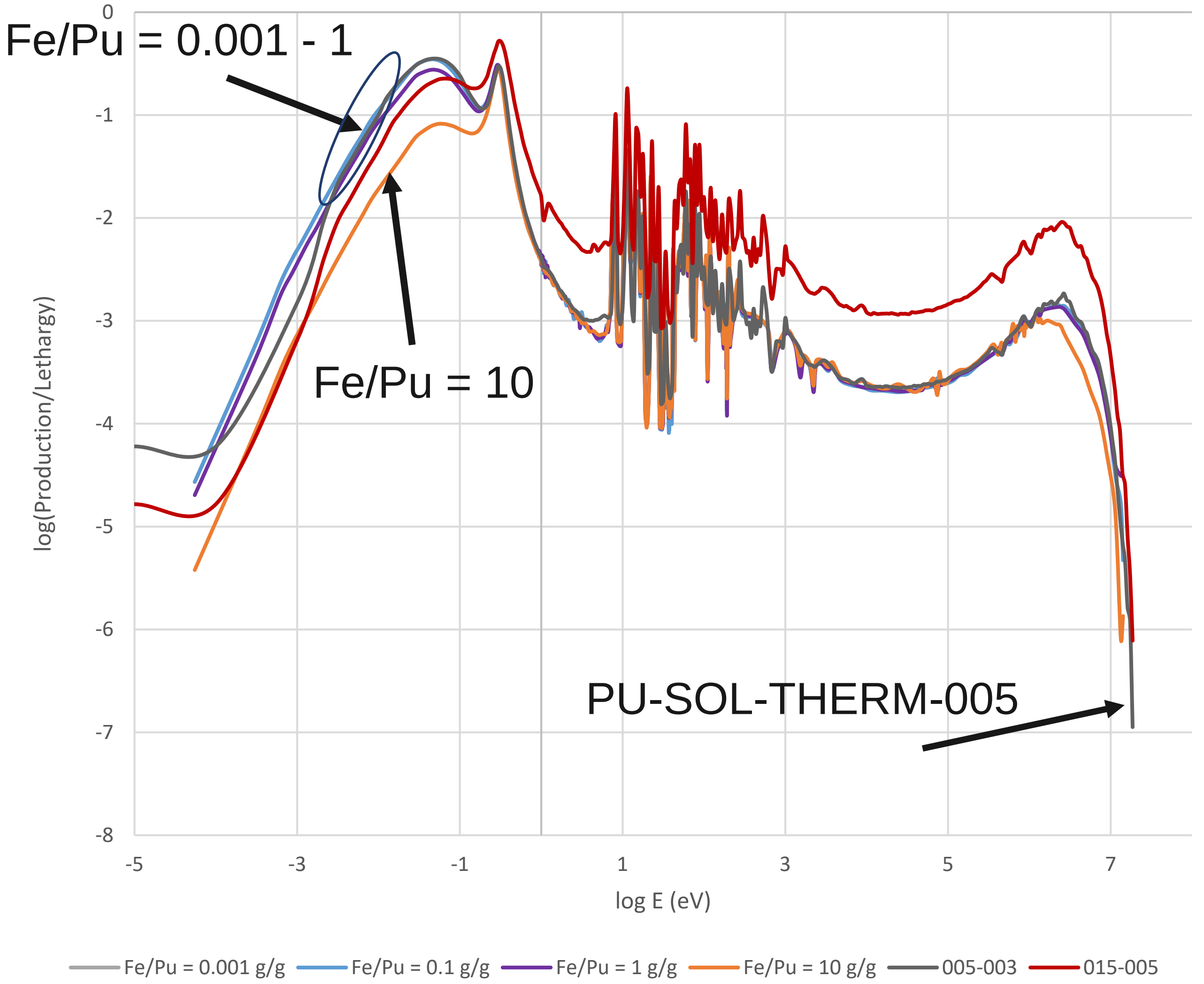
- Increasing negative sensitivity to k-eff with ^{56}Fe increasing concentration



Results

Neutron production comparison to ICSBEP Benchmarks

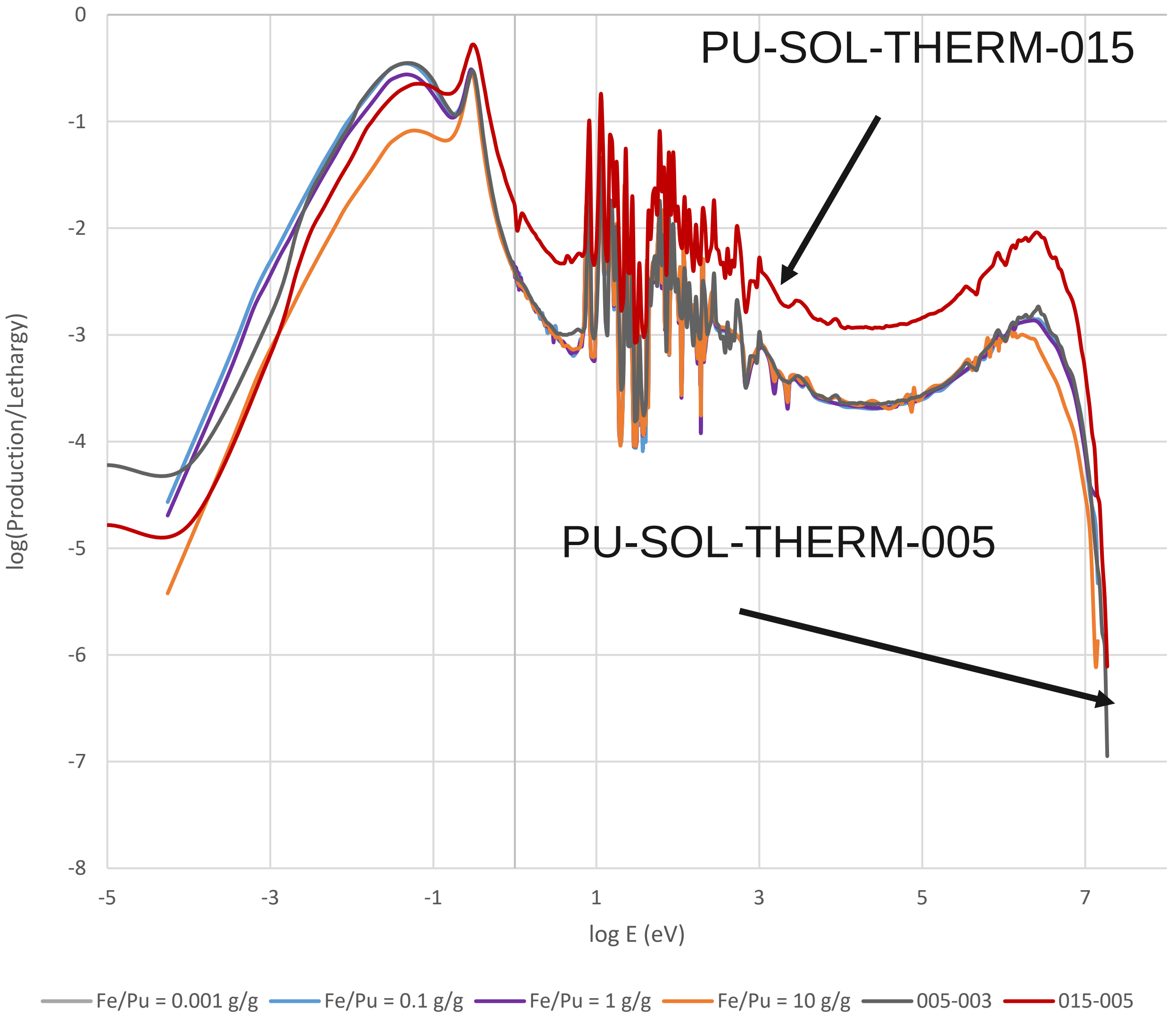
- Fe/Pu = 0.001 – 1 g/g
  - Good fit for neutron production between applications and most benchmarks
- Fe/Pu = 1 – 10 g/g
  - Neutron production between applications and benchmarks begins to diverge in the thermal region

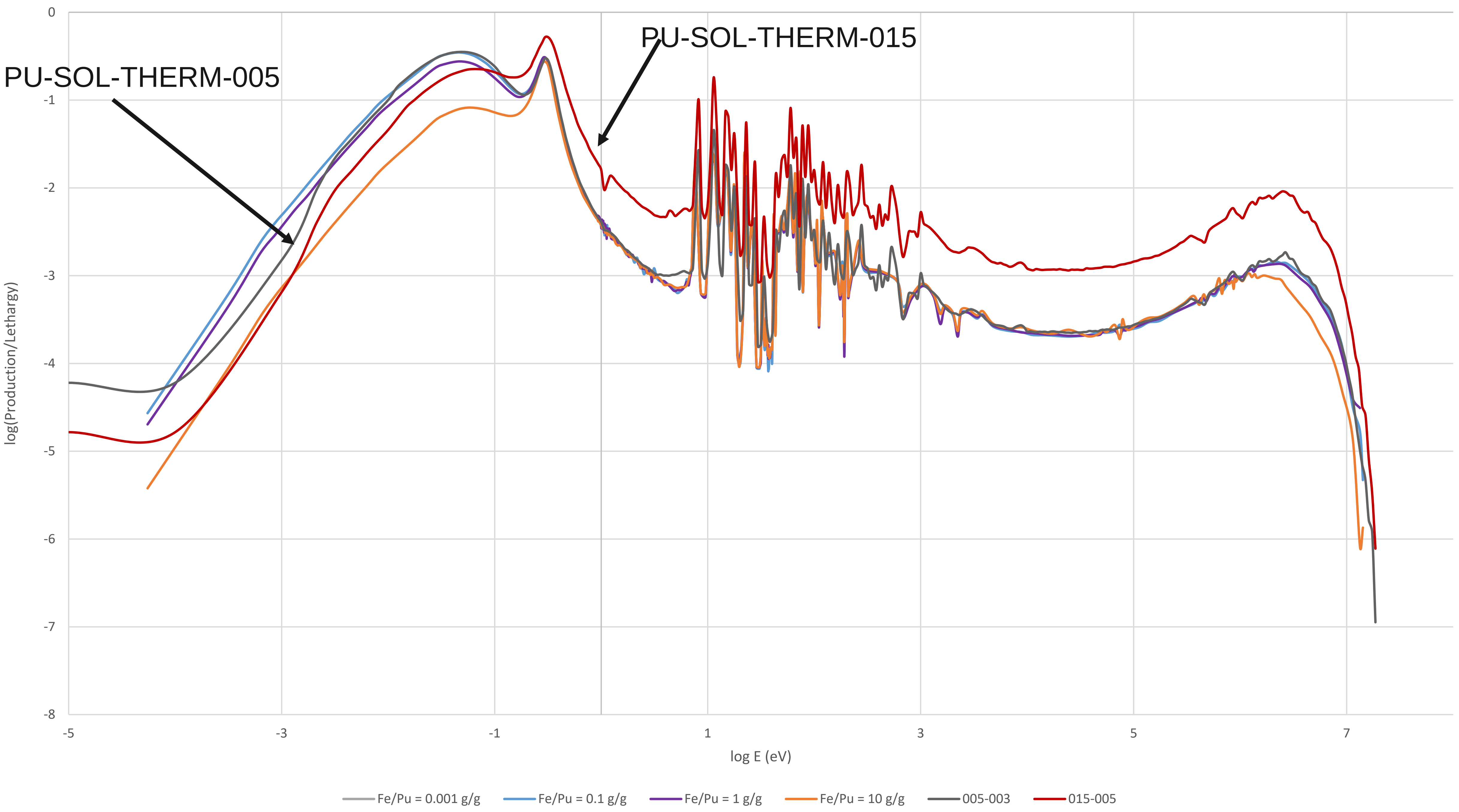


Results

Effect of Dominant Reaction Channel on Applicability of Benchmarks

- PU-SOL-THERM-013 – 017
  - No correlation in the spectra between benchmarks and applications
  - More iron in system than PU-SOL-THERM-001 – 012 benchmarks



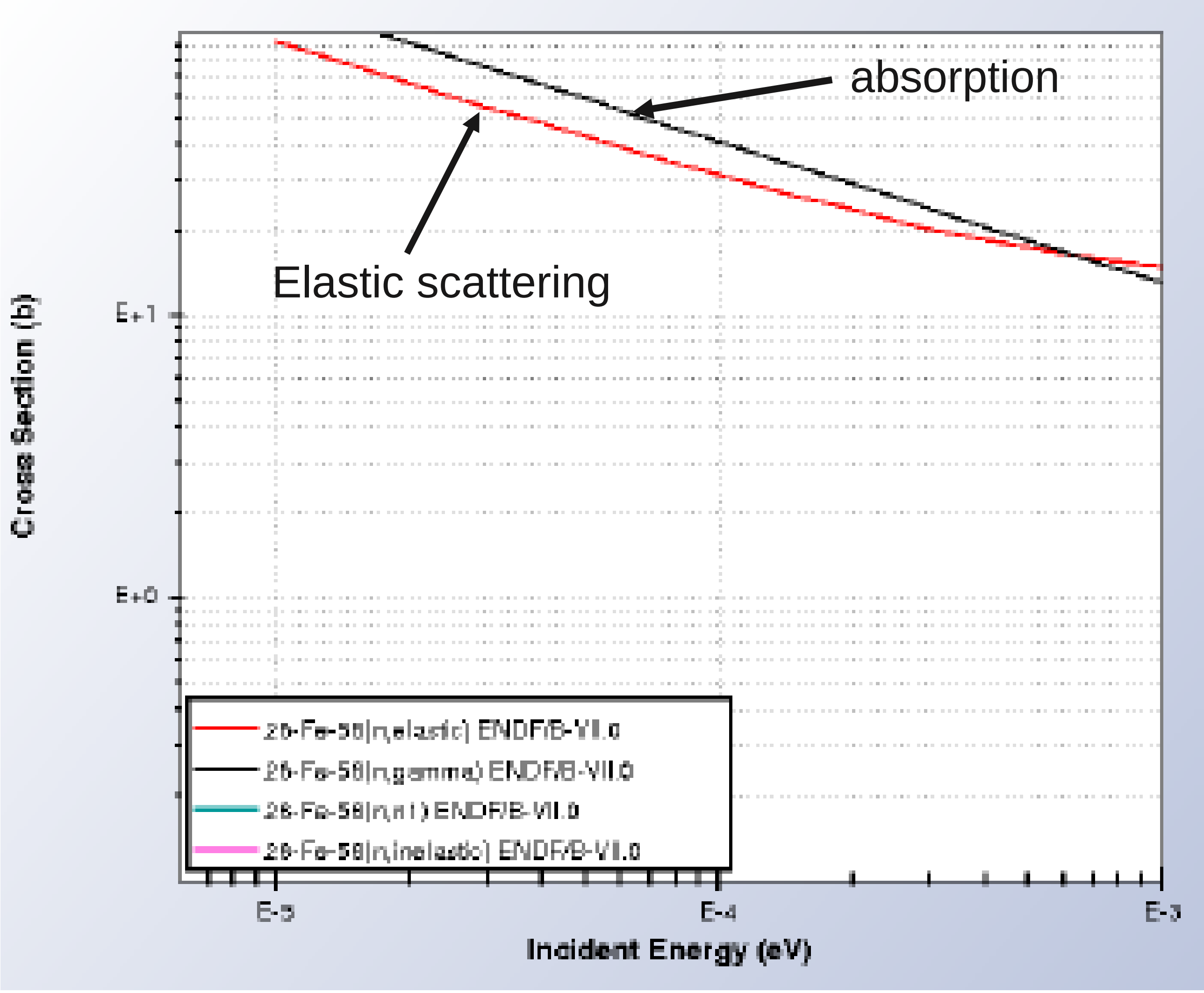


PU-SOL-THERM-015
PU-SOL-THERM-005
log(Production/Lethargy)
log E (eV)
0
-1
-2
-3
-4
-5
-6
-7
-8
-5
-3
-1
1
3
5
7
Fe/Pu = 0.001 g/g
Fe/Pu = 0.1 g/g
Fe/Pu = 1 g/g
Fe/Pu = 10 g/g
005-003
015-005

Results

Effect of Dominant Reaction Channel on Applicability of Benchmarks

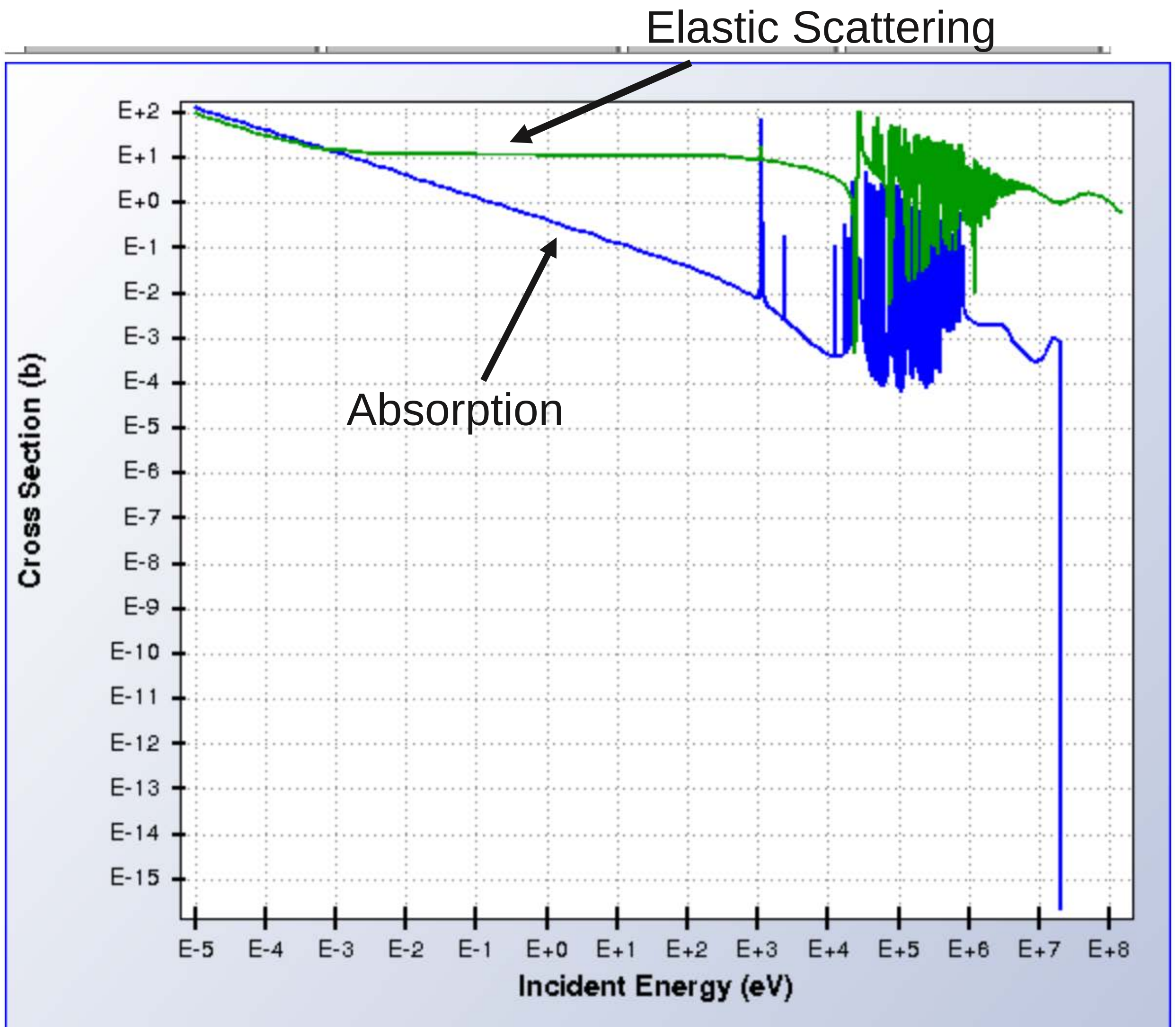
- Absorption is only dominant at thermal energies



Results

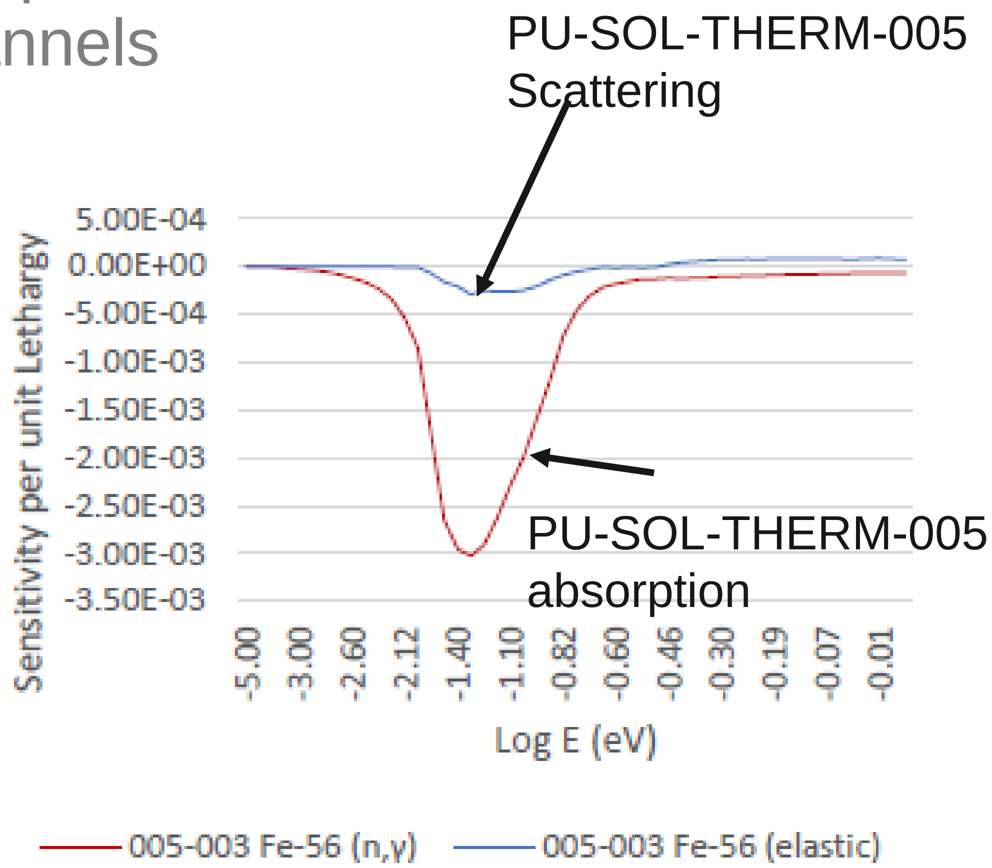
Effect of Dominant Reaction Channel on Applicability of Benchmarks

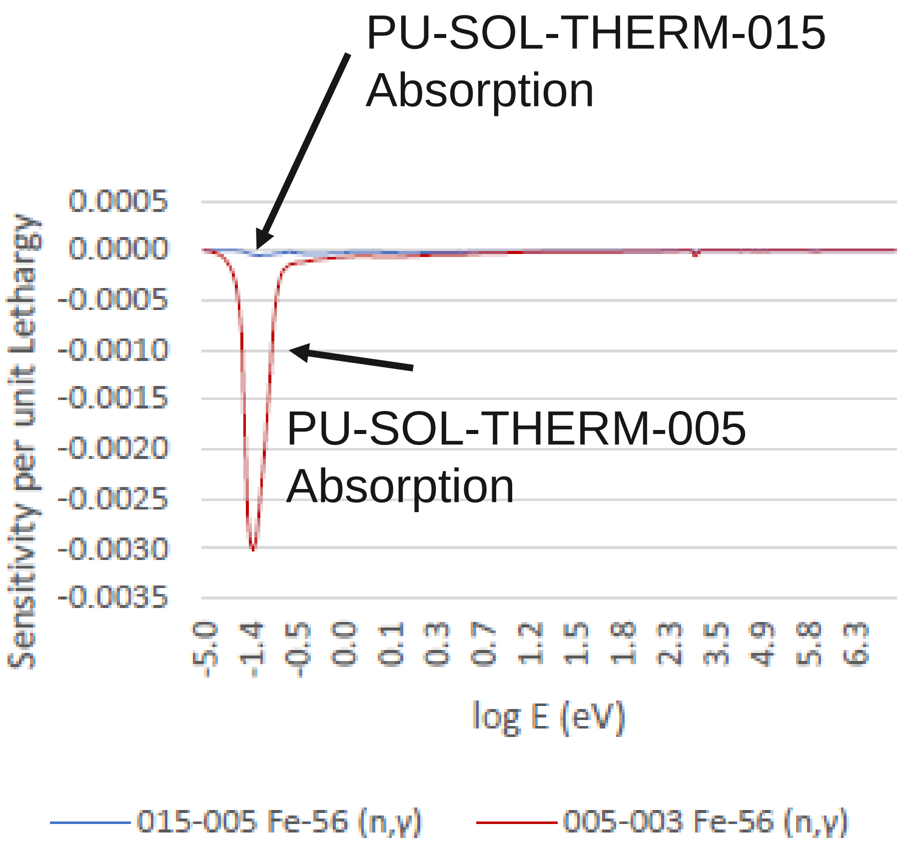
- Dominant reaction channel for ^{56}Fe is elastic scattering not absorption at higher energies (> 10^{-3} eV).
  - When elastic scattering is the dominant rxn channel, sensitivity is inversely proportional to concentration of Fe.



Results

Comparison of PU-SOL Benchmarks and Fe Sensitivity based on Reaction Channels



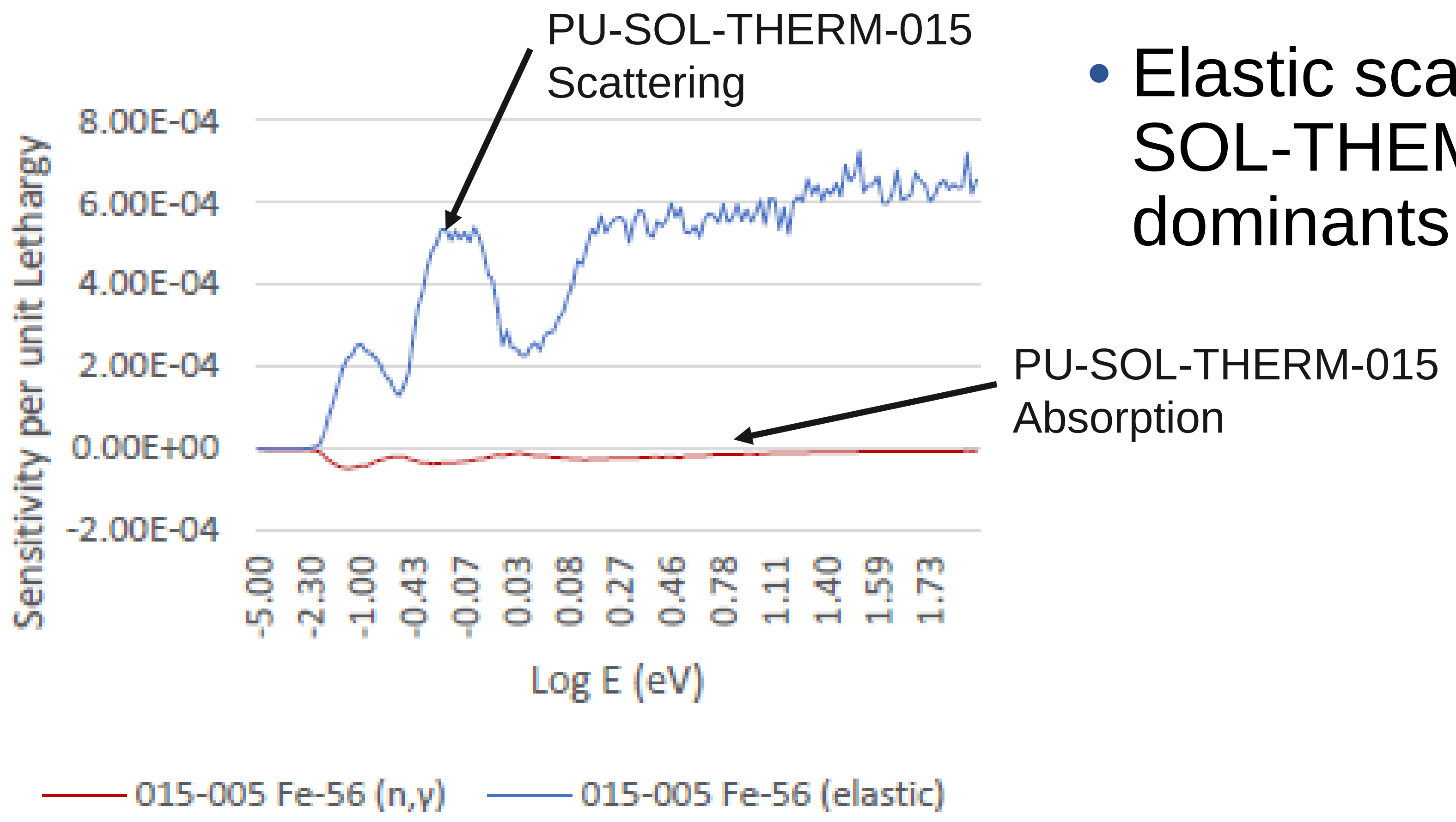


PU-SOL-THERM-001 - 012 dominant reaction channel for Fe sensitivity is absorption

PU-SOL-THERM-013-017 has very little sensitivity neutron absorption with iron

Results

Comparison of PU-SOL Benchmarks and Fe Sensitivity based on Reaction Channels



- Elastic scattering from PU-SOL-THEM-013-017 dominants iron sensitivity.

Don’t let the iron fool you! More iron doesn’t necessarily mean negative sensitivity.

Results

Applicable Benchmarks ($c_k > 0.8$) from TSUNAMI-IP

Fe/Pu Ratio (g/g)	Number of Benchmarks
0.001	64
0.01	64
0.1	58
0.2	39
0.3	0
1	0
10	0

Conclusion/Applicability

- There are applicable benchmarks for the systems utilizing iron in low enough concentrations that criticality is still possible (i.e., Fe/Pu <= 0.2 g/g)
  - This is comparable to the impurity levels of iron in the benchmark experiments
- It is imperative to understand the dominant reaction channels in the application and the selected benchmarks to ensure they match.
  - Do not just assume that because the benchmark has similar nuclides to the application that the benchmark will apply.
- For applications of $Pu(NO_3)_4$ solutions containing iron bearing solids in suspension at sufficient concentration to maintain subcriticality (i.e., Fe/Pu > 0.2 g/g) there do not appear to be any suitable benchmarks.
  - The CSSG Response to Tasking 2014-02 recommends in cases such as these:
    - Designing applicable criticality experiments to support development of a USL; or
    - Setting the USL at 0.8
  - Consider crediting other controls in lieu of a neutron absorber
  - It is not possible to set a defensible USL from the existing ICSBEP database and be in compliance with ANSI/ANS 8.24

Possible Future Work

- Update the Pitzer Method to include $Fe(NO_3)_3$ solutions
  - Allow for a more accurate solution mixture of iron and plutonium nitrate solutions
- Use CIGALES V2.0
  - According to literature this code attempts to fix the underestimation of k_{eff}
  - Compare results
- Model using MCNP6.2 and WHISPER
  - Compare results
- Investigate possibility to extend sub-critical measurements/methods to dilute thermal systems from those used currently for fast systems to propose new, directly applicable benchmark experiments

Questions?