


Nuclear Criticality Safety Division Topical Meeting NCSD 2022

Integral experiments at sandia with molybdenum sleeves for testing ^{95}Mo cross sections in thermal energy spectrum

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Embedded topical meeting at the
 ANS Annual Meeting

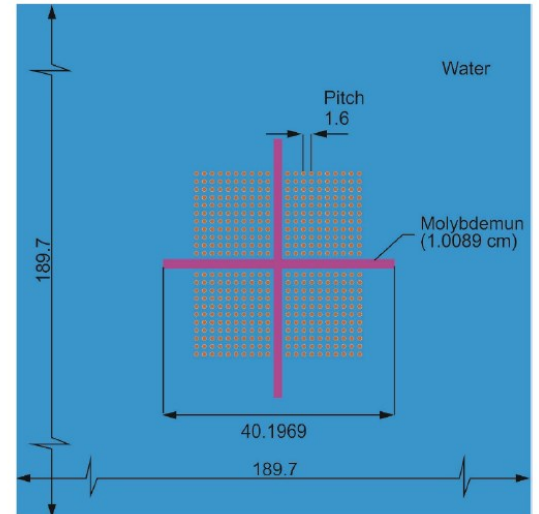
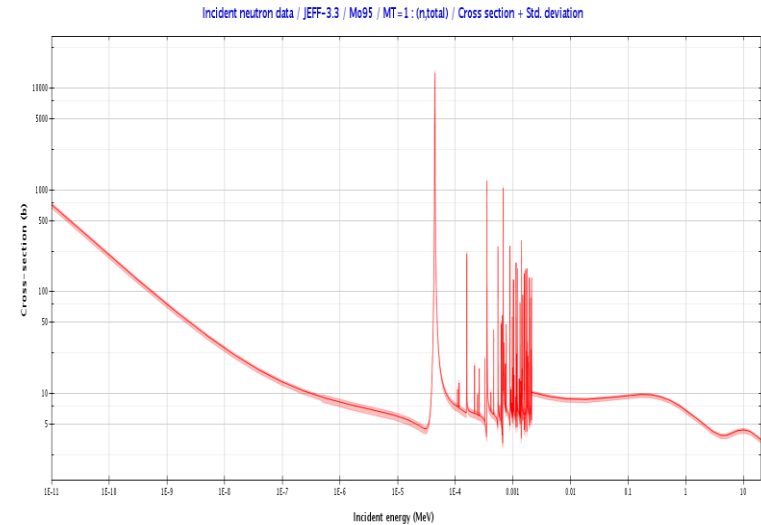
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INSTITUT DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Outline

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- Design of experiments
- Description of experiments with Mo sleeves
- Reproducibility, reference and sampling experiments
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- Predicted results: k_{eff} and experimental uncertainties
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- Conclusions

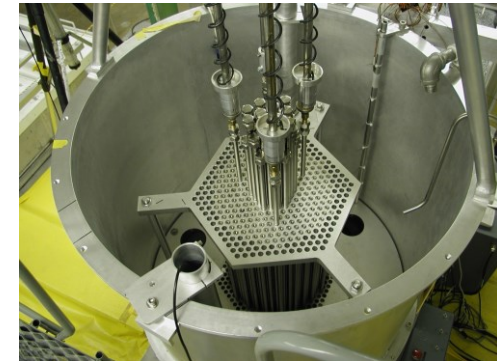
Context

- Molybdenum plays important role in NCS
 - Fission product (BUC)
 - e.g: found in reprocessing plants as a MoZr deposit including Pu and impregnated by dissolution solution
 - Naval and space reactors fuel
 - New research reactor fuel: UMo
- ^{95}Mo , ^{96}Mo and ^{97}Mo isotopes:
 - Main abundant isotopes in molybdenum
- ^{95}Mo has the largest capture cross section
- Few experiments involving molybdenum
 - French MIRTE (LCT-106) with 10 mm Mo screen not sensitive enough to ^{95}Mo capture around 45 eV (first resonance of ^{95}Mo)



Context

- Up to now, IRSN recommends to take margins in NCS calculations to bound the lack of experimental data to validate Mo cross sections
- Collaboration between IRSN and SNL to propose an integral experiment in 2015, approved by NCSP
 - Preliminary design report (CED-1) in 2018
 - Final design report (CED-2) end of 2020
- Material supply study analyzing costs in 2021
- Sleeves ordered by IRSN in April 2021 and delivered at Sandia November 2021
- Experiment planned for FY2022 Q4

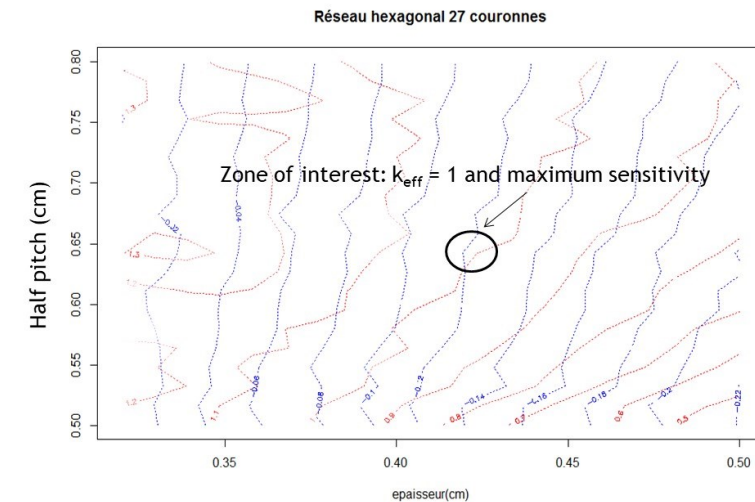
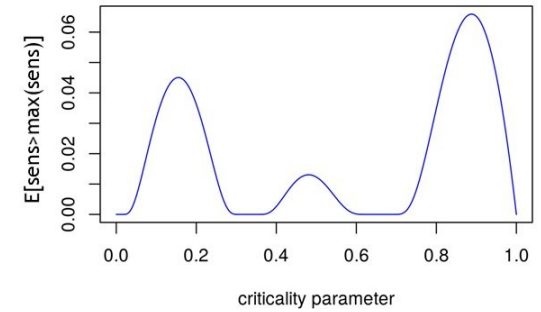
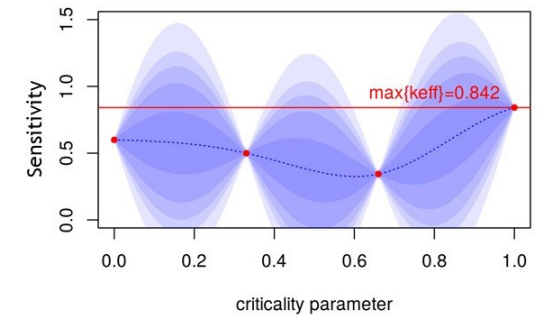


Design of experiments

- Optimization of the experiments with French Prométhée workbench to maximize Mo sensitivities on k_{eff} and to match application cases remaining close to 1
 - Use of EGO, ECEGO algorithms
 - Bayesian approach with constraint on k_{eff}
- Surrogate function Sensitivity (pitch, R_{sleeves})
 - Random function
 - Interpolates calculated points
 - Gaussian predictor mean, sd:

$$E[\text{Sens}(\text{pitch}, R_{\text{sleeves}})] = \text{mean}(\text{pitch}, R_{\text{sleeves}})$$

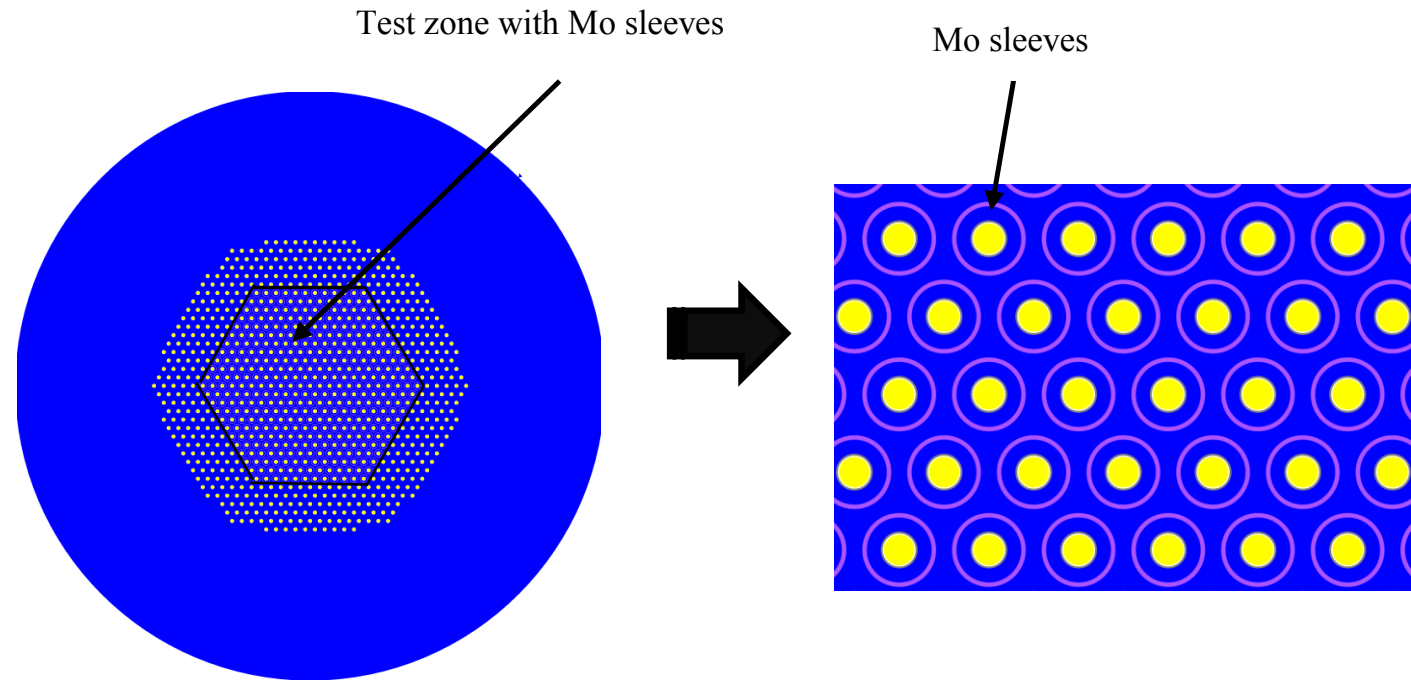
$$\text{Var}[\text{Sens}(\text{pitch}, R_{\text{sleeves}})] = \text{sd}(\text{pitch}, R_{\text{sleeves}})^2$$



↪ Convenient to estimate: $E[\text{Sens}(\text{pitch}, \text{water}) > \max\{\text{Sensitivity}\}] +$

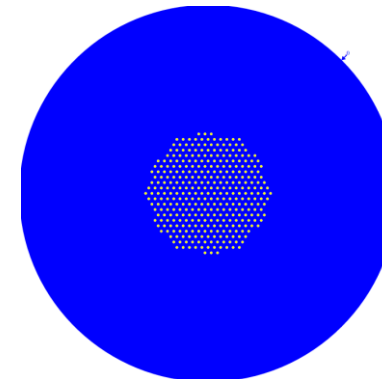
Description of experiments with Mo sleeves

- Use of 877 BUCCX rods
 - 6.9 % ^{235}U enriched
 - Pitch (hexagonal): 1.55 cm
 - New grids to be manufactured
 - Test zone with 397 rods surrounded by Mo sleeves
- Mo sleeves:
 - 0.762 mm thickness
 - External dia: 12.7 mm
 - Height: 50 cm (fissile column of rods)
 - Water can circulate inside sleeves



Reproducibility, reference and sampling experiments

- Reproducibility with complete dismantling of assembly and reassembly
 - Assessment of experimental uncertainties
- Sensitivity of k_{eff} to ^{95}Mo capture is low compared with sensitivity to main reactions (^{235}U fission, ^{238}U capture)
 - Reference experiments necessary to assess bias due to Mo cross sections
 - Hexagonal lattice with 12 rods on the side and 8 rods removed on each edge of the hexagon
- Sampling experiments
 - Assess impact of two lots of rods
 - Assess impact of rods' location



Manufacturing of sleeves

- 400 sleeves manufactured by the PLANSEE company end of May 2021 to September 2021
- Delivered to SNL November 2021
- Measured values (PLANSEE):
 - Height, external and internal diameters (very low uncertainties required by IRSN)
 - Content in Al, Cr, Cu, Fe, K, Ni, Si, C, H, N, O, Cd, Hg, Pb impurities lower than 40 $\mu\text{g/g}$ and lower than 300 $\mu\text{g/g}$ for W
 - Complementary measurements by Gary Harms (SNL)

	Height (mm)	External diameter (mm)	Sleeve thickness (mm)	Internal diameter (mm)	Holes diameter (mm)
Measured values	497.878 \pm 0.094	12.730 \pm 0.021	0.771 \pm 0.045	11.187 \pm 0.080	5.079 \pm 0.023
Specification values	498 \pm 1.5	12.7 \pm 0.1	0.762 \pm 0.076	11.176	5.08 \pm 0.127

Evaluation of experimental uncertainties

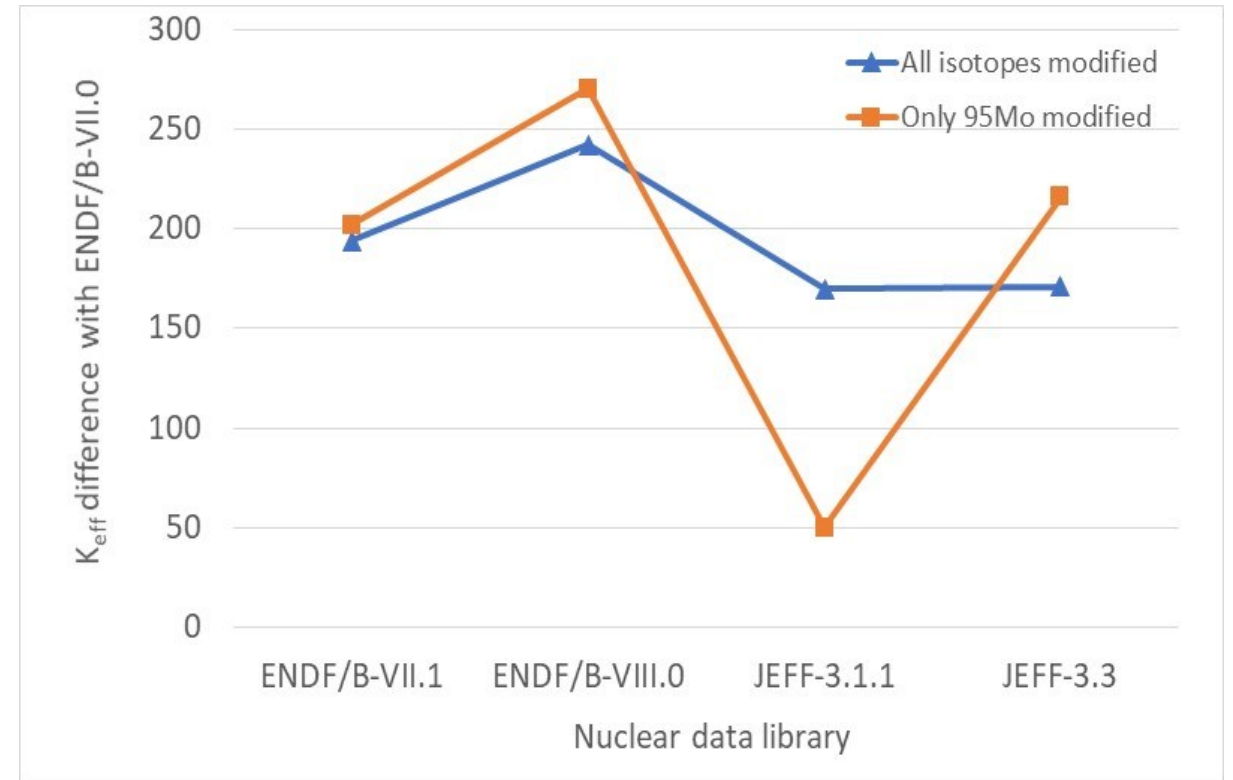
- First estimate

- Main uncertainty comes from the sleeves' thickness and gap sleeve/BUCCX rod
- Total uncertainty estimated to be 72 pcm at 1σ level

	1σ uncertainty (systematic)	Variation	$(k_{\text{eff}}(+\text{variation}) - k_{\text{eff}}(-\text{variation}))/2$	$\Delta k_{\text{eff}} (1\sigma)$
Estimated uncertainty due to 7uPCX rods				55
Sleeves Thickness (cm) – keeping mass constant	0.0013	0.00762	168	29
Sleeves density (g/cm³)	$0.01/\sqrt{3}$	0.01	12	7
Gap sleeve/rod (cm)	0.0002	0.002	362	36
Total				72

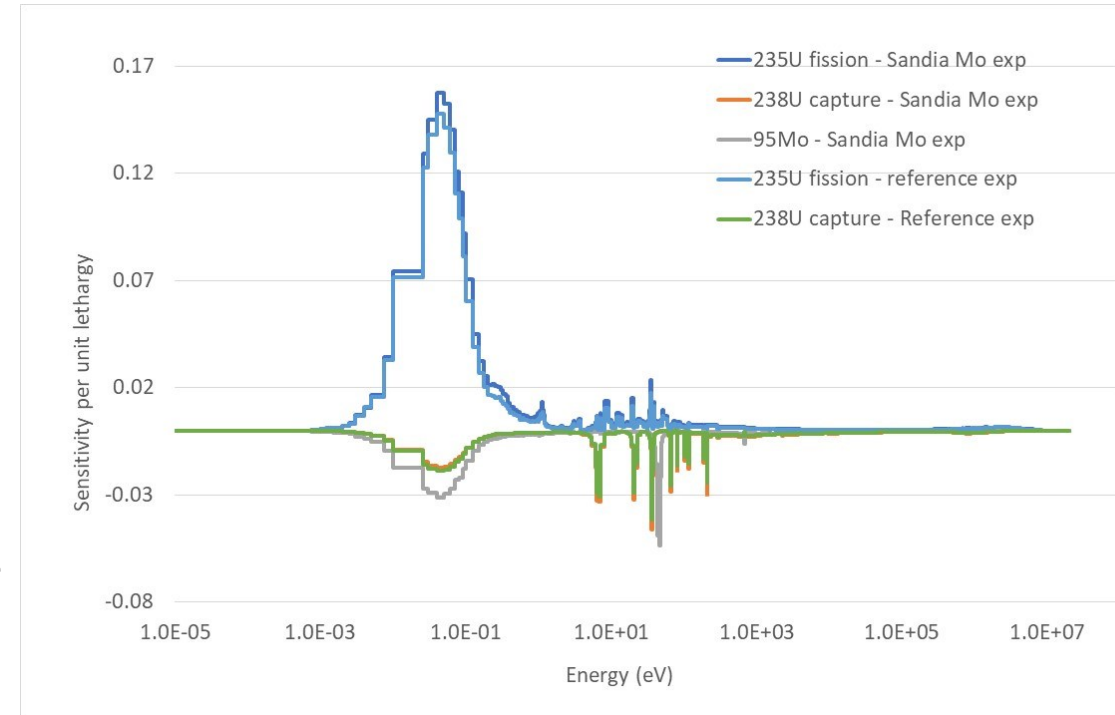
Predicted k_{eff}

- ENDF/B-VII.0: $k_{\text{eff}} = 1.00350 \pm 0.00020$
- ENDF/B-VII.1, ENDF/B-VIII.0, JEFF-3.1.1 and JEFF-3.3 results consistent
- Results with ^{95}Mo are ~200 pcm higher than ENDF/B-VII.0 results, except for JEFF-3.1.1



Comparison to application case

- UPuMoZr industrial case featuring dissolution residues of French reprocessing plant dissolver
 - Bounding composition
 - Density: 2.6 g/cm³
 - Representative of fuel burnt up to 50 GWd/t heavy metal
 - Slightly differs on the ⁹⁵Mo percentage and more on other isotopes

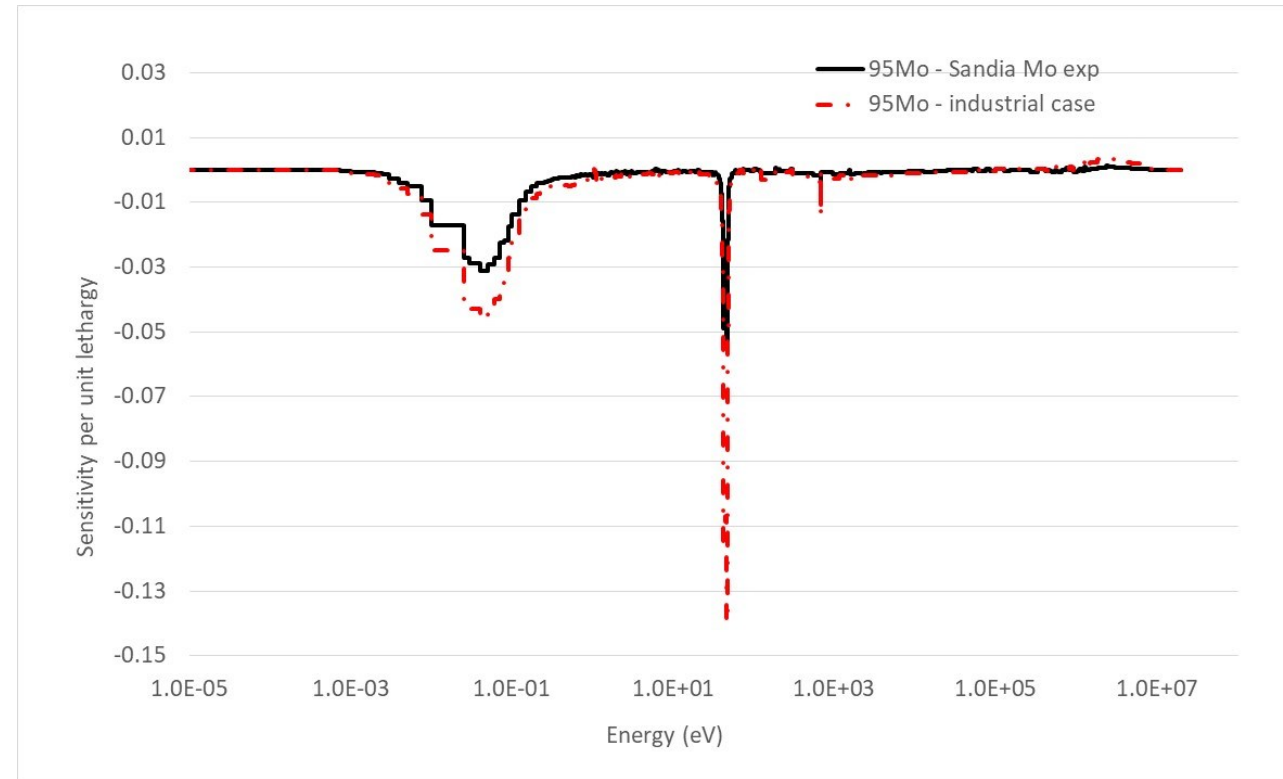


Element	U	Pu	Mo	Zr
Contents in wt. % (approx.)	6	2	63	28
Isotope	²³⁵ U	²³⁸ U		
Enrichment in wt. %	1	99		
Isotope	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
Content in wt. %	57	25	16	2

Isotope	⁹² Mo	⁹⁵ Mo	⁹⁶ Mo	⁹⁷ Mo	⁹⁸ Mo	¹⁰⁰ Mo
Contents in wt. % (application case)	1	21	3	23	25	27
Sandia experiment	14.84	15.92	16.68	9.55	24.13	9.63

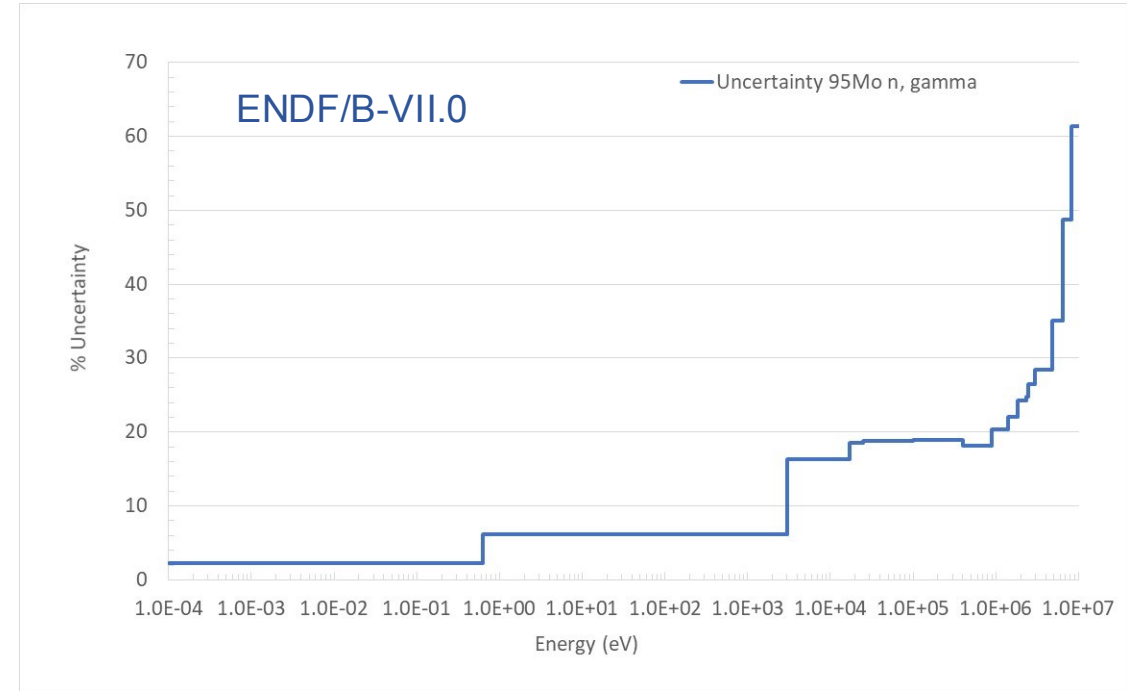
Comparison to application case

- Integral sensitivity to ^{95}Mo total cross section:
 - Application case: $-0.1335 \pm 3.47 \times 10^{-4}$
 - Sandia experiments: $-0.0843 \pm 3.31 \times 10^{-4}$
- Comparison of k_{eff} sensitivity to ^{95}Mo total cross section
 - Close in shape: sensitivity at the same energy (thermal bump) and first resonance of ^{95}Mo at 45 eV addressed
 - Slightly less sensitive than industrial case but better agreement than for previous MIRTE experiments (LCT-106)



Feedback on nuclear data

- Quite low uncertainty on ^{95}Mo capture cross section in thermal energy range
- However, 30 % of the prior uncertainty due to ^{95}Mo capture cross section
- New measurements at J-PARC (2020) and at n-tof, GELINA, RPI on enriched samples will help improving nuclear data
 - New resonance parameters and covariance data with multichannel R-matrix SAMMY software



	Overall uncertainty	Propagated uncertainty due to ^{95}Mo (n, γ) – Using TSURFER
44 group ENDF/B-VII.0	1026 ± 0.200	328 ± 0.05
56 group ENDF/B-VII.1	720 ± 0.226	314 ± 0.06

Conclusions

- New experiments using Mo sleeves designed and accepted by US/DOE NCSP will be performed at SNL in 2022 Q4
- 5 experiments planned including:
 - Reference experiments: nuclear data feedback
 - Reproducibility experiments: check evaluation of experimental uncertainties
 - Sampling experiments: check positioning of rods' uncertainty
- Experiments will provide feedback on ^{95}Mo nuclear data in thermal energy range in keeping with new differential measurements of Mo and new assessment of cross sections

Acknowledgements

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Questions

