Comparative Critical Mass Calculations for NNL and ENDF/B-VIII.0 Zirconium Hydride Thermal Scattering Laws

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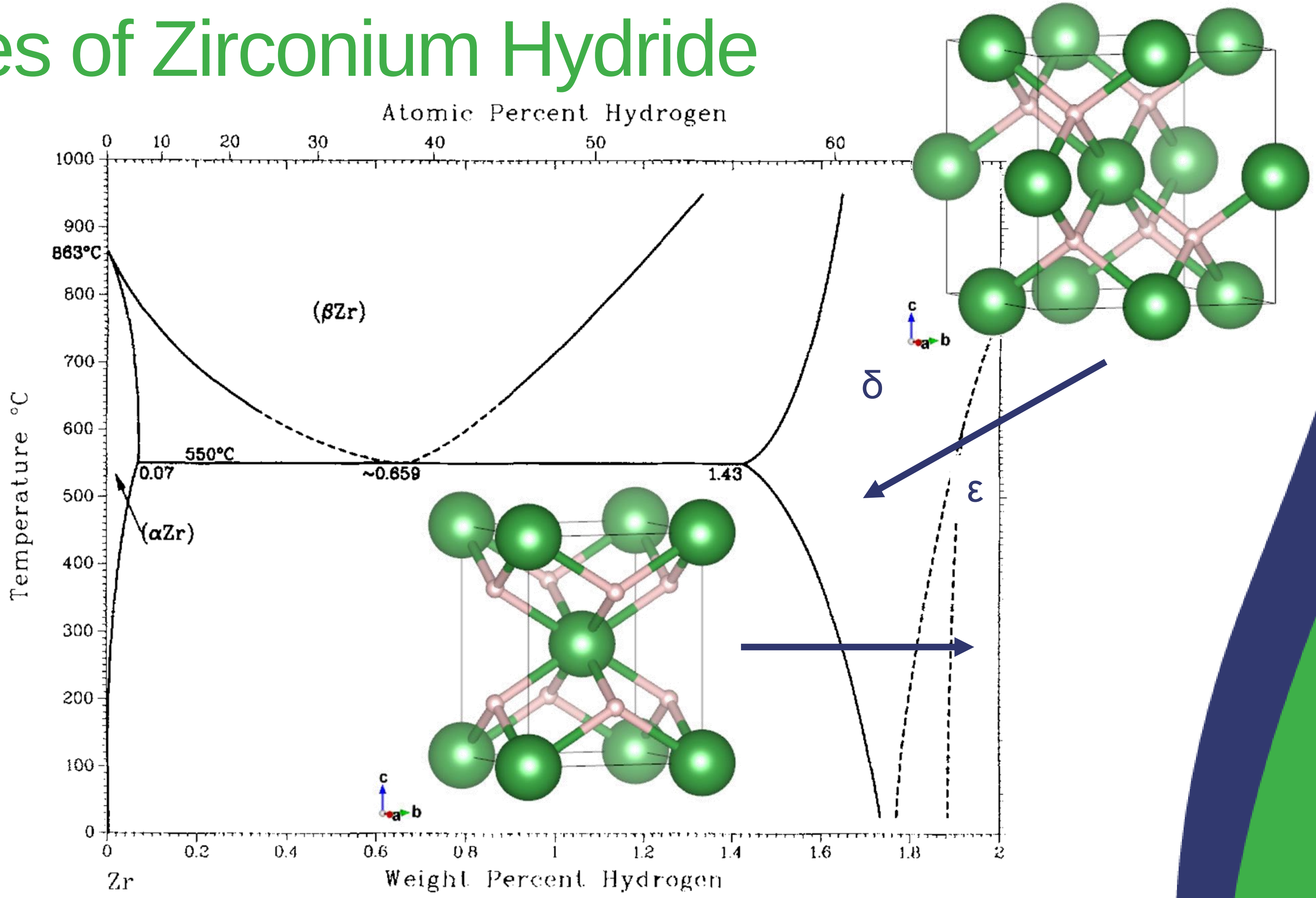
Overview

- Zirconium Hydride (ZrH_x) is a moderator material for:
  - TRIGA reactors
  - SNAP-10A
- The thermal scattering has the behavior of quantum oscillator favorable for moderator feedback
- Occurs as a corrosion product in LWR

- Previously new phase specific TSL evaluations were generated and submitted to ENDF/B-VIII.1
  - Include the effects of crystal structure on elastic scattering of Zr(ZrH_x)
    - ENDF/B-VIII.0 ZrH TSLs neglects crystal structure in elastic scattering
  - Permit flexibility in stoichiometry for use in reactor physics calculations and fuel characterization
  - Expands the TSL material sub-library
    - ENDF/B-VIII.0 ZrH TSLs do not distinguish between material phase
- Current work is preliminary validation & verification testing on the effect of TSLs on critical mass and associated thermal flux

Phases of Zirconium Hydride

- Two phases of concern for reactor applications:
  - δ-phase 1.56 < x < 1.64
    - Face-centered cubic, fluorite prototype crystal
  - ε-phase x > 1.74
    - Body-centered tetragonal, distorted fluorite prototype crystal
- TRIGA reactor use δ-phase
  - Also primary corrosion product
- SNAP-10A used ε-phase



Zuzek, E., Abriata, J.P., San-Martin, A. *et al.* The H-Zr (hydrogen-zirconium) system. *Bulletin of Alloy Phase Diagrams* **11,** 385–395 (1990). https://doi.org/10.1007/BF02843318

Neutron Scattering

- Double differential neutron scattering cross section

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{1}{4\pi k_B T}\sqrt{\frac{E'}{E}}\left[\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)\right]$$

$$S(\alpha,\beta) = S_d(\alpha,\beta) + S_s(\alpha,\beta) \qquad \alpha = \frac{E + E' - \mu\sqrt{EE'}}{Ak_BT}$$

$$S(\alpha,\beta) = \sum_p S^p(\alpha,\beta) \qquad \beta = \frac{E' - E}{k_BT}$$

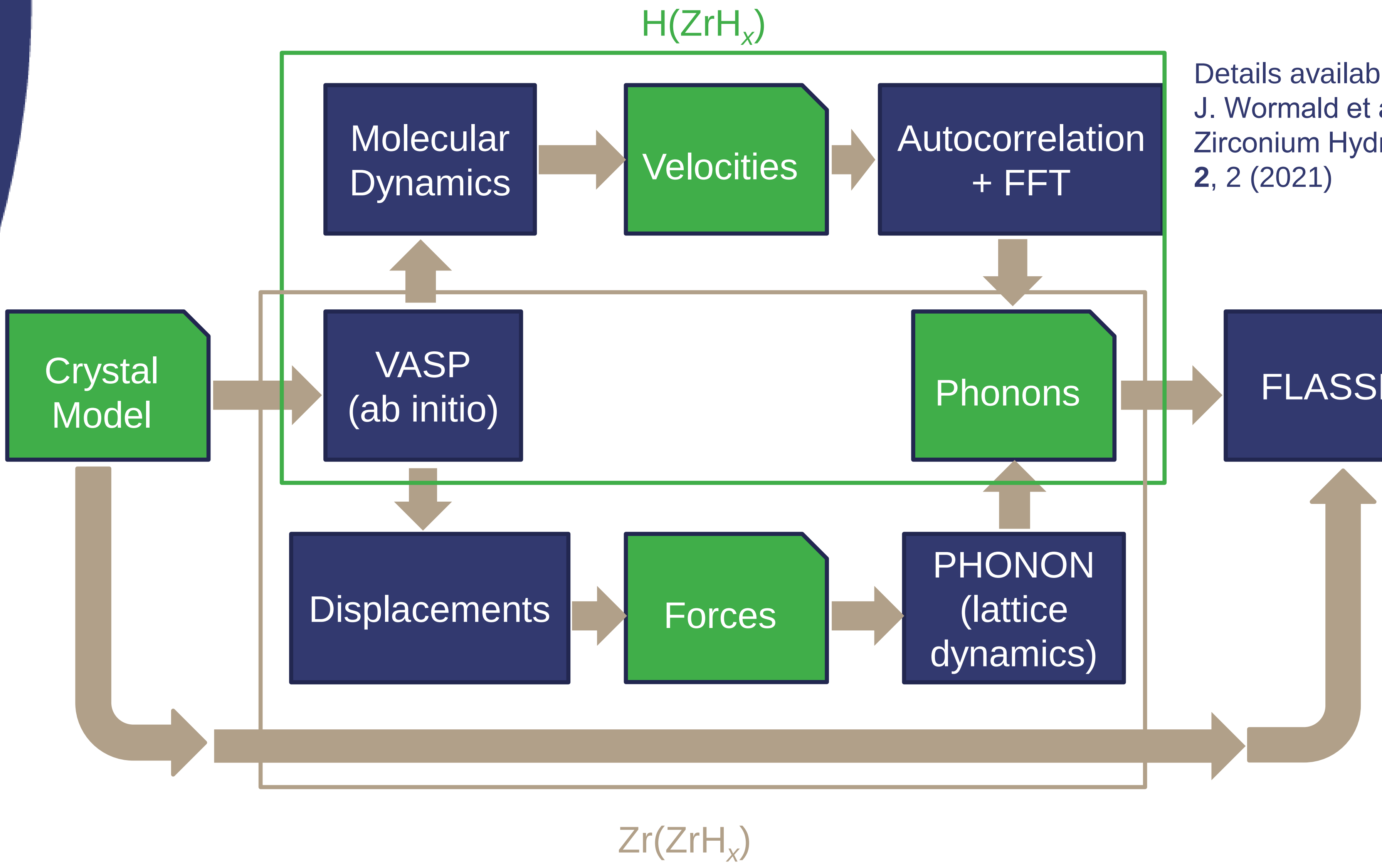
- Phonon Expansion in incoherent approximation
  - Zirconium has no incoherent nuclear potential
  - Crystal effect of hydrogen neglected for stoichiometric flexibility

$H(ZrH_x)$

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{1}{4\pi k_B T}\sqrt{\frac{E'}{E}}\left[\sigma_{coh}S^0(\alpha,\beta) + \sigma_{inc}S_s^0(\alpha,\beta) + (\sigma_{coh} + \sigma_{inc})\sum_{p>0} S_s^p(\alpha,\beta)\right]$$

$Zr(ZrH_x)$

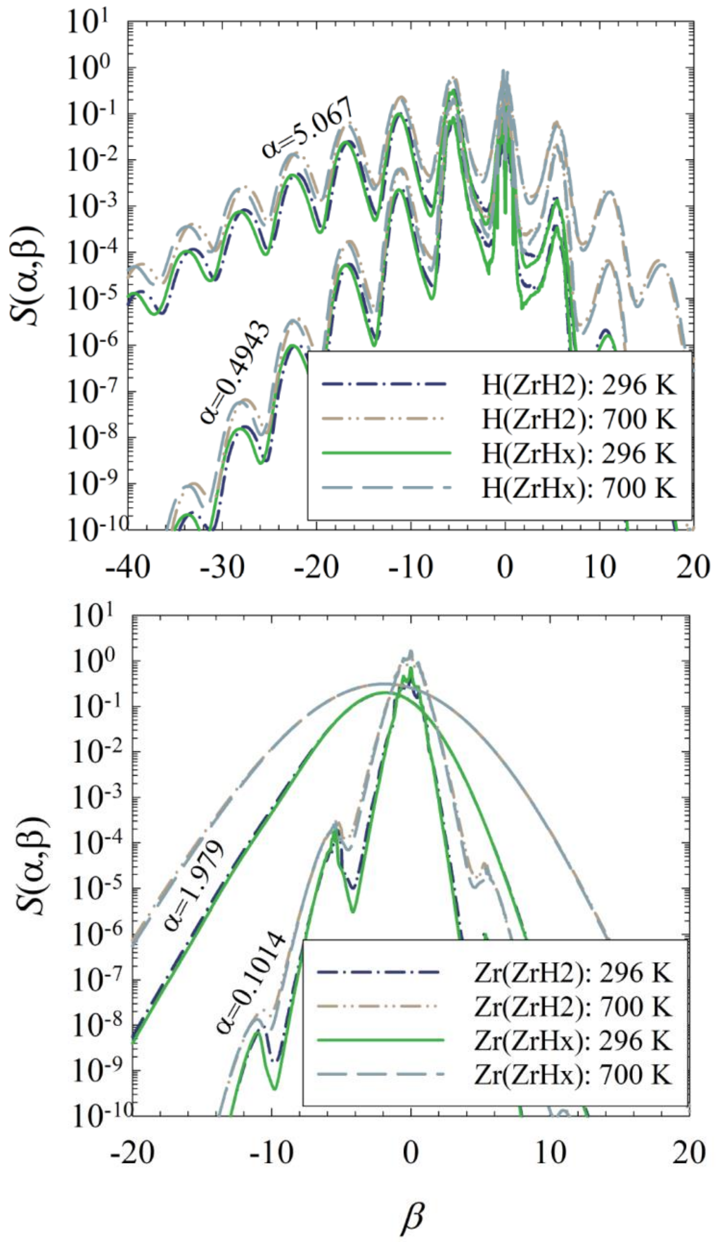
TSL Evaluation Methods



Details available in:
J. Wormald et al., “Generation of the TSL for Zirconium Hydrides from Ab Initio Methods,” *JNE* **2**, 2 (2021)

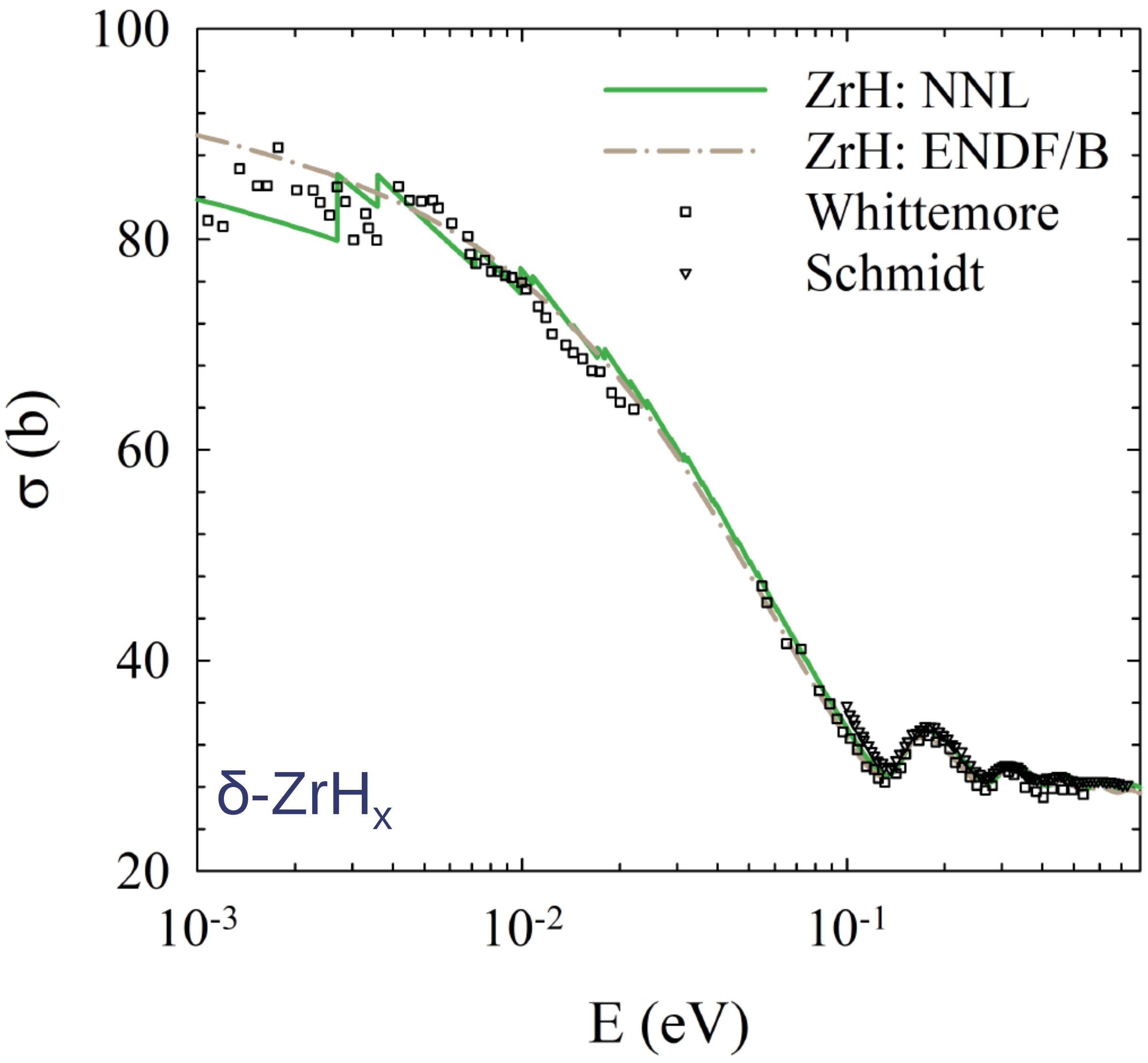
TSL for Different Phases

- TSLs for δ-phase and ε-phase are compared
- δ-H(ZrH_x) and ε-H(ZrH_2) both have strong quantum oscillator effect
  - Oscillations exist for more than 20 phonon orders
  - Acoustic phonons have significant contribution
- δ-phase diverges from ε-phase with increasing phonon order
  - Higher energy phonon spectra in δ-phase relative to ε-phase
- δ-Zr(ZrH_x) and ε-Zr(ZrH_2) are similar



Scattering Cross Section Validation

- Total Scattering Cross Section generated from TSL using NDEX
  - Adaptive energy mesh based on tabulated β-mesh captures oscillations to all tabulated phonon orders**
- Low energy transmission measurements for δ-phase
  - Low energy phonons cause room temperature differences in σ for ε-phase
- Coherent elastic cross section improve agreement with experiment compared to ENDF/B-VIII.0
- Consistent agreement in oscillations between current evaluation and ENDF/B-VIII.0



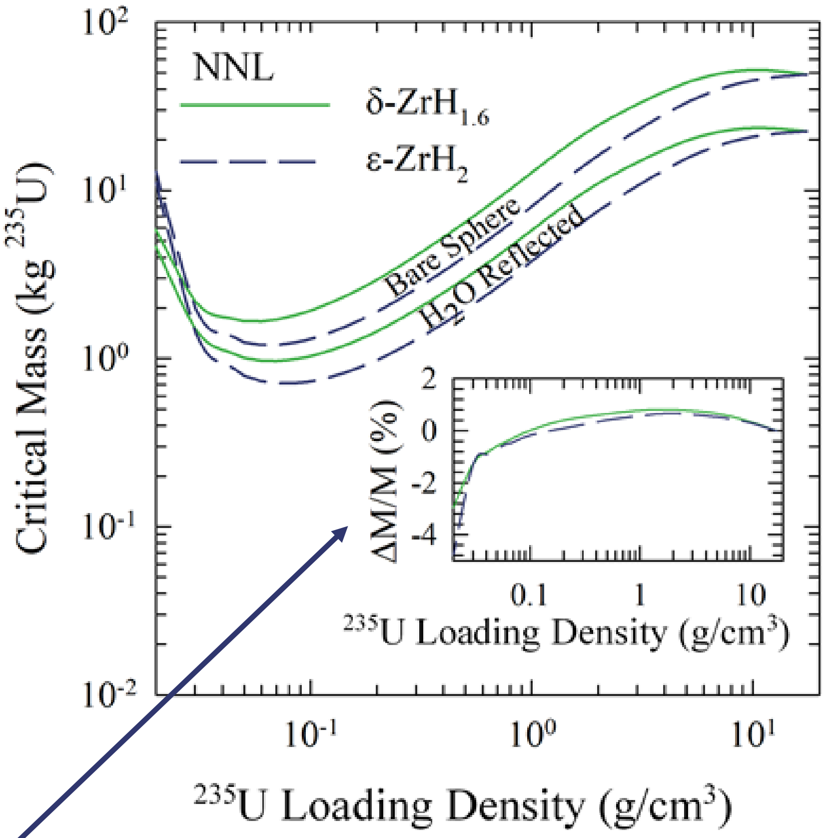
J.L. Wormald et al., *Annals of Nuclear Energy* **149 (2020) 107773

MC21 Critical Mass Calculations

- Critical mass calculated with MC21 for homogeneous ZrH_x and HEU for bare sphere and H_2O reflected
  - ENDF/B-VIII.0 nuclide and $H(H_2O)$ TSL evaluations; processed with NDEX
  - 20 cm thick H_2O reflector with 0.9981 g/cm^3 density @ 1 atm/293.6 K
  - Standard HEU composition with uranium density of 18.82342 g/cm^3
- Critical radius search performed with a 0.001 Δk convergence criterion

Critical Mass Comparison

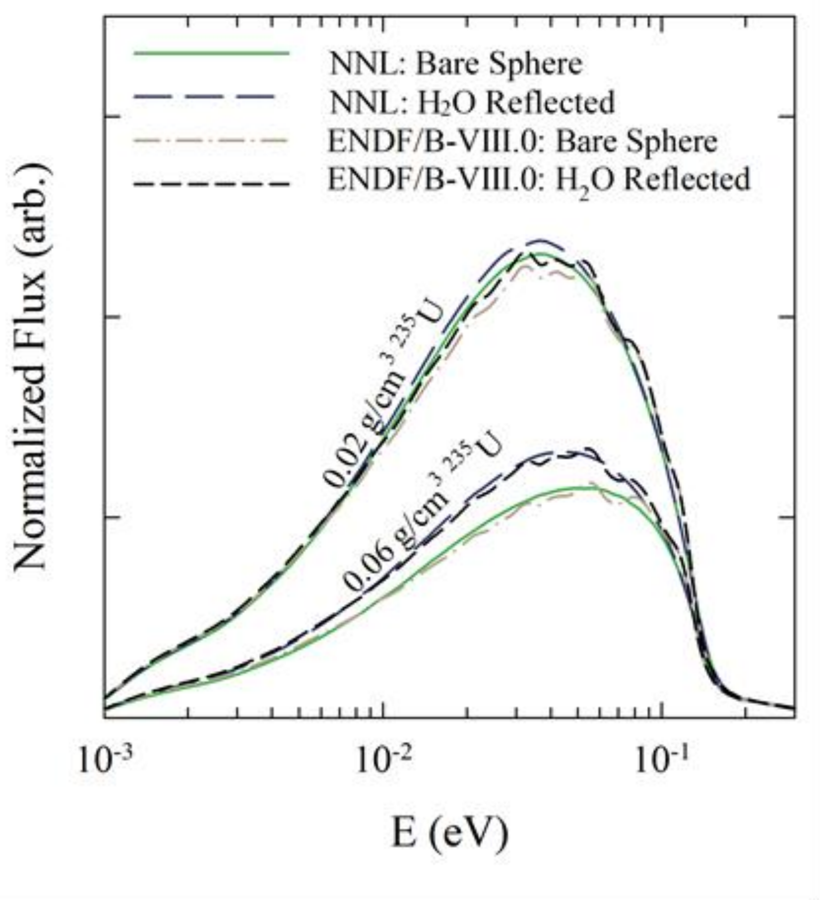
- Critical mass difference in ε-ZrH_2 and δ-$ZrH_{1.6}$ due to the higher hydrogen content
  - H_2O decreased minimum critical mass by 1.5× – 1.7×
- $\Delta M/M$ is approximately the same for H_2O and bare spheres for both ZrH_x phases
- In over-moderated region (<0.04 g/cm^3) difference between NNL and ENDF/B-VIII.0 critical mass exceeds 1% – 5%
  - Driver of changes to critical mass and flux are attributed to differences in hydrogen binding
- Impact of NNL TSL on critical mass for 0.1 – 1 g/cm^3 ^{235}U is less than 1%
  - HEU and high assay LEU (HALEU) TRIGA fuel systems (e.g., in ICSBEP) are in this range
- Expect minimal impact of NNL TSL on predictions of criticality of TRIGA fuel systems



$$\frac{\Delta M}{M} = \frac{M_{NNL} - M_{VIII}}{M_{VIII}}$$

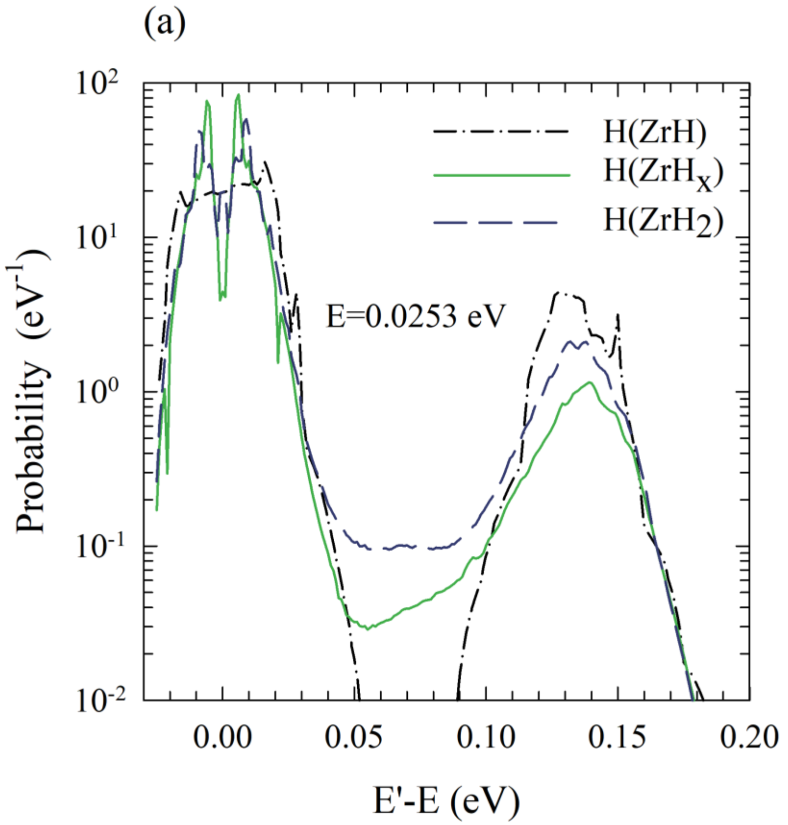
Flux Comparison

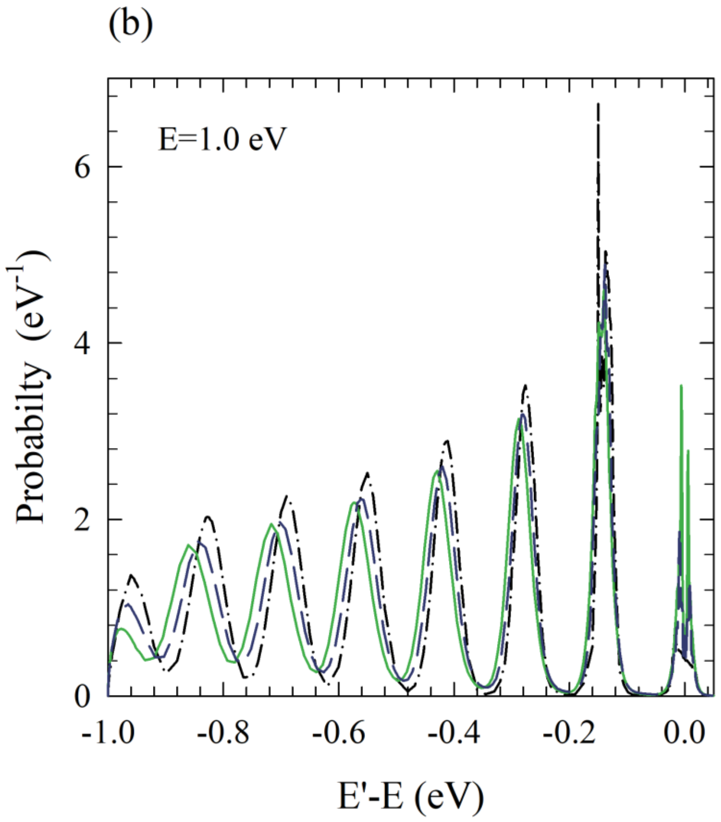
- Thermal flux is compared for δ-$ZrH_{1.6}$ using NNL and ENDF/B-VIII.0 TSLs
  - Flux distributions for ε-ZrH_2 similar to δ-ZrH_x
- Flux distributions for each evaluation version similar at 0.06 g/cm^3 where $\frac{\Delta M}{M}$ similar
  - H_2O reflection increased moderation/flux
- At 0.02 g/cm^3, flux for H_2O reflected and bare sphere begin to converge
- ENDF/B-VIII.0 flux is hardened relative to NNL and contains structure
  - Source of structure yet to be determined
    - May be artifact of TSL evaluation methods



Criticality and Secondary Spectrum

- Differences in NNL and ENDF/B-VIII.0 secondary distributions drive differences in critical mass (especially at low ^{235}U loading density)
  - NNL has reduced probability of high energy upscattering (~0.15 eV)
    - Low energy transfer increased for NNL both in both thermal and epithermal range
    - Effect more pronounced for δ-ZrH_x than ε-ZrH_x
  - Oscillations in NNL secondary distribution are broadened (i.e., anharmonicity) and offset from ENDF/B-VIII.0
    - AIMD captures anharmonicities, suppressing depth of valleys





Conclusions

- Critical mass calculations used for initial validation & verification testing of the NNL ZrH_x and ZrH_2 TSL evaluations compared to the ENDF/B-VIII.0 ZrH TSL evaluations.
- Small differences in the critical mass for homogenous mixtures of HEU and ZrH_x for typical range of ^{235}U loading densities indicate NNL evaluations are likely to have minimal impact on k_{eff} for most ZrH_x moderated systems.
- Changes in Hydrogen crystal binding in ZrH_x is the primary driver of differences in predicted criticality rather than inclusion of coherent elastic scattering in $Zr(ZrH_x)$ and $Zr(ZrH_2)$
- Further testing is in progress including models available in the ICSBEP Handbook.