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Development of an ENDF Thermal Library for SiO_2 and Testing of Criticality Effects

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Objectives

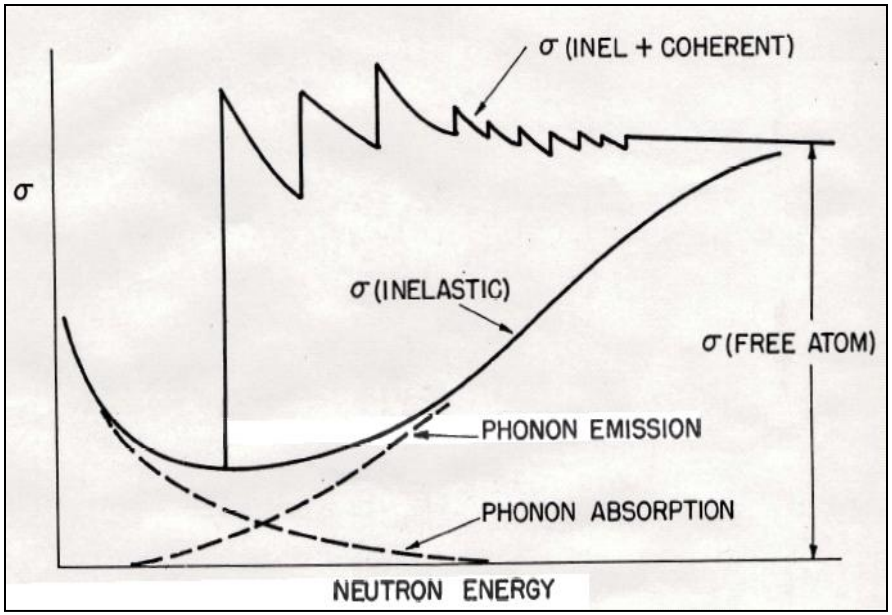
- Generate the partial density of states (DOS) for silicon and oxygen in the α-quartz and β-quartz phases of SiO_2 using an *ab initio* approach
- Utilize the DOS to produce an ENDF-6 format thermal neutron scattering library for SiO_2 with inelastic and elastic cross sections over a standard temperature grid for NNDC publication
- Investigate the sensitivity of k_{eff} of an appropriate ICSBEP benchmark to the use of newly generated thermal SiO_2 libraries versus free-gas libraries with MCNP

Outline

- Introduce the concept of thermal neutron scattering and exhibit the modifications that will be made to a standard free atom library
- Discuss the calculation of inelastic and coherent scattering elastic cross sections as a function of the phonon density of states (DOS)
- Demonstrate the ability to produce a phonon density of states using a first-principles approach with only crystal structure input
- Use an ICSBEP benchmark to evaluate the impact of the new thermal SiO_2 library on k_{eff} compared to the use of traditional free gas libraries
- Explain how this effect results directly from the introduction of a crystal structure with discrete vibrational modes

Thermal Neutron Scattering and Energy Transfer

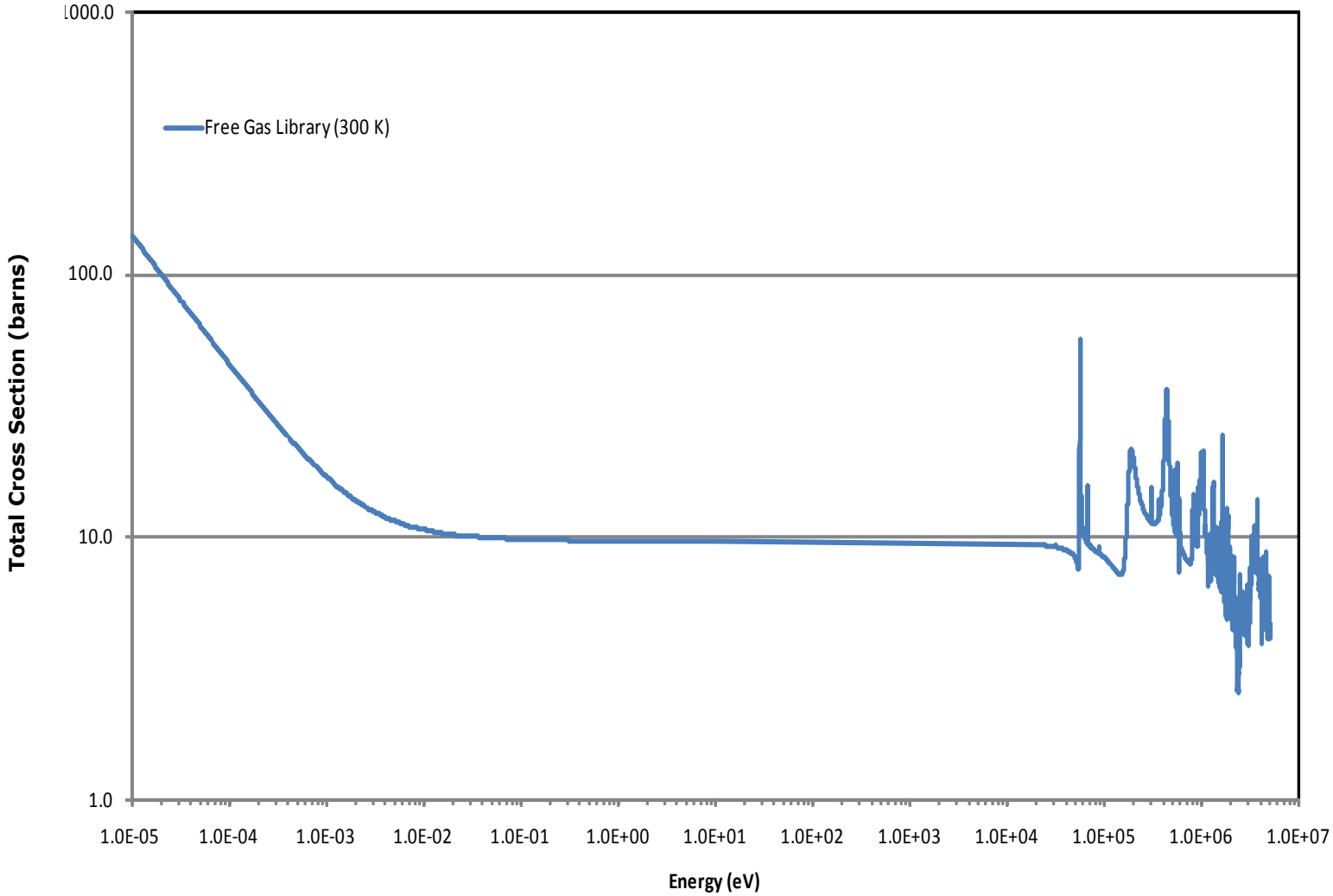
- The de Broglie wavelength ($\lambda = h / p$) of thermal neutrons (< ~ 1 eV) is on the order of the interatomic distances in crystalline solids.
- The energy of thermal neutrons is of the same order as the vibrational excitation modes (e.g., phonons) available in condensed matter.



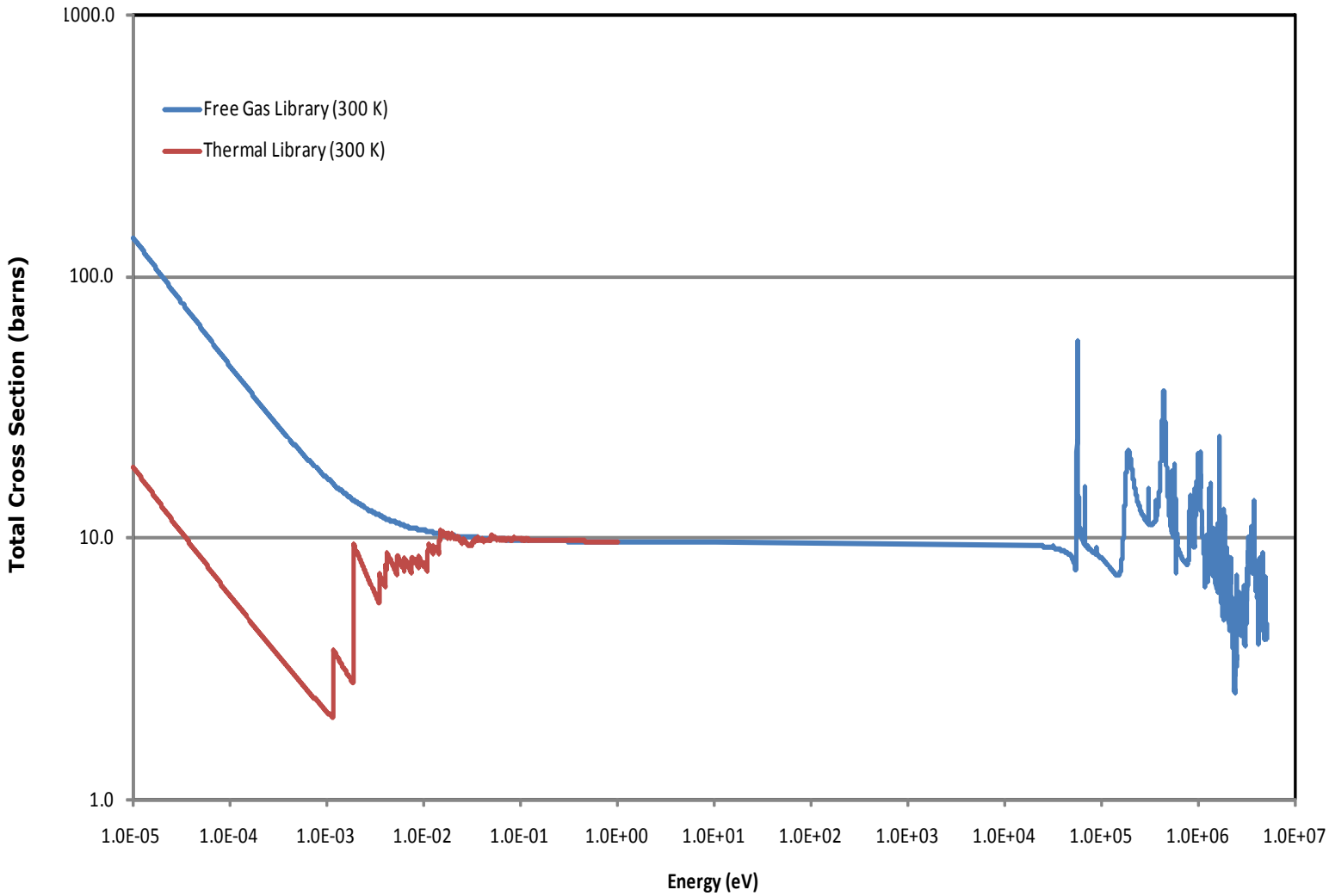
$$\sigma_{TOTAL} =$$

$$\sigma_{INEL} + \sigma_{EL} + \sigma_{ABS}$$

Modifying the Free-Atom Cross Section for SiO_2



Modifying the Free-Atom Cross Section for SiO_2



Calculating the Inelastic Scattering Cross Section

Double-differential cross section can be written in terms of nuclear cross sections and a scattering law:

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi}(k'/k)(\sigma_{coh}S(\boldsymbol{Q},\omega) + \sigma_{incoh}S_s(\boldsymbol{Q},\omega))$$

$$S(Q,\omega) = S_s(\boldsymbol{Q},\omega) + S_d(\boldsymbol{Q},\omega)$$

Simplified NJOY form with $S_d(\boldsymbol{Q},\omega) = 0$:

$$\frac{d^2\sigma_{inel}}{d\Omega dE} = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}}\, e^{-\frac{\beta}{2}} S_s(\alpha,\beta)$$

$$S_s(\alpha,\beta) = \frac{k_B T}{2\pi\hbar}\int_{-\infty}^{\infty} e^{i\frac{k_B T}{\hbar}\beta\hat{t}}\, e^{-\gamma(\hat{t})}\, d\hat{t}$$

$$\gamma(\hat{t}) = \alpha\int_{-\infty}^{\infty}\frac{\rho(\beta)\left[1-e^{-i\frac{k_B T}{\hbar}\beta\hat{t}}\right]e^{-\frac{\beta}{2}}}{2\beta\sinh\left(\frac{\beta}{2}\right)}\, d\beta$$

All that is required to solve for σ_{inel} is $\rho(\beta)$

Calculating the Elastic Scattering Cross Section

$$\frac{d\sigma_{el}}{d\Omega} = \frac{\sigma_{coh}}{E} \sum_{i,\,(E_i<E)} f_i e^{-4WE_i} \delta(\cos\Omega - \mu_i)$$

$$\lambda_j = \int_{-\infty}^{\infty} \frac{\rho_j(\beta) e^{-\frac{\beta}{2}}}{2\beta \sinh\left(\frac{\beta}{2}\right)} d\beta$$

$$W = \frac{1}{3}\left(\frac{\lambda_{Si}}{A_{Si} k_B T}\right) + \frac{2}{3}\left(\frac{\lambda_O}{A_O k_B T}\right)$$

$$f_i \approx \frac{\pi\hbar^2}{m\sqrt{3}ac} \left| \sum_{k=1}^{N} \sqrt{\frac{\sigma_k}{\sigma_{coh}}} e^{-2\pi\phi_k} \right|^2$$

All that is required to solve for σ_{el} is $\rho(\beta)$ and f_i

Ab Initio Calculations

The Vienna *Ab Initio* Simulation Package (VASP) uses DFT to calculate Hellmann-Feynman forces within a given crystal structure by perturbing the system

VASP parameters:

3x3x2 α-quartz supercell (162 atoms) and 2x2x2 β-quartz supercell (72 atoms)

650 eV plane wave cutoff

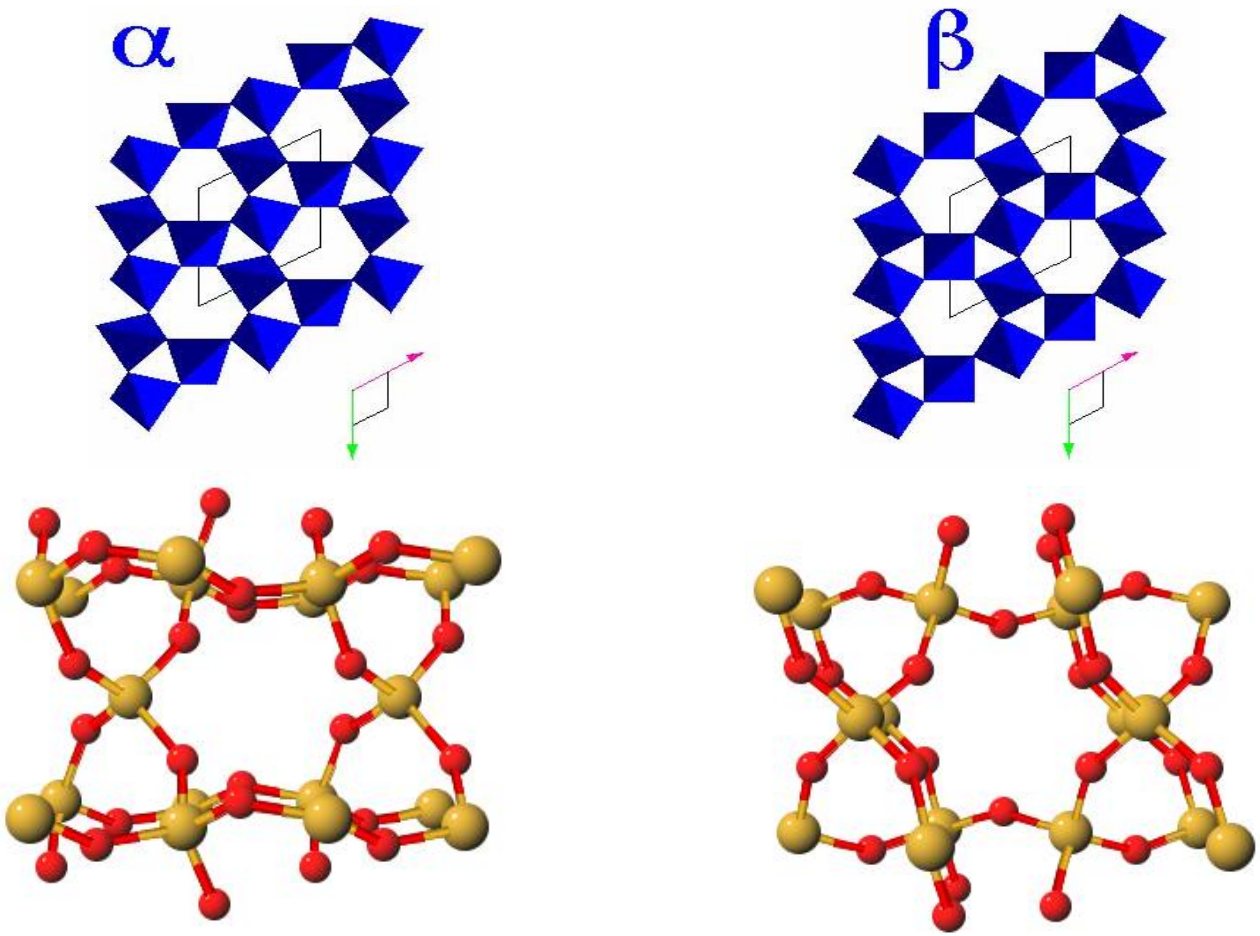
Local density approximation (LDA) used for exchange-correlation energy

PHONON uses lattice dynamics in the harmonic approximation to construct phonon frequency distributions by Monte Carlo sampling of 8 X 10^5 wave vectors in the first Brillouin zone

Structure Parameters for α-Quartz and β-Quartz

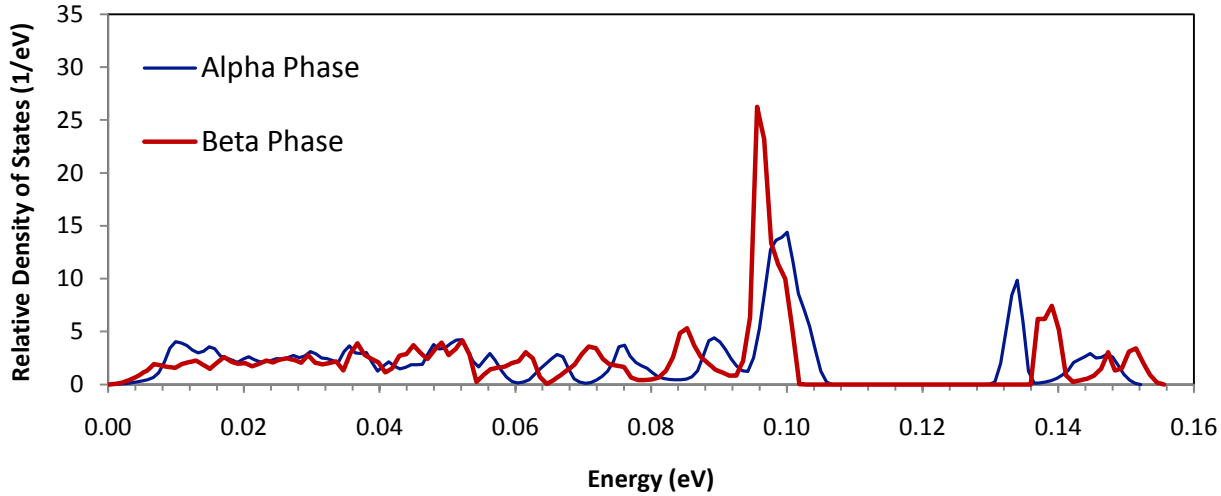
	α-Quartz	β-Quartz
Crystal System	Trigonal	Hexagonal
Lattice System	Hexagonal	Hexagonal
# of Atoms in Unit Cell	9	9
Lattice Parameter a (Å)	4.913	4.997
Lattice Parameter b (Å)	4.913	4.997
Lattice Parameter c (Å)	5.405	5.455
Si-O-Si bond angle (deg.)	144	153
Specific Gravity	2.65	2.53
Stable Temperature (1 atm)	< 846 K	846 K to 1143 K

Structure of α-Quartz and β-Quartz

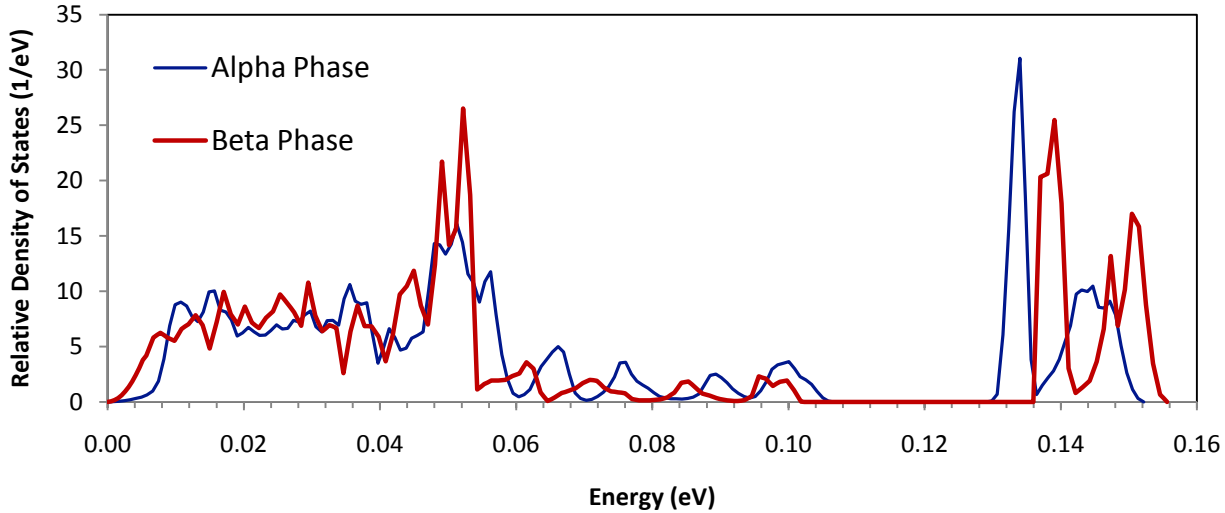


Silicons (yellow), Oxygens (red)

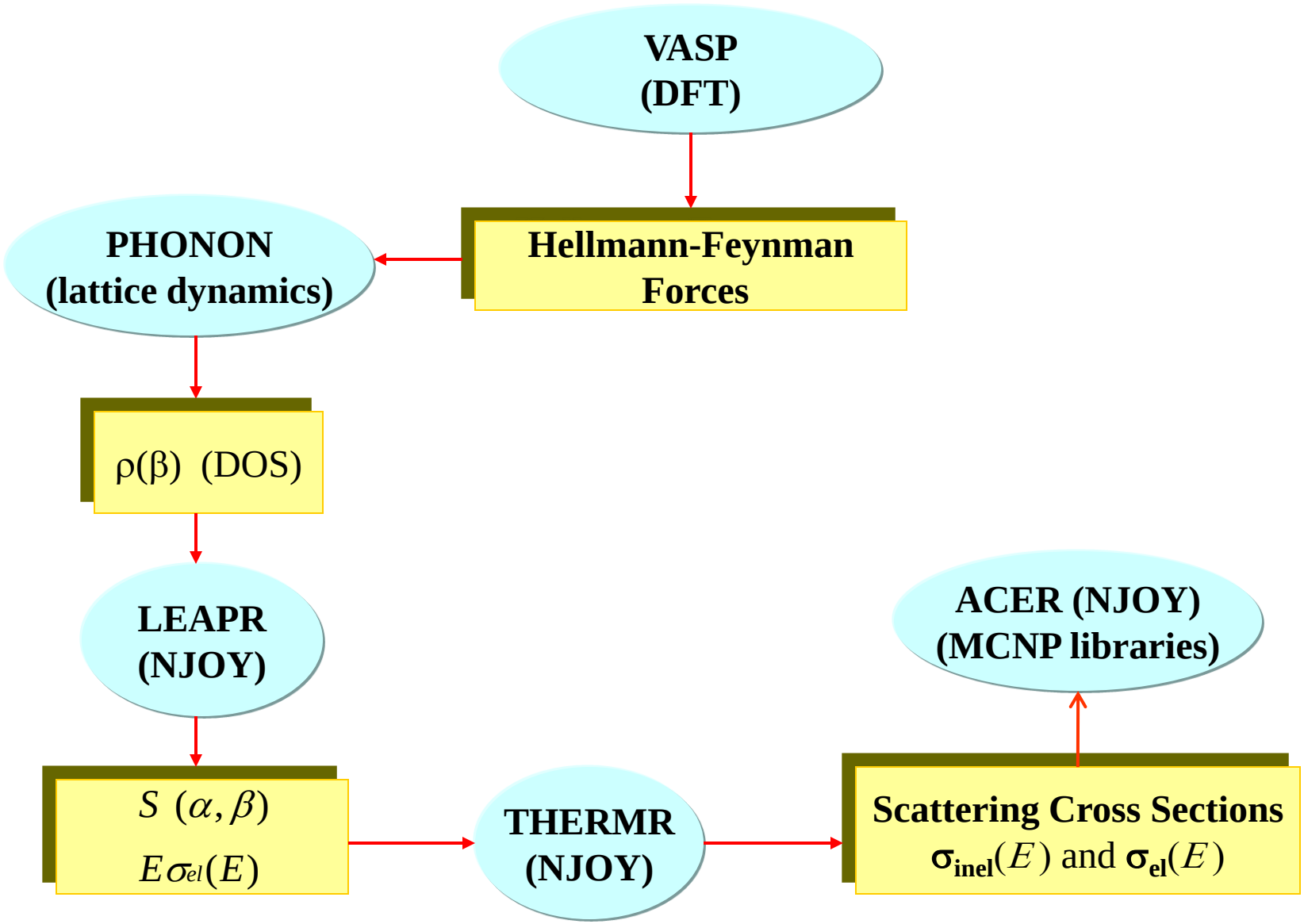
Density of States



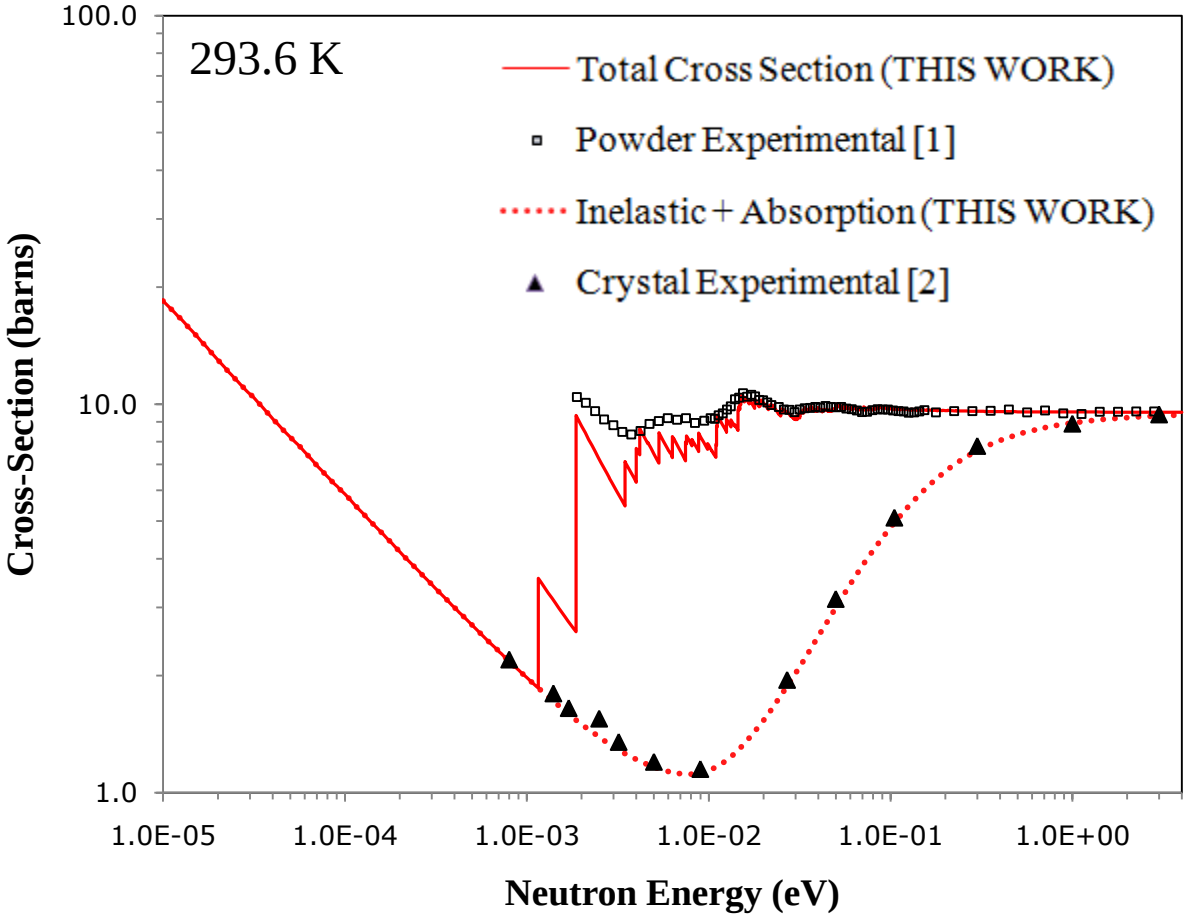
Natural Si in SiO_2



O-16 in SiO_2


VASP
(DFT)
Hellmann-Feynman
Forces
PHONON
(lattice dynamics)
ρ(β) (DOS)
LEAPR
(NJOY)
$S(\alpha,\beta)$
$E\sigma_{el}(E)$
THERMR
(NJOY)
Scattering Cross Sections
$\sigma_{inel}(E)$ and $\sigma_{el}(E)$
ACER (NJOY)
(MCNP libraries)

SiO_2 Calculated Cross Sections Compared to Experimental Values



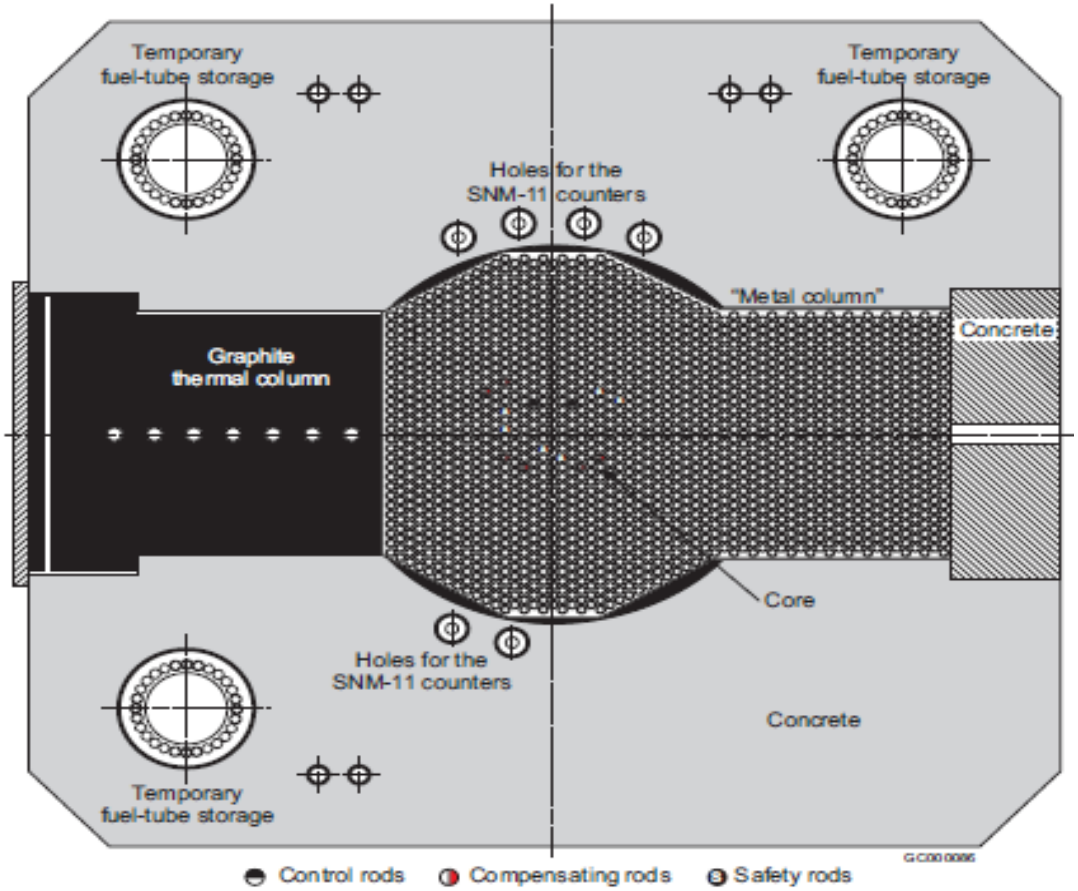
[1] J. J. BLOSTEIN and J. DAWIDOWSKI, "Experimental Neutron Data: Sigma(E) of Vitreous Silica (SiO2) Over the Thermal Energy Range," Private Communication, Inst. Balseiro y Centro Atomico Bariloche, Argentina (1999).

[2] B. M. RUSTAD, et al., "Neutron Crystal Spectrometer with Range Extended to Subthermal Energies," *Review of Scientific Instruments*, **36**, 887 (1965).

ICSBEP HEU-MET-MIXED-005 Benchmark

NEA/NSC/DOC(95)03/II
Volume II

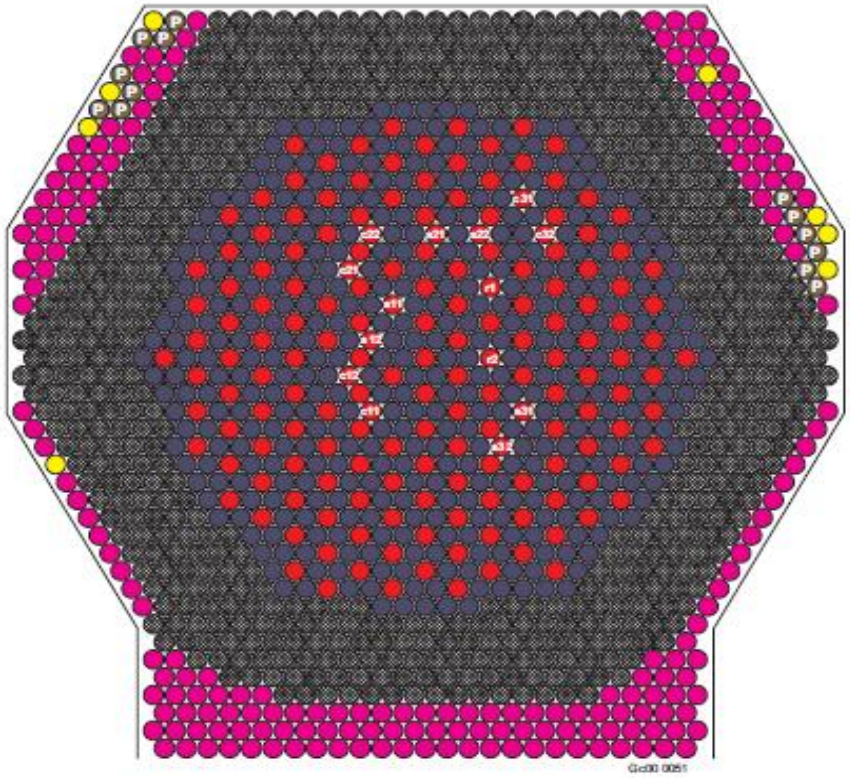
HEU-MET-MIXED-005



BFS-79/3 Configuration

NEA/NSC/DOC(95)03/II
Volume II

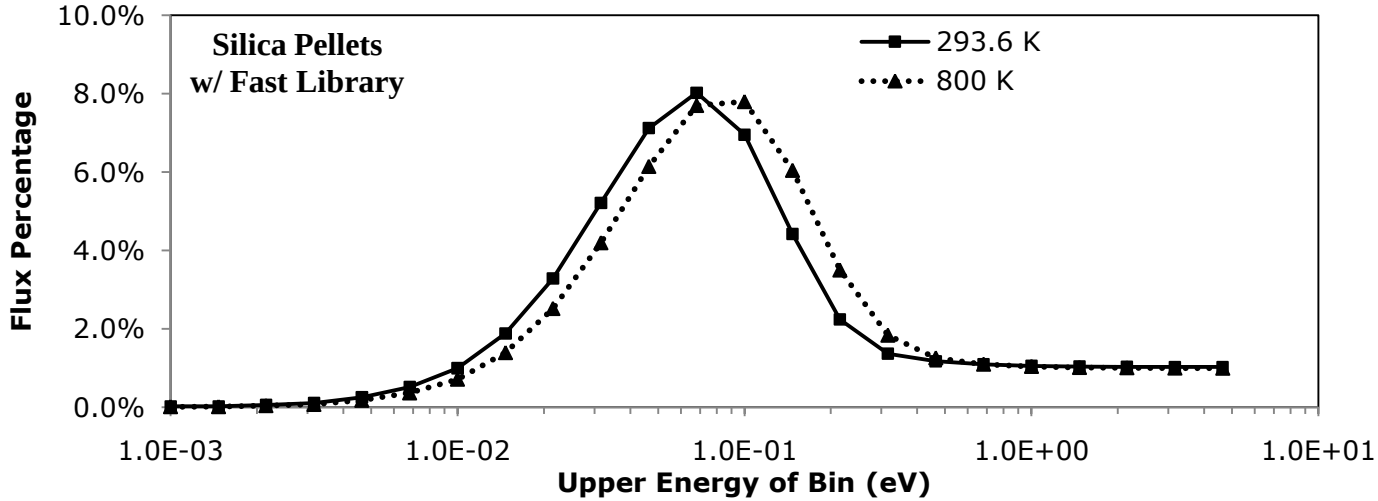
HEU-MET-MIXED-005

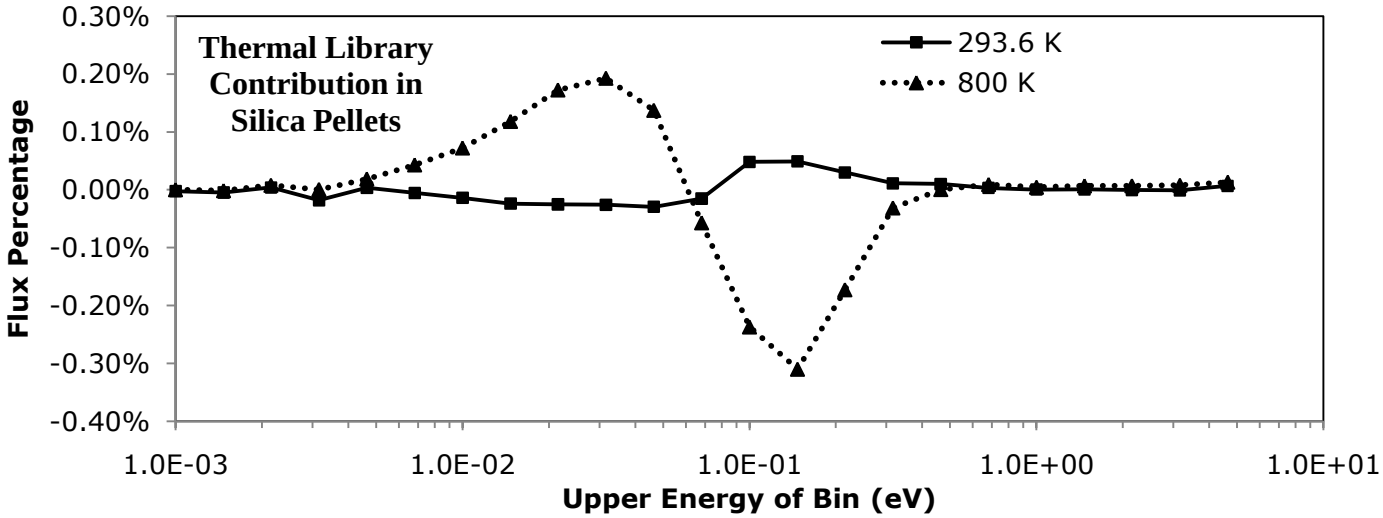


Core:
- f-type tube
  (fb-type tube in numbered control-rod positions)
- p-type tube
- polyethylene dowel

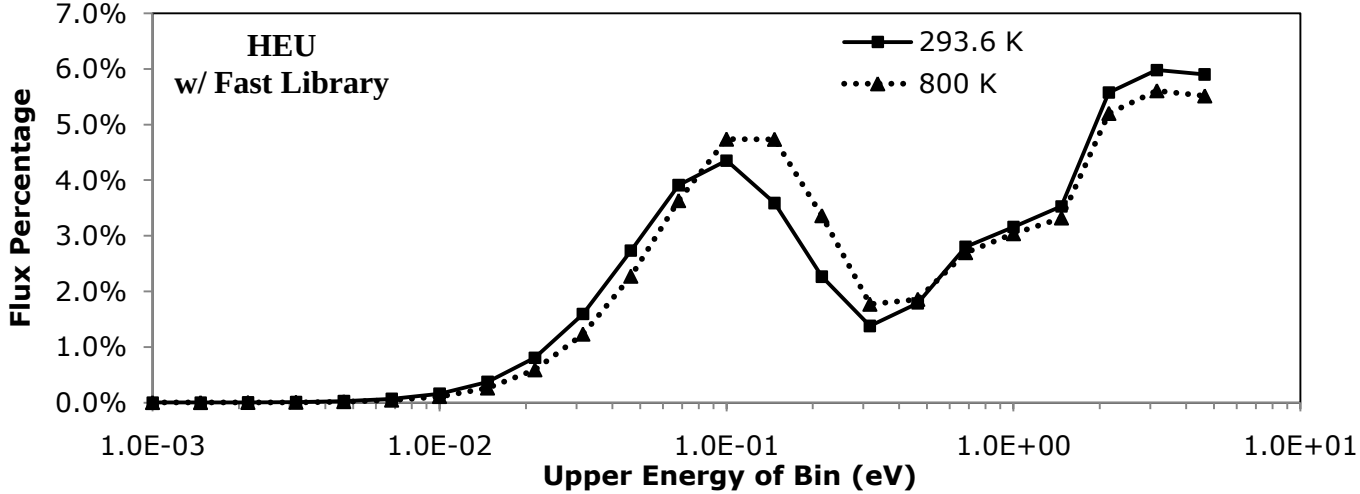
Reflector:
- sd-type tube
- u-type tube
- polyethylene cylinder
- KNK-59
- polyethylene dowel

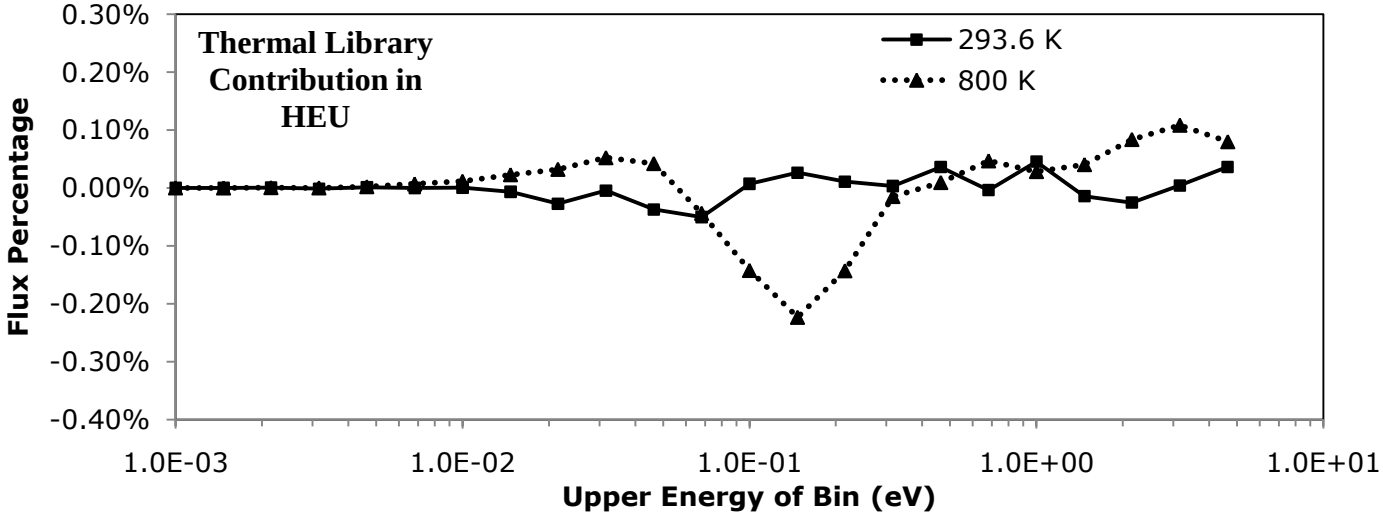
Thermal Neutron Flux in Fuel Tube Silica Pellets





Thermal Neutron Flux in Fuel Tube HEU Pellets

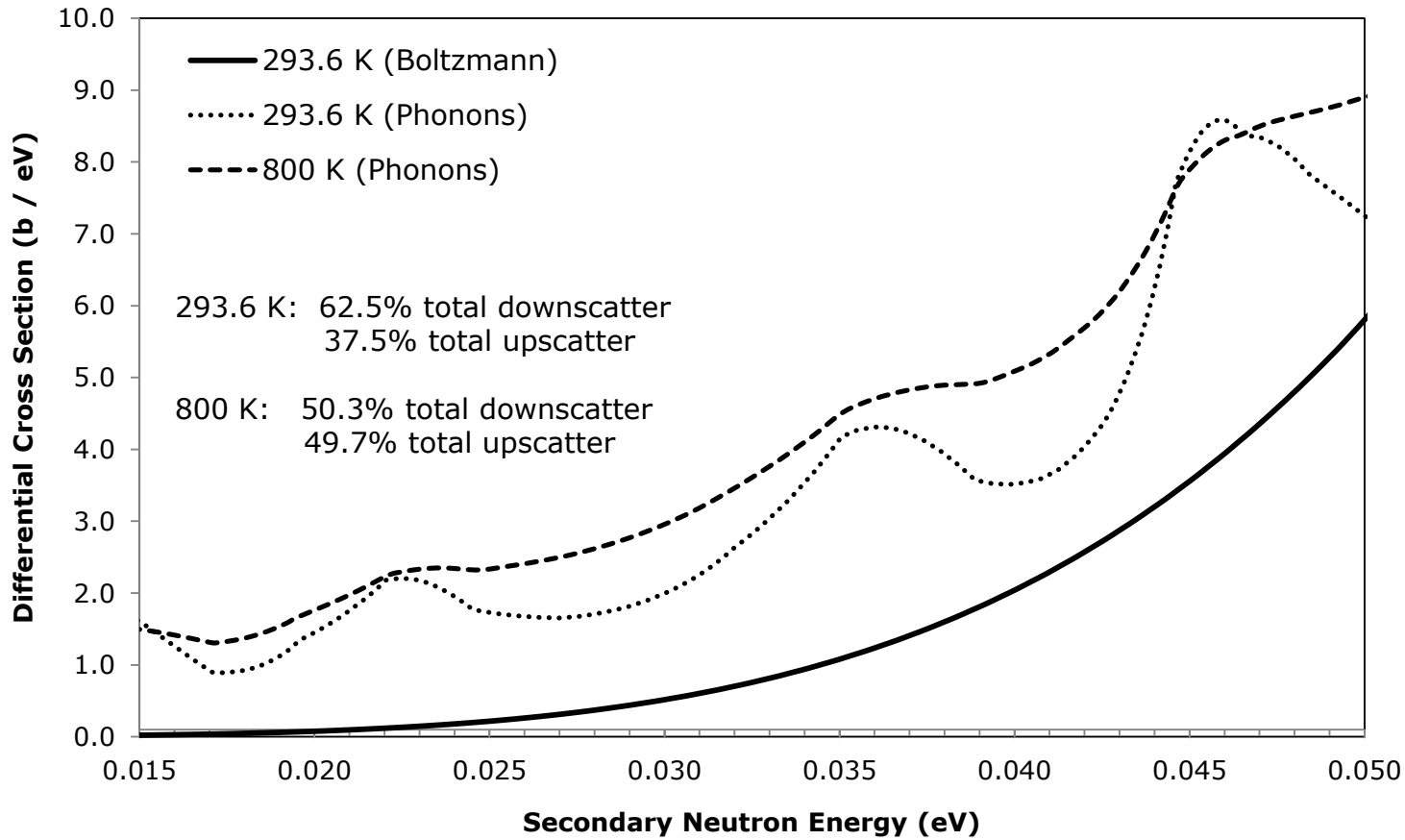




Secondary Neutron Energy Distribution Downscattering Example

E_i = 0.112 eV

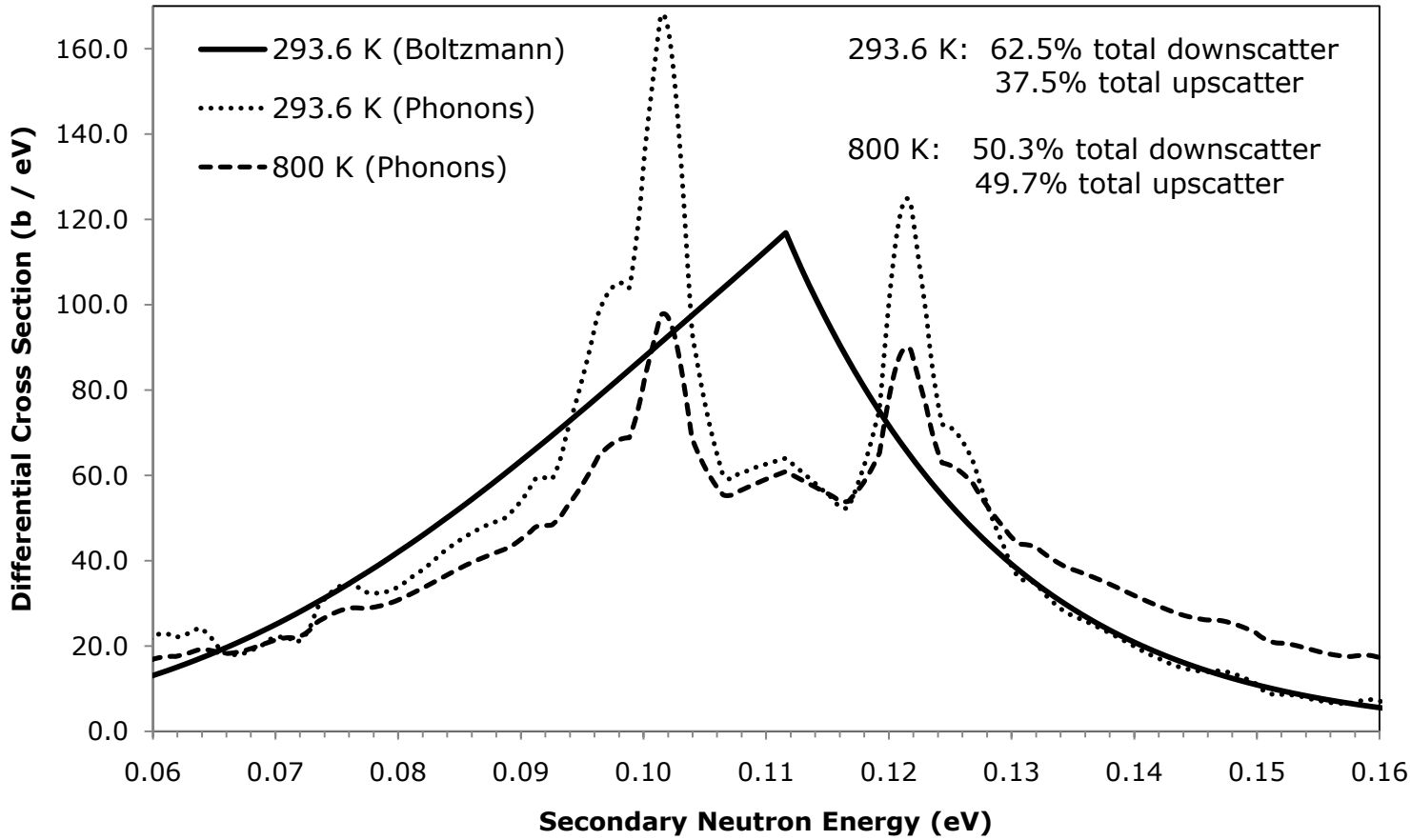
Plots are normalized to 293.6 K (Phonons) such that cross sections integrated over all energies are equal.



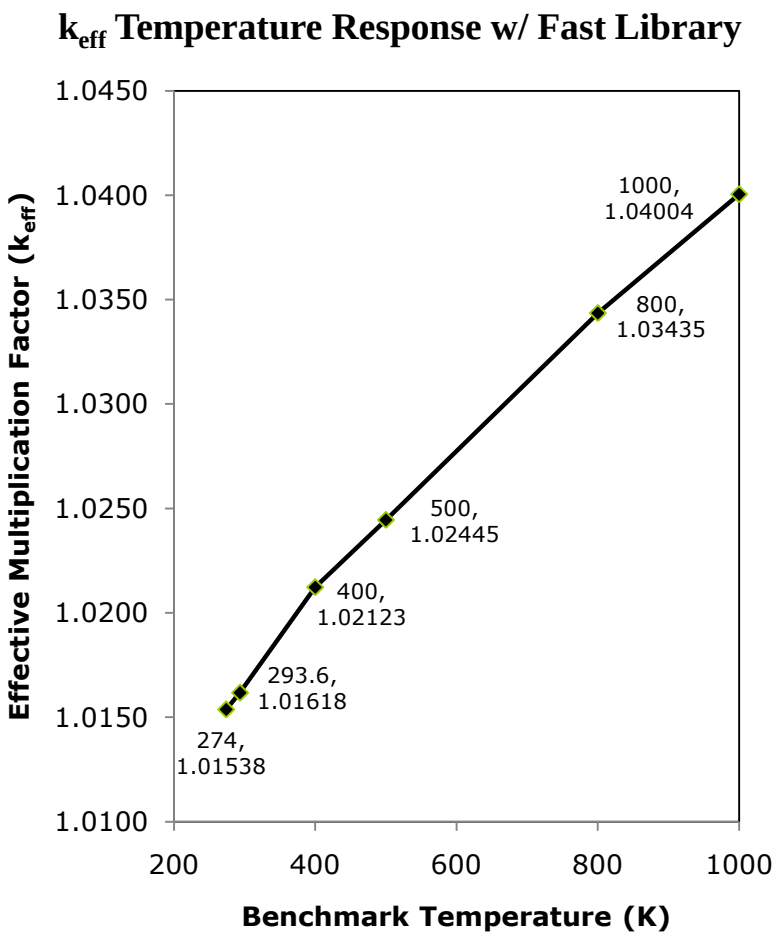
Secondary Neutron Energy Distribution Small Energy Transfer Example

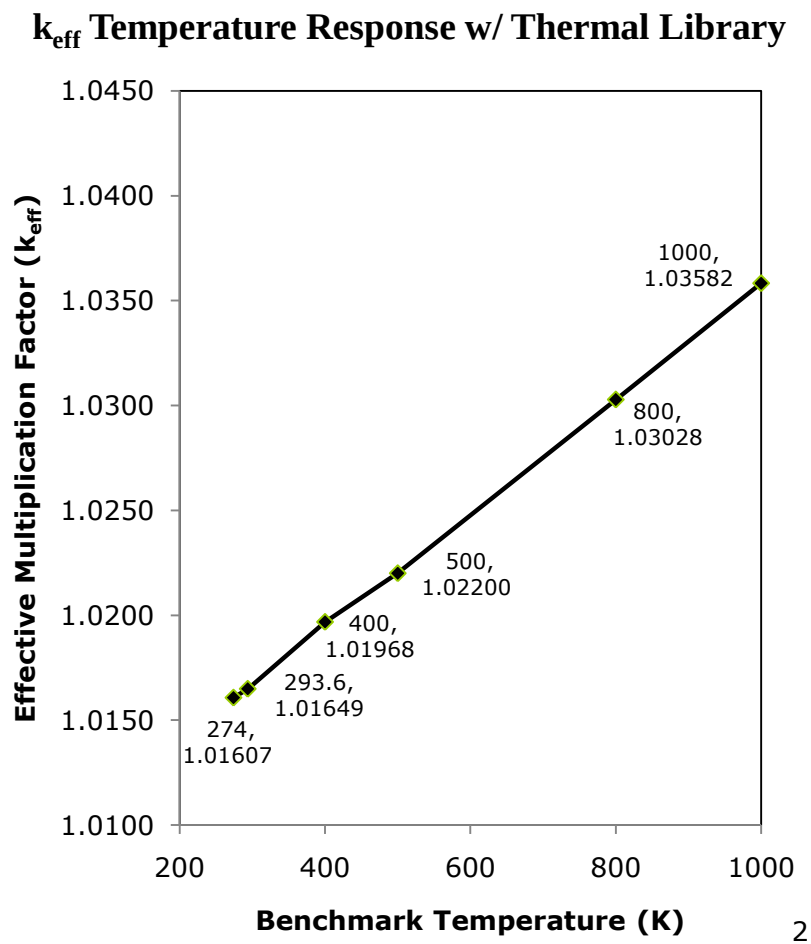
E_i = 0.112 eV

Plots are normalized to 293.6 K (Phonons) such that cross sections integrated over all energies are equal.

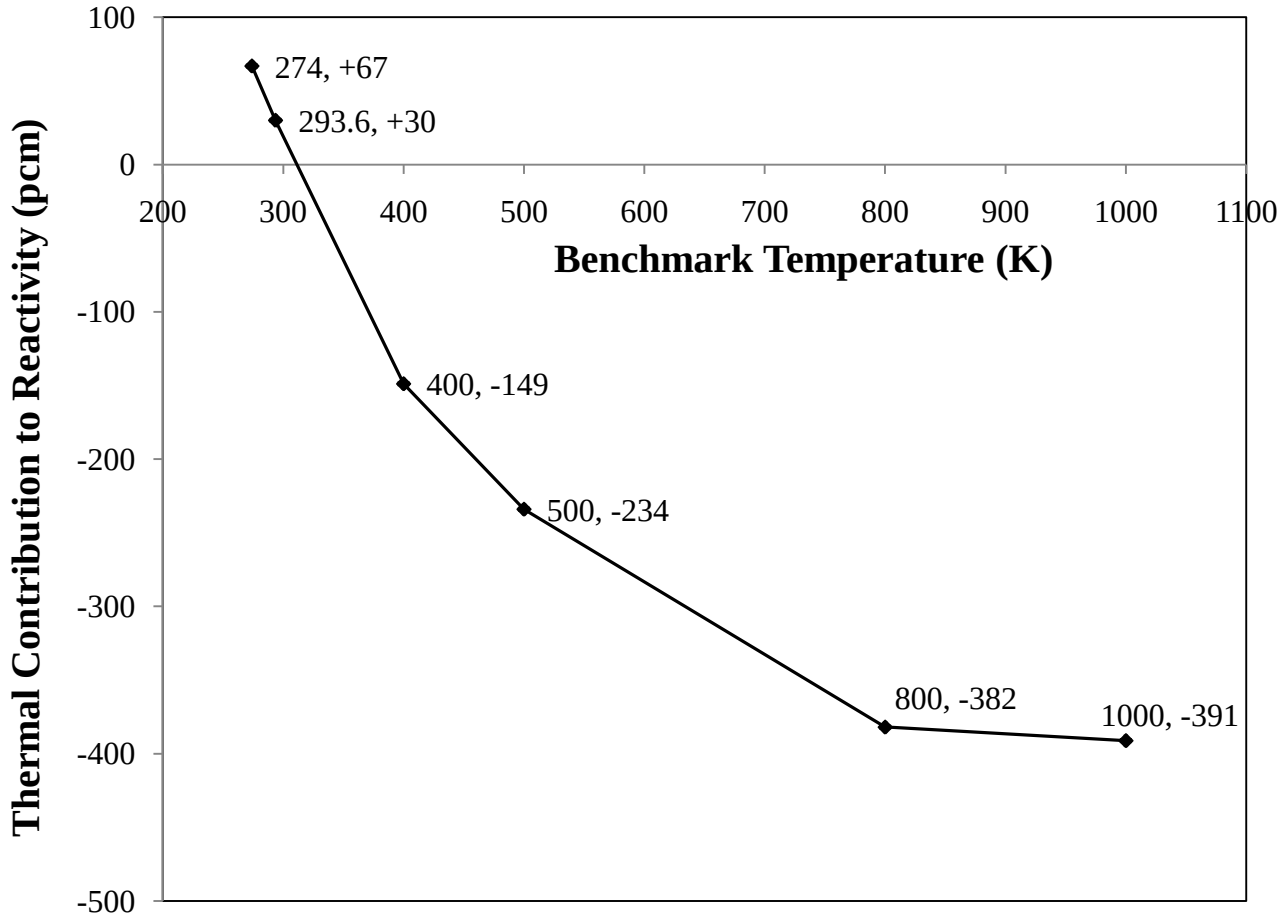


Comparison of k_{eff} Temperature Response with Fast vs. Thermal Library





Thermal Library Contribution to Reactivity (pcm)



Isotopics of Neutron Losses for Selected Fast and Thermal Libraries

	Absorptions						Fission
Temperature / Library	H	U-235	Si	U-238	Al	Other	U-235
293.6 K / Fast	16.941%	8.807%	11.108%	5.494%	5.068%	11.743%	40.839%
293.6 K / Alpha Thermal	16.962%	8.808%	11.089%	5.489%	5.063%	11.740%	40.849%
800 K / Fast	16.529%	9.115%	10.345%	5.662%	4.764%	12.006%	41.579%
800 K / Alpha Thermal	16.613%	9.092%	10.475%	5.626%	4.819%	11.955%	41.420%
1000 K / Fast	16.457%	9.126%	10.161%	5.637%	4.696%	12.100%	41.823%
1000 K / Beta Thermal	16.528%	9.094%	10.290%	5.627%	4.745%	12.066%	41.650%

Summary / Conclusions

- The phonon DOS for SiO_2 generated through a first-principles approach with the VASP / PHONON sequence can produce accurate thermal neutron scattering cross sections. The crystal structure is the only fundamental input required.
- An ENDF-6 format thermal neutron scattering library for SiO_2 was produced over a standard temperature grid (modeling α-quartz and β-quartz) that is ready for submission to NNDC.
- The impact on k_{eff} of accurate thermal scattering treatment of neutrons in SiO_2 in an ICSBEP benchmark is demonstrated.
- The use of a thermal neutron scattering library for SiO_2 may significantly impact criticality calculations for geologic systems, especially in loss of heat removal accident scenarios.