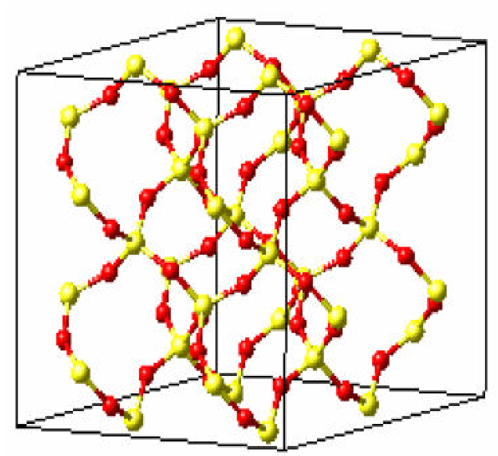
Thermal Neutron Cross-Section Evaluation and Analysis for Silicon Dioxide (Whole Molecule) with Natural Silicon

By:

Jesse C. Holmes
Iyad Al-Qasir
Brian D. Hehr

Department of Nuclear Engineering
North Carolina State University

NCSU Advisor: Ayman I. Hawari
ORNL Advisor: Luiz Leal



SiO2 (alpha quartz)

Objectives

- Produce standard ENDF libraries for the incoherent inelastic and coherent elastic thermal neutron scattering cross-sections on a standard temperature grid for silicon dioxide (alpha quartz).

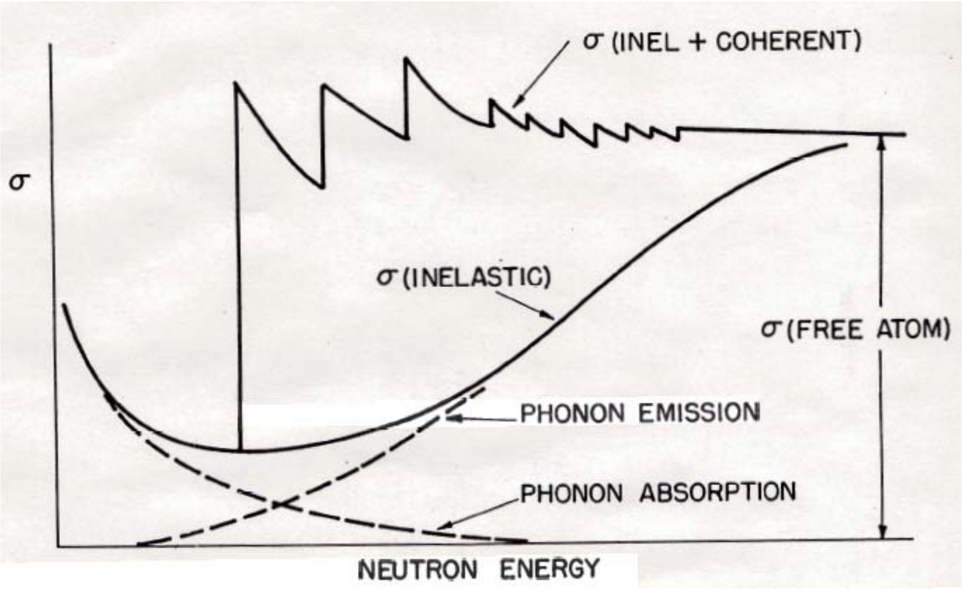
- Investigate the sensitivity of k_{eff} of a test model to the use of free-gas libraries vs. new generated thermal libraries using MCNP.

Outline

- Thermal Neutron behavior
- Incoherent Inelastic Cross-Section (*ab initio* approach)
- Coherent Elastic Cross-Section
- Cross-Section results
- Thermal Neutron Scattering impact on k_{eff}
- Conclusions

Thermal Neutron Scattering and Energy Transfer

- The wavelength of thermal neutrons is comparable to the interatomic distances in solids and liquids.
- The energy of thermal neutrons is of the same order as the excitation modes available in condensed matter (e.g., phonons in crystalline materials).



Developing the Incoherent Inelastic Cross-Section

$$\sigma(E \rightarrow E', \mu) = \frac{\sigma_b}{2kT}\sqrt{\frac{E'}{E}}\,\mathcal{S}(\alpha, \beta)$$

$$\sigma_{bound} = \sigma_{free} * \left(\frac{A+1}{A}\right)^2$$

$$\alpha = \frac{E' + E - 2\sqrt{E'E}\mu}{AkT}$$

$$\beta = \frac{E' - E}{kT}$$

$$\mathcal{S}(\alpha, \beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\beta\hat{t}}\, e^{-\gamma(\hat{t})}\, d\hat{t}$$

$$\gamma(\hat{t}) = \alpha\int_{-\infty}^{\infty} P(\beta)\left[1 - e^{-i\beta\hat{t}}\right] e^{-\beta/2}\, d\beta$$

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)}$$

$$\int_0^{\infty} \rho(\beta)\, d\beta = 1$$

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

Phonon Frequency
$\rho(\beta)$

Lattice dynamics
Force Constants Models

Early Work
-Force constant values are obtained by fitting to thermodynamical experimental data (e.g., specific heat, comperessibility,...)

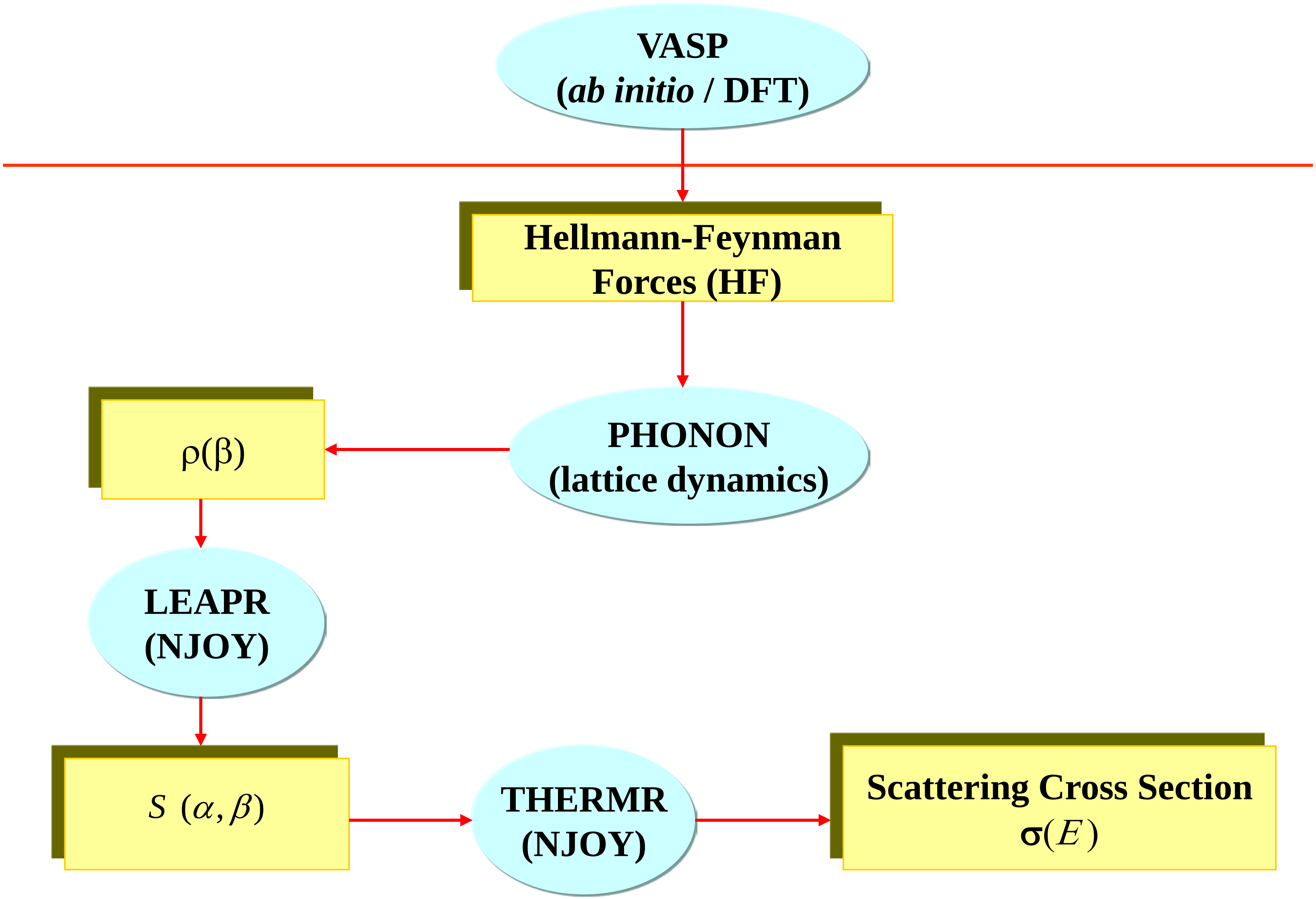
Graphite (1965)
Young-Koppel

Later
(Neutron Scattering)
force constant obtained by fitting to experimental dispersion curves along symmetry directions in the 1st Brillouin Zone

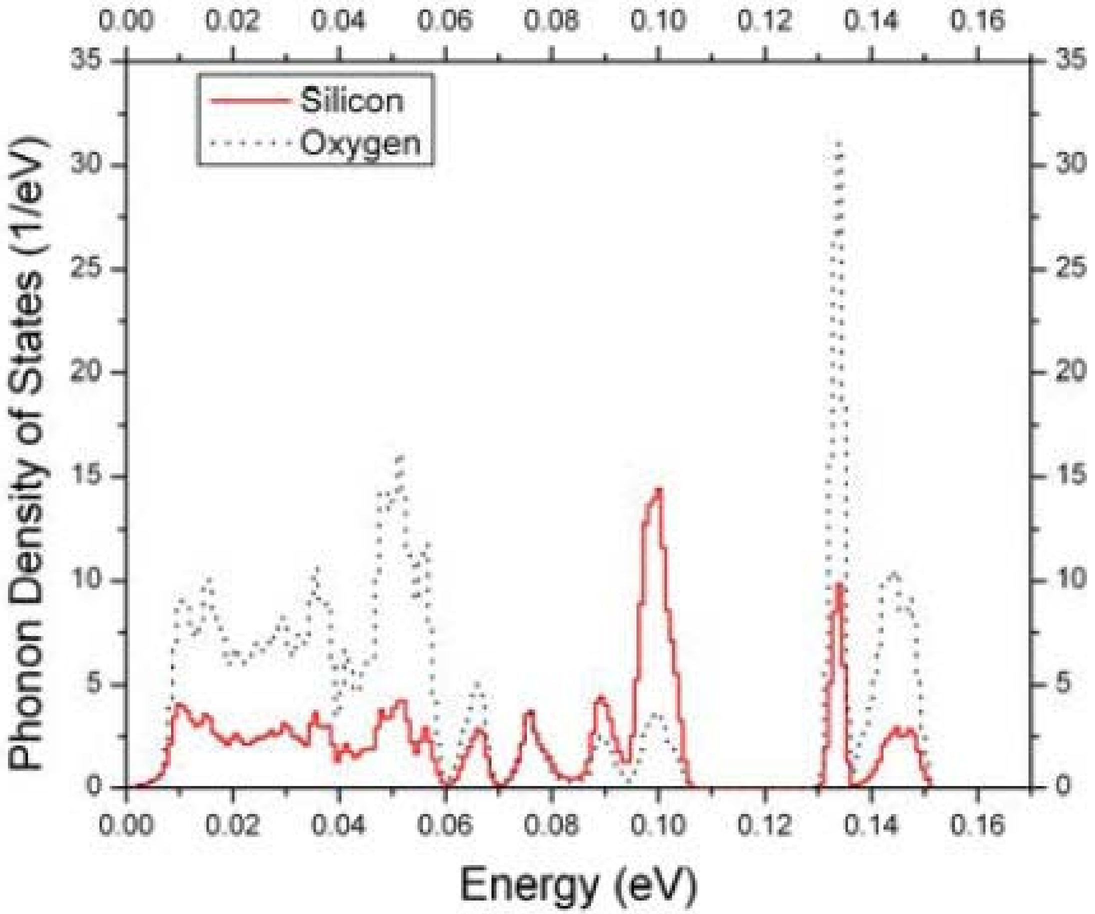
Graphite (1970)
Nicklow et al, (ORNL)

Recently
First Principles
(*Ab initio*)

-Density Functional Theory
-Hellmann-Feynman Theory


VASP
(*ab initio* / DFT)
Hellmann-Feynman
Forces (HF)
PHONON
(lattice dynamics)
ρ(β)
LEAPR
(NJOY)
$S(\alpha,\beta)$
THERMR
(NJOY)
Scattering Cross Section
$\sigma(E)$

Phonon Density Distribution (Density of States)



Ayman I. Hawari and Brian D. Hehr, "Calculation of the Thermal Neutron Scattering Cross-Section of Alpha Quartz," Department of Nuclear Engineering, North Carolina State University, September 2008.

Developing the Coherent Elastic Cross-Section

$$\sigma_{\text{coh}} = \frac{\sigma_c}{E} \sum_{E_i<E} f_i \, e^{-4WE_i}$$

$$E_i = \frac{\hbar^2 \tau_i^2}{8m}$$

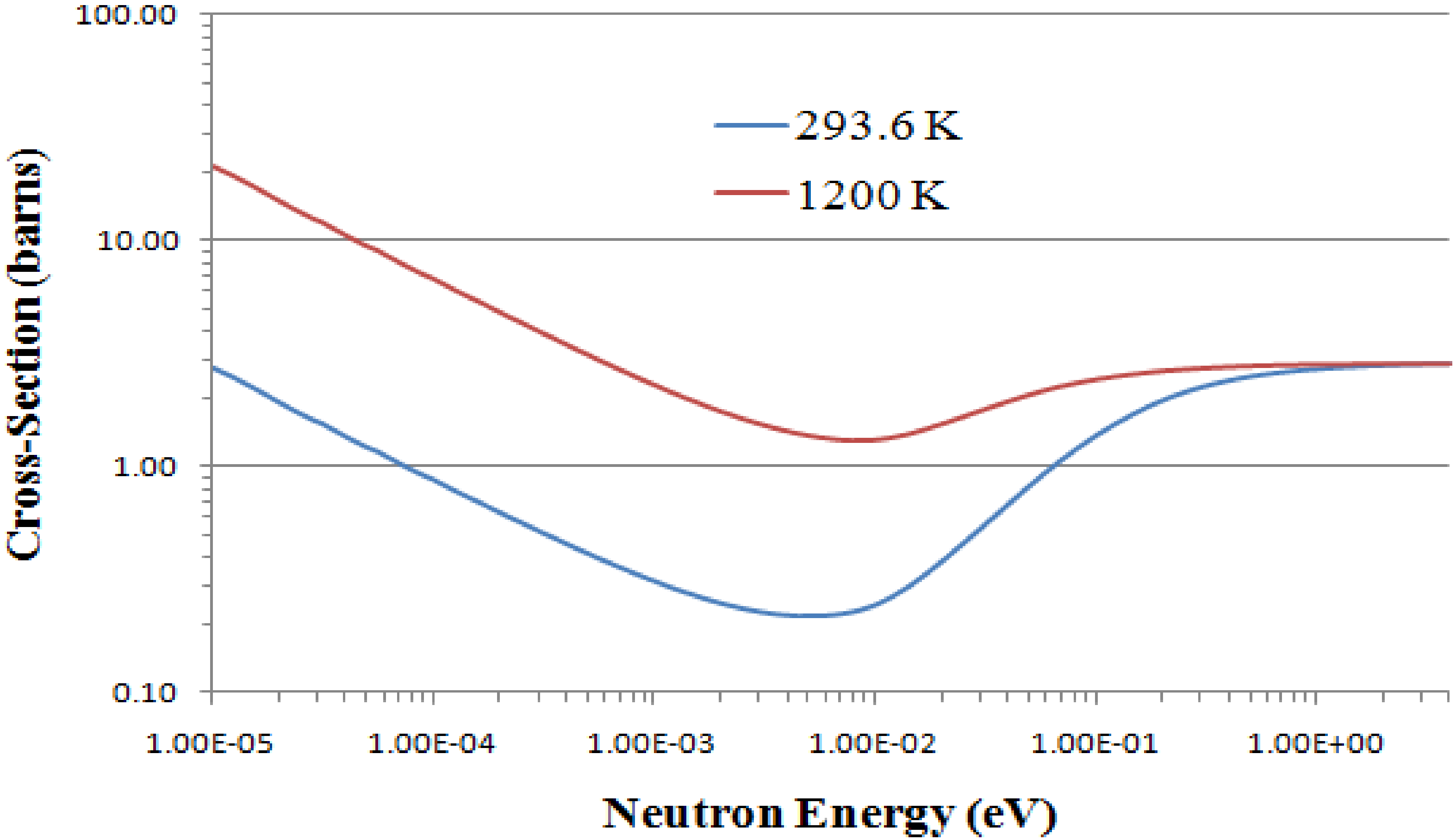
$$\sigma_c e^{-2WE_i} f_i = \left|\sum_{j=1}^{N} \sqrt{\sigma_j}\, e^{-W_j E_i} e^{i2\pi\phi_j}\right|^2$$

$$\sigma_c = \sum_{j=1}^{N} \sigma_j \qquad\qquad \phi_j = \vec{\tau} \cdot \vec{\rho}_j$$

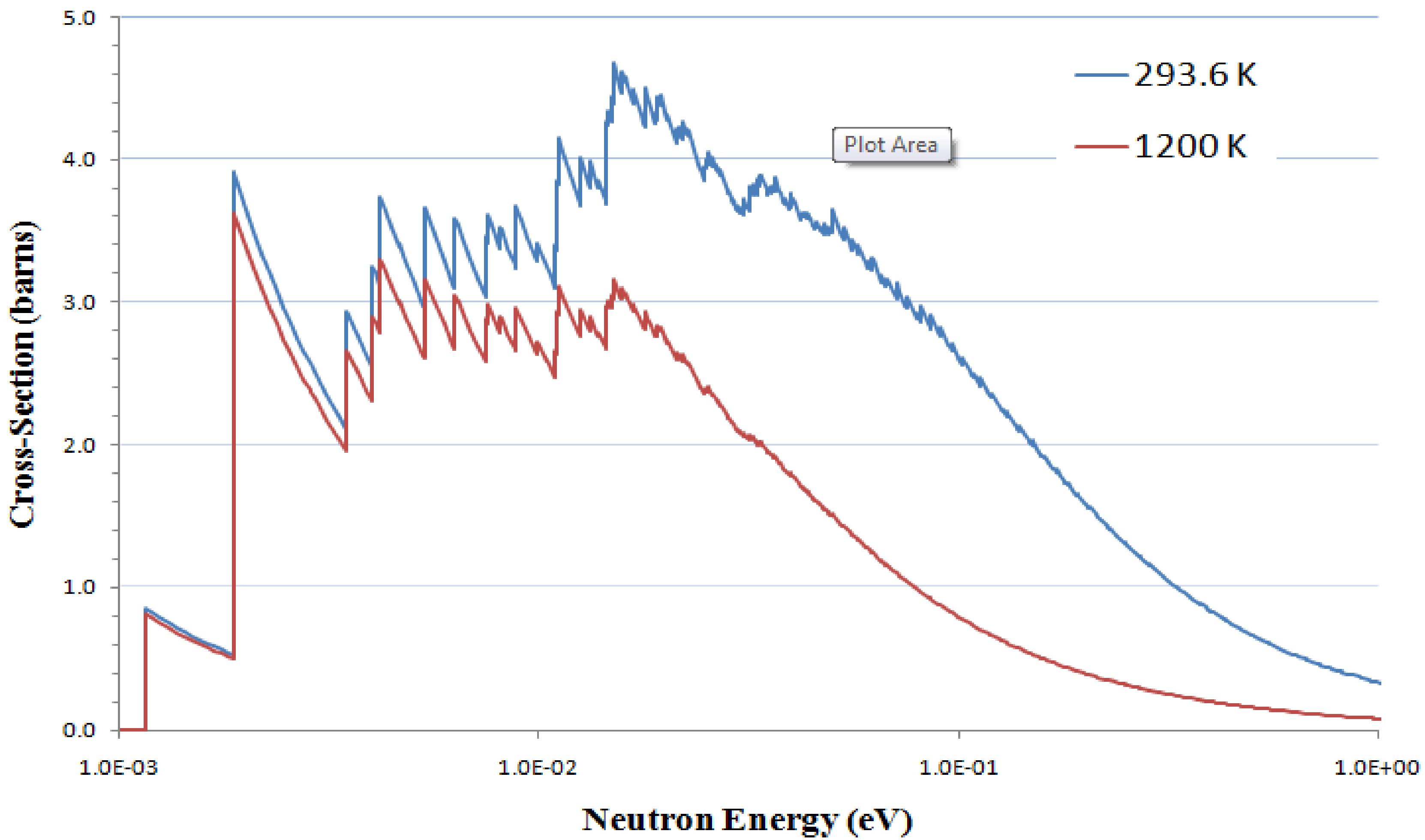
$$|F|^2 = \left|\sum_{j=1}^{N} \frac{\sqrt{\sigma_j}}{\sqrt{\sigma_c}}\, e^{-2\pi\phi_j}\right|^2$$

$$\left(\frac{\tau}{2\pi}\right) = \frac{4}{3a^2}\left(\ell_1^2 + \ell_2^2 + \ell_1\ell_2\right) + \frac{1}{c^2}\ell_3^2$$

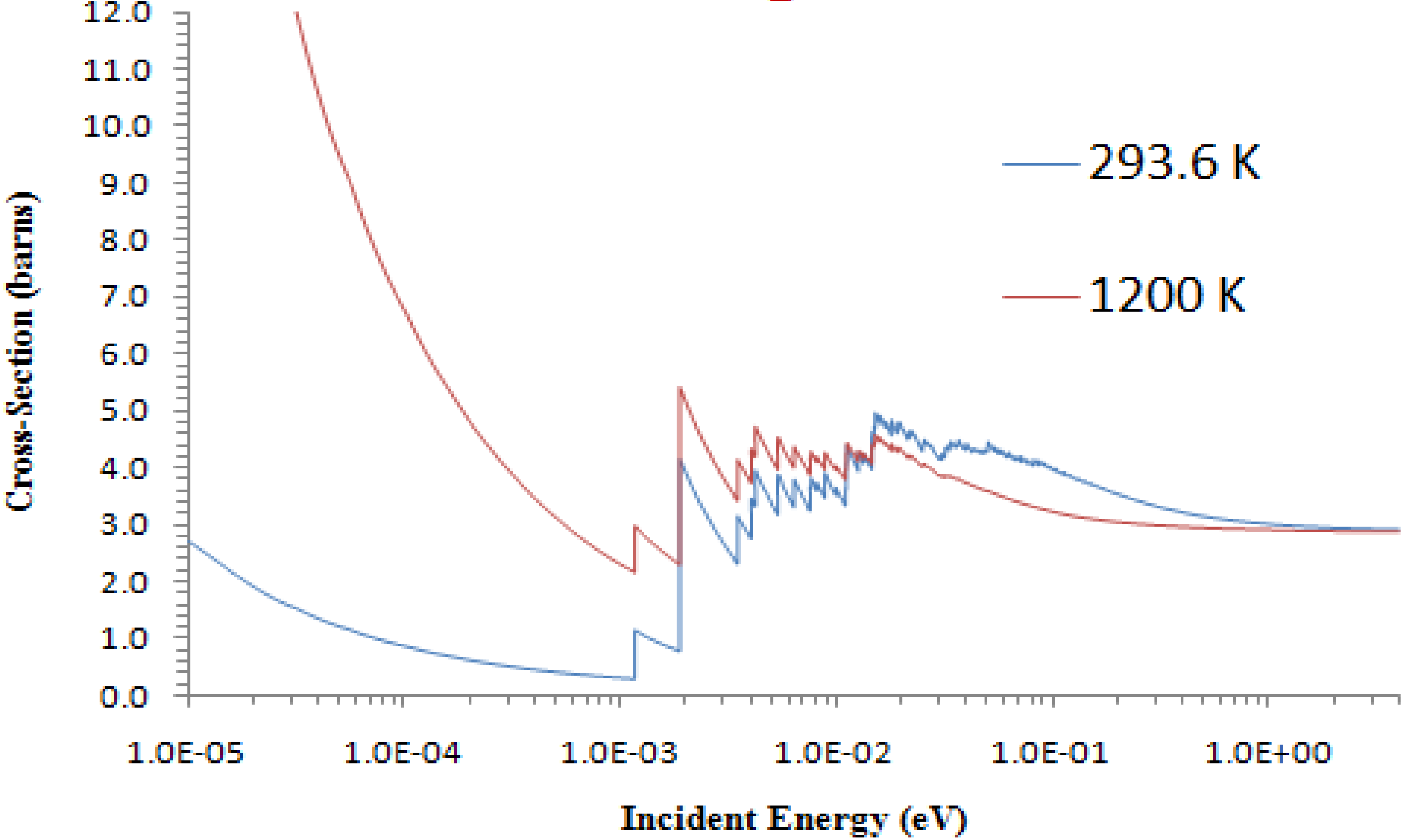
Incoherent Inelastic Thermal Neutron Cross-Sections in Silicon Dioxide



Coherent Elastic Thermal Neutron Cross-Sections in Silicon Dioxide



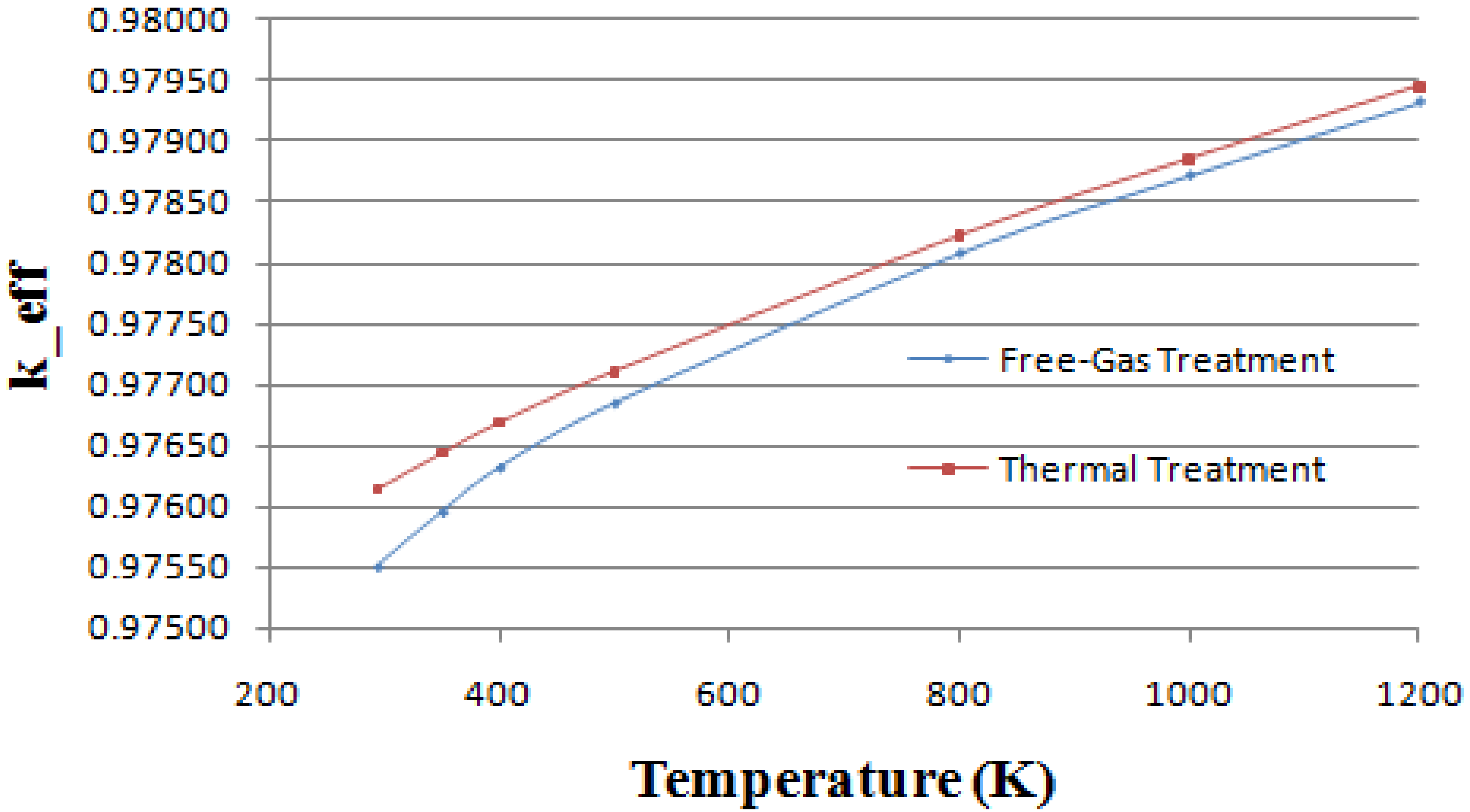
Total Scattering Cross-Section



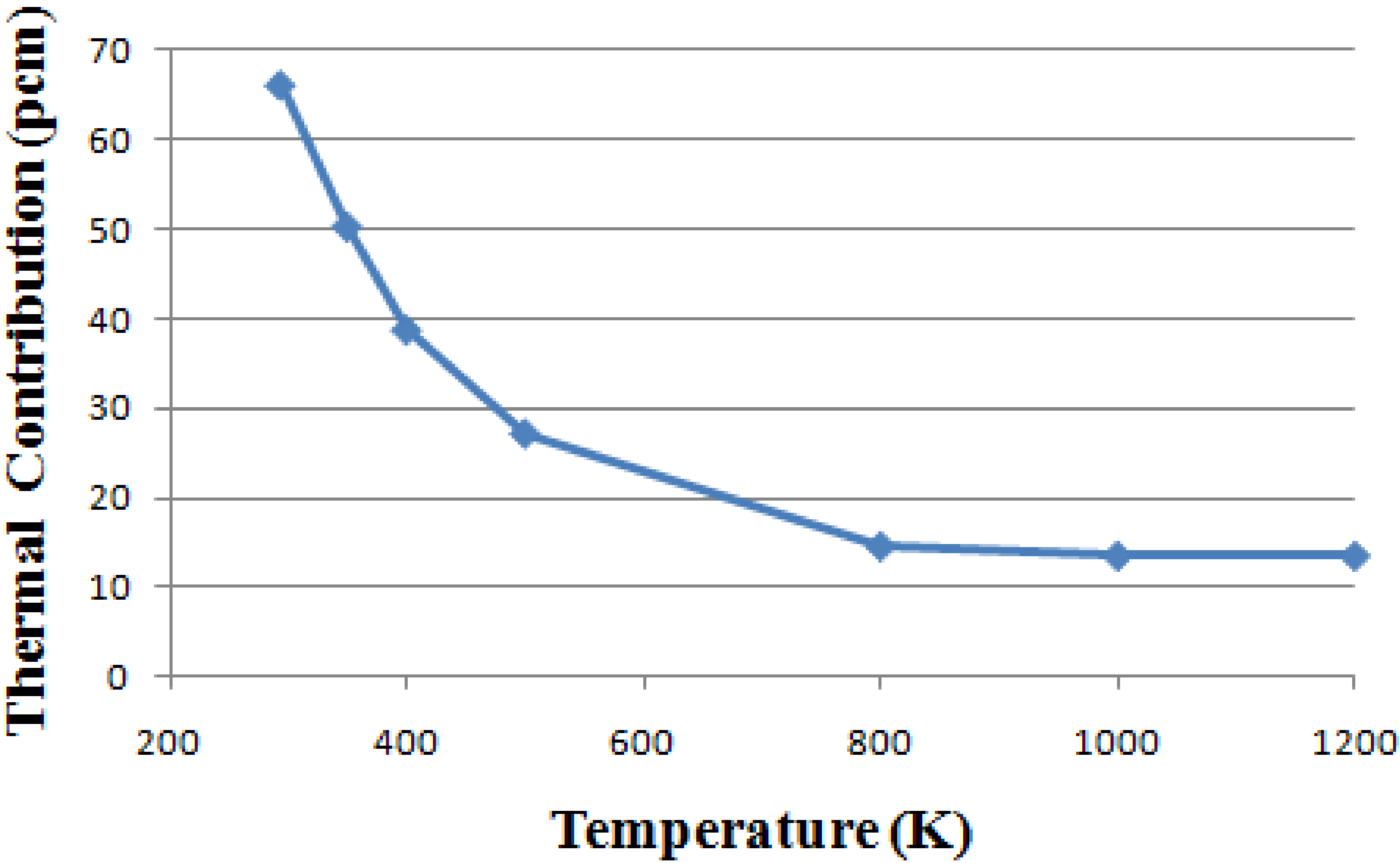
Criticality Modeling with MCNP

- Simple model selected solely to assess the physics of thermal neutron scattering in silicon dioxide.
- Tested k_{eff} over a temperature grid of 293.6, 350, 400, 500, 800, 1000 and 1200 K.
- Evaluated free-gas treatment vs. inclusion of thermal treatment.
- Evaluated inclusion and exclusion of coherent elastic term.
- Analyzed resulting neutron energy spectrums.

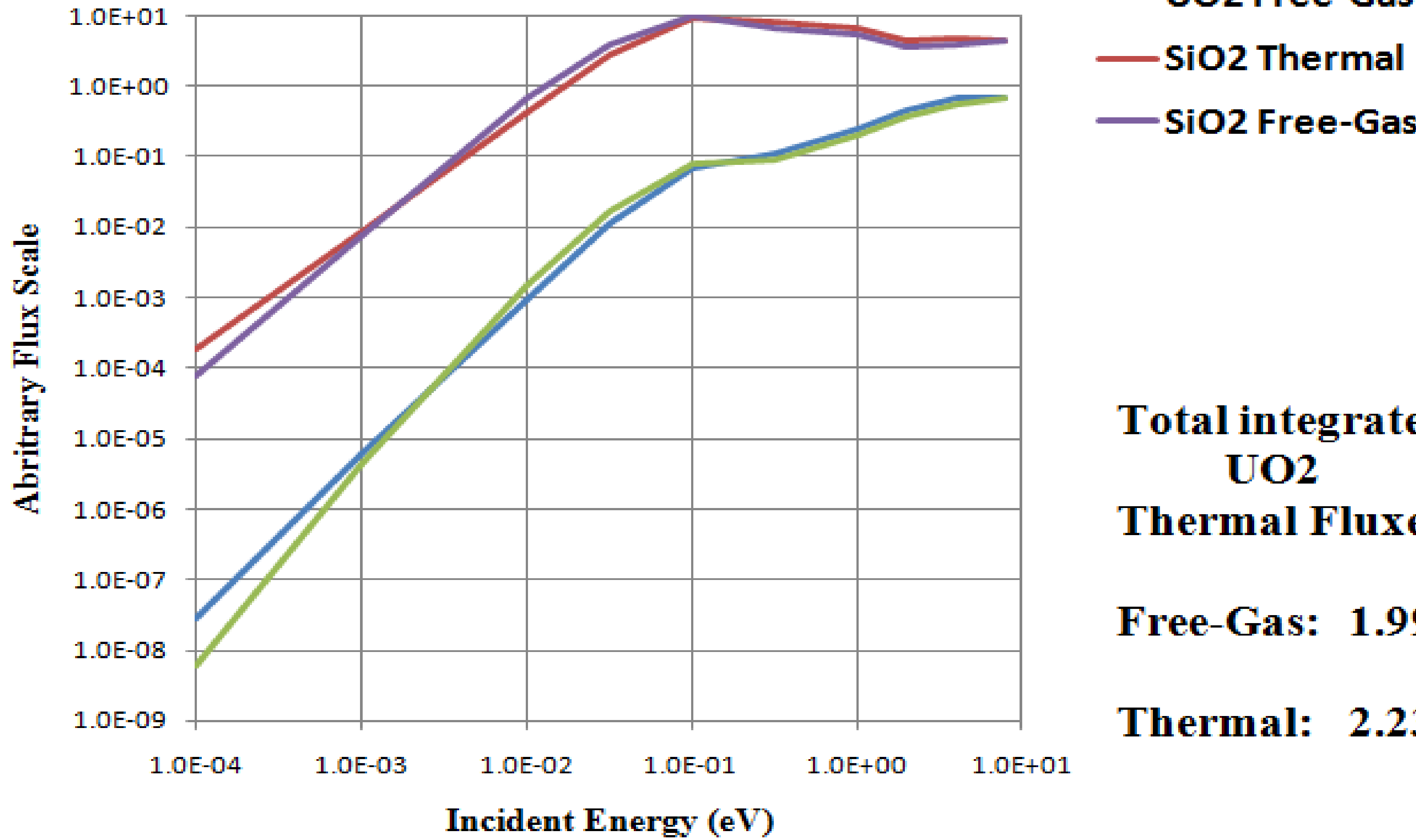
Model k_eff for Thermal vs. Free-Gas Treatment



Contribution to k_eff of Thermal vs. Free-Gas Treatment



Thermal Neutron Flux Spectrum (293.6 K)



Total integrated UO2 Thermal Fluxes:

Free-Gas: 1.99

Thermal: 2.23

Conclusions

- The VASP / PHONON / LEAPR / THERMR sequence was successfully used to produce combined molecule ENDF thermal neutron libraries for incoherent inelastic scattering in silicon dioxide over a standard temperature grid.

- Coherent elastic scattering was successfully added and the resulting thermal scattering cross sections for SiO_2 are ready for ENDF/B VII publication.

- Testing of a criticality model in MCNP using the combined SiO_2 thermal library demonstrated a positive reactivity contribution over using free-gas libraries for Si and O, with the thermal contribution increasing with decreasing temperature.

- The coherent elastic cross-section component for SiO_2 was found to have no significant reactivity contribution in the criticality model.