Project No. 09-770

Accurate Development of Thermal Neutron Scattering Cross Section Libraries

Reactor Concepts R&D

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FINAL TECHNICAL REPORT

Project Title: ACCURATE DEVELOPMENT OF THERMAL NEUTRON SCATTERING CROSS SECTION LIBRARIES

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Project Objective: The objective of this project is to develop a holistic (fundamental and accurate) approach for generating thermal neutron scattering cross section libraries for a collection of important neutron moderators and reflectors. The primary components of this approach are the physical accuracy and completeness of the generated data libraries. Consequently, for the first time, thermal neutron scattering cross section data libraries will be generated that are based on accurate theoretical models, that are carefully benchmarked against experimental and computational data, and that contain complete covariance information that can be used in propagating the data uncertainties through the various components of the nuclear design and execution process. To achieve this objective, computational and experimental investigations will be performed on a carefully selected subset of materials that play a key role in all stages of the nuclear fuel cycle.

Background: The interaction with thermal neutrons is influenced by chemical binding and structure effects. As an example, for crystalline materials, slow (Energy ≤ 1 eV) neutrons have de Broglie wavelengths that are comparable to the inter-atomic spacing of the scattering material. As a result, coherent and incoherent scattering effects become possible. In

addition, the kinetic energy of slow neutrons is comparable to the energy levels that can be excited in a scattering event (e.g., the vibrational levels in crystals). This implies that elastic and inelastic scattering processes are allowed. Moreover, it also implies that thermal neutron scattering cross sections will reflect the dynamics of the structure of the scattering material. Therefore, based on standard neutron scattering theory, the double differential thermal neutron scattering cross sections are usually expressed as a multiplicative combination of the nuclear scattering cross sections and a quantity known as the scattering law.

For the materials of interest to this work, examination of the thermal neutron scattering cross section libraries in the Evaluated Nuclear Data File (ENDF/B-VII.1) results in the following observations: (1) The Be metal libraries remain similar to the ENDF/B-VII release, (2) The graphite libraries are also similar to those in the ENDF/B-VII releases. This treatment completely ignores the fact that a particularly important class of graphite, i.e., "reactor-grade" graphite, is the one implemented in nuclear reactor applications, and (3) In the case of SiO_2, ENDF/B type libraries do not exist and need to be generated and validated. For all of the above materials, the ENDF/B-VII.1 release does not include any thermal neutron scattering cross section covariance information. Therefore, performing sensitivity and design studies that include the impact of the thermal neutron scattering cross section uncertainties is currently impossible.

Status: The following major achievements were made during this project

1) The development of a first-of-a-kind "reactor-grade" graphite library

   Ideal graphite consists of planes (sheets) of carbon atoms arranged in a hexagonal lattice with 4 atoms per unit cell. Strong covalent bonding exits between intraplaner atoms, while the interplaner bonding (i.e., between the carbon sheets) is of the weak Van der Waals type. Alternatively, reactor-grade graphite represents a multi-phase material where graphite ideal crystals are embedded in a carbon binder matrix. Most strikingly, the density of reactor-grade graphite is usually in the range of 1.5 g/cm^3 to approximately 1.8 g/cm^3, while ideal graphite is characterized by a density of nearly 2.25 g/cm^3. This difference in density is manifested as a significant fraction of porosity in the structure of reactor-grade graphite. However, this structural feature of graphite is not captured in the process of generating the inelastic thermal neutron scattering cross sections. Consequently, to account for the impact of porosity on the phenomenon of thermal neutron scattering in graphite, a classical molecular dynamics (MD) approach was implemented to calculate the

phonon spectrum. In this case, an atomisitic model of graphite with various degrees of porosity was constructed. The model was introduced into MD code systems that were especially established to model graphitic structure and to extract the phonon spectrum that is required for thermal neutron scattering cross section calculations. Figure 1 below shows the calculated total inelastic thermal neutron scattering cross section in comparison to ideal graphite data and experimental measurements.

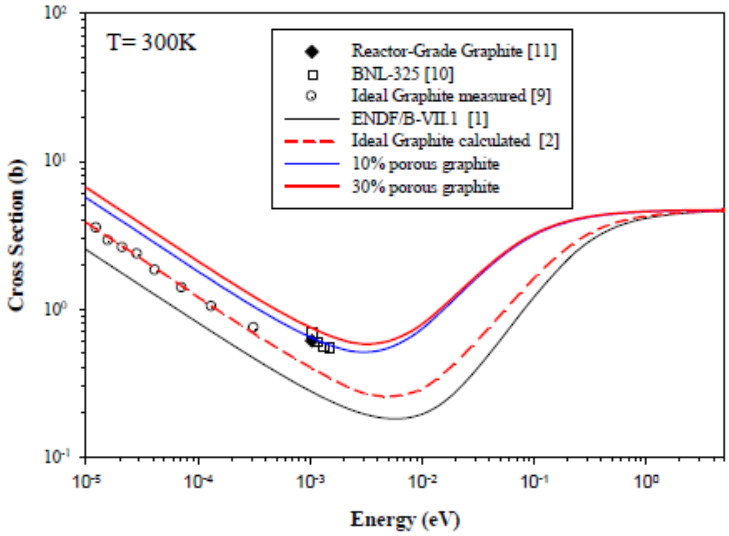

Fig. 1. The total inelastic thermal scattering cross sections of graphite based on various measurements and calculations. The porous data represent the calculations performed in this work to simulate reactor-grade graphite.

2) Experimental investigation of thermal neutron scattering in “reactor-grade” graphite

Elastic and inelastic neutron scattering experiments were performed to investigate the structure and dynamics of reactor-grade graphite. These experiments were conducted at the powder neutron diffraction facility of NCSU and the SEQUOIA instrument of the spallation neutron source (SNS) at ORNL. Sample sof NBG-10 graphite were used. The powder diffraction measurements showed deviations from the ideal graphite structure, which required the development of a special Rietveld code for the interpretation of he collected diffraction patterns. The SEQUOIA measurements were processed to extract a measured phonon spectrum. The measured spectrum clearly revealed a diminished graphite peak in the optical region and

enhancement of the acoustical (low energy) region. This is consistent with the expected behavior for porous graphitic structures. Figure 2 below shows the measured phonon spectrum for reactor-grade graphite.

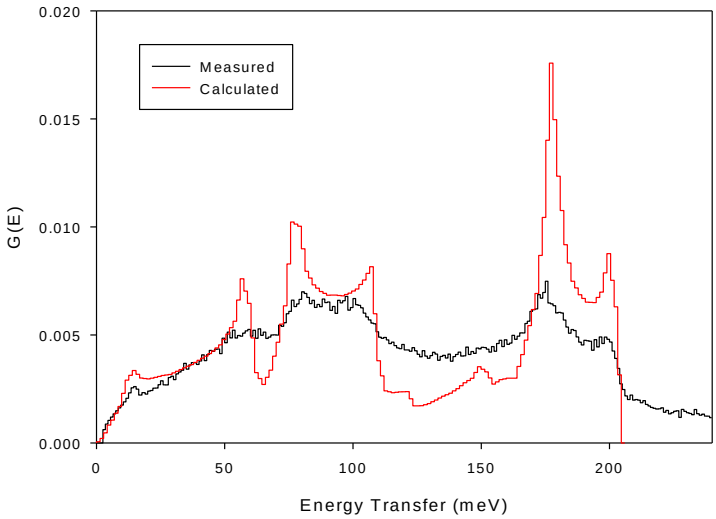

Fig 2. The measured phonon frequency for reactor grade graphite as compared to the calculated phonon frequency for ideal graphite.

3) Ab ignition generation of the thermal neutron scattering law for silicon dioxide (SiO_2)

Silicon carbide (SiC) is an important nuclear material. It is a major constituent of various nuclear fuel concepts including TRISO and fully ceramic microencapsulated (FCM) fuel. In addition, advanced nuclear reactor systems that include SiC as a major constituent of the core have been proposed. More than 250 polymorphs of SiC have been identified. The predominant phase in a reactor environment is the cubic 3C (β) SiC polytype, which represents a zinc blend structure that belongs to space group 216 ($F\bar{4}3m$). In this work, the VASP ab initio package and the PHONON code were used to generate the phonon frequency curve for Si and C in SiC. The phonon frequency information was used in the LEAPR/NJOY code system to generate the elastic and inelastic thermal neutron scattering cross sections for SiC. The figure below shows the total inelastic thermal scattering cross sections.

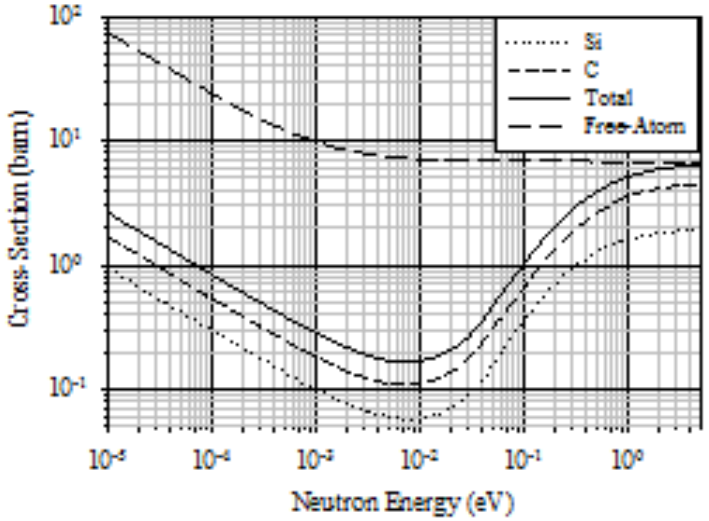

Fig 3. The total inelastic thermal neutron scattering cross sections for SiC at a temperature of 300 K..

4) The generation of the thermal neutron scattering library for beryllium (including coherent inelastic effects)

   In this project, thermal neutron scattering cross section libraries for beryllium were generated at various temperatures. The libraries accounted for coherent inelastic scattering effects, which are missing in the ENDF/B-VII.1 libraries that are generated using the incoherent approximation.

5) The development of a novel approach for calculating the covariance matrix in thermal neutron scattering cross sections

   The phonon frequency spectrum may be viewed as a probability density function of atomic vibrational energy states that exist in a material. Probable variation in the shape of this spectrum may be established that depends on uncertainties in the physics models and methodology employed to produce such a spectrum. Through Monte Carlo sampling of perturbations from the reference phonon spectrum, an $S(\alpha,\beta)$ covariance matrix may be generated. In this work, density functional theory and lattice dynamics in the harmonic approximation were used to calculate the phonon DOS for hexagonal crystalline graphite. This form of graphite is used as an example material for the purpose of demonstrating procedures for analyzing, calculating and processing thermal neutron inelastic scattering uncertainty information. Several sources of uncertainty in thermal neutron

inelastic scattering calculations are examined, including sources which cannot be directly characterized through a description of the phonon frequency uncertainty, and their impacts are evaluated. Covariances for hexagonal crystalline graphite S(α,β) data are quantified by coupling the standard methodology of LEAPR with a Monte Carlo sampling process. The S(α,β) covariance matrix can be propagated to generate covariance data for integrated cross sections, secondary energy distributions, and coupled energy-angle distributions. This approach enables a complete description of thermal neutron inelastic scattering cross section uncertainties. The figure below shows an example of a covariance matrix.

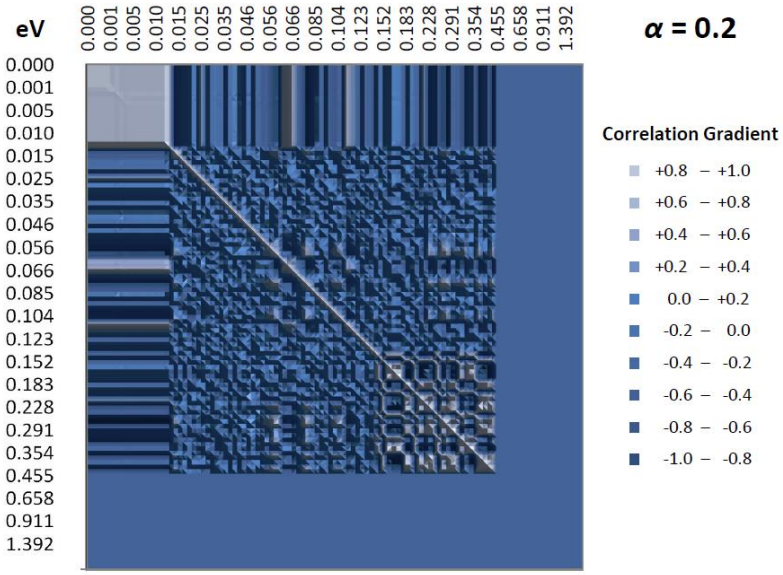

Fig. 4. An example covariance/correlation matrix in S(α,β) for graphite. The greatest region of uncertainty is shown to be in the low energy range.

Patents/Publications/Presentations: The following representative publications have been produced during the duration of this project

1) A. I. Hawari, V. H. Gillette, "Inelastic Thermal Neutron Scattering Cross Sections For Reactor-Grade Graphite," Nuclear Data Sheets, 2014. (in press)

2) J. C. Holmes, A. I. Hawari, "Generation of an S(α,β) Covariance Matrix by Monte Carlo Sampling of the Phonon Frequency Spectrum," Nuclear Data Sheets, 2014. (in press)

3) A. I. Hawari, A. I. Kolesnikov, Q. Cai, J. C. Holmes, P. D. Ferguson, “Inelastic Neutron Scattering Analysis of Reactor Grade Graphite,” Embedded Topical Meeting on Nuclear Fuels and Structural Materials for the Next Generation Nuclear Reactors, Reno, NV, 2014.

4) Y. Zhu, J. L. Wormald, A. I. Hawari, “Thermal Neutron Scattering Cross Sections for Silicon Carbide,” *Transactions of the American Nuclear Society*, 108, 2013.

5) J. C. Holmes, A. I. Hawari, “Generation of an S(□,β) Covariance Matrix by Monte Carlo Sampling of the Phonon Frequency Spectrum,” *Transactions of the American Nuclear Society*, 107, 2012.

6) A. I. Hawari, V. H. Gillette, “Thermal Neutron Scattering Cross Sections For Reactor-Grade Graphite,” *Transactions of the American Nuclear Society*, 106, 2012.

7) J. C. Holmes, I. I. Al-Qasir, A. I. Hawari, L. C. Leal “Development of an ENDF Thermal Library for SiO_2 and Testing of Criticality Effects,” *Transactions of the American Nuclear Society*, 104, 2011.

Milestone Status Table:

The milestone status table is given in the project record at www.neup.gov .

Budget Data (5/5/2014):

Budget data are given in the project record at www.neup.gov .