Project No. 09-822

Optimizing Neutron Thermal Scattering Effects in Very High Temperature Reactors

Reactor Concepts R&D

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FINAL TECHNICAL REPORT

Project Title: OPTIMIZING NEUTRON THERMAL SCATTERING EFFECTS IN VERY HIGH TEMPERATURE REACTORS

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Project Objective: This project aims to develop a holistic understanding of the phenomenon of neutron thermalization in the VHTR. Neutron thermalization is dependent on the type and structure of the moderating material. The fact that the moderator (and reflector) in the VHTR is a solid material will introduce new and interesting considerations that do not apply in other (e.g., light water) reactors. The moderator structure is expected to undergo radiation induced changes as the irradiation (or burnup) history progresses. In this case, the induced changes in structure will have a direct impact on many properties including the neutronic behavior. This can be easily anticipated if one recognizes the dependence of neutron thermalization on the scattering law of the moderator. For the pebble bed reactor, it is anticipated that the moderating behavior can be tailored, e.g., using moderators that consist of composite materials, which could allow improved optimization of the moderator-to-fuel ratio.

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.

In this project, the thermal neutron scattering data libraries for beryllium carbide were generated based on ab initio atomistic simulations. The libraries were generated both in ACE (continuous energy) format that is suitable for use in Monte Carlo codes and in ISOTX multi-group format that is useable in deterministic codes. In addition, detailed analysis of the impact of irradiation on the microstructure and in turn the scattering law of reactor-grade graphite. Testing was also initiated of the reactor-grade graphite libraries using slowing-down benchmark models and data. In general, the libraries and measured data during this project provided novel information, which motivated the publication of several papers as listed below.

Status: The following major achievements were made during this project

1) Experimental and computational examination of the structure of irradiated reactor-grade graphite

   Reactor-grade graphite represents the primary neutronic (moderator and reflector) and structural core material in the VHTR concept. Upon exposure to the neutrons produced in fission the atomic structure of graphite is disrupted and radiation damage is created. In addition to the potential impact on structural properties, in this case, the disruption in structure is expected to impact the neutron thermalization properties of graphite. This is due to the fact that neutron thermalization is a microstructure dependent phenomenon. In this work, neutron scattering and positron annihilation analysis were used to examine the structure of irradiated graphite. The observations were then related to the computational findings. In particular, samples of NBG-10 graphite that were irradiated to a total neutron fluence of 4.9×10^{21} neutrons/cm^2 at 280 °C and 4.7×10^{21} neutrons/cm^2 at 700 °C were examined (at the positron beam laboratory of the NCSU PULSTAR reactor) and compared to unirradiated samples. Positron annihilation analysis showed that for the samples irradiated at 280 °C, clusters composed of multiple vacancies are prevalent. However, at

700 °C mono-vacancies dominate. Molecular dynamics radiation cascade analysis (Fig. 1) showed that vacancies represent localization sites for low frequency phonons. Consequently, the existence of such clusters was found to enhance the graphite phonon spectra at low incoming neutron energy and increase the probability of down scattering for thermal neutrons. These conclusions were directly supported by neutron scattering experiments that were performed using the SEQUOIA instrument of the spallation neutron source (SNS) at ORNL. Figure 2 below shows the measured phonon spectrum for irradiated reactor-grade graphite. As it can be seen, the optical peak 180 meV is completely eliminated in the spectrum, while the lower energy region below 100 meV is enhanced. In particular, the region below 10 meV shows significant enhancement, which is important as thermal neutron scattering is highly sensitive to the structure of this region. Furthermore, it is anticipated that irradiated graphite may exhibit some sp^3 type bonding as opposed to the pure sp^2 bonding of unirradiated graphite.

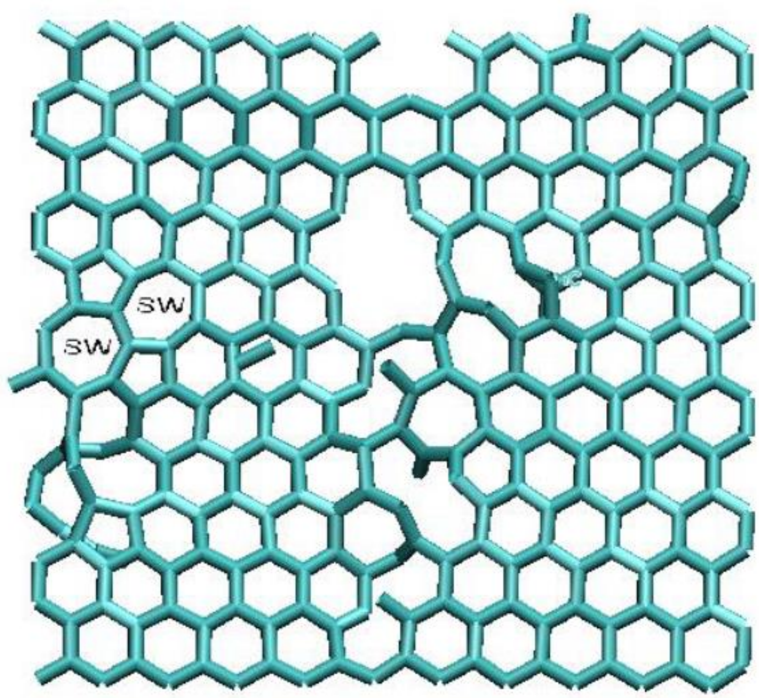

Fig 1. Vacancy clusters in a graphite plane as simulated using classical molecular dynamics. Low energy acoustical phonons localize at such clusters.

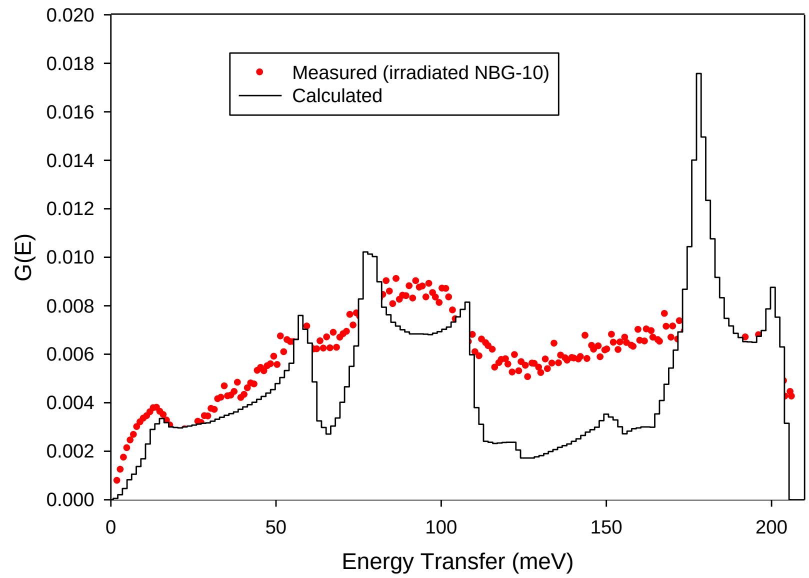

Fig 2. The measured phonon frequency spectrum for irradiated reactor grade graphite as compared to the calculated phonon frequency spectrum for ideal graphite.

2) Ab initio generation of the thermal neutron scattering law for beryllium carbide (Be_2C) and the cross section associated libraries

In this work, first-of-kind cross sections for the mixed moderator material beryllium carbide (Be_2C) were produced. Beryllium carbide belongs to the cubic, antifluorite class of crystallographic structures (space group 225) with each beryllium atom surrounded by four carbon atoms in a tetrahedral arrangement. The lattice parameter is of length 4.342 Å. The ab initio code VASP and the lattice dynamics code PHONON were used to generate the phonon spectrum for beryllium carbide. Using this spectrum the NJOY code system was used to generate the thermal neutron scattering libraries both in the ACE (continuous energy format) and the ISOTX (multi-group energy) format. Figure 3 below shows the total inelastic cross section for C and Be in beryllium carbide calculated at 300 k. The comparison between the ACE and multi-group generated data shows consistency and indicates that both data sets should be equivalent when used in their corresponding computer codes. For example the ACE libraries are usually implemented in Monte Carlo transport codes (such as MCNP) and the ISTOX libraries should be usable in the deterministic type of transport codes.

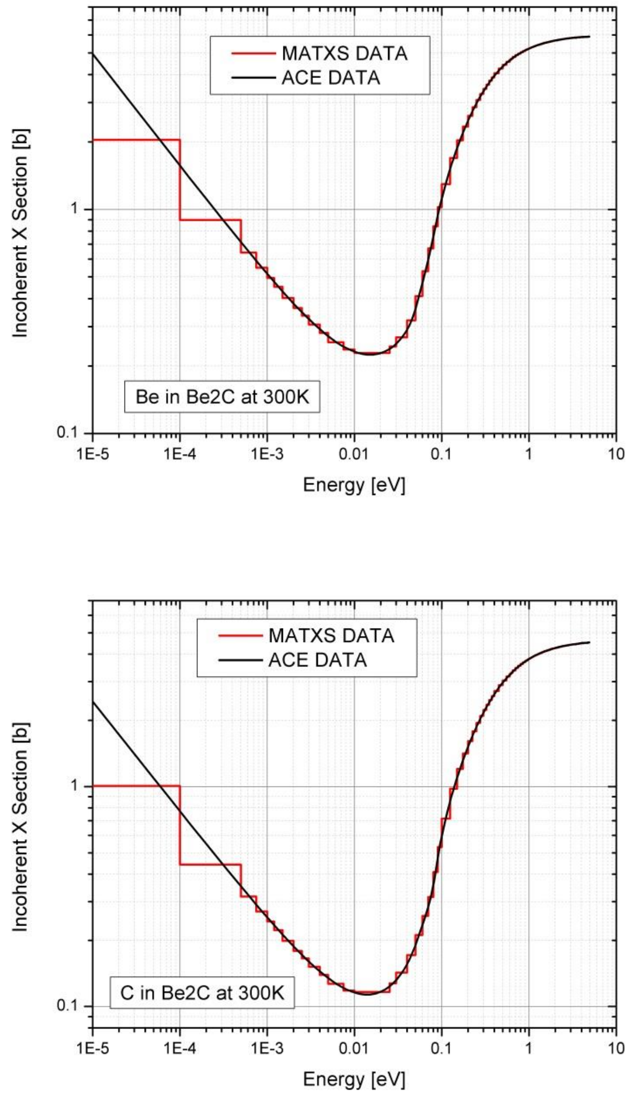

Fig 3. The total inelastic thermal neutron scattering cross sections for Be and C in beryllium carbide at a temperature of 300 K.

3) Initial assessment of reactor-grade graphite cross section libraries in neutronic simulations

   During this project, MCNP Monte Carlo simulations were performed to test the "reactor-grade" graphite thermal neutron scattering cross section that were generated. The Monte Carlo analysis simulated a past experiment that was performed at the ORELA facility of Oak Ridge National Laboratory. The collected data in that experiment represented time dependent slowing down spectra of an ORELA neutron pulse that is injected into a 70x70x70 cm^3 graphite pile. Previous simulations of the experiment showed differences between the predicted time dependent signal and the measured signal in detectors located around the pile reaching 15-20%. These experiments used the standard ENDF/B-VII $S(\alpha,\beta)$ data libraries for graphite. In this work, the simulations were conducted using graphite data libraries that reflect its porous structure and should therefore more faithfully represent the reactor-grade type of this material. In this case, the agreement between the simulated signals and the measured signals improved by approximately 50%. However, the data indicates that further improvements may be expected by further the structure and porosity levels of graphite.

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

1) B. D. Hehr, Ayman I. Hawari, “Generation of Thermal Neutron Scattering Libraries for Beryllium Carbide”, Proceedings of the 1st International Nuclear and Renewable Energy Conference (INREC10), Amman, Jordan, 2010.

2) Q. Cai, Ayman I. Hawari, “Neutron Powder Diffraction Study of Reactor Grade Graphite,” Embedded Topical Meeting on Nuclear Fuels and Structural Materials for the Next Generation Nuclear Reactors, Chicago, IL, 2012.

3) M. Liu, Ayman I. Hawari, “Positron Characterization of Neutron Irradiated Reactor Grade Graphite,” Embedded Topical Meeting on Nuclear Fuels and Structural Materials for the Next Generation Nuclear Reactors, Reno, NV, 2014.

4) 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.

Milestone Status Table:

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

Budget Data (6/13/2014):

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