Neutron Resonance Radiography and Application to Nuclear Fuel Materials

Yuxuan Zhang,1 Kristian G. Myhre,1 Hassina Z. Bilheux,1 Anton S. Tremsin,2 Jared A. Johnson,1 Jean-Christophe Bilheux,1 Andrew J. Miskowiec,1 Rodney D. Hunt,1 Louis J. Santodonato,1 Jamie J. Molaison1

1Oak Ridge National Laboratory, P. O. Box 2008, Oak Ridge, TN 37831, USA, zhangy6@ornl.gov
2Space Sciences Laboratory, University of California at Berkeley, 7 Gauss Way, Berkeley, CA 94720, USA

INTRODUCTION

Neutron imaging techniques have been used for decades to nondestructively map features throughout an entire sample volume including internal structures, porosity, and defects. The types of obtainable information have been dramatically increased by recent advancements in pulsed neutron sources and time-of-flight (TOF) detectors. Spatially resolved crystallographic and isotopic information can now be obtained using these TOF (or energy dependent) neutron imaging techniques made possible by the unique way neutrons are produced at an accelerator-based neutron source.

The Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL) produces neutrons by bombarding a heavy metal target with protons that are accelerated by a linear accelerator. Neutrons are “spalled” or knocked from the nucleus of a target material (in this case mercury) to accelerations of very high energy (MeV), therefore the metal target needs to be surrounded by moderators to kinetically “cool” (i.e., remove energy) the relativistic neutrons. The ‘cooled’ neutrons are then transported from the moderator to the sample area where they are measured using spatially and temporally resolved detectors. The data recorded can be then analyzed using a technique known as the TOF technique.

An illustration of the TOF neutron imaging technique is shown in Fig. 1. In this figure, \( T_0 \) indicates the time when neutrons are generated by pulsed protons, \( T \) is the time when neutrons are recorded at the detector, \( t_m \) is the time neutrons spend in the moderator, \( t_s \) is the response time of detector (time resolution), \( L \) is the known source-to-detector distance, and \( t_{tof} \) is the neutron travel time over \( L \).

For a specific moderator and instrument setup, \( t_m \) can be fitted using the Ikeda–Carpenter function,\(^1\) and \( t_s \) can be experimentally determined. Thus, by acquiring a series of time-stamped radiographs that are timed relative to \( T_0, t_{tof} \) can be extracted using Eq. (1).

\[
T - T_0 = t_m + t_{tof} + t_d
\]

Further, \( t_{tof} \) can be used to derive the neutron energy using Eq. (2), where \( m \) is the mass of the neutron, \( v \) is the speed of the neutron, and \( E \) is the neutron energy.

\[
E = \frac{1}{2} m v^2 = \frac{m}{2} \left( \frac{L}{t_{tof}} \right)^2
\]

By converting the time stamp into neutron energy, two energy dependent imaging techniques can be applied in different ranges of neutron energy. In the thermal/cold range (i.e., for neutrons with energies less than 0.025 eV), Bragg-edge imaging\(^2\)\(^–\)\(^8\) is optimized for crystalline materials and can provide information such as (hkl) plane orientations and phase evolution. In the epithermal range (i.e., for neutrons with energies higher than 1 eV), the energy variation of neutron cross sections is isotope specific with sharp resonances present for isotopes of nearly all heavy elements and many of the light ones.\(^9\) This advantage can be readily used to spatially identify the isotopic content in bulk nuclear materials, which are mostly opaque to other imaging techniques (e.g., X-ray radiography). This technique, known as neutron resonance radiography,\(^10\)\(^–\)\(^12\) has been investigated at other accelerator-based neutron facilities, including Japan Proton Accelerator Research Complex (J-PARC),\(^13\)\(^–\)\(^14\) ISIS Neutron and Muon Source,\(^15\)\(^–\)\(^17\) Geel Linear Accelerator (GELINA),\(^18\) and Los Alamos National Laboratory (LANL).\(^19\)\(^–\)\(^22\)

This work is the first time that neutron resonance radiography was implemented at ORNL’s SNS. Results are presented from proof-of-principle measurements conducted at the SNS Spallation Neutrons and Pressure Diffractometer (SNAP) beamline. This includes measurements of Au, Ag, Cd, Co, Hf, In, and W metallic foils. The energy dependent behavior of the foils was measured simultaneously as a function of neutron energy using the TOF technique. Following the foil demonstration, an investigation of tristructural-isotropic (TRISO) fuel kernels was also conducted. TRISO fuel kernels are of high interest for use as an advanced fuel form in several nuclear reactor designs. The results of this work show the potential of this technique at
SNS to characterize nuclear materials in ways not previously possible with other analysis methods.

EXPERIMENTS AND METHODS

Time-stamped radiographs were acquired using a microchannel plate detector (shown in Fig. 2(a)) developed by Tremsin et al.\(^{23}\) Metallic foils (Ag, Au, Cd, Co, Hf, In, and W) were stacked and mounted on a thin Al plate before being placed against the microchannel plate detector. Aluminum was chosen as a backing because it does not have neutron resonances and is highly transparent to neutrons. The foils overlapped and occupied the whole field of view of the detector. A schematic of the foil stack within the field of view (2.8 cm × 2.8 cm) is shown in Fig. 2(b).

Fig. 2. (a) Photograph of a microchannel plate detector, (b) drawing of the foil stack within detector field-of-view.

Utilizing the results from the foil experiments, TRISO fuel kernels were explored using neutron resonance radiography. These measured TRISO fuel kernels (~2.6 mm in diameter) contain UO\(_3\) with natural uranium.

RESULTS

Neutron radiographs of the measured foils are shown in Fig. 3. In this figure, the radiographs, with element names and energies labeled, are image frames at corresponding energies that neutron resonances occur for each foil. A radiograph labeled as ‘white beam’ (bottom right) is also obtained at the energies corresponding to cold neutron sources (<0.025 eV) to demonstrate what the conventional neutron imaging (total attenuation based) would see for the same samples. In each foil region, as shown in Fig. 2(b), attenuation as a function of image frames were extracted and converted to an attenuation vs. energy spectrum as shown in Fig. 4. In Figs. 3 and 4, it can be clearly seen that the resonance attenuation peaks are energy-specific and unique to an element, thus can be treated as fingerprints to locate interested element spatially using TOF neutron imaging technique. It is important to note that, the resonance peak of indium is not very pronounced due to the insufficient material quantity, which explains why only a light shadow was observed in the In foil region (Fig. 3). In addition to the spatial elemental identification purpose, fitting resonance peaks individually allows the material amount and temperature to be quantified.

Fig. 3. Radiographs with different foils shown at given neutron energies corresponding to a resonance peak for each metallic foil. At the bottom right is a radiograph based on total attenuation, which is the most commonly used neutron imaging contrast mechanism at reactor sources.

Fig. 4. Resonance peaks for each foil. These resonances were acquired simultaneously.

Since the resonance characteristics depend on the reactions between neutrons and nuclides, the elemental sensitivity of neutron resonance radiography can also be extended to isotopes, which could be extremely helpful in the characterizations of nuclear fuel. Therefore, following the foil measurements, TRISO fuel kernels were explored in this work. A total cross-section vs. neutron energy plot of \(^{235}\)U and \(^{238}\)U using data on Evaluated Nuclear Data File (ENDF/B-VII.1)\(^{25}\) database is shown in Fig. 5. As can be seen, separate resonance peaks for \(^{238}\)U and \(^{235}\)U can be
identified. However, the $^{238}$U resonance peaks are more pronounced than those of $^{235}$U. The measured resonance spectrum of these TRISO fuel kernels is shown in Fig. 6 along with a radiograph corresponding to the image frame at ~20.85 eV (where $^{238}$U resonance presents). In the measured attenuation vs. energy plot, resonance peaks of $^{238}$U can be successfully identified, but, because of the inherently low level of $^{235}$U enrichment in natural uranium, $^{235}$U resonance peaks were not observed.

Fig. 5. Plot of total cross-section vs. neutron energy generated using data from ENDF database.

Fig. 6. Neutron transmission radiograph (~20.85 eV) where darker contrast indicates the TRISO kernels (left) and resonance spectrum showing $^{238}$U peaks (right).

SUMMARY

Neutron resonance radiography, a technique capable of spatially resolving isotopic/elemental information, has been successfully implemented at SNS using the SNAP beamline. In this work, we were able to visually identify metallic foils made of different elements. Additionally, unirradiated nuclear fuel material ($^{238}$U), has been successfully measured. It is noticed that the amount of the active material, which has a resonance reaction with neutrons at certain energies, is very crucial to provide enough contrast in these measurements. Therefore, in order to provide an idea of how applicable this technique is to a specific sample, an open-source Python library called ImagingReso has been developed to simulate a resonance spectrum based on sample compositions.

ACKNOWLEDGMENTS

This work is sponsored by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory, managed by UT-Battelle LLC for the US Department of Energy. This research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by Oak Ridge National Laboratory.

REFERENCES


