Nondestructive examination of uranium oxide kernels using energy-resolved neutron imaging

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INTRODUCTION

Neutron radiography is one approach to nondestructively characterizing a wide range of sample types, including nuclear fuel materials. Tomographic data can be obtained when radiographs are collected at multiple rotation angles. Traditional neutron radiography can offer a wealth of structural information including porosity, defects, and shape information. A relatively new suite of neutron radiography modalities makes use of energy-resolved data, which is typically obtained using a time-gated detector and pulsed neutron source. This type of neutron imaging is called Energy-Resolved Neutron Imaging (ERNI). One form of ERNI makes use of neutrons from roughly 1 eV to 1000 eV (epithermal) to obtain spectral data in the neutron resonance region. This technique is called Neutron Energy Resonance Imaging (NERI) and provides isotopic composition information. Numerous isotopes, including those of uranium and gadolinium, have large resonance cross sections in the epithermal neutron energy region. NERI can then be used to map individual uranium, gadolinium, and other elements through a sample.

Oak Ridge National Laboratory (ORNL) has started development of NERI capabilities at the Spallation Neutron Source (SNS) with the last few years. Initial studies have been focused on developing the technique through study of spherical metal oxide kernels used to produce tristructural-isotropic (TRIS) microspheres. Preliminary results were presented previously and demonstrated that uranium and gadolinium could be selectively imaged with NERI within mixed uranium-gadolinium oxide spheres. This paper presents new results from further data analysis as well as comparison of NERI results with scanning electron microscopy analysis of the same samples.

Recent efforts to develop NERI capabilities at Oak Ridge National Laboratory's (ORNL) Spallation Neutron Source (SNS) have largely focused on the study of advanced nuclear fuels. Specifically, tomographic mapping of elements within spherical metal oxide kernels used to produce tristructural-isotropic (TRISO) microspheres has been carried out. This paper presents recent results from NERI measurements of mixed uranium/gadolinium oxide TRISO kernels. Uranium is used as a fissile fuel and gadolinium is used as a burnable neutron poison. The distribution of the two elements within the TRISO kernels is important for fuel design. Scanning Electron Microscopy with Energy-dispersive X-ray Spectroscopy (SEM-EDS) is typically utilized for elemental mapping of TRISO kernels. However, SEM-EDS requires the destructive cross-sectioning of samples thereby allowing only one two-dimensional slice of an individual kernel to be investigated as well as potentially damaging and altering the sample itself. Neutron-based imaging techniques are non-destructive and probe the entire sample volume, thereby avoiding both issues. Other radiation-based imaging techniques, such as X-ray radiography, have a limited capability to penetrate high-Z materials.[11] NERI and other neutron-based imaging techniques therefore offer a unique capability to study high-Z materials including nuclear fuels.

EXPERIMENTAL

The uranium and gadolinium oxide spheres were produced using an internal gelation sol–gel method.[12] The spheres contained roughly 69 wt% uranium, 15 wt% gadolinium, and 16 wt% oxygen. The spheres had a diameter of about 3 mm. The spheres were dried prior to loading into special vanadium cans for the neutron imaging measurements (vanadium is transparent in the epithermal neutron energy region).

The Spallation Neutrons and Pressure Diffractometer (SNAP) instrument at the SNS was used to perform the NERI measurements. The SNAP beamline has a direct view of the decoupled H_2 moderator and provides the required wavelength discrimination to perform NERI measurements. A total of 36 radiographs were collected by rotating the sample in 5° steps over the course of 0° to 175°. Each angle was measured for 2 hours. The radiographs were collected using a micro-channel plate detector developed by Tremsin *et al.* The time stamp of each radiograph was correlated to a neutron energy using the neutron travel time from the mercury target to the neutron detector. This results in a neutron energy spectrum at each pixel. Because of the low count rate for a single pixel, it is necessary to sum across multiple pixels to calculate a NERI spectrum with a reasonable signal-to-noise ratio. The NERI spectrum contains peaks associated with a sharp increase in neutron absorption by a specific isotope. It is then possible to sum the data corresponding to that peak and map individual isotopes.

Multiple peaks resulting from the same isotope may be summed to increase the signal-to-noise ratio. Likewise, data for multiple isotopes of the same element may be summed if only the elemental distribution is of interest. In this study, two data sets were produced with one mapping the uranium distribution and the second mapping the gadolinium distribution. The radiographs identified for each element were then reconstructed using Octopus Imaging Software's Octopus Reconstruction software tool, to produce element-specific three-dimensional datasets.

RESULTS

Figure 1 shows an example spectra obtained through the NERI measurements. The radiographs correlating to each of the indexed peaks were summed together to produce element-specific radiographs. Element-specific tomographs were then produced from the radiographs (36 for gadolinium and 36 for uranium) by the Filtered Back Projection technique. The resulting dataset was loaded into Object Research Systems Inc., Dragonfly visualization software to produce three-dimensional visualizations. Example screenshots of the visualizations are provided in Figure 2.

The NERI measurements qualitatively show an inhomogeneous density profile for both gadolinium and uranium within the spheres. There is a higher density for both elements at the edges of the spheres compared to the middle. Information such as this can be utilized to explore the effects different processing parameters have on the internal material composition. It can be seen that NERI is a useful technique for exploring nuclear materials, such as the kernels used to produce TRISO microspheres.

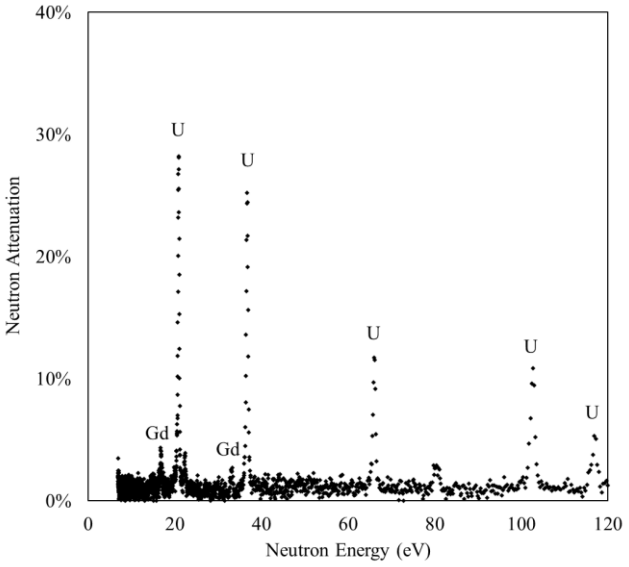

Fig. 1. Experimental NERI spectra of uranium/gadolinium oxide spheres from 0 eV to 120 eV. Uranium peaks are marked with a U and gadolinium peaks are marked with a Gd.

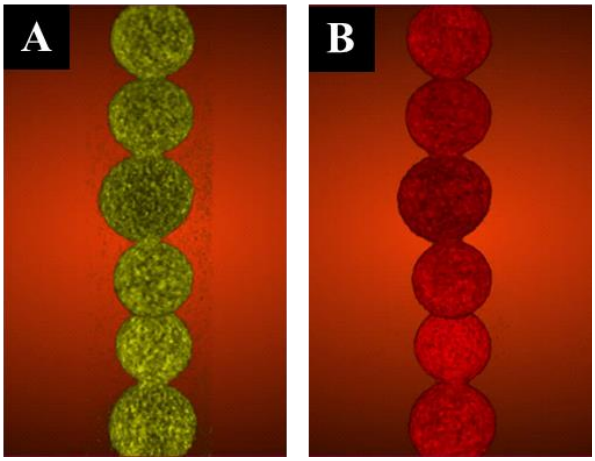

Fig. 2. Snapshots of 3D visualizations of gadolinium (*left*, A) and uranium (*right*, B) distributions within the uranium-gadolinium spheres.

SUMMARY

NERI was used to map in three dimensions the uranium and gadolinium content throughout mixed uranium/gadolinium oxide spheres. New results include showing that the NERI results match well with SEM studies. This work helps to lay a foundation for the more frequent use of NERI to study nuclear fuel materials at the SNS.

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