NEUTRON RADIOGRAPHY AND FISSION MAPPING MEASUREMENTS OF NUCLEAR MATERIALS WITH VARYING COMPOSITION AND SHIELDING [1]

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ABSTRACT

Neutron radiography and fission mapping measurements were performed on four measurement objects with varying composition and shielding arrangements at the Idaho National Laboratory's Zero Power Physics Reactor (ZPPR) facility. The measurement objects were assembled with ZPPR reactor plate materials comprising plutonium, natural uranium, or highly enriched uranium and were presented as unknowns for characterization. As a part of the characterization, neutron radiography was performed using a deuterium-tritium (D-T) neutron generator as a source of time and directionally tagged 14 MeV neutrons. The neutrons were detected by plastic scintillators placed on the opposite side of the object, using the time-correlation-based data acquisition of the Nuclear Materials Identification System developed at Oak Ridge National Laboratory. Each object was measured at several rotations with respect to the neutron source to obtain a tomographic reconstruction of the object and a limited identification of materials via measurement of the neutron attenuation. Large area liquid scintillators with pulse shape discrimination were used to detect the induced fission neutrons. A fission site map reconstruction was produced by time correlating the induced fission neutrons with each tagged neutron from the D-T neutron generator. This paper describes the experimental configuration, the ZPPR measurement objects used, and the neutron imaging and fission mapping results.

INTRODUCTION

This paper describes neutron radiography and fission mapping measurements of fissile objects performed by the Nuclear Materials Identification System (NMIS), a laboratory system at Oak Ridge National Laboratory (ORNL). The measurements were made at Idaho National Laboratory's (INL's) Zero Power Physics Reactor (ZPPR) facility. INL created a group of objects that were assembled from ZPPR reactor plate materials comprising plutonium, natural uranium, or highly enriched uranium [1]. The objects were presented as unknowns for characterization.

NMIS serves as a development platform for various time-coincident radiation measurements. The system has been developed to do coincidence counting measurements of fissile materials. NMIS typically makes coincidence measurements using fast detectors and coincidence windows down to 1 nanosecond wide. It can perform active coincidence measurements with deuterium-tritium (D-T) neutron sources, ^{252}Cf fission sources, or gamma-ray sources; it can perform

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passive coincidence measurements of neutrons or gamma rays. NMIS measurement results are in the form of time correlations (time-varying coincidence), multiplicities, tomographic images, and fission density maps (images).

NMIS has been performing fast neutron radiography and tomography measurements since 2004. Fission mapping, a new capability to image the fission locations in an object, is under development. The images made at INL were the first NMIS fission mapping measurements of objects containing fissile material; prior measurements had been done on objects containing depleted uranium (DU) (fissionable).

MEASUREMENT TECHNIQUES

For these measurements NMIS employed several types of imaging using its 14 MeV neutron source: transmission radiography, transmission tomography, and induced-fission emissions mapping (imaging).

The system performs combined transmission and induced-fission imaging; Figure 1 shows its components. A D-T neutron generator, on the left side of the figure, creates interrogating 14 MeV neutrons by the $d + t \rightarrow \alpha + n$ reaction. Each neutron is created with an associated alpha particle that travels in the opposite direction. A pixelated alpha-particle detector is positioned so as to record the emission time and direction (by pixel) of interrogating 14 MeV neutrons emitted toward the object being inspected. There is uncertainty in the recorded direction due to the finite resolution of the pixelated alpha-particle detector and the finite source size. Figure 1 shows the neutron generator enlarged to illustrate its internal functioning; the actual DT generator fits inside a 7.5 cm diameter tube whereas the inspected object might typically be a 55 gallon drum.

Figure 1. Schematic diagram of a combined transmission and induced-fission imaging system based on a D-T neutron generator with an associated alpha-particle detector. The D-T generator is enlarged in this view to show the alpha particle coincident with each interrogating neutron.

The D-T neutron generator used in the measurements was a Thermo-Fisher Scientific API120 with an embedded alpha-particle detector. The generator's tritium target was reduced to 5 mm to decrease the size of the neutron source and to increase the imaging resolution. Consequently, the neutron production is reduced to 4×10^7 neutrons/second. The alpha detector is a cerium-doped yttrium aluminum perovskite (YAP:Ce) scintillator; it is viewed by Hamamatsu H9500

16 × 16 pixel photomultiplier tube (PMT). NMIS uses one 16-pixel row of the PMT to detect the alpha particle associated with an emitted neutron, thereby tagging the neutron’s direction (pixel number) and emission time (pixel pulse time).

The radiograph measurement is very similar to an x-ray: the system produces an image of the degree to which the object attenuates 14 MeV neutrons. NMIS uses the time and direction tagging of the neutrons to eliminate some neutrons that have scattered within the inspected object. Scattered neutrons that nevertheless arrive at some transmission detector blur the image. Like an x-ray, the radiograph is very useful, but in these measurements, which needed to be done quickly, they were used only to learn the position and overall shape of the object in the container. Thus they could be done quickly at low resolution and short measurement times.

Figure 2. NMIS scans in ways similar t tomographic scanners. The turntable (c the scanned container (e.g., a drum); th vertically positions the neutron generat

The transmission tomograph measurement is very similar to a medical computed tomography scan in that it images the interior of the inspected object. Figure 2 shows the NMIS configuration where transmission is being measured by the D-T neutron source (left) and the array of 32 2.5 × 2.5 × 10.2 cm fast plastic scintillators (right). The image resolution, which would otherwise be 32 pixels, is extended to 128 pixels by measuring the 32 detectors in 128 positions; the array is shifted by ¼ of the detector face width to create 4 times more detector positions than detectors. While this cannot truly quadruple the resolution, it works well in practice. NMIS images a single two-dimensional (2D) horizontal plane through the object; multiple images can be stacked to produce a 3D view. Like the radiograph, the image shows the degree to which the object attenuates 14 MeV neutrons, and the alpha detector is used to eliminate some scattered neutrons. The tomographic measurement requires that the object be scanned from many angles; in these measurements the objects were rotated to span 360° in increments of 6° or 12°.

The induced-fission emissions image measurement uses a form of emission tomography [2]. Like transmission tomography, it images the interior of the object, but the image reflects the intensity of induced-fission neutron emission from each pixel in the image. The fission imaging technique uses the fission detector array (shown in Figure 3) to count the neutrons produced by the induced fissions in the inspected object. These measurements used eight 25 × 25 × 8 cm scintillation detectors. The array needs to be close to the measured object for the sake of efficiency and far away from the source to reduce exposure to the noninterrogating portion

of its neutron emissions. The position chosen is as close to the object as practical on the side opposite the source. NMIS uses a form of multipl
gated multiplicity singles and doubles. That is, m
time interval in which induced fission neutrons a
the fission detectors. This interval begins around
neutrons have passed through the inspected objec
about the time that induced fission neutrons havi
through the detectors. Most of the gamma rays fo
the detectors before the time interval starts. NMIS
liquid scintillators to further reduce gamma-ray c
scintillators and rely solely on time of arrival to d

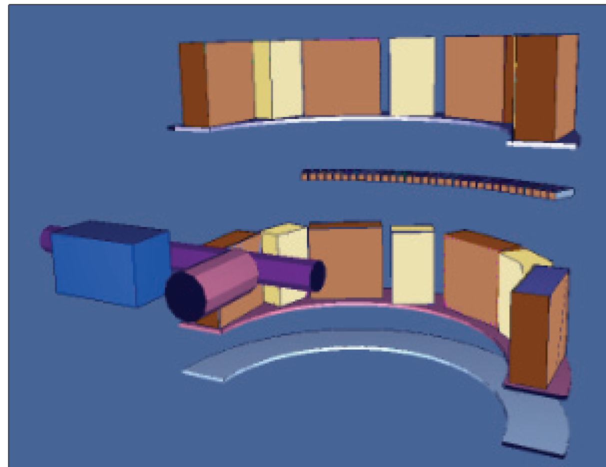


Figure 3. The fission detector array is two rows of four 25 × 25 × 8 cm scintillation detectors (dark brown) and plastic shielding (light brown) to decrease crosstalk. The D-T generator (front left) and transmission detector array (between the fission detector rows) are also shown.

The fission imaging relies totally on the D-T generator's embedded alpha detector to determine the direction the interrogating neutron took and thereby what part of the object was interrogated. The embedded alpha detector has only 16 pixels, so the resolution of the fission image will be limited. In addition, some neutrons will have scattered out of the object region indicated by their original direction when they induce fission, which will also blur the image.

NMIS can produce a fission mapping image from the multiplicity singles or the multiplicity doubles [2]. The advantage of using singles is that there are many more late singles counts than coincident late doubles counts, so the measurement can be done faster or with more statistical precision. (The fission detectors' neutron efficiency is about 2% for a ^{252}Cf source in air at the center of the target position.) The advantage of using doubles is that the probability that a double comes from fission is higher than the probability that a single comes from fission. For example, a single could be a D-T generator neutron that has scattered in such a way that it arrives at the fission detectors in the multiplicity counting time window.

NMIS makes the transmission and fission emissions image measurements simultaneously during a scan, imaging the same horizontal plane. The measurement requiring the most time is the induced fission doubles, followed by the induced fission singles. The time used for a scan was about 2 hours. This time could be shortened if a higher-flux DT/ associated particle imaging (API) generator were available. Although such a generator is thought to be possible with existing technology, none has been built yet. The scan time was conservatively long in all but one case. Given the long scan time, we selected one height to image a horizontal plane through the object.

MEASURED OBJECTS

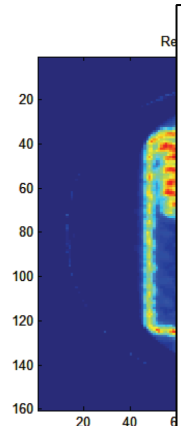
Each measured object is a collection of ZPPR fuel plates configured by INL personnel [1]. Each measured object consists of some fissile material of interest shielded by some other material(s), all of which are contained in a cubical aluminum box (Figure 4). The interior dimensions

of the aluminum box are 8 ×8 × 8 in.; the walls are ⅜ in. thick. The container serves to conceal the materials inside; the contents were not disclosed until all measurements were complete.

ORNL measured unknown objects IO 7, 9, and 10. ORNL also measured object 3, which was declared to contain plutonium-aluminum (PuAl) plates in a sealed aluminum clamshell container instead of an IO box.

MEASUREMENT RESULTS

Figure 5 shows the transmission image of object 3. The object was in an aluminum clamshell containing PuAl plates with dimensions of 2 × 2 × ⅛ in. The image is a good illustration of the transmission imaging resolution. The clamshell was initially declared to be full of PuAl plates. However, the NMIS measurement showed that the container was half full. Thus the object was actually not known, and the measurement was a double-blind test of the measurement systems.



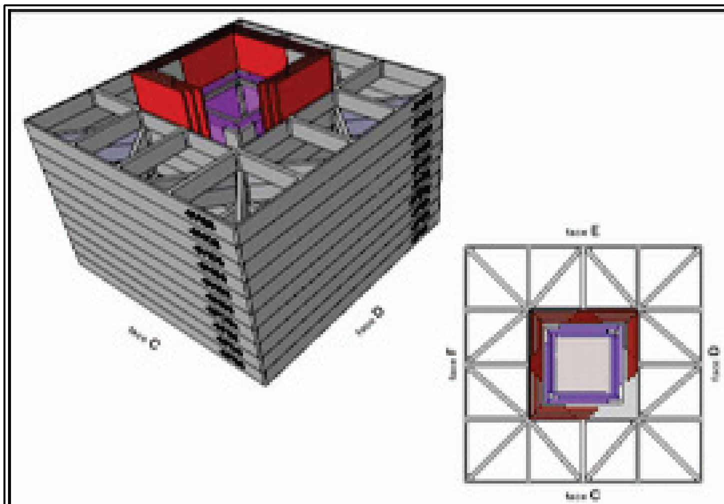
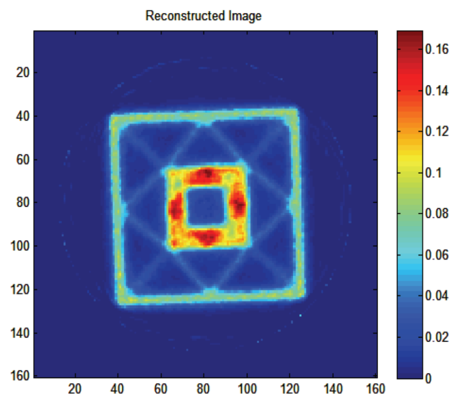


Figure 6. The configuration of IO 9 is an interior Pu box (purple), surrounded by an HEU box (red), and aluminum support structure/spacers (gray). One corner of the HEU plates is removed to show the Pu interior. All was contained in an aluminum IO box (interior dimensions: 8 ×8 × 8 in.).

IO 9 is a configuration of HEU shielding Pu. Figure 6 shows a 3D view of the object, partially assembled, and a top view of a horizontal layer. One HEU corner is removed to reveal the inner Pu box. Other details of note are the Al pieces in corners of the Pu box and the alternating Al and HEU plates in the HEU box.

The tomographic scan of IO 9 consisted of 60 projections (measured every 6° around the object) that collected 102 minutes of data.

The transmission imaging result in Figure 7 shows the aluminum support structure present at the height imaged and the fissile material boxes in the center. It shows the lower-attenuating aluminum pieces in the corners of the Pu box. The HEU box has aluminum plates interleaved between the HEU plates everywhere except at the center of the box sides; the transmission image shows the difference in attenuation between the Al + HEU and HEU regions.



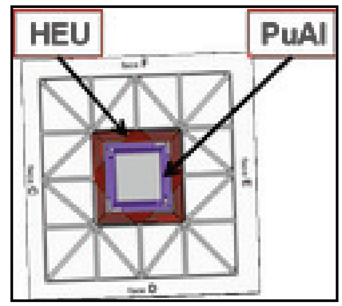


Figure 8 shows the fission mapping results for doubles and singles as well as a composite of the singles result and the transmission image. The doubles and singles results are similar if allowance is made for the poorer counting statistics of the doubles data. Although limited to 16 pixels (alpha detectors), the singles result identifies the fissile area very well; the transmission image has 128 pixels.

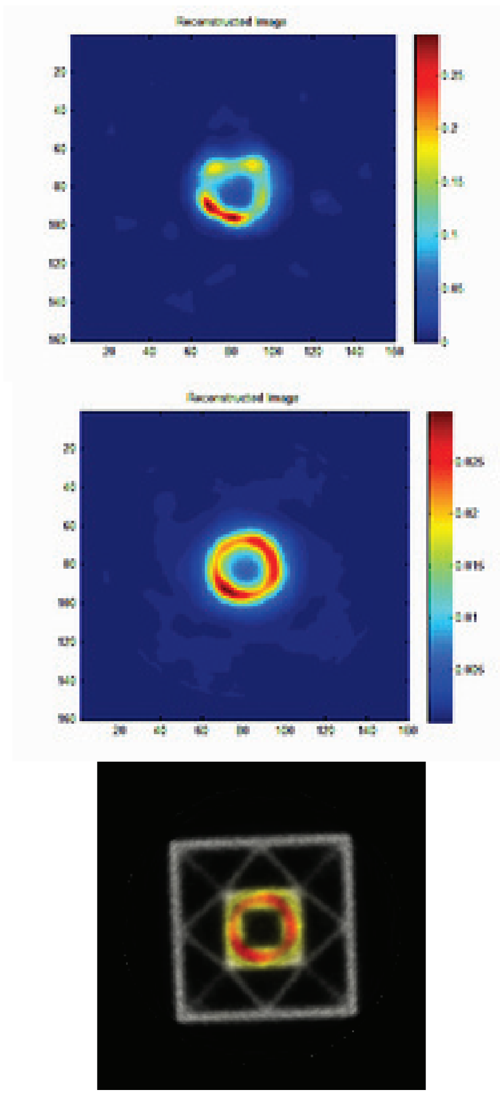


Figure 8. IO 9 fission mapping results from doubles (top), singles (middle), and singles overlaid on the transmission image (lower).

IO 7 is a configuration of HEU shielded by DU (Figure 9). There are Al pieces in corners of the HEU box. While the top view shows the Al support framework from several levels, NMIS scanned at a height with no supports.

The tomographic scan of IO 7 consisted of 60 projections (measured every 6° around the object) that collected 123 minutes of data.

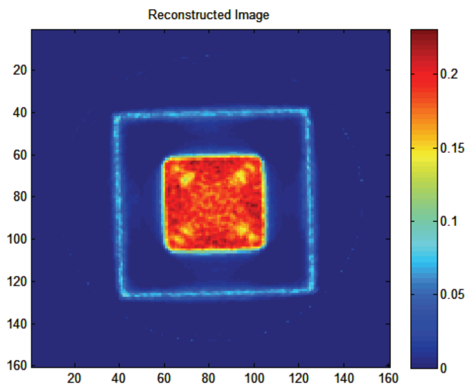
The transmission imaging result in Figure 9 shows the fissile material boxes in the center. The lower-attenuating aluminum pieces in the corners of the fissile materials box are visible.

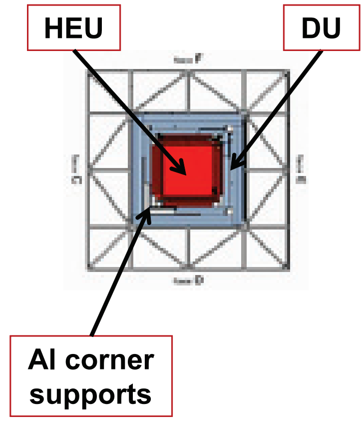
Figure 10 shows the fission mapping results for doubles and singles as well as showing the singles result overlaid on the transmission image. As expected, the fission map is peaked in the HEU location in the center. The DU is fissionable by high-energy neutrons and will show about half the density of fissions of HEU.

IO 10 is a configuration of high-density polyethylene (poly) and DU shielding Pu (Figure 11). The Pu box is hollow, and there are Al pieces in its corners. The poly is surrounded by an Al framework, which is mostly hollow. In the drawing it appears to be solid because the edge of the frame is shown.

The tomographic scan consisted of 30 projections (measured every 12° around the object) that collected 51.2 minutes of data. Compared with the other measurements, this measure acquired half of the projections (lower resolution) and half of the measurement time (poorer statistics).

The transmission imaging result (Figure 11) clearly shows the object's major components. The lower attenuation of the Al pieces in the Pu box corners is also visible. The Al framework around the poly is so sparse that it images as a gap around the poly.





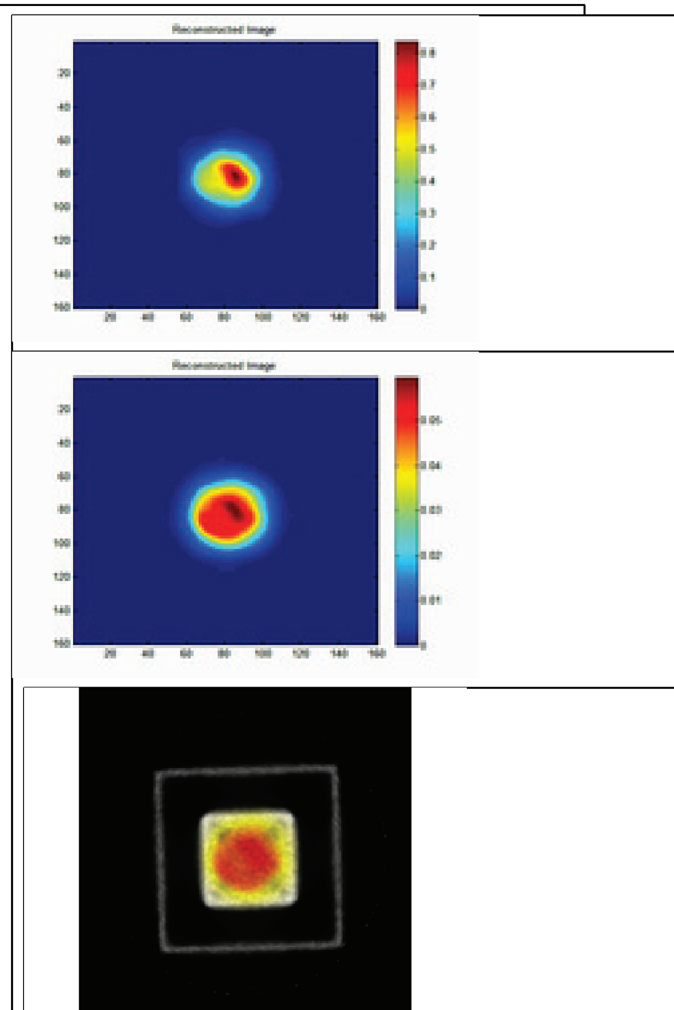


Figure 10. IO 7 fission mapping results for doubles (top), singles (middle), and singles overlaid on the transmission image.

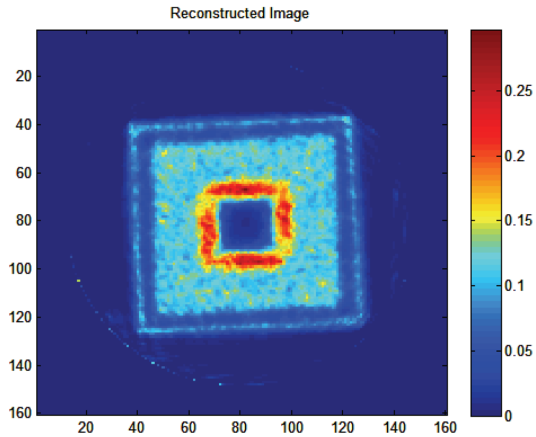


Figure 12 shows the fission mapping results for singles as well as showing the singles result overlaid on the transmission image. The poly shielding is an effective shield of 14 MeV neutrons entering the object and an even more effective shield of outbound induced fission neutrons. This shielding and the halving the acquisition time limited the detection of induced fission doubles to the extent that the fission mapping result for doubles was not useful. The fission map for neutron singles attributes fissions to much of the plastic volume. Two phenomena can contribute to this result. First, the singles fission mapping method will mistakenly accept a D-T source neutron as evidence of fission when the neutron has been widely scattered (by the hydrogen in the poly) so that its late arrival at a fission detector falls within the fission multiplicity time window.

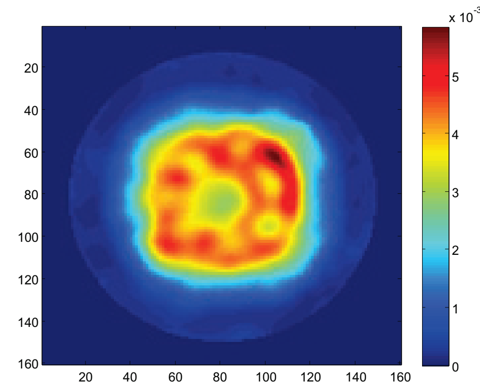


Figure 12. IO 10 fission mapping results for singles (top) and singles overlaid on the transmission image.

A measurement long enough to produce a converged doubles result would avoid this effect. Second, D-T source neutrons entering the poly region can be scattered into the fissile material, where they will likely induce fission. The fission is attributed to the poly region because the D-T generator's API detector is reporting the initial direction of the neutron (toward the poly region). Since transmission imaging is able distinguish the poly and fissile metal regions, and it is the scattering between them that contributes to this result, an enhanced analysis that uses the transmission result and accounts for scatter should be beneficial.

SUMMARY

ORNL took advantage of an opportunity to test neutron imaging on fissile objects at INL. ORNL's NMIS system currently performs tomographic transmission/attenuation imaging; tomographic emission (induced-fission radiation) imaging is under development. These were the first measurements of fissile materials performed with this new capability.

The measurements demonstrated the resolution of the transmission imaging. The ability to detect small aluminum parts among the fissile metal plates demonstrated the ability to contrast different materials in close proximity.

The measurements demonstrated the system's ability to map fissions to regions in the interior of an object. Performance was good for most measurements, although improvements are possible. The low resolution of the D-T API imaging detector (16 pixels) limits the fission map resolution (although overlaying the fission image on the higher-resolution transmission image aids the interpretation). The fission detector array employed has a fission neutron efficiency of about 2%, which limits the measurement of induced-fission neutron doubles. Gains could be made by increasing their solid angle, either by moving the detectors closer to the object or by adding detectors. The increased efficiency would improve the measurements of objects shielded with hydrogenous materials as it would improve the fission mapping doubles result. Enhancing the analysis to better account for neutron scattering would improve the ability to distinguish between fissile material regions and adjacent regions of nonfissile material that scatter D-T source neutrons into fissile material region.

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