EXAMINING ELEMENTAL DISTRIBUTIONS IN THREE DIMENSIONS USING TWO-DIMENSIONAL MICRO-XRF IMAGING

Christopher G. Worley, Patrick T. Martinez, Kevin J. Kuhn, and Lav Tandon
Los Alamos National Laboratory, MS G740, Los Alamos, NM 87545

ABSTRACT

Micro-X-ray fluorescence was used to nondestructively examine a metal coupon described by the submitter to be an alloy of aluminum and uranium. In addition to collecting two-dimensional elemental distribution maps of the coupon surface, three-dimensional elemental information was inferred based upon attenuation differences between the high energy U L and low energy U M X-rays. Only aluminum was detected on one end of the coupon, and no uranium was observed in the surface-sensitive U Mα MXRF image from any region of the coupon surface except along a cut edge facing the detector. Uranium was detected in the higher energy Lα image everywhere except on the pure aluminum end. Thus, the data indirectly indicated that the sample consisted of aluminum cladding surrounding a uranium layer. The sample was then remounted to image a cut edge, and the aluminum cladding geometry was confirmed.

INTRODUCTION

Micro-X-ray fluorescence (MXRF) is often used to map elemental spatial distributions across a surface and near the surface in two dimensions (Nichols et al., 1987; Janssens et al., 2000). Confocal MXRF uses the focal point overlap of capillary optics aligned with the source and detector to provide nondestructive three-dimensional depth profiling in addition to lateral elemental distribution information (Ding et al., 2000; Kanngießer et al., 2003; Vekemans et al., 2004). However, a two-dimensional (2-D) mapping MXRF instrument can also be used to determine certain 3-D information by using multiple elemental X-ray lines having different energies and escape depths (e.g. L and M lines).

A 2-D mapping MXRF instrument was used in the present study to nondestructively characterize a metal coupon composed of aluminum and highly enriched uranium. The coupon was intended to be used as a test sample to demonstrate the quantitative capabilities of multiple actinide analytical chemistry techniques, which required extracting a specimen followed by destructive chemical analyses. Prior to the specimen extraction, the sample was nondestructively examined with MXRF to determine the chemical composition and the presence of any heterogeneity. This information was then used to determine the best course of action to take for extracting the specimen for the destructive analyses.

The uranium and aluminum distribution was mapped across the coupon to look for any spatial variations in these matrix constituents, and the sample was determined to be markedly heterogeneous. Images collected using both the uranium L and M X-ray lines revealed depth-dependent information about the uranium distribution based on the difference in absorption
length between these two lines. To directly confirm the 3-D spatial distribution, the sample was then remounted to image a cut edge.

EXPERIMENTAL

Instrument hardware. An EDAX Orbis PC MXRF instrument was used for this work. All data were acquired using a polycapillary optic producing a 30 μm diameter X-ray beam (full width at half maximum intensity measured at the Mo Kα energy). The instrument uses a 50 W rhodium X-ray anode and a 30 mm² silicon drift detector. The source was operated at 45 kV and 950 μA to collect the images and spectra. For many of the analyses, source filters were used to improve the signal-to-background and remove the source Rh L scatter peaks. All analyses were performed with the instrument chamber at atmospheric pressure due to the radioactive nature of the sample.

Instrumental Data Collection Parameters. Full spectral mapping was used to collect the MXRF images whereby a complete spectrum was collected at each image analysis point. Background-subtracted net intensity images were used. Parameters used to collect the MXRF data in figures 2 and 3 were as follows: 25 μm Al source filter, 370 ms live time per analysis point, 30.19 mm x 11.27 mm imaged area, and 612 x 228 image points. Each spectrum in figure 4 was collected for 30 s of live time using the 25 μm Al source filter. The image in figure 7 was collected using the following parameters: 1 s live time per analysis point, 2.36 mm x 1.85 mm imaged area, and 128 x 100 image points. The image in figure 8 was collected using the following parameters: no source filter, 100 ms live time per analysis point, 29.99 mm x 18.37 mm imaged area, and 512 x 400 image points. (For display purposes, the original MXRF image was cropped in the y direction to remove the extra black background.) The spectra used for fundamental parameters quantification were acquired for 60 sec live time using a 250 μm Al source filter to remove interfering diffraction peaks.

Sample Preparation. A metal coupon containing aluminum and highly enriched uranium was submitted for MXRF analysis. The edges had been cut by a sheet metal shear prior to receipt. For horizontal surface imaging, the coupon was sealed in a plastic bag with a window cut out and 4 μm thick Prolene (Chemplex) taped over the window. The coupon was analyzed through the Prolene window. To analyze the coupon cut edge, the sample was mounted between two pieces of foam inside a plastic cup with the cut edge facing up toward the open end of the cup, and the sample was sealed in the cup with 4 μm thick Prolene.

RESULTS

The submitter described the sample coupon as an alloy of highly enriched uranium and aluminum and requested MXRF be used to examine any possible elemental heterogeneity. A piece was to be extracted from the coupon for various analytical methods to demonstrate the quantitative capabilities of these techniques. To nondestructively determine the extent of any uranium heterogeneity before cutting the sample for the other analyses, the coupon surface was imaged by MXRF. Figure 1 is a picture of the coupon with a red box denoting the region imaged by MXRF. (The light colored rectangle surrounding the sample is tape used for mounting the sample containment bag to the instrument stage.)
Figure 1. Visible image of sample coupon.

Figure 2 is an overlay of the uranium Lα (red) and aluminum Kα (blue) images collected from the red boxed region in figure 1. The pink shaded area indicates overlap of these two constituents. No uranium was detected on the far left side of the coupon; only aluminum was detected. Figure 3 compares the uranium Lα (left) and uranium Mα (right) images. No signal was detected in the more surface-sensitive uranium Mα image (~3.2 keV) from any area of the coupon except along a cut edge facing the detector (top edge in the figure). This edge had been cut with a sheet metal shear exposing the interior material. In contrast to the U Mα map, uranium was detected in the higher energy Lα map (~13.6 keV) except on the left end of the coupon which contained aluminum and a few impurities (see later discussion). The uranium Lα absorption length in aluminum is ~350 μm, whereas the U Mα absorption length is only ~5 μm. Analysis of the reverse side of the coupon revealed the same effect. Thus, these results indirectly indicated that the sample consisted of aluminum cladding surrounding a uranium layer, and the U Mα X-ray energy was insufficient to penetrate the aluminum cladding.

Figure 2. Overlay of coupon aluminum Kα (blue) and uranium Lα (red) MXRF images.
Figure 3. Coupon uranium Lα image (left) and uranium Mα image (right).

Figure 4 shows point spectra collected from the top cut edge (red spectrum) and from the uncut middle of the coupon (blue spectrum). A prominent uranium M peak series was detected from the cut edge facing the detector, but no uranium M peaks were present in the spectrum collected from the middle of the coupon. Since a buried uranium layer would be exposed to the detector on the cut edge, and the energies of the U M peaks were likely too low to penetrate the aluminum cladding, this was further evidence of a uranium layer located inside aluminum cladding. A comparison of the uranium Lα peak intensity from the cut edge and the coupon middle indicated that although the 13.6 keV Lα X-rays were energetic enough to penetrate the aluminum cladding, the intensity was attenuated approximately five fold passing through the cladding.

Figure 4. Point spectrum from coupon cut edge (red) and uncut middle (blue).
To confirm the presence of aluminum cladding surrounding a uranium layer and to measure the uranium and aluminum layer thicknesses, the sample was remounted between two pieces of foam (dark regions in figure 5) to image a cut edge (silver colored area in figure 5). Figure 6 is a visible image of a section of the coupon cut edge with a red box indicating where the sample was imaged by MXRF. As predicted a uranium-containing layer (green U L\(\alpha\) image in figure 7) was detected sandwiched between two aluminum layers (red in figure 7). The uranium M\(\alpha\) image (blue in figure 7) did not reveal the full uranium layer thickness because the aluminum smeared when the coupon edge was cut and covered part of the exposed uranium layer. This smeared aluminum absorbed the lower energy U M\(\alpha\) X-rays, and only the higher energy U L\(\alpha\) X-rays could penetrate it. Based on measurements made using the U L\(\alpha\) image, the uranium layer was determined to be ~550 \(\mu\)m thick.

![Figure 5. Coupon remounted in a cup with cut edge facing up.](image1)

![Figure 6. Visible image of a section of the coupon cut edge.](image2)
The entire coupon cut edge was then imaged to view the complete uranium layer (green in figure 8). In addition to detecting the aluminum (red in figure 8) and uranium layers, no uranium was detected on the far left side of the sample, corroborating what was observed when the top surface was imaged (figure 2). (Note: The aluminum intensity from the bottom layer was less than that from the top layer because the cut edge was not perfectly flat, and the top and middle surfaces were closer to the polycapillary X-ray beam focal plane than the bottom surface plane.)

**Figure 7.** Overlay of MXRF aluminum K\(\alpha\) (red), uranium L\(\alpha\) (green), and uranium M\(\alpha\) (blue) images acquired from the red boxed area in figure 6.

**Figure 8.** Overlay of MXRF aluminum K\(\alpha\) (red) and uranium L\(\alpha\) (green) images acquired from the entire cut edge of the coupon.
Spectra were also acquired from the uranium and aluminum layers for elemental quantification using a standardless fundamental parameters approach. The uranium layer only contained ~9 wt % uranium. The remaining balance was mostly aluminum with ~0.2 wt % of miscellaneous impurities. The boundary between the smeared aluminum overlayer (created when cutting the edge as previously discussed) and the exposed uranium layer was clearly visible using the instrument high magnification camera. Uranium Mα X-rays were not detected from the area where the aluminum overlayer was present because these X-rays were too low in energy to penetrate the aluminum. However, prominent U M X-ray peaks were detected from the exposed uranium layer. Thus, this exposed region was used to collect the point spectrum to determine the elemental composition of the uranium-containing layer. The aluminum layer contained ~98.7 wt % aluminum, no uranium, ~0.6 wt % calcium, ~0.5 wt % iron, and ~0.2 wt % of miscellaneous constituents. Based upon the MXRF imaging results, this aluminum end was removed, and the remaining sample (demonstrated by MXRF to be a relatively uniform three layer material) was dissolved for the additional chemical analyses.

CONCLUSION

A coupon containing aluminum and highly enriched uranium was submitted for nondestructive analysis using a 2-D imaging MXRF instrument to look for any elemental heterogeneity prior to extracting a piece for destructive chemical analyses. Comparison of the uranium Lα and Mα images indirectly indicated that the uranium was buried beneath aluminum cladding. The sample was then remounted to image a cut edge to confirm this inferred geometry, and a uranium-containing layer was detected surrounded by the aluminum cladding. The uranium layer was quantified using standardless fundamental parameters, and it was found to contain mostly aluminum and only ~9 wt % uranium. In summary, this work demonstrated the ability to nondestructively determine 3-D elemental distribution information using a conventional 2-D imaging MXRF instrument.

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REFERENCES


