LA-UR-23-32563

Approved for public release; distribution is unlimited.

Title:	X-ray Analysis of Actinides using hiRx
Author(s):	Patterson, Brian M. Hunter, Bryan Keeney Cordes, Nikolaus Lynn White, Joshua Taylor van Veelen, Arjen
Intended for:	Report
Issued:	2023-11-03

Los Alamos NATIONAL LABORATORY

X-ray Analysis of Actinides using hiRx

Brian M. Patterson, Bryan K. Hunter, Nikolaus Cordes, Joshua White, Arjen van Veelen

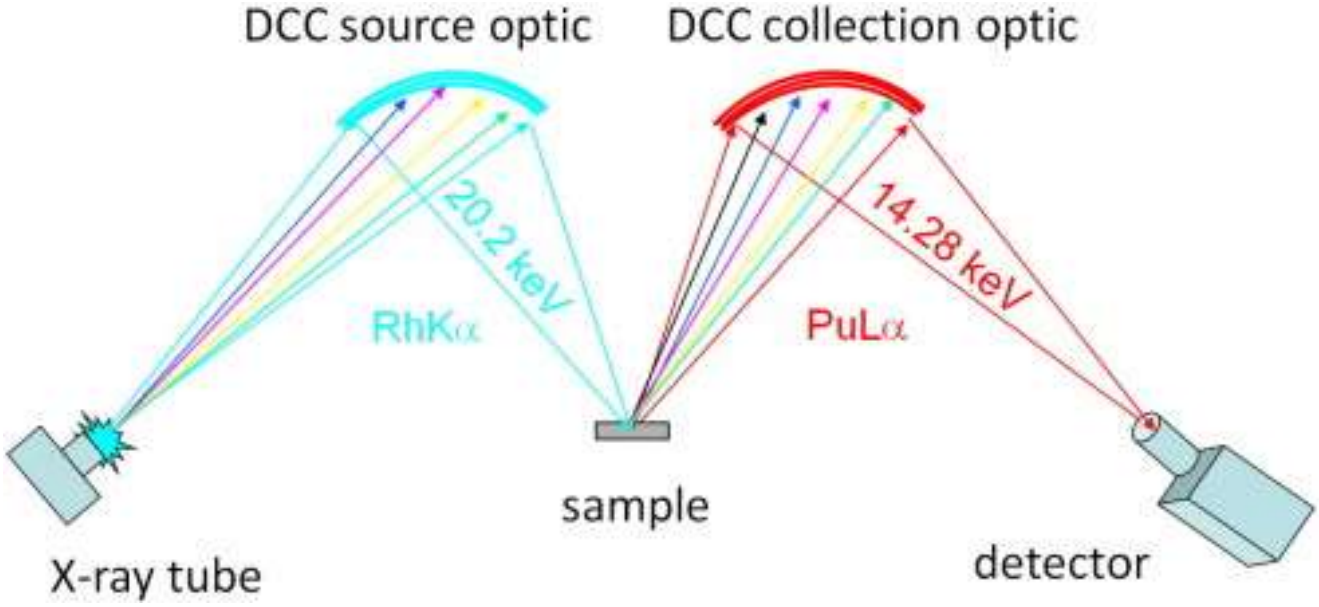
November 2, 2023

Background: Reconstitution

- A novel X-ray fluorescence instrument was constructed in C-div a few years ago.
- Instrument designed to map specimens, monochromatically, for Pu, U, Y/Sr, Np, Th.
- This is achieved by using monochromatic X-ray optics, with the benefit of increased signal-to-noise is when compared to instruments with polychromatic X-ray beams.
- Due to a retirement, the instrument sat dormant for several years. MST-DO provided funding to move the instrument; TED funding resurrect it and train a student in its use, alignment, and data processing.

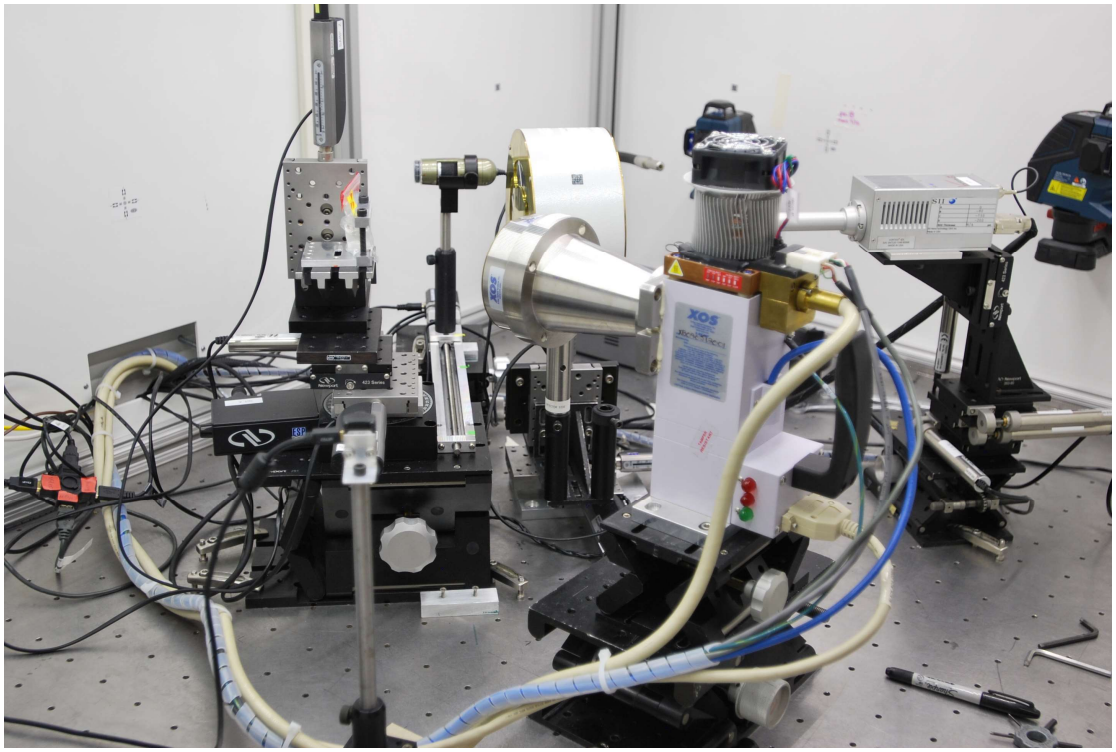
- **Demonstration goal: Locate particles and impurities; map the particles.**

Example samples mounted on glass slices and wrapped in Kapton tape.



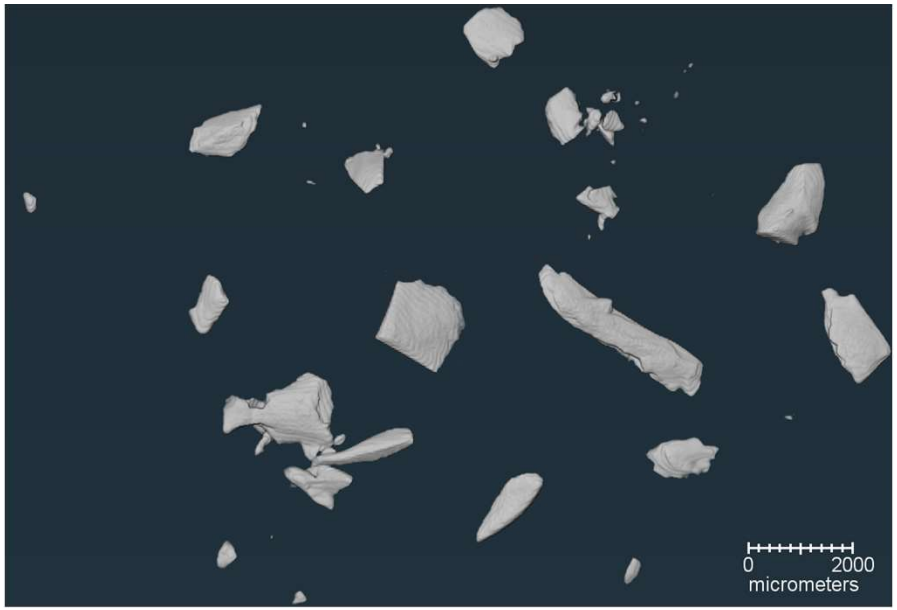
Demonstration

- Sample:
  - Samples which were already in the possession of MST-8/-16 were transported to 35-213.
  - The mounting and moving of the samples required the student to become proficient in working with the radiological control technician.
- Instrument:
  - Motorized translation stages were remounted to simplify homing and alignment between optics.
  - The optics were successfully realigned.
  - Elemental spectra and maps were collected.

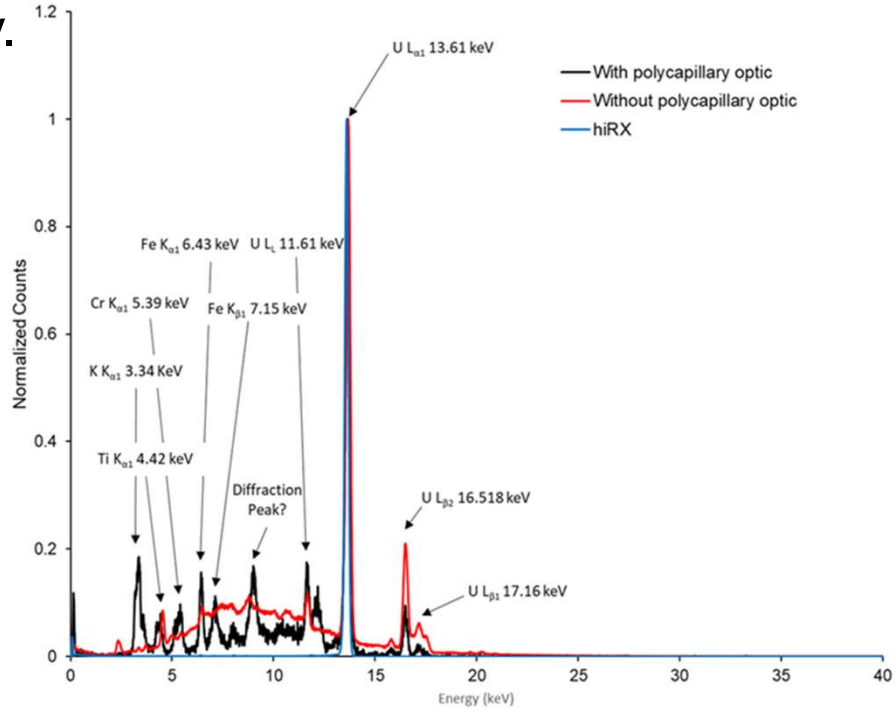


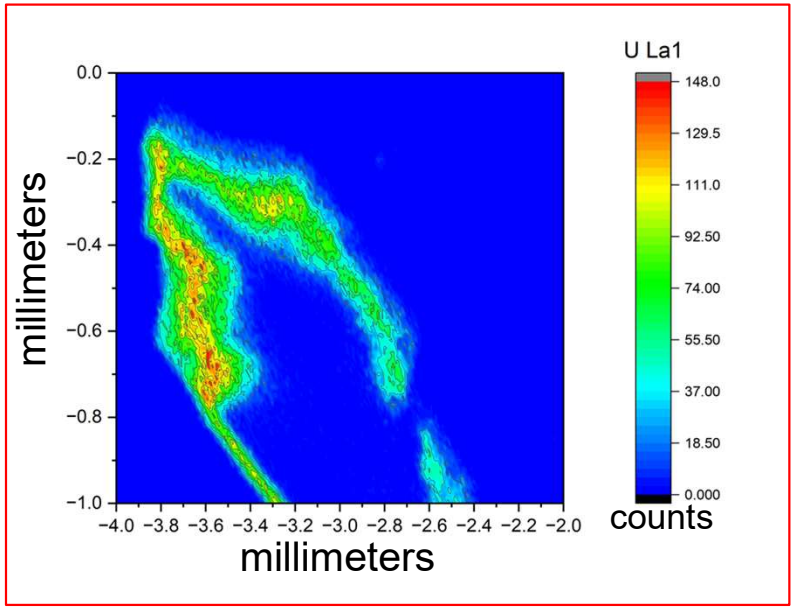
TED Funding Results

- A sample of a mixed ceramic oxide particles was imaged with X-ray CT and mapped with XRF and the reconstituted hiRx system.
- An overlay of the elemental spectrum (below), demonstrating the power of the technique to remove spectral noise. Notice that the blue spectrum (collected on the hiRx) is free of any other signal, greatly increasing the instruments sensitivity.



3D X-ray CT rendering of the particles.

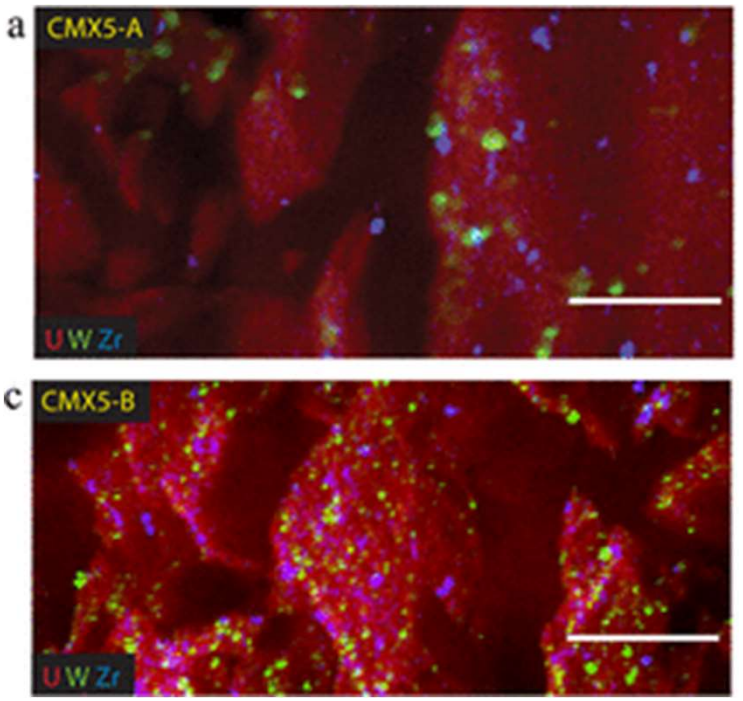


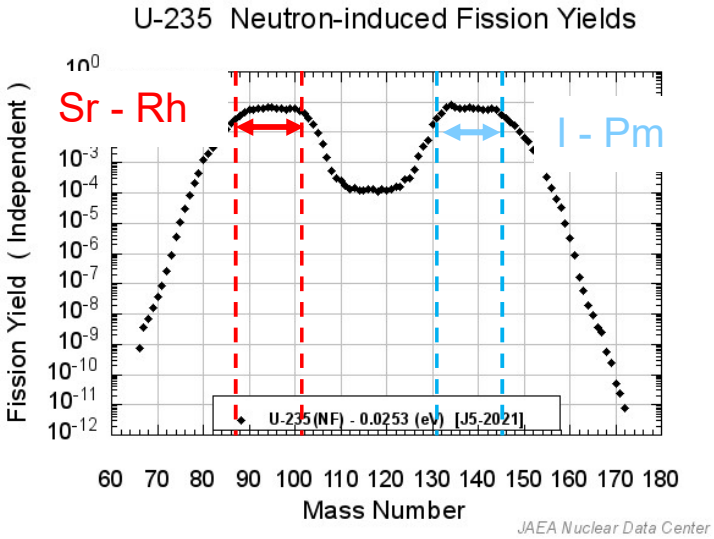


2D map of the uranium signal vs. location. Note the lack of image noise.

Impact

- The resurrection of this high-resolution X-ray mapping instrument has potential impact for nuclear fuel processing and safeguards.
  - Possible integration into safeguards required for the Low-Enriched Fuel Fabrication Facility (LEFFF)
- Integrating the instrument with 3D imaging techniques will allow for nondestructive investigations of nuclear fuel-cladding interactions and other post-irradiation examinations
  - Fuel migration into aluminum claddings of U-Mo research reactor plate fuel
  - Pu in spent nuclear fuels
  - Investigations of Pu and other elements in soil samples
  - Synchrotron experiments can be used to increase resolution (spatial and spectral)
- The demonstration of this capability could lead to further technique development and broader community adoption for the detection of high-profile elemental species that are previously difficult to obtain.
- Identification of particles/impurities in complex matrices (e.g. soils) where origin is of importance, such as nuclear forensics, site cleanups, etc..
  - Examples of overlapping emission lines: U Lα and Rb Kα, and Sr Kα and Pu Lα.





Future directions (experimentally)

Laboratory characterization:

- Brighter X-ray source (Sigray)
  - Microparticle embedded in diamond; carousel for different anode particles
  - 10's of microsecond dwell instead of multi seconds
  - Sigray Attomap XRF instrument with the ability to include our optics ($1.3 M); on weapons investment list.
  - Sigray QuantumLeap XAS where we will build in our mapping stage and optics to obtain both maps and chemistry.
- Integration of full spectral elemental maps with X-ray CT images.
  - Python programming

Synchrotron Characterization

- Detector optic to beamline (2-ID @APS or 4-BM @NSLS II)
  - 3D mapping using monochromatic synchrotron beam

Submit a simple technique paper to Microscopy Today magazine.

Future directions scientifically

- We will submit an ER proposal and obtain synchrotron beamtime to XRF map materials using the optics.

- Develop a compact portable spectrometer with various crystals that can be mounted on any beamlines at synchrotrons around the world. Our goal is to transform any standard imaging/XAS beamline in a high energy resolution imaging/spectroscopy beamline

- Study dilute dopants, impurities, and/or fission gas products in fuel/metals etc.
  - Research questions: are these correlated, hotspots, particles, or diffuse in the matrix? Correlations and structural functional relationships?

Appendix 1: Chart

- Chart summary of project requested by Program Manager Duncan McBranch

Monochromatic X-ray fluorescence: Non-destructive identification and mapping of trace actinides in soils and ceramics for nuclear processing and forensics applications

BACKGROUND & MOTIVATION

Identifying trace contaminants on actinides can be difficult.

- Spectral contamination due to X-ray scatter or natural radiation reduces the signal to noise ratio (SNR).
- Spatial information and correlation of contaminates may be critical to forensic investigations

INNOVATION

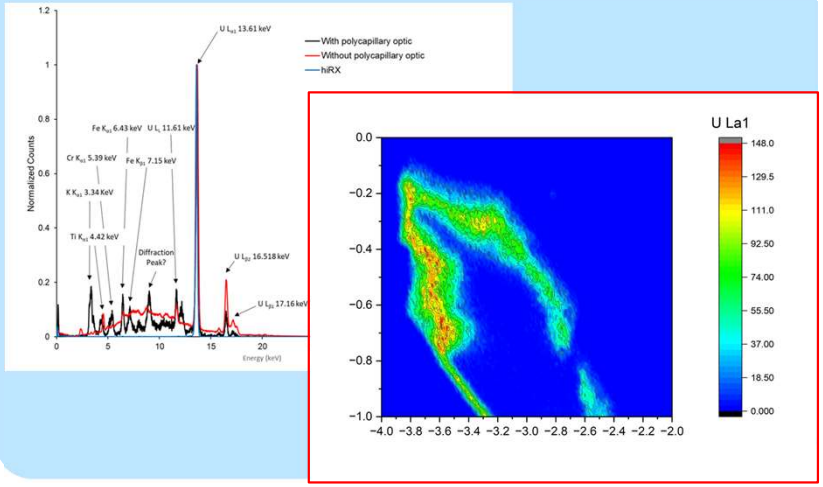
Monochromatic X-ray fluorescence (hiRx)

- Specialized optics (doubly curved crystals) can spatially and spectrally confine the beam.
- Only one wavelength of X-rays are transmitted to the detector, filtering out any scatter or off energy signals; greatly improving SNR and lowering detection limits.
- Confined beam also critical for mapping the location of contaminants. Resolution is approximately 50 micrometers.

DESCRIPTION

We have demonstrated 2D elemental mapping of uranium in particles along with 3D X-ray CT imaging.

- Specialized optics only transmit the signal for the element of interest.
- The beam is ~50 micrometers in size; the sample is rastered under the beam to create an elemental map.
- X-ray CT is also used to create a 3D image of the particles.
- Technique was previously demonstrated with plutonium
  - McIntosh, K.G., et al., *Laboratory-based characterization of Pu in soil particles using micro-XRF and 3D confocal XRF.* J. Analy. Atom. Spect., 2015.
- Graphic is a 2D map based upon the elemental signal of uranium. Red and black spectra shows the signal from the sample with a lot of scatter in the baseline. The blue spectrum shows only uranium signal with no collected baseline.



ANTICIPATED IMPACT

Novel method of X-ray analysis

- Technique is not commercially available for actinide analysis.
- Potentially useful for:
  - Trace actinide element correlation analysis
  - Forensics
  - Waste
- Current instrumentation is slow, hours to days to map a sample.
  - Brighter X-ray sources and more sensitive detectors are available.

PATH FORWARD

What is your path to mission benefit?

- Reach out to PM's to further develop technology.
- Example of next steps:
  - Develop automated map processing; correlate to other imaging techniques
  - Upgrade source and detector.
  - Examine technology at a synchrotron

Potential End Users:

- IAEA, Next-gen Safeguards, Weapons program

Point of Contact: Brian M. Patterson, MST-7, (505) 606-0831, bpatterson@lanl.gov

Los Alamos NATIONAL LABORATORY