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Title:	Improved Accuracy of Spent Fuel Pu Assay
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Form 836 (7/06)

Improved Accuracy of Spent Fuel Pu Assay

George J. Havrilla,

Los Alamos National Laboratory

X-ray optics and synchrotron sources offer new opportunities for actinide characterization. X-ray optics provides intense focused x-rays for excitation using low power x-ray tubes. The use of doubly curved crystals (DCC) for monochromatic excitation and collection of emitted x-rays from the specimen has resulted in the development of a new prototype instrument, hiRX, high Resolution X-ray. hiRX is based on monochromatic wavelength dispersive X-ray fluorescence (MWDXRF) technology, which uses DCCs to enable selective and sensitive analyses of selected actinide elements. Synchrotron excitation offers monochromatic excitation with high intensity at high energy. Ultra high energy X-ray fluorescence UHEXRF has been demonstrated to detect uranium through 1.3 mm of Zircaloy shielding. The experiment uses 117 keV excitation and detects the U Ka line at 98.428 keV. Detection sensitivity is below 1 microgram through the 1.2 mm Zircaloy shielding. Both of these new approaches offer direct, sensitive analyses of actinides for safeguards applications.

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Office of Nonproliferation and Verification Research and Development

Global Safeguards Programs Review Meeting

GS2011

Improved Accuracy of Spent Fuel Pu Assay

May 10, 2011

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Improved Accuracy of Spent Fuel Pu Assay

Technology Roadmap

New approach for Pu assay by direct analysis using monochromatic wavelength dispersive X-ray fluorescence (MWDXRF) – NA22 R&D

- FY 09 – demonstrate MWDXRF feasibility, secure continued development – exceeded target of 10 ppm
- FY 10 – build MWDXRF prototype instrument for Th and U, obtained less than 1 ppm detection limits
- FY 11 – Advanced concepts for Pu assay using ultra high energy XRF; UHEXRF for through wall composition analysis – new paradigm

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2

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Technology Roadmap

- FY 11 - Advanced Concepts – synchrotron experiments exceeded expectations, demonstrated feasibility, justifies ultra high energy optic development for laboratory-based instrumentation
- FY 12 – demonstrate UHE oxidation state determination
- FY 13 – demonstrate UHE isotopic measurement
- FY 14 and beyond – transition technology to laboratory-based instrumentation, demonstrate UHEXRF capabilities and transfer technology for safeguards applications

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Technology Transfer to NA24 - hiRX

Successful demonstration of MWDXRF transitioned to **hiRX** (high resolution X-ray) with NA24

- FY 11 - demonstrate Pu detection limit at 1 ppm or less
  - Identify any spectral interferences for Pu detection
  - Preliminary sampling protocol using low volume sample deposition
  - Identify potential partners for lab-based instrumentation use

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Project overview

- Direct analysis of Pu using hiRX based on MWDXRF technology with the overall objective of improving the accuracy of Pu assay for application to nuclear fuel reprocessing safeguards

Goals

- Implement new X-ray optic technology for sensitive detection of Pu for safeguard applications
- Attain a sensitivity of at least 10 ppm for Pu detection
- Develop new sampling methodology to reduce sample size, improve safety and accuracy of Pu measurements

Improved Accuracy of Spent Fuel Pu Assay

Technical approach – Monochromatic wavelength dispersive x-ray fluorescence uses a novel X-ray optic technology to generate monochromatic excitation of the specimen and another optic to selectively collect the emitted x-rays of the target elements and direct them to the detector.

Key features

- Doubly curved crystal optics for excitation and collection
- Small spot excitation – several hundred micrometers
- Small sample requirements – 200 microliters or less
- Collection optic to reject background and collect only analyte signal

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Deliverables FY09 - completed

- Breadboard feasibility
- Analytical Sensitivity - exceeded 10 ppm target

Deliverables FY 10 – completed

- Prototype instrument built at LANL
- Detection of thorium and uranium at 10 ppm – exceeded
- Annual report

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Deliverables FY 11 – in progress

- UHEXRF of U will be demonstrated with a target of 10 ppm detection limit along with measured attenuation of U XRF signal with selected container wall materials.
- UHEXANES proposal for APS beam time submitted.
- One publication UHEXRF of U
- One presentation will be prepared on UHEXRF of U.

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Description of capability improvement to be addressed by project success - Safeguards issues:

- How much Pu has been recovered in a reprocessing facility? This will be achieved by direct measurement of plutonium using MWDXRF.
- The increased accuracy of the Pu assay can be applied to other Safeguards issues including:
  - **whether a reprocessing operation is consistent with safeguards?**
  - **how much Pu is unaccounted for in a safeguarded facility?**
- Each one of these issues can benefit from a more accurate Pu determination which directly affects the accuracy of the Pu assay for the facility.

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9

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Interaction with potential users:

JNFL

- Presented INMM work to JNFL visitors at LANL
- Presentation at Rokkasho, Japan
- significant interest in potential applications throughout plant, especially glass operation

JAEA – Mitutoshi Suzuki, Tech Devl & Support Office

IAEA

- Alain LeBrun – Department of Safeguards
- Andrew Monteith – Department of Safeguards
- IAEA Global Safeguards Symposium Nov 2010
- Participation in cooperative research program (CRP) on "Micro-analytical techniques based on nuclear spectrometry for environmental monitoring and material studies"

NA24

- Alex Sunshine, Steve LaMontagne and Kevin Veal

Potential for in-line analyses and portable, handheld instrumentation for survey applications and immediate identification of elements

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10

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Technical challenges for MWDXRF and SR experiments:

- achieving the desired detection limits with high dried mass solids for MWDXRF
- SR data analysis, over 11,000 spectra collected in 4 days
- Doing XRF on an XRD beam line

hiRX not yet deployed – need to develop laboratory-based instrument for hiRX

Need to initiate UHE optics development for UHEXRF lab-based instrument development

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11

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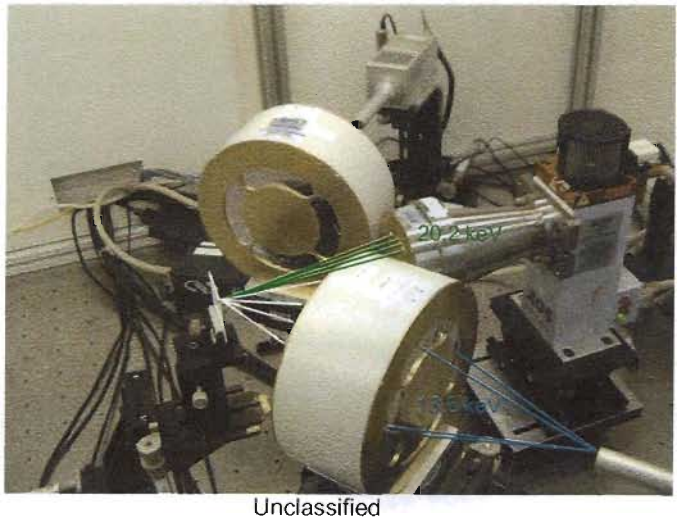
Top View of prototype hiRX with Th and U DCC optics



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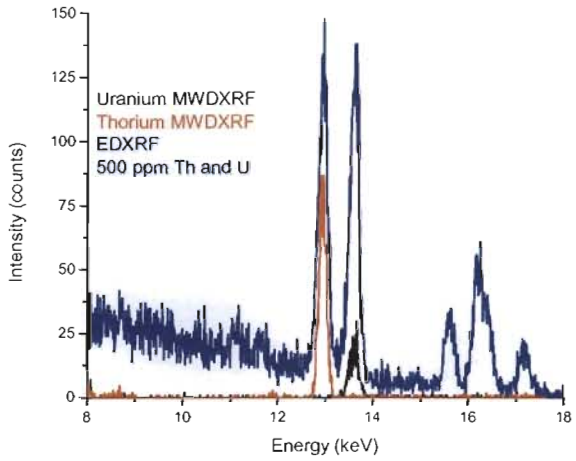

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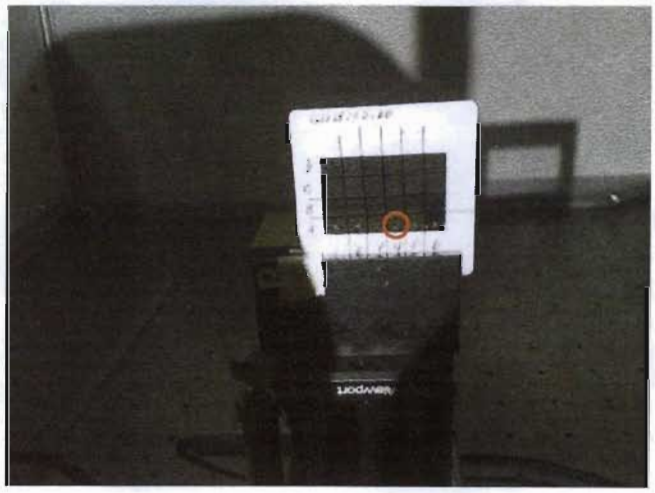

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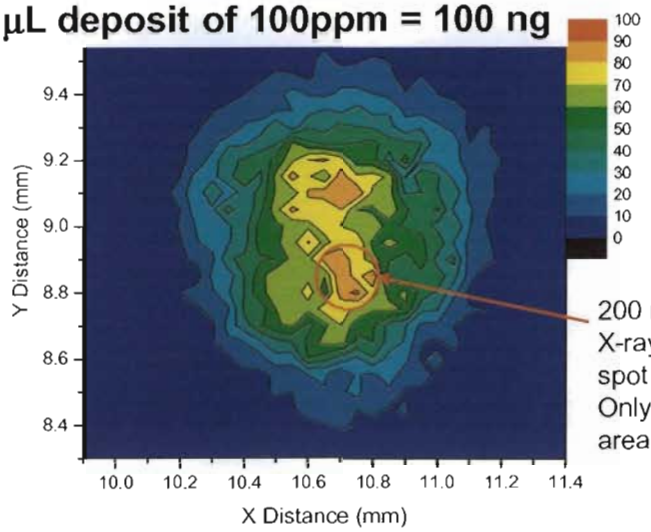
dU 1 μL deposit of 100ppm = 100 ng



200 micrometer X-ray excitation spot
Only ~4% of the area is excited

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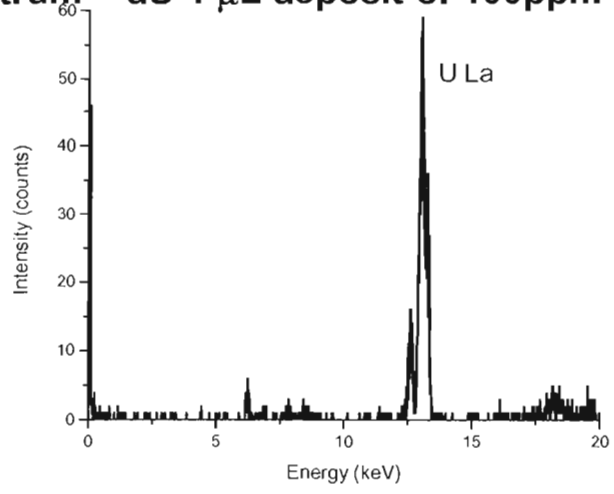
Spectrum – dU 1 μL deposit of 100ppm 100 ng



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Advanced Concepts

- Ultra high energy XRF – using excitation at 117 keV and higher to excite Ka X-ray emission of actinides, U, Pu, Cm
- Little to no spectral overlap
- Sensitive lines
- Energy penetration of container walls
- Direct nondestructive elemental analysis

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17

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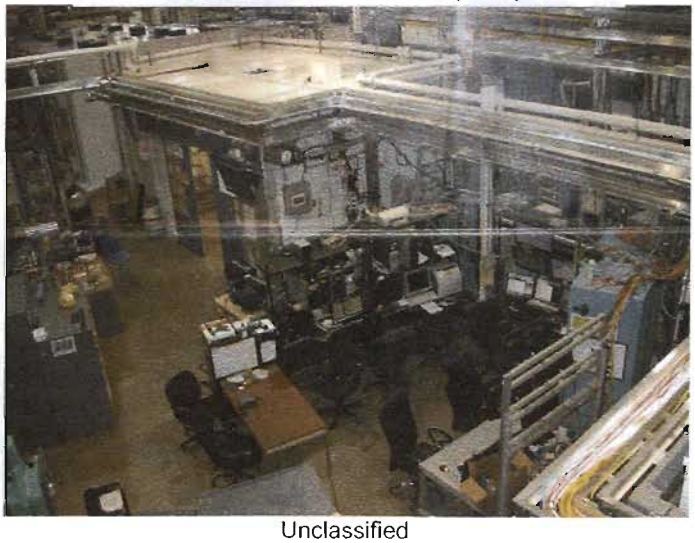
ANL Advanced Photon Source (APS) 1-ID-C



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18

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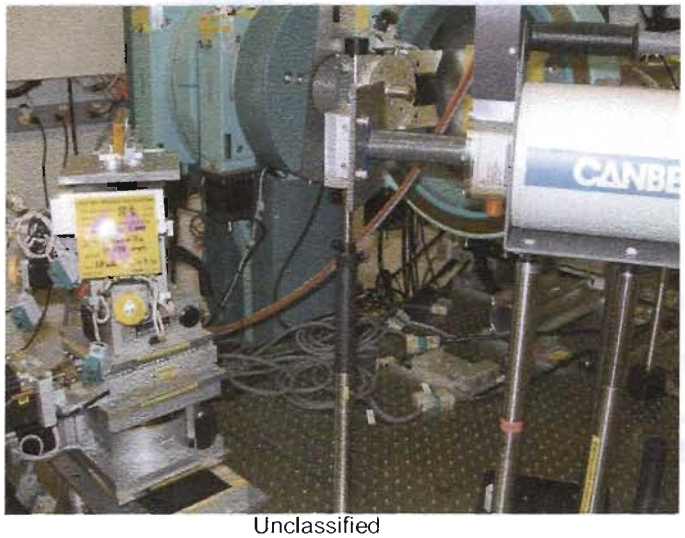
Experimental Setup



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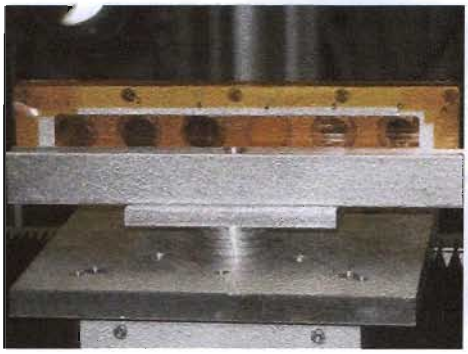
Sample holder



Zircaloy shield in place

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Samples Analyzed

Pellets of dUO_2 mixed with stearic acid

- 90, 60, 40% dUO_2

Dried spot deposits of aqueous dU solution

- 10 µg dU
- 5 µg dU
- 1 µg dU

dU metal

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21

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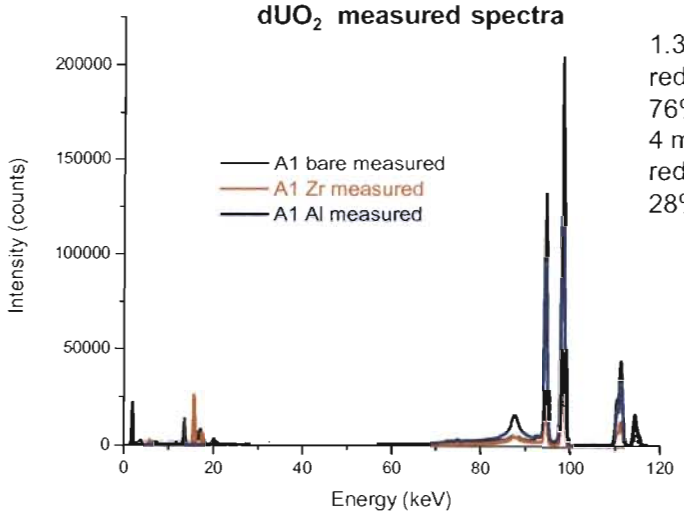
dUO_2 measured spectra



1.3 mm Zircaloy reduces signal by 76%
4 mm Al 6061 reduces signal by 28%

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22

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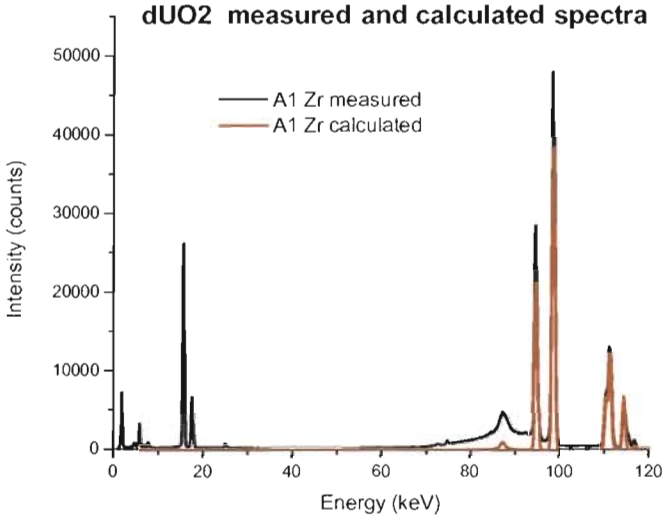
dUO2 measured and calculated spectra



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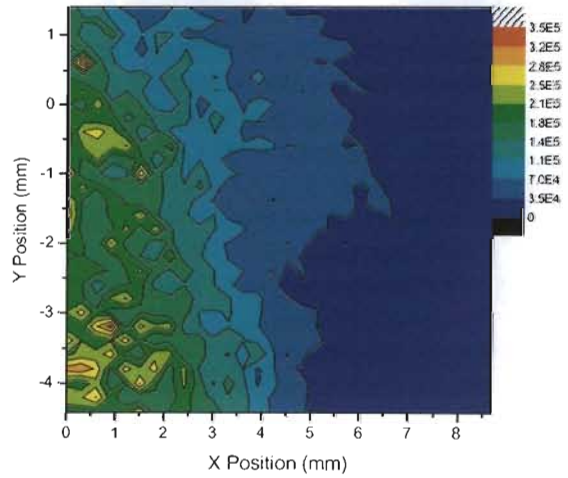
41% dUO_2 pellet map through 1.3 mm Zircaloy with 1 mm excitation spot



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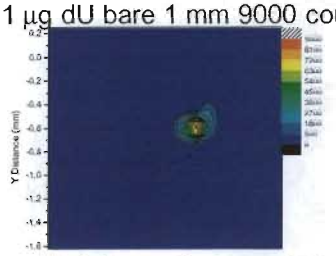
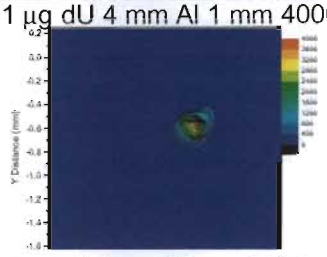
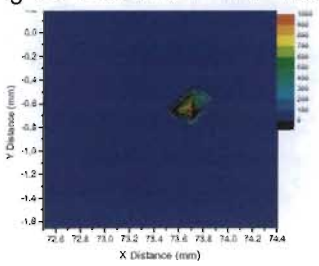
1 µg dU bare 1 mm 9000 counts



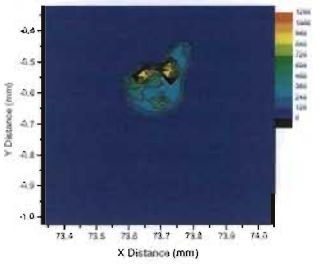
1 µg dU 4 mm Al 1 mm 4000 counts



1 µg dU 1.3 mm Zr 1 mm 1000 counts



1 µg dU bare 25 µm 1200 counts



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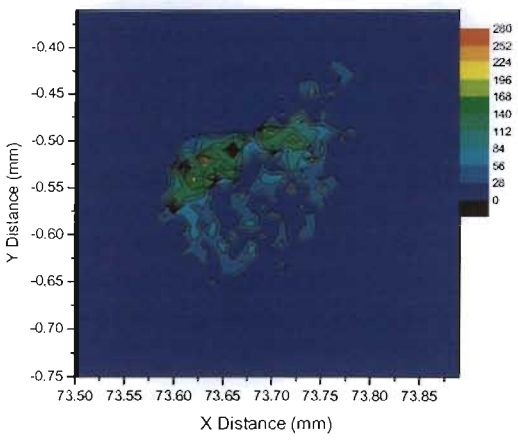
1 µg dU 1.3 mm Zr 17 µm 280 counts



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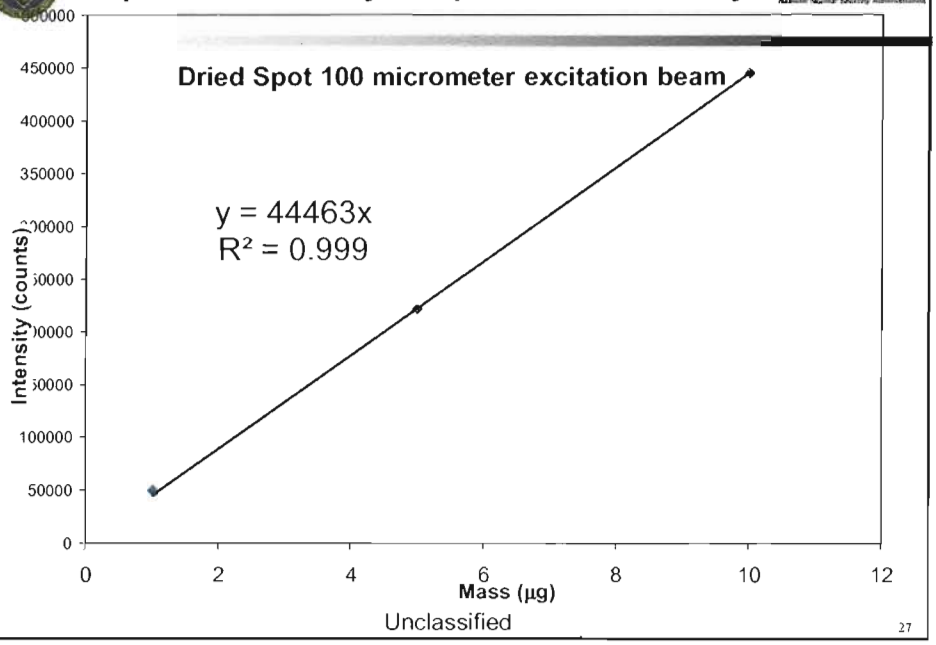

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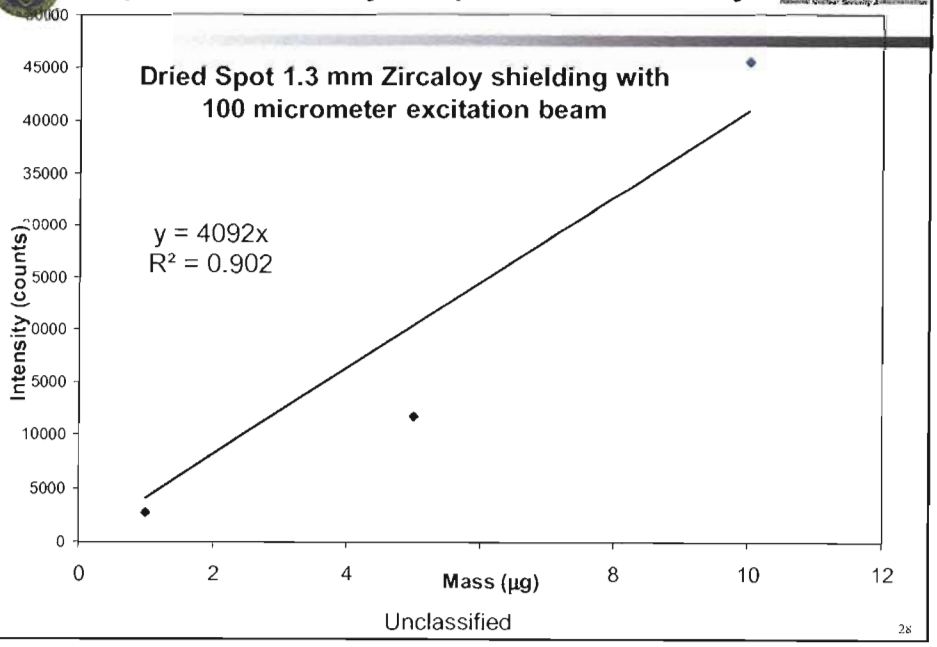

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Advanced Concepts Summary

- Achieved sensitive detection of uranium (10's of nanogram level) through container walls two times thicker than typical nuclear fuel cladding
- Demonstrated preliminary direct quantitative capabilities. Could cover at least 4-5 orders of magnitude, sub-microgram to weight percent
- Applicable to actinide elements of interest, Pu, Am, Np Cm, etc.
- Demonstrated UHE XRF to justify development of x-ray optics for laboratory instrumentation development

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29

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Improved Accuracy of Spent Fuel Pu Assay

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- Matthew Cusack

University of Washington

- Tim Elam

Argonne National Laboratory, Advanced Photon Source

- Sarvjit Shastri
- Ali Mashayekhi

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30