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Title: Ultra High Energy X-ray Fluorescence: A New Paradigm for Actinide Characterization of Spent Fuel

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Ultra High Energy X-ray Fluorescence: A New Paradigm for Actinide Characterization of Spent Fuel

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Ultra high energy X-ray fluorescence (UHEXRF) has been demonstrated as a feasible means for characterizing actinide content in spent nuclear fuel. Depleted uranium (dU) samples have been characterized using the 1-ID-C beam line of the Advanced Photon Source (APS) at Argonne National Laboratory. The dU samples included both pressed pellets of UO2 and dried spots of an aqueous uranium solution. The excitation energy was 117 keV which is above the absorption edge for the U Ka line. The XRF emission of the U Ka1 line at 98.428 keV was detected using a liquid nitrogen cooled high purity Ge detector. The samples were measured with and without a 1.2 millimeter Zircaloy shield in front of the samples which is twice the thickness of the normal fuel pin cladding. Although there was a decrease in U Ka XRF signal, sufficient intensity was obtained with as little as 5 live second dwell time resulting in several hundred counts for the smallest samples. The pressed pellets covered a concentration range from 0% to 90% dUO2 and the dried spots were 10, 5 and 1 microgram respectively. Elemental maps of the dried spots showed the heterogeneity of the dried material. This also illustrates the potential for sub-microgram sensitivity. The significance of this achievement demonstrates the feasibility of direct actinide composition measurement through the nuclear fuel cladding. Such measurements would reduce the uncertainty obtained with passive measurements such as gamma spectroscopy and neutron counting. Although this work was done with a synchrotron, it is conceivable that this measurement could be accomplished in the laboratory using a high power x-ray tube source and appropriate x-ray optics to provide quasi-monochromatic x-rays for excitation. The demonstration of uranium detection through container walls nondestructively, offers new opportunities for applying this technology for on-line through pipe characterization, field analyses of samples and increased reliability for safeguards measurements.

ULTRA HIGH ENERGY X-RAY FLUORESCENCE: A NEW PARADIGM FOR ACTINIDE CHARACTERIZATION OF SPENT FUEL

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Overview

- Introduction – UHEXRF
- Experimental
- UHEXRF spectrum of uranium
- Pressed pellets
- Dried spots
- Summary
- Future Direction

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Objective

- The objective of this work is to develop an advanced concept for Pu assay through container walls
- This is the 1st year of a three year development program which includes determination of oxidation state and isotope

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UHEXRF

- Ultra High Energy X-ray Fluorescence – energy range above 80 keV
- Utilize the high energy K lines of the actinides for characterization and quantification

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Advantages/Disadvantages

- Kα and Kβ lines for actinides
- Simple spectra, less likelihood of spectral interferences
- Deep critical depth of penetration
- High energy – commercial sources not common
- No tube line to provide monochromatic energy using DCCs

Experimental Concept

- Utilize monochromatic UHE excitation for actinide characterization
- Demonstrate through container wall XRF
- Direct and accurate quantification of actinides using UHEXRF
- Demonstrate feasibility of UHEXRF for laboratory-based instrumentation development

Experimental

- Monochromatic excitation at ~117 keV – synchrotron radiation, Advanced Photon Source, beam line 1-ID-C, 50-150 keV, $\Delta E/E = 1.4 \times 10^{-4}$, photon flux 7×10^{9} photons per second

APS 1-ID-C Beam Line

Experimental Setup

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Sample Holder and Shielding

Sample holder

Zircaloy shield in place

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Samples

Pellets of dUO_2 mixed with stearic acid

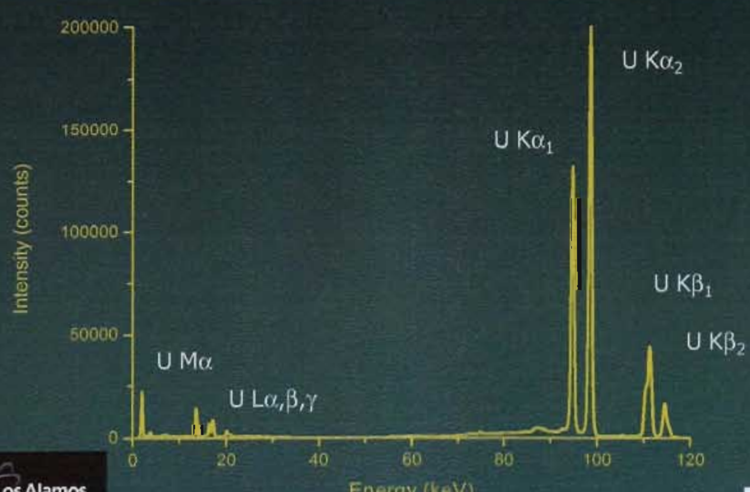
- 90, 60, 40% dUO_2

Dried spot deposits of aqueous dU solution

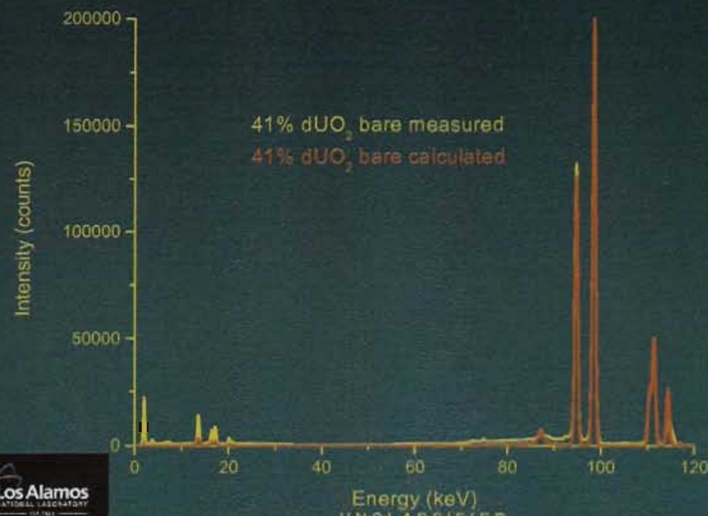
- 10 μg dU
- 5 μg dU
- 1 μg dU

dU metal

Pellet Spectrum dU 40%



Comparison of measured and calculated spectra



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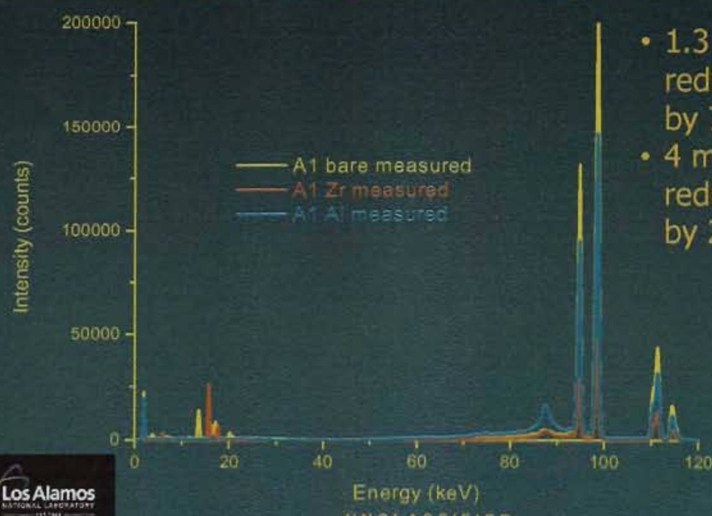
Comparison Spectra



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

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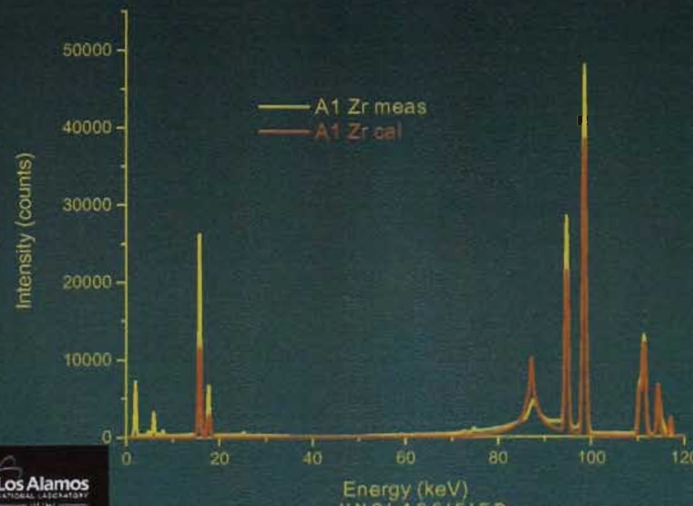
Comparison Zircaloy shielding measured and calculated



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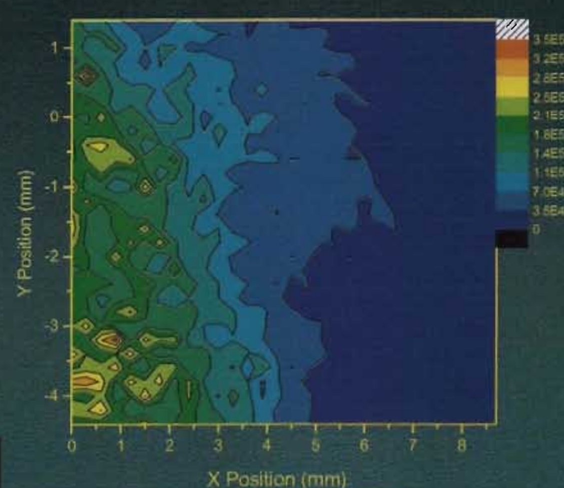
Pellet Partial Map through 1.3 mm Zircaloy



- 41% dUO_2
- 1 mm excitation spot

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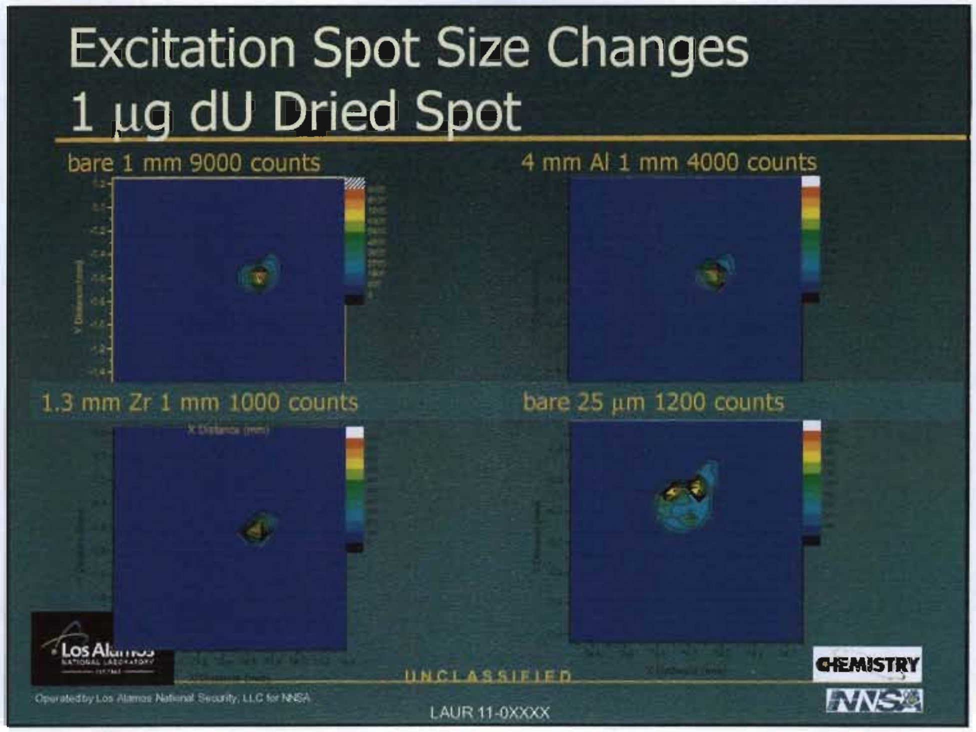
Excitation Spot Size Changes
1 µg dU Dried Spot
bare 1 mm 9000 counts
4 mm Al 1 mm 4000 counts
1.3 mm Zr 1 mm 1000 counts
bare 25 µm 1200 counts
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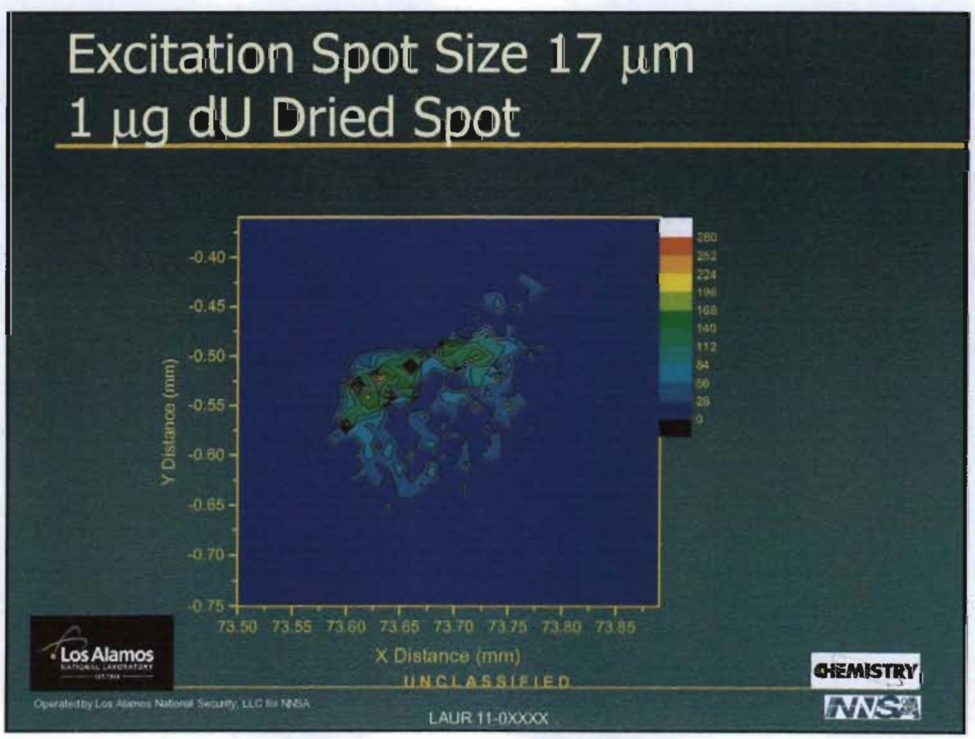
Excitation Spot Size 17 µm
1 µg dU Dried Spot
-0.40
-0.45
-0.50
-0.55
-0.60
-0.65
-0.70
-0.75
Y Distance (mm)
73.50 73.55 73.60 73.65 73.70 73.75 73.80 73.85
X Distance (mm)
280
252
224
196
168
140
112
84
56
28
0
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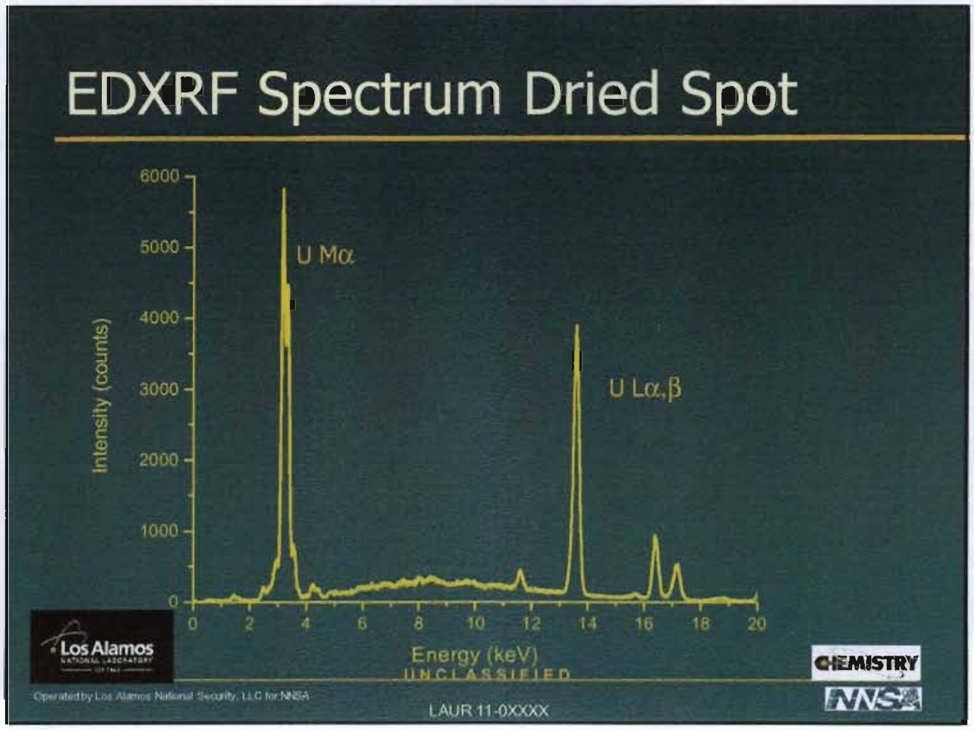
EDXRF Spectrum Dried Spot
Intensity (counts)
6000
5000
4000
3000
2000
1000
0
U Mα
U Lα,β
0
2
4
6
8
10
12
14
16
18
20
Energy (keV)
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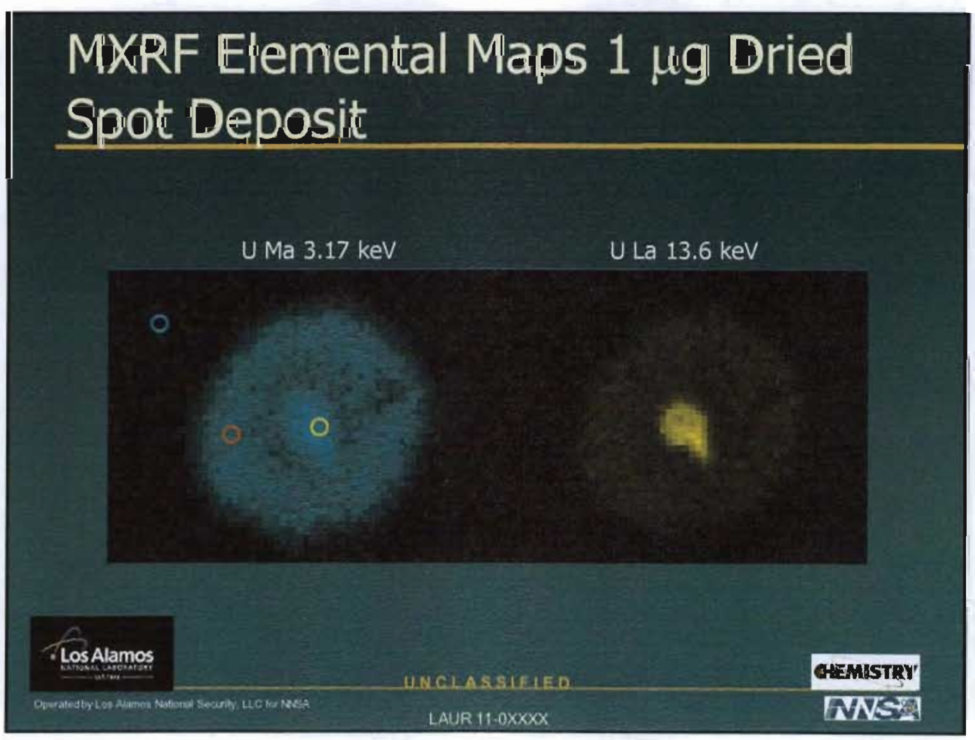
MXRF Elemental Maps 1 μg Dried Spot Deposit
U Ma 3.17 keV
U La 13.6 keV
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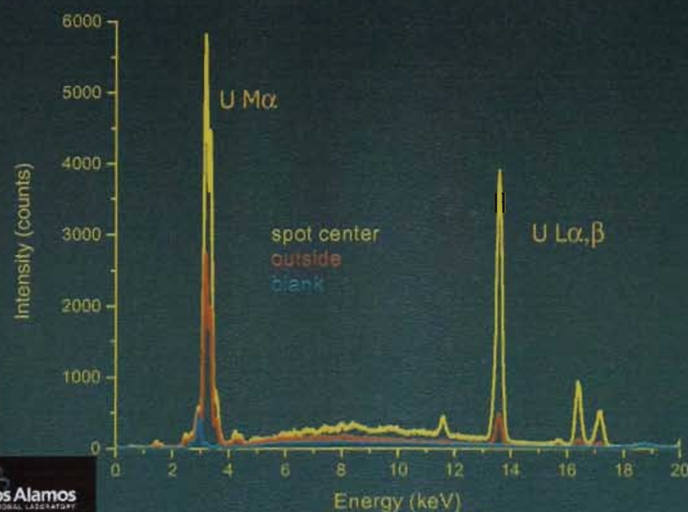
EDXRF Spectrum 1 μg Dried Spot Deposit



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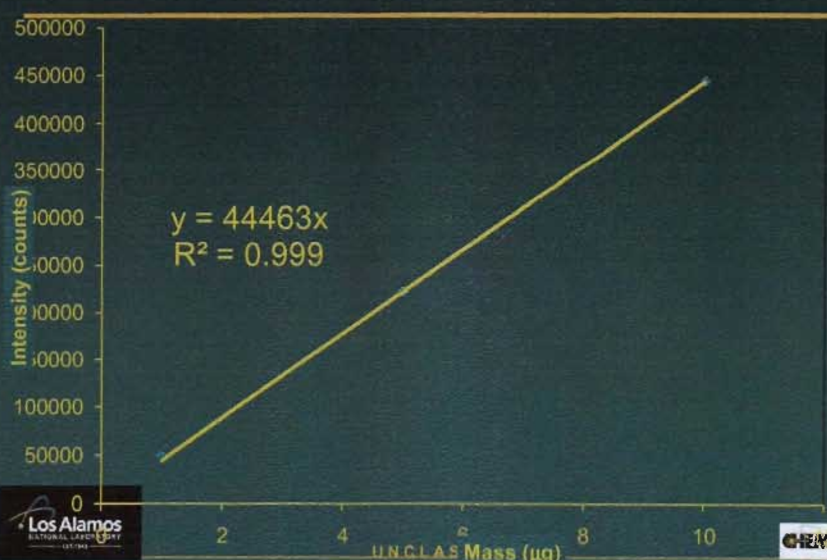
Dried Spot 100 μm Excitation Beam



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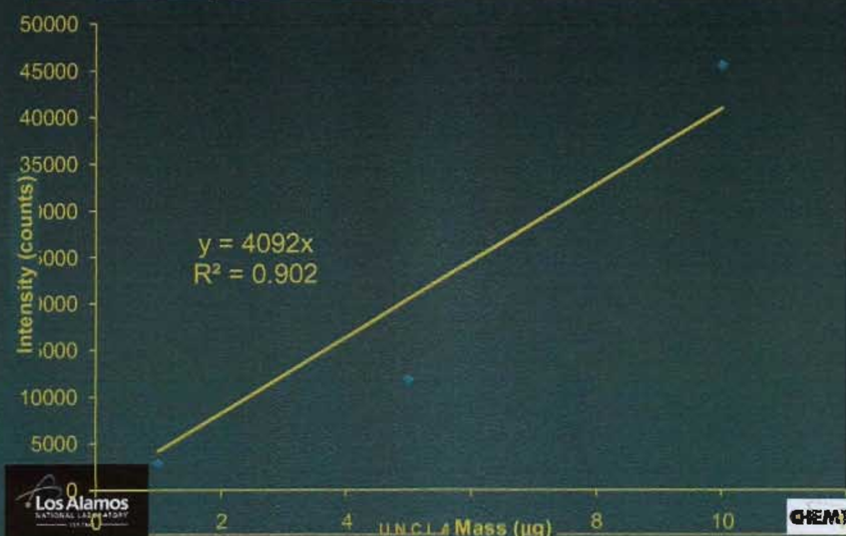
Dried Spot with 1.3 mm Zircaloy Shielding, 100 μm Excitation Beam



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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, Cm
- Demonstrated UHEXRF to justify development of x-ray optics for laboratory instrumentation development

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Acknowledgements

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