Laser ablation mass spectrometry for interrogating nuclear materials

Peter S. Boone[1,2], William P. Mason[1], Peter Hosemann[1], David G. Weisz[2], Brett H. Isselhardt[2]

[1] University of California, Berkeley, [2] Lawrence Livermore National Laboratory

Nuclear Science & Security Consortium

Introduction

Our goal is to characterize irradiated targets and fuel samples to understand a material's production, process history, and material effects under irradiation. Pre-irradiated materials are safer to handle so we chose depleted uranium for this phase of method development. We have generated data using laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS) in tandem with electron microscopy (SEM/EDS). Generally, information available from ICPMS can aid in nuclear material analysis through the relationships of the isotopic signatures, or trace element analysis and process history. **Our experimental objectives are to create a W calibration curve, quantify W in the bulk and inclusion, spatiotemporally resolve inhomogeneity in surface features, and measure select isotopic ratios in W and U.**

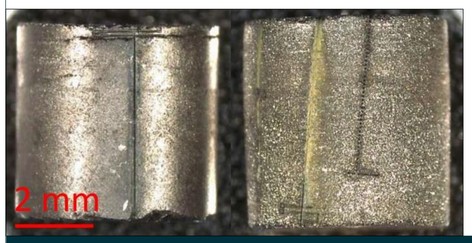

Figure 1. Pellets of depleted uranium oxide showing signs of laser damage after LA-ICPMS analysis; Sample 2, discussed later, is on the right.

Experimental Method

Five cylindrical samples of sintered depleted-UO_2 alloy (Fig. 1) were compared. Two samples contained declared impurities of W metal. MS was done with a modified Thermo XSERIES ICPMS. LA utilized a Spectra-Physics Spitfire emitting at 845 nm with 100 fs pulse width, yielding 800 mW at the sample. The focused beam spot is nominally 50 µm in diameter. During ablation In a sealed chamber, He carrier gas was flowed over the sample (Fig. 2). Post-ablation characterization involved optical and SEM low-magnification imaging, SEM imaging of laser damage, and EDS maps, line scans, and quantification.

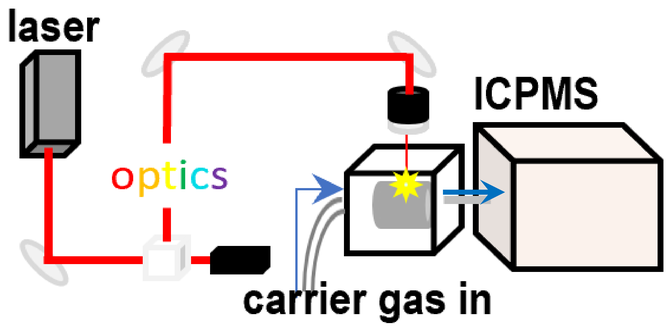

Figure 2. Basic schematic of an LA-ICPMS system.

Results I

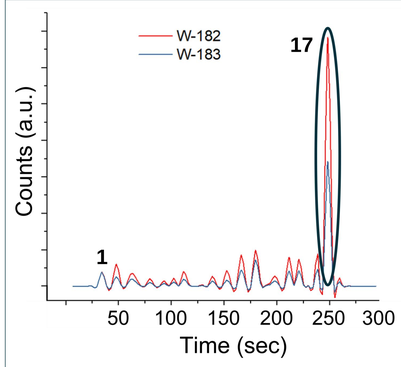

Figure 3. Sample 2 ^{182}W and ^{183}W MS data show a signal spike at Crater 17.

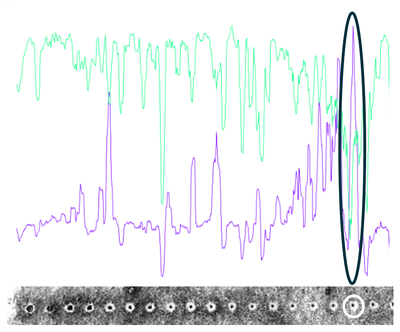

Figure 4. An EDS line scan shows a spike in W signal at Crater 17 (white circled crater).

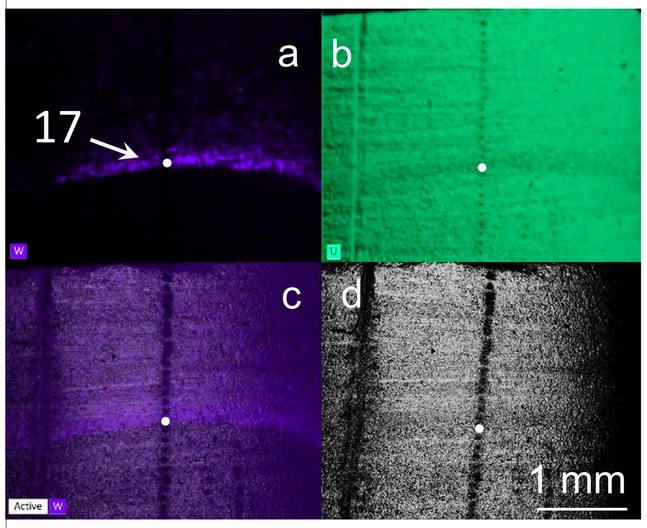

Figure 5. EDS maps a-d: W (purple), U (green), BSE with W, BSE alone. A near-surface W inclusion is clearly visible in a and c. The feature is less apparent in b and d, which show W to be anticorrelated with U as also seen in Figure 4.

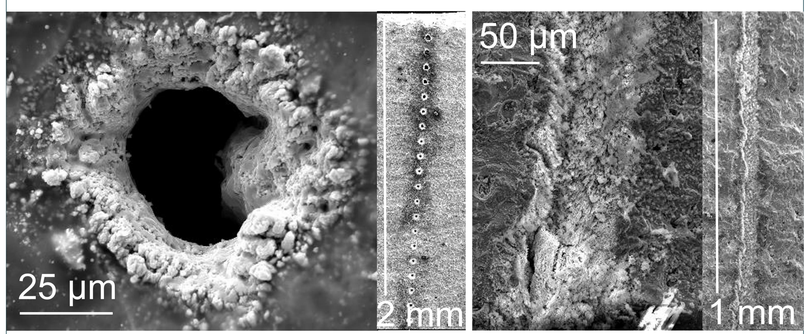

Figure 6. Comparison of laser damage from crater- and trench-type analyses. Both are approximately 75 µm in diameter and tend to yield comparable data.

Results II

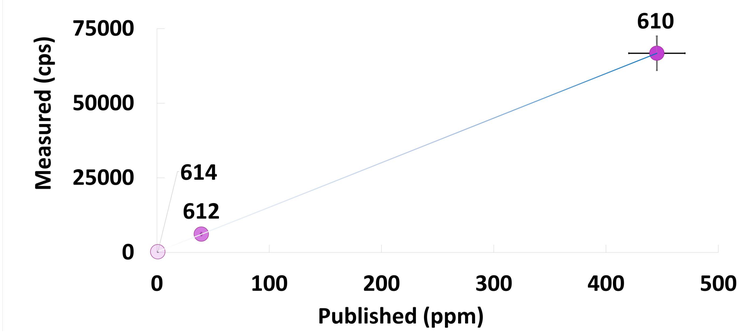

Figure 7. W calibration curve from NIST glass ICPMS data. Measured analyte concentrations are extrapolated from this plot. Error bars are visible only on the NIST 610 data point.

Sample	Expected W (ppm)	Measured W (ppm)	Measured/ Published
NIST 614[1]	0.85±0.024	0.59±0.05	0.690
NIST 612[2]	39.55±0.78	40.29±3.50	1.007
NIST 610[2]	445.3±25.0	445.28±38.2	1.000
Sample 2, Crater 17	11,000±1,000	10,348±889	0.941

Table 1. W concentration data from NIST glass and Sample 2, Crater 17; the 11,000 ppm expected value is taken from EDS data.

Sample	$^{238}U/W_{total}$	$^{238}U/^{235}U$
1	784356	128
2	33	197
3	23	212
4	45853	218
5	189811	208

Table 2. Subset of ICPMS data. As expected, Samples 2 and 3 exhibit a $^{238}U/W_{total}$ ratio 3-4 orders of magnitude greater than the others. Here, uncertainties (< 10%) are neglected.

Measured $^{182}W/^{183}W$	Natural $^{182}W/^{183}W$	%Diff
1.87±0.16	1.85	1.01±0.09

Table 3. Measured W isotope ratio data from Figure 3 is within 1% of values seen in natural W.

Conclusions

- We successfully used laser ablation ICPMS to fulfill our present research objectives, demonstrating its applicability to scientific and technical issues concerning the nuclear security community at large.
- Confirmatory techniques provide significant value by giving confidence in the conclusions, as evidenced by the obvious correlation of signals observed in EDS and ICPMS data.

Acknowledgments

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References

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