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Laser Induced Breakdown Spectroscopy Diagnostics for Nuclear Debris

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Abstract: This paper demonstrates the suitability of Laser-Induced Breakdown Spectroscopy (LIBS) for nuclear debris analysis by presenting LIBS elemental maps of surrogate nuclear debris and isotopic measurements of lithium, a nuclear fuel, via LIBS and chemometrics. © 2020 The Authors

1. Introduction

Research and development in the area of the technical nuclear forensics has found renewed vigor since 2010 when Congress enacted US public law 111-140, the “Nuclear Forensics and Attribution Act [1].” This act highlighted the importance of precision and timeliness of debris analysis in a post-detonation environment. In the case of a hostile nuclear detonation by an unknown adversary, the scientific community will face extreme time pressure to provide analytical results to national decision makers. Hence, a need exists for rapid chemical and isotopic analysis of nuclear debris, preferably in a field deployable form.

This paper demonstrates that it is possible to conduct multi-elemental mapping and isotopic analysis in nuclear debris. Additionally, gallium – a key plutonium alloy material and forensics signature – was mapped and quantified in a plutonium surrogate material. Finally, lithium isotopic quantification was achieved in powders of Lithium Hydroxide Monohydrate (LiOH-H₂O). Lithium is commonly used in thermonuclear weapons because of its unique nuclear properties. Hence, its isotopic ratios can provide another key forensics signature. All of these measurements were taken with LIBS systems capable of configuration for field use.

2. Experimental Details

2.1 Uranium Mapping in Surrogate Nuclear Debris

Melt glass samples were prepared based on the urban debris formulations proposed by Giminaro *et al.* [2]. This formulation involved replicating the bulk elemental constituents likely to be found in urban debris and melting them into a glassy matrix as described by Nizinski *et al.* [3]. A Nd:YAG laser operating at the 532-nm wavelength (10 ns pulse width, 80 mJ per pulse) was then focused via a plano-convex lens into the sample box to ablate the material as shown by Shattan *et al.* [4]. Light generated from 60 laser pulses from each spot was directed into a Princeton SpectraPro 2300i spectrometer through a focusing lens. Despite the inherent richness of LIBS spectra generated from samples containing both iron and uranium, isolated emission lines in the 585-595 nm region were identified for iron (Fe I 585.6 nm), sodium (Na I 589.0 nm) and uranium (U I 591.5 nm). These lines were used to simultaneously map iron and uranium through a center cross section of the debris sample as shown in figure 1.

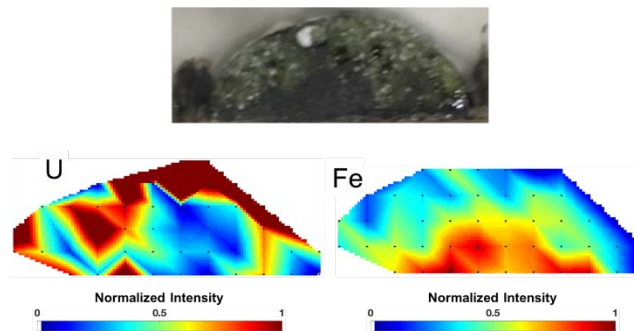


Fig. 1. Center cross-section concentration maps for (A) uranium and (B) iron calculated from the U I (591.539 nm)/Na I (589.592 nm) and Fe I (585.595 nm)/ Na I (589.592 nm) peak ratios.

2.2 Gallium Mapping in Surrogate Plutonium Material

Cerium and gallium alloy samples were made using a Thermo Scientific Thermolyne (Model Number FD1545M) resistive heating furnace with premeasured cerium and gallium precursor materials as described by Rao *et. al.* [5] to create plutonium alloy surrogates with cerium concentrations ranging from 0-3 weight percent as shown in figure 2a. A commercially available SciAps Z500-ER was then used to collect spectral data from the alloy samples. The Z500-ER is an industrial HH-LIBS instrument that uses a 5 mJ/pulse, 1064-nm Nd:YAG laser at a repetition rate of 10 Hz to ablate the surface of a sample. Four emission lines (Ga I 287.4 nm, Ga I 294.4 nm, Ce II 394.3 nm, and Ce II 413.8 nm) were identified for use in building calibration curves. Univariate and multivariate calibration techniques were then applied which yielded a limit of detection better than 0.25% weight percent gallium for the handheld. Elemental maps of the gallium concentration in the samples were constructed as shown in figure 2b.

2.3 Lithium Isotopic Determination

Solid samples were prepared using of 95% $^6\text{LiH-H}_2\text{O}$ and $^7\text{LiH-H}_2\text{O}$ powders (Sigman-Aldrich) which were pressed into pellet samples in a stainless steel dye at 10,000 psi for 60 seconds. A 532 nm Nd:YAG, (6 ns pulse width, 12 mJ/pulse) was used to ablate the samples. Light collected with a McPherson 350 mm Czerny-Turner Spectrometer revealed an isotopic shift of 0.016 nm for the strong Li I 669.8 nm emission line. PCR and PLS algorithms were then able to yield calibration curves across all concentrations.

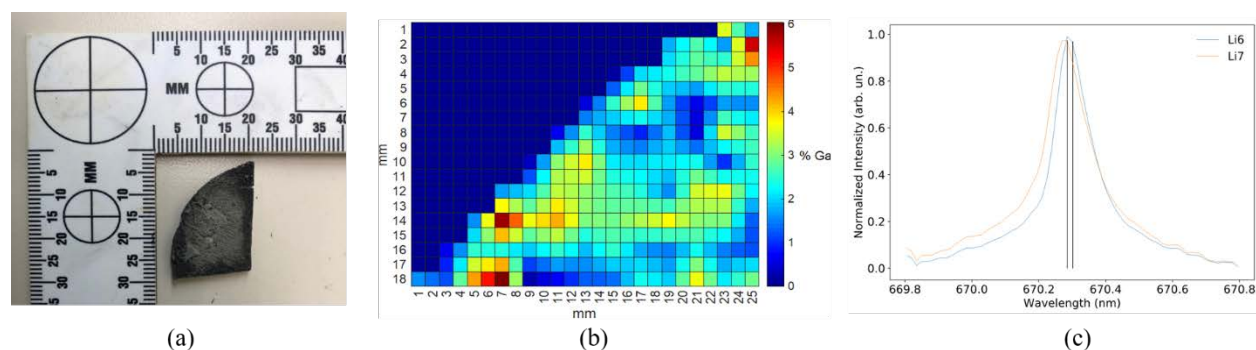


Fig2. (a) Photograph of the 3% Ga cerium alloy sample used in the mapping analysis and (b) surface Ga concentration map of the same sample. (c) Isotopic shift of the Li I 669.8 nm emission line experimentally measured to be 0.016 nm. In pressed pellet samples.

3. Conclusions

Laser-Induced Breakdown Spectroscopy has been demonstrated as a useful technique for field nuclear forensics applications. LIBS elemental maps have been constructed for actinides and key minor elements commonly found actinide metal alloys. Isotopic determination of a sample of material usable as a nuclear fuel was also demonstrated. These techniques could aid in rapid nuclear forensics analysis in post nuclear detonation scenarios.

4. References

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