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Abstract. This work identifies analytical lines in laser-induced plasma for chemical analyses of major elements found in surrogate nuclear debris. These lines are evaluated for interferences and signal strength to insure they would be useful to measure relative concentrations. Compact, portable instruments are employed and can be included as part of a mobile nuclear forensics laboratory for field screening of nuclear debris and contamination. The average plasma temperature is measured using the well-established Boltzmann plot technique, and plasma's average electron density is determined using empirical formulae based on Stark broadening of the H-alpha line. These measurements suggest existence of partial local thermal equilibrium.

1. Introduction

Broadband, portable laser-induced breakdown spectroscopy (LIBS) systems are promising to detect localized concentrations of actinides and fission products with high forensics value in field samples that are collected following a nuclear detonation with little to no sample preparation. Handheld LIBS systems can be useful in locating uranium contamination during nuclear facility remediation [1], and spectra of actinide oxides in mixed-oxide, pressed pellets have been previously analyzed [2].

This paper demonstrates that it is possible to identify useful analytical lines using LIBS of nuclear debris. The selected atomic lines are appropriate for elemental identification, relative concentration monitoring, and possibly quantitative analysis. Non-radioactive samples of surrogate debris are prepared based on proposed recipes, and applying processes communicated in the literature [3,4]. Surrogate debris "blanks" are put together with the major bulk ingredients common to these recipes. The addition of small amounts of particular elements serves the purpose of isolating the new spectral response due to the presence of each element.

2. Material Preparation

Melt glass samples and blanks are prepared based on the urban debris formulations [4]. Figure 1 displays typical samples prepared for experiments with notable variations in morphology based on the presence of different analytes. Once formed, each bead was then set in an epoxy resin (Epoxicure 2, Buehler) and lightly hand polished to remove any residual epoxy from the bottom of the mounted sample.
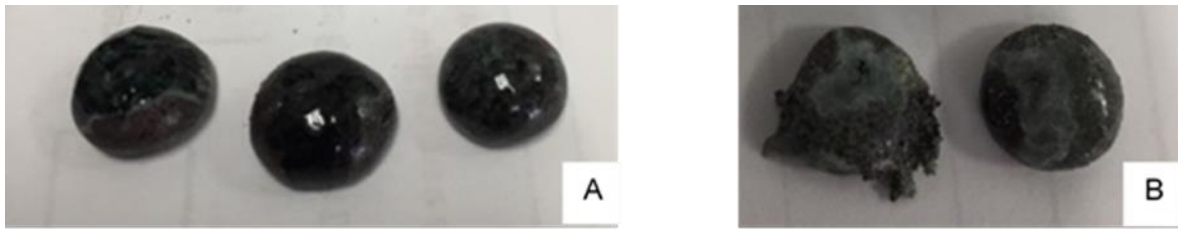
Figure 1. (A) Blank samples, and (B) phosphorus samples.

Table 1 summarizes the ingredients. Blank-samples (composition 84.8% SiO2, 4.5% NaO, 10.7% Fe2O3) contain the base ingredients that are critical to establishing a consistent matrix and morphology. Subsequently, samples are then processed by adding one analyte to the base ingredients for each sample. The set of samples show differences in the spectral response caused by the presence of the new analyte.

Table 1. Sample formulations for analytical line identification.

Sample	Base Ingredients (%)	Analyte (%)	Analyte Form
Aluminium	82.5	17.5	Al_2O_3
Calcium	91.8	8.2	CaO
Potassium	95.3	4.7	KOH
Magnesium	96.4	3.6	MgO
Sulphur	99.9	0.1	S
Barium	99.9	0.1	BaO
Manganese	99.8	0.2	MnO
Phosphorus	99.9	0.1	$Ca_3(PO_4)_2$

3. Experimental Setup

A combined laser-spectrometer-detector arrangement (TSI Insight LIBS System™) is used. It includes a Nd:YAG laser operated at the wavelength of 266 nm, with 6 ns pulse width and 52 mJ per pulse. The laser ablation micro-plasma is collected via a fiber optic probe and channeled into a spectrometer bank (Ocean Optics LIBS 2500) showing a 200-800 nm combined spectral range, and a resolution of 0.1 nm.

All of the optics in the system are locked in place. The instrument accumulates sample spectra from 20 different locations on each sample. For the experiments reported here, laser ablation is initiated 10 times at each location, and internal software averages resultant spectra to deliver a representative record for that location. The spectrometer's detector is set for a light collection gate with of 10 µs and a time delay of 2 µs. Radiometric and wavelength calibrations are accomplished using standard equipment.

4. Results

Spectra from the blank sample are compared to the spectra from each sample containing a specific analyte. In this analysis, the new peaks are identified that are not present in the blank sample spectra. These peaks are scrutinized for chemical interferences, resolvability, and strength of signal. After spectral normalization and background continuums subtraction, the most analytically useful peaks were identified for each element.

The interference free analytical lines identified include: 248.327 nm (Fe I), 279.800 nm (Mg I), 280.270 nm (Mg II), 288.158 nm (Si I), 328.560 nm (Na II), 373.486 nm (Fe I), 394.401 nm (Al I), 396.152 nm (Al I), 403.076 nm (Mn I), 440.475 nm (Fe I), 455.403 nm (Ba II), 521.038 nm (Ti I), 542.999 nm (Fe II), 553.548 nm (Ba I), 561.863 nm (Fe I), 585.745 nm (Ca I), 588.995 nm (Na I), 589.592 nm (Na I), 616.217 nm (Ca I), 656.279 nm (H I), 766.429 nm (Fe I), 766.490 nm (K I), 769.897 nm (K I).

The average plasma temperature is determined using the well-established Boltzmann plot technique [5]. The average electron density is evaluated using empirical formulae based on Stark broadening [6]. The experiments present challenges given the complexity of the sample matrix, the relatively low laser peak power from 6 ns, 50 mJ pulses, and the available spectrometer resolving power, $R = \lambda/\Delta\lambda = 4000$.

Nonetheless, the average temperature for time windows of 2 µs and 12 µs following ablation, $T_e = 6{,}200 \pm 800$ K, and the electron density, $N_e = 0.76 \pm 0.2 \times 10^{17}$ cm^{-3}, are inferred for effectively optically-thin plasma conditions at the selected time delays. These results suggest partial local thermal equilibrium in this plasma [7]. Figure 2 shows the hydrogen alpha line, H_α, and the Boltzmann plot constructed from the recorded data. The measured H_α spectra are likely due to residual moisture in the laboratory air at standard ambient temperature and pressure conditions.
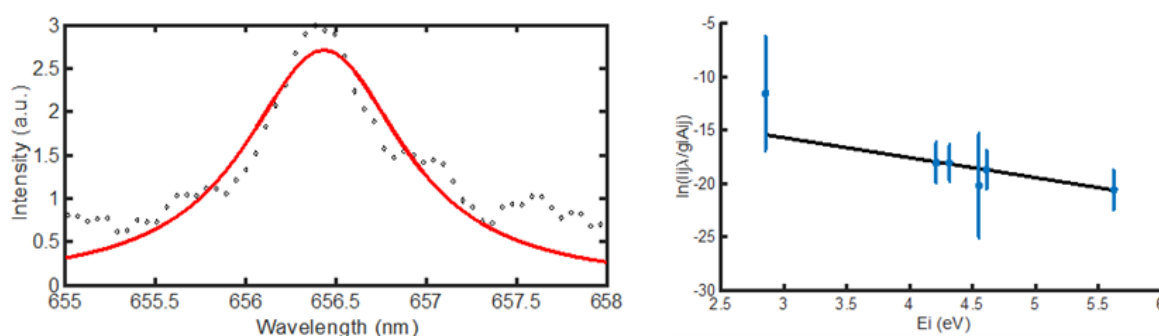

Figure 2. H_α and fitted Lorentzian (left) and Boltzmann plot (right).

5. Conclusions

The portable LIBS system allowed us to identify interference-free lines for all major non-radioactive elements present in a leading surrogate nuclear debris formulation. The plasma temperature and electron densities are determined from Boltzmann plots and empirical formulae of the primarily Stark broadened Hα, respectively. The results suggest portable LIBS systems may be useful for field identification of key elements in nuclear debris. Further work will include analysis of radioactive debris and optimization of spectra post-processing. Future challenges include assessments of the line-of-sight average measurements and of the statistical variations from different samples or sample regions.

6. Acknowledgments

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