



Laser-induced breakdown spectroscopy measurements of uranium and thorium powders and uranium ore

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ABSTRACT

Laser-induced breakdown spectroscopy (LIBS) was used to analyze depleted uranium and thorium oxide powders and uranium ore as a potential rapid in situ analysis technique in nuclear production facilities, environmental sampling, and in-field forensic applications. Material such as pressed pellets and metals, has been extensively studied using LIBS due to the high density of the material and more stable laser-induced plasma formation. Powders, on the other hand, are difficult to analyze using LIBS since ejection and removal of the powder occur in the laser interaction region. The capability of analyzing powders is important in allowing for rapid analysis of suspicious materials, environmental samples, or trace contamination on surfaces since it most closely represents field samples (soil, small particles, debris etc.). The rapid, in situ analysis of samples, including nuclear materials, also reduces costs in sample collection, transportation, sample preparation, and analysis time. Here we demonstrate the detection of actinides in oxide powders and within a uranium ore sample as both pressed pellets and powders on carbon adhesive discs for spectral comparison. The acquired LIBS spectra for both forms of the samples differ in overall intensity but yield a similar distribution of atomic emission spectral lines.

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1. Introduction

Laser-induced breakdown spectroscopy (LIBS) has been described in several review articles [1–3] and books [4–6]. Briefly, LIBS is an atomic emission technique that uses a focused, typically nanosecond, laser pulse to form plasma on the surface of the sample. The plasma atomizes, excites, and ionizes the material within the focal volume of the laser pulse. The early stages of the plasma are dominated by bremsstrahlung emission. Very little, if any, atomic or molecular information is observed over the continuum during this time. After a delay time of nanoseconds to microseconds, the plasma begins to cool and expand. The excited species relax, emitting light that is characteristic of electronic transitions within the atom. The emitted light is collected and then analyzed using a spectrometer. The resulting spectrum is used to determine the elemental composition of the sample. Every element in the periodic table emits a unique spectral signature allowing for accurate identification via comparison to atomic emission data in the literature. The actinide elements have very complex emission spectra due to the high density of electronic states and associated quantum physics and photo-dynamics yielding thousands of emission lines [7–10].

Currently, the method for analyzing special nuclear material or suspected nuclear material found in the field is to swab or collect the sample, package, and ship to an accredited laboratory. The samples are then prepared for analysis and analyzed using traditional analytical analysis techniques [11]. This process may take several months and thousands of dollars to complete. The rapid identification of nuclear materials in situ is therefore a very attractive alternative to the current methods. Thus far, LIBS has been used to identify uranium, thorium, and plutonium in solid pellets [12–14], metals [15], in solutions [16], and dried on substrates such as filter paper [17]. Recently, several simulated fuel pellets containing UO_2/PuO_2 and $\text{UO}_2/\text{PuO}_2/\text{AmO}_2/\text{NpO}_2$ were successfully analyzed using LIBS [18].

The use of LIBS to analyze uranium and thorium powders as a potential rapid in situ analysis technique in nuclear production facilities or in-field forensic applications is of great interest. Powders are notoriously difficult to analyze using LIBS since ejection and removal of the powder occur in the laser interaction region. Less material is analyzed per laser shot compared to dense pellets and less stable plasmas are formed. Therefore, the S/N in powder data is typically lower than in dense pellets. However, semi-quantitative investigations are still possible. Powders, such as inorganic and organic NIST powders containing magnesium [19] and cerium oxide [20], have been analyzed using LIBS. The samples were placed on an adhesive disc in an attempt to keep the powder stationary. The analysis of powders in

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Table 1

Approximate chemical composition for uranium ore BL-5 provided by Natural Resources Canada, Minerals and Metals Sector.

Constituent	Weight %
Si	22.0
U	7.09
Al	6.0
Fe	5.9
Ca	4.0
Na	3.6
C	1.9
Pb	1.5
Mg	1.5
K	0.4
Ti	0.4
S	0.3
V	0.1
P	0.07
Mn	0.05
Zr	0.04
Sr	0.03
Cr	0.01
Th	0.004

bulk has also been demonstrated by placing iron oxide powder in a vial containing a mechanical barrier that stirs and flattens the powder as the sample rotates, maintaining the same sample-to-lens distance for more reproducible plasmas and spectra [21]. However, a large quantity of sample is not always available, especially in forensic applications. Therefore, powders adhered to an adhesive tape or some other method used to immobilize the powder are more likely required to accomplish the experimental needs of small sample sizes and rapid analysis found in the field. The capability of analyzing powders could also be used for rapid analysis of intermediate products during chemical

synthesis without the need for the addition of a binder and a high pressure press to form a solid pellet. Another appealing capability of this technique would be for environmental sampling. The instrument could be used to directly analyze a contaminated area or a small sample could be transferred onto an adhesive tape and analyzed in the field for the presence of actinides. The identification of trace contamination on a surface where an attempt was made to clean or hide evidence of illegal enrichment or unapproved use of special nuclear material would also require a rapid in-field technique to identify the material. Finally, LIBS analysis could be used to determine the presence of nuclear material if release or exposure of radioactive material occurred within a nuclear facility. The strong interest in using LIBS for nuclear material analysis is that all elements in the periodic table can be detected whereas only radioactive material can be detected in traditional nuclear counting analytical techniques. For example, a contamination site may contain radioactive material as well as RCRA (Resource Conservation and Recovery Act) metals (arsenic, barium, cadmium, chromium, lead, mercury, selenium, and silver). LIBS would be able to detect both the radioactive material as well as the RCRA metals whereas traditional nuclear counting analytical techniques would only detect the radioactive material.

Here we demonstrate the analysis of depleted uranium and thorium powders and uranium ore as a potential rapid in situ analysis technique in nuclear production facilities, environmental sampling, and in-field forensic applications. This is the first report of the analysis of actinide powders using LIBS to our knowledge.

2. Experimental

The laser excitation source used in these experiments was a Big Sky Ultra 100 pulsed Nd:YAG laser operating at 1064 nm with 8 ns pulses and a variable attenuator producing pulse energies up to 100 mJ at 20 Hz. A laser pulse energy of approximately 6 mJ at 20 Hz was used for all experiments. A 2.5 cm diameter plano-convex focusing

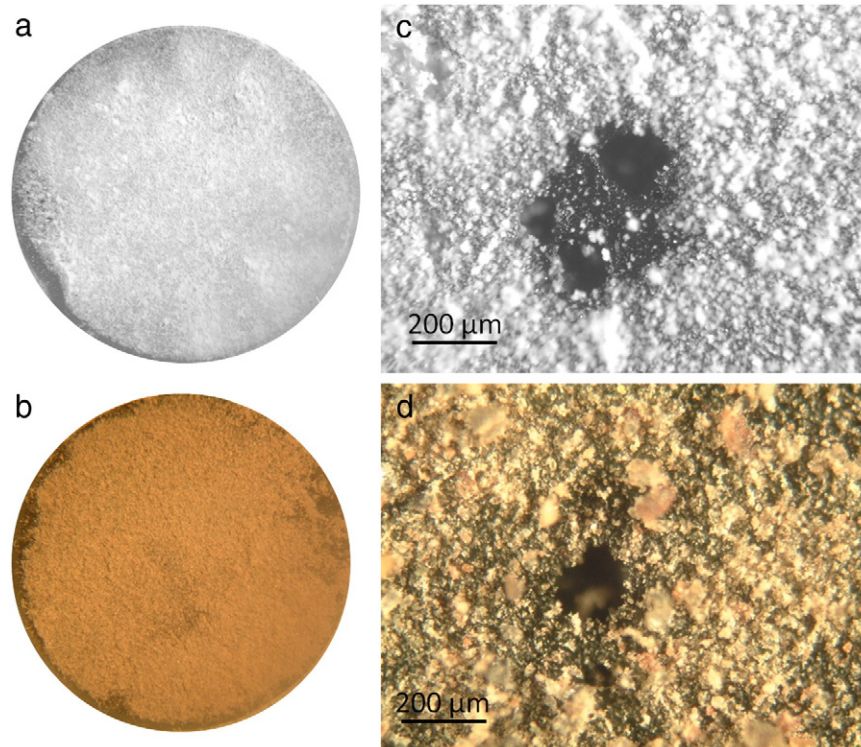


Fig. 1. (a) Thorium oxide powder and (b) uranium ore were placed on 12 mm diameter carbon adhesive discs. A single laser shot ablation crater is shown in (c) thorium oxide powder and (d) uranium ore.

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