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Rapid Nuclear Forensics Analysis via Laser Based Microphotonic Techniques Coupled with Chemometrics

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Abstract

Nuclear forensics (NF) is an important tool for analysis and attribution of nuclear and radiological materials (NRM) in support of nuclear security. The critical challenge in NF currently is the lack of suitable microanalytical methodologies for direct, rapid and minimally-invasive detection and quantification of NF signatures. Microphotonic techniques can achieve this task particularly when the materials are of limited size and under concealed condition. The purpose of this paper is to demonstrate the combined potential of chemometrics enabled LIBS and laser Raman spectromicroscopy (LRS) for rapid NF analysis and attribution. Using LIBS, uranium lines at 385.464 nm, 385.957 nm and 386.592 nm were identified as NF signatures in uranium ore surrogates. A multivariate calibration strategy using artificial neural network was developed for quantification of trace uranium. Principal component analysis (PCA) of LIBS spectra achieved source attribution of the ores. LRS studies on UCl_3 , $UO_3(NO_3)_2 \cdot 6H_2O$, $UO_2SO_4 \cdot 3H_2O$ and UO_3 in pellet state identified the bands associated with different uranium molecules as varying in the range of $(840 \text{ to } 867) \pm 15 \text{ cm}^{-1}$. Using this signature, we have demonstrated spectral imaging of uranium under concealed conditions.

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1. Introduction

The illicit trafficking of nuclear and radiological materials (NRM) led to the emergence of a new branch of science in the 1990's known as "nuclear forensics" (NF) [1, 2]. NF is the analysis of intercepted illicit NRM to

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provide evidence for attribution. NF and nuclear forensic interpretation are important for monitoring nuclear safeguards in support of nuclear security [3, 4]. Microphotonic techniques are attractive for NF analysis as they have the ability to perform rapid, minimally invasive and non-destructive analysis with high sensitivity. In this paper, we discuss the utility of laser-induced breakdown spectroscopy (LIBS) and laser Raman spectromicroscopy (LRS) as examples of such techniques in the field of NF. LIBS is an emission spectroscopic technique, in which the radiation emitted by the micro plasma formed on focusing a powerful laser beam onto the sample is used to fingerprint the elements associated with the spectral peaks [5, 6, 7]. On the other hand, LRS uniquely identifies and images specific chemical compounds and microstructure in a given sample based on molecular vibrations following little or no sample preparation [8, 9]. Although these techniques have potential to identify and quantify at stand-off distance the elemental, molecular, and isotopic composition of NRM, their practical utility is limited by the complexity of the spectra in air at atmospheric pressure and the interpretative challenges of the multivariate data [10]. For instance, colored compounds are difficult to analyze by LRS because of intense fluorescence, laser heating and diversity of oxidation states and deviations from stoichiometry; and for LIBS the analyzes gets difficult due to the matrix effects which are driven by the strongly coupled transient laser-matter interactions. The LIBS spectra are optically thick and therefore trace element spectral peaks are masked and buried by the strong background continuum. These limitations may to a large extent be overcome by using multivariate chemometrics to reduce the data dimensionality and to extract and model the subtle NF signatures in aid of NF attribution [5].

1.1. The Problem

Traditional nuclear forensic science makes use of analytical techniques such as gamma-ray spectrometry, alpha spectrometry, mass spectrometry and microstructural techniques, which were developed for applications related to the nuclear fuel cycle [1]. Unfortunately, most of these techniques are time-consuming and destroy the integrity of the sample. The critical challenge in NF currently is the lack of suitable microanalytical methodologies for direct, rapid and non-invasive detection and trace analysis of nuclear forensic signatures in NRM. The ability to rapidly detect and measure trace NF signatures in NRM such as uranium (U) and its compounds in samples of limited size, typical in NF, at stand-off distance and mostly under concealed conditions without destroying their integrity has significance in nuclear security. Additionally, it could prove useful in combating illicit trafficking of NRM and fight against nuclear terrorism that may employ improvised nuclear devices (IND) and/ radiological dispersal devices (RDD).

Doucet *et al.* (2009) determined the isotopic ratio of partially resolved uranium-235/uranium-238 and hydrogen/deuterium isotope shift lines in air at atmospheric pressure using partial least squares (PLS1) regression on LIBS spectra for nuclear safeguard inspection [11]. Cremers *et al.* (2012) developed a LIBS system with lightweight and compact high-resolution spectrometers to rapidly detect different ratios of isotopes of H and U and highly enriched samples of ^6Li and ^7Li to monitor radiological, nuclear and explosive (RNE) threats [12]. Chinni *et al.* (2009) performed a detailed study of uranium detection on surfaces and mixed in bulk materials (e.g., soil) by LIBS for application to activities associated with environmental surveillance and detecting weapons of mass destruction (WMD) [13]. These demonstrate the applicability of LIBS in air and atmospheric pressure for comprehensive in-field NF analysis for a variety of NRM both in bulk and on surfaces (samples of limited size), as well as the benefits of coupling multivariate chemometrics with the method.

The high collection efficiency of the microscope optics and the multi-scanning capability of the micro-Raman spectrometer were utilized to characterize uranium oxide particles and a range of uranium compounds [14, 15]. It was concluded that the use of *in situ* Raman provides a powerful technique to study the mechanism of transformation and the stability of uranium oxides under oxidizing conditions [15]. Raman spectroscopy was used on 95 samples comprising mainly of uranium ore concentrates as well as some UF_4 and UO_2 samples, in order to classify uranium compounds for nuclear forensic purposes using PCA for the first time by Ho [16]. The use of LRS in the study of uranium compounds gets limited due to fluorescence and ignition of the colored samples. However, these limitations can be overcome to a great extent by the use of low energy laser and chemometrics. While low laser energy reduces the fluorescence and burning of the colored samples, chemometrics helps to extract the required information from the weak Raman signal arising due to low energy laser.

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