NUCLEAR FORENSICS VIA MACHINE LEARNING LASER BASED SPECTRAL ANALYSIS AND IMAGING

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INTRODUCTION & BACKGROUND TO THE STUDY

The limitations of classical nuclear forensic analysis methods calls for innovative approaches for rapid noninvasive detection and accurate quantification and attribution of illicit trafficking of nuclear and radiological materials against nuclear security threat.

This is enabled by combining machine learning and laser based spectroscopy and spectral imaging techniques which we are developing to elucidate trace isotopic, molecular and elemental (trace impurities) composition, as well as the microstructure (as each step in the fuel cycle creates and/or modifies these signatures) of nuclear materials.

We focus on analysis of samples of limited sample size for responding to environmental releases of NORM and illicit trafficking activities in our region, which is a hub for trade in radioactive 'conflict' minerals and counterfeit nuclear materials, with high possibility of radiological dispersal devices (RDD) and improvised nuclear devices (IND).

Key advantages of the approach: small samples (mg) can be evaluated with minimal sample preparation; samples can be remotely analyzed very rapidly (ms-seconds) and method can utilize multivariate calibration and exploratory analysis.

Attribution (origin, method of production, probability that more of the material exists, transit route, and means by which administrative control over the material was lost) i.e. especially enabled by this approach. Multivariate interpretation is the crucial factor in this exercise.

While Laser Induced Breakdown (LIBS) reveals the atomic (and sometimes molecular and isotopic) emission spectra of elements in micro-plasma obtained from samples, laser Raman microspectroscopy reveals the molecular configuration by active vibrational spectra of polyatomic ions in samples as well as structure and morphology. These methods are targeted for their versatility, high sensitivity, speed, simple operation and *in situ* capabilities.

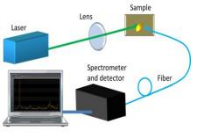
MATERIALS & METHODS

Current Analytical Challenges

- Standard nuclear forensics methods are limited by the complexity of samples and of the often multivariate signature interpretation.
  Mostly destructive, and consuming large amounts of sample.
  Complex matrix and contamination during sampling and treatment make determination of especially trace REE complicated.
- Information that must be interpreted (using expert knowledge) is often not meaningful (often subjective) to the attribution assessment.

The Practical Approaches

- Machine learning (ML) is used to reduce the analytical complexity and increase the information gained.
  Attribution is being achieved and will be improved via ML fuzzy logic and exploratory analysis/modeling.
- Need for spectral resolution of system are significantly relaxed via computational intelligence (ML) techniques



♦Minimally invasive. ♦Rapid. ♦Simultaneous analysis. ♦No chemical treatment. ♦Sample shape irrelevant

- **LIBS 2500 PLUS (Ocean Optics) - Q-Switched Nd: YAG**
- **Confocal Laser Raman Microspectroscopy.**
- **ML techniques for data compression and modeling**

We analyzed and imaged uranium oxide as well as several U-bearing ores and radiological (HBRA) samples and also interpreted natural variability between uranium ore-bearing rocks and soils from HBRA.

SELECTED RESULTS & DISCUSSION

The chemical composition of uranium oxide was found, via two unique peaks, to be an indicator to which part of the nuclear fuel cycle the material belongs.

The two nuclear forensic signatures we successfully used to reconstruct the composition of high background radiation area (HBRA) ore samples.

We have successfully designed a hyperspectral imaging (HSI) scheme for non-contact analysis of nuclear forensic traces, to obtain both high (l μm diffraction limit) spatial and spectral information from small micron scale samples such as 'hot' aerosol particulates .

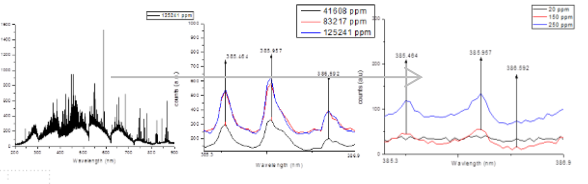
This enables us to analyze the chemical and/ structural composition of nuclear traces and simultaneously visualize their spatial distribution.

Table1. Identification of lines for quantitative analysis of U using LIBS

U-I 356.659 nm
U-II 367.007 nm
U-II 383.146 nm
U-II 385.464 nm
U-II 385.957 nm
U-II 386.592 nm
U-II 417.159 nm

LIBS has demonstrated potential for the detection and quantification and evaluation of characteristics of trace evidence found at a nuclear/radiological crime scene as signals from ppm concentration are obtained.

Interest in LIBS lies in analysis of REE – to determine e.g., the degree of enrichment, and trace elements (varying concentrations of impurities exist in every material) present.



Figs 5. LIBS spectra obtained from UO_3 in cellulose. The figs show (a) the spectrum obtained, (b) regions of spectral interest of selected lines delineated at major concentration and (c) analytical line responses at ppm level U.

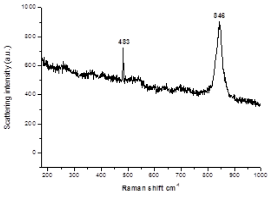


Fig 1. Typical Raman spectrum of a UO_3 sample.

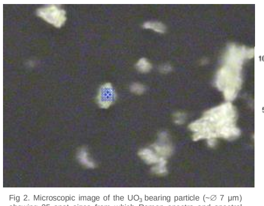


Fig 2. Microscopic image of the UO_3 bearing particle (~∅ 7 μm) showing 25 spot sizes from which Raman spectra and spectral images of the 483 and 846 Cm^{-1} peaks were imaged to construct HSI for chemical imaging.

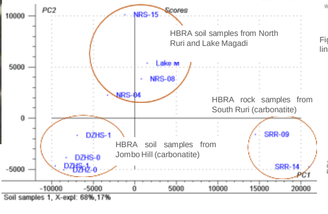


Fig 3 PCA score plot of HBRA soils and rock samples analysed by LIBS. The hypothesis of presence of an underlying uranium-rich rock in the Lake Magadi basin is strengthened by this result.

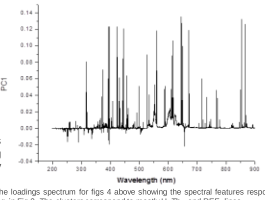


Fig 4. The loadings spectrum for figs 4 above showing the spectral features responsible for the clustering in Fig 3. The clusters correspond to mostly U, Th, and REE lines..

Fig 6. Sample slide used for spectral mapping of individual particles.

Microstructural features from laser Raman spectromicroscopy are very informative: shape of particles or the surface structure gives hints of the sample origin. E.g. surface roughness may also be needed to determine when the material was last chemically processed in relation with altitude of enrichment.

CONCLUSIONS AND PROSPECTS

- ❑ The combined utility of ML-assisted laser spectral and imaging techniques via LIBS and laser Raman microspectroscopy provides complementary information and adds novelty to a comprehensive analytical picture in nuclear forensics: ML extracts subtle relevant information from the complex spectra/images and affords multivariate data reduction as well as exploratory analyses of the nuclear forensic investigation. Fusing data from the two techniques is essential for the success of nuclear forensics investigations and subsequent attribution.
- ❑ The obtained PCA multivariate models have capacity for constraining the geological models of uranium deposits as well as for genetically discriminating new uranium discoveries.
- ❑ The challenge for the future is to develop and apply ML tools for data interpretation that provide combined and credible determinations of locations and methods of materials production.
- ❑ Analysis of the temporal behavior of spectra will give insight in the chemical changes within specimens, which can be used for age estimation purposes.

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