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Rapid Nuclear Forensics Analysis Via Laser Based Spectroscopy and Spectral Imaging Coupled With Chemometrics

Nuclear Forensics (NF) involves the laboratory analysis of intercepted nuclear and radiological material (NRM) for nuclear attribution (origin and intended use). The aim of NF analysis is to establish the relationship between seized NRM and their attribution to monitor and strengthen nuclear security. The critical challenge in NF at the moment is the lack of direct, rapid and non-invasive analytical techniques for detection, microanalysis and imaging of NRM. Laser based spectroscopic techniques namely Laser Induced Breakdown Spectroscopy (LIBS) and Laser Raman Spectromicroscopy (LRS) combined together has the potential to overcome these limitations to a great extent. LIBS is an emission spectroscopic technique which fingerprints the element associated with the spectral peaks. On the other hand, LRS identifies the molecular bands associated with specific chemical compounds and microstructure in a given sample based on molecular vibrations following little or no sample preparation. The practical utility of these techniques is limited by the complexity of the spectra in air at atmospheric pressure and (multivariate) data interpretation. Coupling LIBS and LRS with chemometrics enables data dimensionality reduction besides extracting subtle NF signatures from the spectra and images. Uranium lines at U II 385.464 nm, U II 385.957 nm and U II 386.592 nm were identified as the NF signatures of uranium for rapid detection of trace uranium in uranium ore surrogates. Principal Component Analysis performed on the LIBS spectra of the uranium-bearing mineral ores collected from different regions of Kenya (South Ruri, Magadi, Coast) successfully grouped the samples to their mineral mines (origin). A multivariate calibration strategy for the quantification of uranium bearing mineral ores was developed using artificial neural network (ANN) (feed forward back-propagation algorithm) utilizing spectral feature selection and making use of (ii) resonant uranium lines (iii) weak uranium peaks (to demonstrate the power of ANN to model noisy LIBS spectra for trace quantitative analysis). NF signatures of uranium molecule were identified using multi-photon (λ⊠= 532 nm, 785 nm) LRS in uranium trioxide, uranyl nitrate, uranyl sulphate and uranium chloride bound in cellulose at 848 cm-1, 865 cm-1, 868 cm-1 and 861 cm-1 respectively. These molecular signatures were utilized to infer the presence of uranium compounds in the various HBRA soils. Spectral imaging performed on a pellet spiked with trace level of uranium in cellulose (to mimic uranium trafficked in concealed condition) displayed the distribution of uranium on the pellet surface. Thus, LIBS and LRS coupled with chemometrics have the potential to detect trace level of uranium in NRM rapidly under concealed condition without destroying the integrity of the sample.

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