Nuclear Forensics by Means of Multiphoton Laser Raman Spectromicroscopy

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Abstract

The possession and handling (including illicit trafficking) of nuclear materials out of statutory control has led to the emergence of a new branch of science known as “nuclear forensics” (NF). NF aims at identifying and attributing unknown nuclear and radiological material (NRM) in support of nuclear security. The critical analytical challenge in NF is the lack of methods for direct, rapid and non-invasive (non-destructive) detection and characterization of especially concealed or tiny amounts of NRM to aid in accurate attribution. Traditional NF techniques (radiochemical, radiometric) have limitations in this regards. We are developing machine learning (ML) enabled multiphoton laser Raman spectromicroscopy to identify the nature, attribute source, intended use and mode of production of NRM rapidly, directly and non-invasively. Laser Raman spectromicroscopy (LRM) uniquely identifies and images samples based on molecular vibrations following little or no sample preparation. Although the technique has high versatility, its practical utility is limited by the complexity of the samples and (multivariate) data interpretation. These limitations may be overcome by combing LRM with machine learning techniques to help reduce the data dimensionality and mine and extract the required information from the spectra and images. We have analyzed powdered and peletized UO$_3$ using optimized multi-photon laser ($\lambda = 532$ nm, 785 nm) Raman spectromicroscopy. The identified nuclear forensic signatures were 483 cm$^{-1}$ and 843 cm$^{-1}$ (using 5 mW power Laser-532 nm with an exposure time of 15 s and 15 accumulations); and 381 cm$^{-1}$, 411 cm$^{-1}$, 527 cm$^{-1}$ and 843 cm$^{-1}$ (using 0.4 mW power Laser-785 nm with an exposure time of 20 s and 20 accumulations). We further examined UO$_3$ spiked at trace levels in a cellulose matrix to simulate uranium concealed in an organic powder and studied its detectibility by LRM. This was achieved by performing multispectral imaging of the above NF signatures from UO$_3$ particles of diameter 0.1 µm, 0.25 µm and 5 µm to study their sub-microm distribution with respect to particle size, and compared with those obtained from uranium ores from High Background Radiation Area (HBRA). We outline the spectral imaging methodology and novel utility of machine learning techniques we used for multivariate image analysis (MIA) to extract key morphological and chemical properties from the images and show how this achieves rapid nuclear forensic analysis and attribution of the samples.