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Editorial: Applications of spectroscopy and chemometrics in nuclear materials analysis

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Editorial on the Research Topic
Applications of spectroscopy and chemometrics in nuclear materials analysis

Optical analysis techniques, including spectroscopy and image analysis, have many advantages when applied to the study of nuclear materials. They require small sample sizes, can be performed remotely, and can be proceduralized through consistent practice. Most importantly, they provide a wealth of information by generating multivariate data. For example, ultraviolet–visible–near-infrared absorbance spectroscopy of actinides in aqueous and organic solutions is dependent on the oxidation state, anionic complexation, and temperature. These variables are important for solution-based separation processes, and sensitivity to these factors, combined with online monitoring, can drive the efficiency and control of these processes (Tse et al., 2020; Tse et al., 2024). The morphology and chemical composition of actinide particles can also provide a vital clue to the mechanisms by which the particles were formed, providing forensic information on the origins of the particles (Mayer et al., 2012; Burr et al., 2021).

The extraction of the desired information from multivariate data is the domain of chemometric analysis. Chemometrics determine the empirical relationships between chemical or physical variations in a sample set and observed data. This method can be used to compensate for the variations, for example, by providing consistent concentration measurements of an actinide despite variations in matrix conditions. It can also be used to estimate the matrix conditions. The power of chemometrics rests in being able to avoid assigning specific mechanisms for the cause/effect relationship; merely including the cause over a range of levels consistent with those expected for subsequent samples is sufficient.

This Research Topic of seven papers illustrates the power of combining optical methods (spectroscopy or image analysis) with chemometrics to characterize nuclear materials in the context of process analysis and answering questions of provenance. It also provides several examples of how successfully an analysis can be achieved when there is limited knowledge of the samples (matrix conditions cannot be fully replicated) or limited resources to create a full calibration set.

Ly et al. presented a domain adaptation framework to improve the robustness of convolutional neural networks to provide faster and more consistent characterization of surface structures using scanning electron microscopy (SEM) images. The new approach is

more suitable for nuclear forensics because it uses only unlabeled test samples to adapt a pretrained model, which supports data sensitivity.

Crouse et al. introduced a novel blind source separation algorithm to preprocess infrared absorbance spectra acquired online so that the process spectra looked like the training set. This method improved the nitrate quantification accuracy by helping the model cope with unexpected sources of variation in process spectra in tank samples at the Savannah River Site.

McNamara et al. probed UF_6 hydrolysis intermediates by cooling the reaction to cryogenic temperatures to reduce the rate of hydrolysis using *in situ* Fourier transform infrared spectroscopy. Cryogenic layering was used to monitor the generation of reaction intermediates and elucidate the chemistry governing UF_6 hydrolysis. Several previously unobserved bands were attributed to the formation of intermediates containing U–O–U bonds, which were directly converted to UO_2F_2.

Sadergaski et al. built partial least squares regression models for Np(III/IV/V/VI) in HNO_3 using spectra corresponding to true Bi-chemical mixtures and spectra generated from additive combinations of pure component spectra. The performance of each modeling strategy was similar, suggesting that the spectral features from the potential interionic interactions of the Np ions were negligible. This methodology can be leveraged to efficiently build predictive chemometric models in multivalent metal nitrate systems for online monitoring applications.

Lamson McCombs et al. developed a Bayesian implementation of seemingly unrelated regression to successfully predict HNO_3 concentration in a variety of processing conditions based on morphological features of PuO_2. This work suggested that different levels of HNO_3 may produce detectable differences in PuO_2 particles by SEM images. The proposed technique processes SEM images using Morphological Analysis for Material Attribution software and then analyzes the data using functional seemingly unrelated regression. This technique is promising for the analysis of special nuclear materials in nuclear forensics applications and can be generalized for applications to other materials.

Villa-Aleman et al. developed versatile double-walled cells to enable the analysis of Pu-bearing compounds in a radiologically clean spectroscopy laboratory using advanced techniques, including Raman spectroscopy, diffuse reflectance infrared Fourier transform spectroscopy, diffuse reflectance spectroscopy, and gamma spectroscopy. New signatures in the conversion of Pu oxalates to PuO_2 were observed by describing the molecular properties of the material, including calcination, crystallinity, purity, and age dating since the last calcination.

Sadergaski et al. tested a generalized optimal design approach for selecting calibration set samples in a two-factor design space for the quantification of U(VI) and HNO_3 by Raman spectroscopy. A nonlinear support vector regression model outperformed a more traditional partial least squares regression model in terms of prediction performance and maintained an optimal number of samples in the training set to minimize time and materials.

These seven articles are published in the Research Topic, "*Applications of Spectroscopy and Chemometrics in Nuclear Materials Analysis*." Each article highlights the growing interest in spectroscopy and chemometric methods for the analysis of nuclear materials in solid, liquid, and gas forms for advanced nuclear forensics, process monitoring, and fundamental science applications. Future studies focusing on modeling morphological features (e.g., SEM images) and chemical information (e.g., Raman spectroscopy) using sensor fusion strategies are encouraged. The editors of the Research Topic are open to further communication.

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