Development of In Situ Monitoring and Data-Driven Modeling for Complex Systems: Case Study on Simulant Mixtures of Nuclear Waste

Stefani Kocevska

Directed by Drs. Martha Grover and Ronald Rousseau

Approximately 90 million gallons of nuclear and chemical waste are stored at the Hanford and Savannah River Sites in the United States. One of the main challenges associated with nuclear waste treatment and stabilization is the complexity of the waste, which calls for extensive sampling during processing. In this thesis, the use of Process Analytical Technology (PAT), including in-situ infrared and Raman spectroscopy, is demonstrated for Real-Time In-Line Monitoring (RTIM) of simulated nuclear waste. In-situ monitoring will facilitate the continuous operation of the waste treatment facility and help reduce employee exposure to hazardous working conditions. The work in this thesis bridges the gap between spectroscopy data and concentration outputs. A flexible spectra-to-composition modeling framework is developed to address the varying complexity in the simulated waste mixtures. The initial work incorporates linear multivariate regression models to quantify the concentrations of process-relevant (target) species in the waste system. As the complexity of the waste system increases, advanced signal separation preprocessing techniques are incorporated in the modeling framework. The goal of the additional steps is to identify the contributions of the target species in complex systems, which allows for increased robustness of the spectra-to-composition model. Another aspect of the thesis work is the analysis of non-linear phenomena occurring in Raman spectra, centered around the nitrate peak which exhibits peak shifting during certain conditions. The overall work in this thesis enables robust and efficient concentration quantification of target chemical species in complex mixtures, enabling real-time monitoring of nuclear waste.