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Development of a Thin-Layer Electrochemical Sensor for Molten Salt Reactors and Fuel Cycle Processes

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ABSTRACT:

This proposed project seeks to develop and prototype a molten salt thin-layer electrochemical sensor (TLES) and model capable of accurately (<1% error) measuring the concentration of special nuclear material (SNM) at both high (>10 wt.%) and low (~100 ppm) levels with potential interferents present in environments relevant to molten salt reactors (MSR) and nuclear fuel cycle processes. Achieving this objective will provide a new, near-real-time, *in situ*, measurement tool for more effective process monitoring and material control and accountability (MC&A).

Thin-layer electrochemical concentration measurements have been well-developed in aqueous solutions for both stagnant and flowing conditions. The advantage of a TLES is that a single probe can measure both high and low concentrations. Both voltametric and bulk electrolysis can be performed with a TLES in near real-time and *in situ*, due to the small volume, without affecting the bulk properties of the molten salt. Voltammetry (e.g., CV, NPV) is a powerful class of methods in which a potential is scanned or stepped in various patterns to produce a current response (often a peak) which can be related to concentration. A low limit of detection can be achieved with voltammetry. However, voltametric analyses are based on Fick's laws of diffusion which necessitates dilute concentrations. Bulk electrolysis exhaustively reduces (or oxidizes) the species of interest and relates the amount reduced (or oxidized) to the total charge (Q). It does not rely on Fick's laws of diffusion which removes the dilute concentration limitation.

The potential applied is used to selectively detect certain species. Each species has a certain reduction potential. When that potential is reached for a species in the salt, it will reduce and generate an electrical signal. In traditional (i.e., semi-infinite linear diffusion) electroanalytical measurements, overlap can occur when one species reduces after another because the response of a species does not decay close to zero. However, the exhaustive reduction that occurs in the TLES in both voltammetry and bulk electrolysis results in the signal for each species decaying to nearly zero. Hence, each species can be detected in series with minimal signal overlap, which improves selectivity of the electroanalytical measurements.

The versatility, sensitivity, and selectivity of the TLES makes it a promising tool to support MC&A and safeguards in a variety of molten salt reactor designs, fuel fabrication processes, and fuel recycling processes. The development of the TLES will directly support's DOE-NE's mission of advancing nuclear power in the U.S. by enabling rapid and accurate MC&A in advance reactor designs and fuel cycle facilities. However, the transition of the TLES from aqueous, low-temperature environments to high-temperature, molten salt environments presents unresolved challenges, such as compatibility of materials, radiation effects and handling of nuclear material. This work will focus on developing a robust TLES that can withstand the harsh environments of molten salts. Furthermore, the measurement uncertainties of the developed TLES will be quantified to inform evaluations of the expected standard error of inventory differences when utilizing the TLES.