BENCHMARK ON CU-NI CO-DEPOSITION IN CuSO$_4$/NiSO$_4$ SOLUTION BY THREE-DIMENSIONAL MASS TRANSPORT AND ELECTROCHEMICAL MODEL

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Abstract

Highly radioactive isotopes remain within spent nuclear fuels even one million years after discharge from the reactors. To end this problem in a manageable period, pyroprocessing has explored to achieve sufficient throughput with inaccessible high temperature and strong radiation. Such remote operations, especially at uranium electrorefining, are challenges for repeated experiments that also generate a pile of radioactive wastes. Computational analysis is a plausible option to reduce trials and errors to find optimized designs and operating conditions of the electrorefiner.

To allow simulations to replace electrorefining experiments to the extent possible, we have developed a three-dimensional multi-species electrochemical model. Commercial computational fluid dynamics software was coupled with user-defined electrochemical modules. Ion transport equation governs overall mass transport and concentration distribution while concentration-modified Butler–Volmer equation controls electrochemical reactions at electrodes. This model was already validated by molten-salt electrorefining experiments using EBR-II spent fuels in cooperation with the Idaho National Laboratory. At that time, we benchmarked 80 hours long variations of overall cell potential only due to a lack of position-dependent information. This, it is needed to examine the model’s capability to predict position-dependent potential and current.

Here, we further validated the model with well-proven aqueous systems which experimentally measured more specific results other than cell potential. Cu-Ni co-depositions in CuSO$_4$ and NiSO$_4$ solution were experimented by using both a corrosion cell and a rotating cylindrical Hull cell. First, transfer coefficients, exchange current densities, and other materials properties of Ni$^{2+}$ and Cu$^{2+}$ in sulphate solution was estimated by cathodic potentiodynamic polarization at the static corrosion cell. Second, the co-deposition experiment was galvanostatically conducted without stirring in the corrosion cell. Third, the co-deposition was also experimented in the rotating Hull cell. Fourth, simulated cathodic overpotential changes were validated to measured results at the corrosion cell. Fifth, cathodic overpotential distributions at the Hull cell were benchmarked with computational results, as a function of position, cathode rotating speeds, and applied current densities.

Benchmark tests at various operating conditions and two different geometries generally showed good agreement between simulations and experiments. Despite minor discrepancies at detailed potential variations, these could be improved by using more accurate electrochemical properties and considering current efficiency, nucleation overpotential, and other missing effects.
Therefore, the multi-species co-deposition model could be a computational design tool to predict current densities and overpotentials for electrorefiners.