Benchmark of Mark-IV electrorefiner using 3-D computational electrochemo-hydrodynamic model*

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Abstract

In contrast to active experimental studies on electrorefining as a key process in pyroprocessing, computational simulations have shown limited progress due to complexity of multi-species modelling. Despite of its vital necessity to 3-D approach to increase the throughput and purity of recovered uranium in low cost means, there is little study in this scope.

As a part of 3-D computational electrochemo-hydrodynamic model development, applied potential (i.e. the summation of anode overpotential, cathode overpotential and ohmic drop) is benchmarked with Mark-IV electrorefiner experimental results provided by Idaho National Laboratory. The experimental results were gavanostatically carried out with two cruciform anode baskets and one cylindrical solid cathode in LiCl-KCl electrolyte at 773 K for 80 hours equivalent to 20 million coulombs of charge transferred. The solid cathode was switched with fresh one after 39 hours operation.

It is highly significant to decide the appropriate effective electrode area profile by incorporating complexity of anode dissolution behaviour because overpotential and ohmic drop are functions of current density that is largely influenced by electrode area. Several anode profiles which have various final effective areas were examined. The profile with the final area same as anode dissolution basket showed the best benchmark results with 0.0636 RMSD value, which is about seven times smaller than the case of smooth cylinder model whose final area converged to almost zero.

The reason why the above case showed the good agreement may be the formation of conductive rough metal layer on the anode basket by disproportionate reaction of zirconium ($Zr^{4+} + Zr \leftrightarrow 2Zr^{2+}$). The metal layer is composed of zirconium and few percent of other elements. However, more benchmarking and validation processes are necessary to apply this computational model to use engineering design and analysis for commercial electrorefiners because there is still large uncertainty in electrorefining process such as cathode dendrite growth, porosity effect, material properties, and complexity of multi-element electrochemical.

* The full paper being unavailable at the time of publication, only the abstract is included.