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Modeling the UREX+3a Process using Aspen Plus Coupled with AMUSE

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INTRODUCTION

A plant level simulation of the UREX+3a separations process has been developed using AMUSE for solvent extraction calculations coupled with Aspen Plus for other operations. AMUSE, an Excel based application developed at Argonne National Laboratory [1], performs a rigorous calculation of countercurrent solvent extraction processes using thermodynamically based distribution coefficients specifically designed for nuclear separations. Aspen Plus [2] models simulate other separations plant operations such as head end assembly chopping and dissolution, product solidification, acid recovery, off-gas treatment and waste water treatment.

METHODOLOGY

Aspen Plus is the basic simulation software. It provides a direct link to Microsoft Excel that was used to automatically couple solvent extraction calculations using AMUSE into the simulation. Coupling AMUSE solvent extraction calculations with Aspen modeling of the rest of the plant provides a rigorous simulation of the UREX+3a process that can be easily modified to simulate other variations of the UREX+ process. The preliminary results presented in this report can be used for qualitative assessment of UREX+ flowsheet options, as well as an evaluation of the model itself.

MODEL DESCRIPTION

The UREX+3a process separates spent fuel into several fractions suitable for either recycling or disposal using a series of solvent extraction processes. The process model uses 171 chemical species while the AMUSE calculations typically require about 34 ions (anions and cations) to define stream compositions. This set of ions represents 28 metals, H^+ and five anions. These 28 metals as elements, ions, nitrate salts and oxides account for 110 chemical species. Organic and inorganic solvents, gases, and miscellaneous chemical species complete the list of components.

Front End Operations

An assembly model links to an Excel spreadsheet where the user provides full isotopic compositions for fuel, clad and hardware. The reduced 34 element composition used in the Aspen/AMUSE model is generated automatically. A chopper model uses fixed split fractions to separate off-gas and fines from the fuel, clad and hardware streams. A model of the nitric acid fuel dissolution process uses fixed stoichiometric equations to convert metals into ionic species and off-gas. Models of washing hardware discharged from the chopper and cladding hulls discharged from the dissolver to remove residual fuel material use fixed stoichiometry to convert metals into ionic species and off-gas. Wash solution is sent to the dissolver while the washed solids are waste streams.

Solvent Extraction

The UREX, CCD-PEG, TRUEX and NPEX operations call AMUSE to perform solvent extraction calculations with parameters and feed stream composition from Aspen entered automatically. After the AMUSE calculation has completed, product stream compositions are passed back to the Aspen model. Because the TALSPEAK solvent extraction process uses fixed distribution coefficients, it was modeled directly in Aspen Plus using the Aspen countercurrent solvent extraction unit operation model. All of the solvent extraction models include product stream washing and recycle of wash solution and solvent.

Solidification Processes

The solidification processes are similar and use Aspen unit operations to model calcining products from the solvent extraction. Typically this process consists of: evaporation using a rigorous fractionation column calculation; calcination with the Aspen Rgibbs reactor module that calculates the equilibrium product composition by minimizing Gibbs free energy; separation of the output stream into gaseous and solid product streams; mixing of the gaseous product stream and evaporator overheads; and condensation of the mixed gas stream into liquid and gaseous waste streams.

Recovery and Waste Streams

Nitric acid recovery is calculated using an Aspen distillation column model that generates an overhead stream that is essentially water and a concentrated nitric acid bottoms stream slightly contaminated with residual metals.

Off-gas streams are combined and treated by: Thermal Catalytic Oxidation to destroy organics; Selective Catalytic Reduction to convert NO_x into N_2 and water; and caustic scrubbing to remove SO_2 from the offgas. The gas is condensed and effluent streams discharged from the unit.

Waste water streams are collected in the waste water treatment operation. The stream is filtered, organics are removed by activated carbon, and salts are removed by reverse osmosis.

Spent solvent from the solvent extraction processes and raffinate and product washing are collected in the organic waste treatment operation.

RESULTS

The model predicts that 55 feed streams and 14 output streams will be generated by separations plant operation. On the basis of one metric ton of initial reactor fuel, the model predicts a plant throughput of approximately 200 metric tonnes of material. Approximately half is treated waste water. Another 30% is gas emissions arising from feed to the calcination furnaces. The gas stream is treated for discharge to the environment. About 5% of the throughput is product material. Another 10% is recovered organics and acid that may be recycled. The remaining 5% is contaminated waste that requires disposal.

While these results are preliminary, the model has successfully simulated operation of the UREX+3a separations process. Coupling AMUSE to Aspen Plus provides rigorous solvent extraction calculations directly within the plant simulation, greatly increasing the accuracy of the model. Many areas, such as acid recycle, can be optimized to improve performance and reduce material usage and waste generation. The rigorous plant simulation model resulting from this work provides a framework to conduct such studies. The model is easily modified to simulate other variations of the UREX+ process.

REFERENCES

1. R. A. LEONARD and M. C. REGALBUTO, *Solv. Extraction and Ion Exchange.,* **12(5),** 909 (1994).
2. *Aspen Plus® 11.1 User Guide*, Aspen Technology Inc., 2001.