Modeling and Simulation of Nuclear Fuel Recycling Systems

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The effective utilization of nuclear power through the use of a sustainable fuel cycle will require the development of efficient systems that address issues of cost, safety, waste, and proliferation. Computing power has grown tremendously over the past several decades, as has the capability of scientific codes to simulate complex systems. This growth, coupled with increased interest in an expected resurgence of nuclear power, provides great opportunity for the application of advanced modeling and simulation for responsible development of future nuclear energy systems.

Expected potential benefits of modeling and simulation of nuclear reprocessing systems include the following:

- Reduced cost of process development by guiding and minimizing the amount of experimental and piloting work required
- Optimized system designs, with technically supported reduced design margins
- Development of new chemical processes with lower cost and waste generation
- Reduced risk of material diversion by providing accurate predictions of materials streams

This article provides a brief overview of the application of modeling and simulation for separations processes relevant to nuclear fuel recycling. This is a subset of the applications for modeling and simulation of the nuclear fuel cycle; the reader is directed to references of systems analysis fuels, waste forms and near-field repositories for discussion of those areas.
The references in this article consist of open-literature publications; therefore, discussion of developments and application of nuclear process model technology is limited to that which is publicly available. The reader is also directed to reports of recent workshops which provide more detailed information on the current state of understanding and opportunities for the future.

Background

The panel report on predictive modeling and simulation from the recent Basic Energy Sciences Workshop on Basic Research Needs for Advanced Nuclear Energy provides a concise description of modeling and simulation and its applicability to nuclear energy systems:

> Modeling and simulation is now considered to be the third branch of science, bridging experiment and analytical theory. Through modeling one incorporates the most relevant theories and concepts developed by the full range of scientific and engineering disciplines. Through simulation one exploits leading-edge computational methods, algorithms, and platforms to obtain results unattainable by any other means. Together modeling and simulation enhance understanding of known systems, provide qualitative/quantitative insights and guidance for experimental work, and produce quantitative results that replace difficult, dangerous, or expensive experiments. These advantages are well suited to basic research for (Advanced Nuclear Energy Systems) because of the experimental difficulties posed by radioactive materials and harsh environments.

While it is understood that modeling and simulation will not supplant experimental testing, the value of modeling and simulation has long been recognized for multiple tasks in the development, design, and operation of reprocessing systems. For example, a 1979 paper on the SEPHIS code for transient simulation of countercurrent solvent extraction indicated it could be used to “(1) guide flowsheet optimization studies and thus minimize the amount of experimentation required to establish a particular set of operating conditions; (2) aid in a nuclear criticality analysis of the solvent extraction plant; (3) analyze the transient response to startup or

shutdown operations and optimize methods of control; (4) study the effects of a process upset caused by component failure, process solution error, or change in feed characteristics, and the return of the process to normal operation on correction of the problem; (5) aid in maintaining an exact inventory of security-sensitive materials for nuclear safeguards purposes; (6) help maintain process control in an automated solvent extraction plant."

A memorandum from 1990 lists the following potential uses envisioned for a process simulator of a full reprocessing plant:

- Operator training
- Plant licensing
- Safeguards studies
- Process and/or chemical flowsheet design confirmation
- Safety studies
- Process diagnostics
- Process monitoring
- Sensitivity studies
- Modeling destination of minor streams having environmental impact
- Process instrumentation studies
- Surge capacity studies

Modeling an aqueous reprocessing plant to a sufficient level of realism to accomplish the tasks listed above is a significant undertaking. As detailed elsewhere in this course, the overall process involves many interconnected steps, each of which entails complex physical and chemical phenomena. Fuel disassembly involves mechanical processes (chopping, clad removal, filtration), chemical dissolution in strong acid, and feed clarification. The fuel solution is then passed through several stages of solvent extraction in order to separate several fission product and actinide streams. Multiple solvent extraction processes are required to accomplish this separation, each using different additives and components in the organic phase, as well as different acid concentrations in the aqueous phase. The separated streams containing the isolated species are further processed and solidified to produce materials meeting specifications for fuel and waste forms. Supporting systems, including those for solvent recovery and off-gas treatment, are also integral parts of a complete plant. Safety and environmental considerations require (1) monitoring of volatile fission product and organic gaseous releases, (2) careful evaluation of component inventories throughout the system, (3) strict attention to nuclear criticality safety in actinide solutions with widely varying component inventories, and (4) control systems that are based on realistic models of processes.

Previous Work

Development of models of aqueous reprocessing systems has historically focused primarily on the solvent extraction steps, which constitute the main separations in the process. The goal is to predict the performance of countercurrent extraction processes, in which the constituents of dissolved spent nuclear fuel are separated by selectively transferring metal ions between aqueous solutions and organic solvents containing complexing agents in a series of fluid-contacting devices. The separations performance is governed by a complicated interplay between reaction kinetics, interfacial mass transport, fluid dynamics, and thermodynamics in highly nonideal, multicomponent, multiphase chemical systems.

Starting in the 1960's, computer models were developed to convert equilibrium distribution data and material balance equations into stagewise calculations for predicting steady-state concentration profiles in solvent extraction processes. A significant amount of effort was subsequently spent during the following two decades to improve the predictive capability of several different model families, including SEPHIS, PUMA, SOLVEX, and AMUSE. Development of these codes has been primarily focused on the partitioning of uranium and plutonium in PUREX systems; however, other systems have been addressed to some extent. AMUSE, initially developed for the TRUEX flowsheet, has been modified to provide predictions of PUREX, UREX, SREX, and, with input of appropriate experimental data, flowsheets for other processes, including CSSX, CCD-PEG, and TALSPEAK. SEPHIS modules have been written for THOREX, BUTEX, and a process for co-extraction of plutonium and neptunium; in addition, preliminary blocks have been added to calculate extraction coefficients or provide coefficient data bases for UREX, TRUEX, FPEX, and TALSPEAK. The models have been of significant value in advancing the development of nuclear separations technologies, guiding experimental development work, and serving as the basis for safety analyses and for accountability in safeguards development. While significant efforts have been aimed at improving predictions through adding features to account for issues such as oxidation reaction kinetics, partitioning variation with ionic strength and temperature, solvent degradation, non-ideal fluid contacting, etc., the existing models are still limited in capability. These models, which are based on equilibrium predictions from correlations to experimental data, do not predict the partitioning of a wide range of trace or non-key species and are not highly accurate under conditions (temperature, concentration, etc.) outside the data ranges for which the correlations were developed. New models for prediction of equilibrium partitioning in solvent extraction continue to be developed.

Little information exists in the published literature regarding the development of full plant simulations. During the 1980's, under the Consolidated Fuel Reprocessing Program, Oak Ridge National Laboratory prepared a complete plant simulation to run in the Advanced System for Process Engineering (ASPEN) simulator. The model tracked up to 52 components throughout a preconceptual plant design containing 32 systems and approximately 700 streams, including fuel cleaning and storage; disassembly and shearing; dissolution and feed preparation; hulls drying; feed clarification; feed preparation and accountability; solvent extraction (codecontamination, partitioning, uranium purification, plutonium purification); solvent extraction ancillary systems (concentration, backcycle, storage, high-activity waste concentration, solvent recovery); process support (acid and water recovery and recycle, process steam, and sump); product conversion; cell atmosphere cooling and purification; process off-gas (vessel off-gas, dissolver off-gas iodine recovery and noble gas recovery); and vitrification and vitrification off-gas treatment. The large size of the simulation relative to the computers of the time required that it be broken down into three segments that were executed separately to achieve a steady-state material balance for the complete plant. This comprehensive model provides an outline of the types of processes involved in a reprocessing plant and a view of the complexity needed in code development for realistic simulations. Recently, Savannah River National Laboratory and Argonne National Laboratory have collaborated on efforts toward reestablishing plant-level plant modeling, coupling current versions of ASPEN with AMUSE.

Significant advances have been made in molecular-level modeling and simulation. An area where this has impacted the development of nuclear separations technology is in the computer-aided selection of sequestering agents for design of extraction solvents. Identification of optimal ligands through experimental development involving synthesis and testing is time-consuming. Computer screening of candidate molecules through electronic structure and force field models has been validated by experiment and can significantly reduce the experimental effort needed.

Recent progress has been made toward computer-generated design of molecules, in which fragment libraries, structure-generating algorithms, and binding affinity evaluation are combined to yield improved candidate ligands. This approach has considerable value in the near to mid term for application to the persistent issue of Am/Cm separations.

Current Status

The status of codes for modeling and simulation of nuclear reprocessing systems in the United States reflects the relative lack of activity in the area over the past two decades. Currently available computer codes that have been developed by the U.S. Department of Energy for dynamic modeling of countercurrent solvent extraction processes specific to nuclear fuel reprocessing efforts are summarized in a recent report. The authors conclude there has been little relevant work in the United States over the past two decades in developing advanced modeling and simulation tools for reprocessing systems. That analysis indicates there are only two simulation codes – SEPHIS and SOLVEX – with dynamic capability and validated to any significant degree against actual data from operating fuel separation processes. The authors also recognize the merit of the steady-state tool AMUSE and its current use among flowsheet developers. As discussed by the authors of the review, each of these codes has significant limitations.

In addition to those established models, a recent effort to develop a Safeguards Performance Model is worth noting. This model, developed using Simulink (a simulation software package that runs under MATLAB), enables the transient analysis of material flow in a reprocessing plant for evaluation of accountancy systems. The current model includes simple descriptions of processes in five submodels – the front end and four solvent extraction processes – and was used to demonstrate the capability of alternative instrumentation approaches to improve materials accountability. This modeling effort indicates the flexibility of newer computational tools, which are expected to provide opportunity for expansion in simulation capability.

In summary, current reprocessing models provide only qualitative predictions of process performance. Empirical models of chemical behavior for major components are used to provide overall descriptions of various reprocessing strategies. Many species are not modeled well, or not at all. The models usually assume chemical equilibrium conditions are met instantly, and do not sufficiently incorporate mass transfer and reaction kinetics. Very few reaction rate constants are known, and where transient conditions are simulated, they are often assumed or selected heuristically. The current models are unable to answer many questions involving interphase transport and equilibria, such as precipitation from solution of micellization, third-phase formation, radiolysis, or determining oxidation states, where multiple possibilities exist. Hence, in order to support both detailed design and safe operation, the improvement of reprocessing models requires improved chemistry modeling, including both equilibria and kinetics. The development of new processes that can produce fuel and waste form materials meeting stringent specifications while also meeting environmental, safety, accountability, and cost constraints demands the development and use of modern, sophisticated modeling tools in concert with experimental development and testing for the design and optimization of reprocessing systems.

Looking Forward — Opportunity to Employ Advanced Modeling and Simulation

Recent workshops and studies have evaluated the research and development needs for advanced nuclear energy systems. These studies point to several key areas where advanced modeling and simulation can play an important role in enabling understanding and providing useful tools for practical implementation.

The workshop on Basic Research Needs for Advanced Nuclear Energy Systems, sponsored by the Department of Energy's Office of Science in 2006, identified several key topical areas for advancement of understanding related to reprocessing systems. It is recognized that modeling and simulation conducted in concert with fundamental experiments will be needed to develop understanding in these areas. The reader is directed to the workshop report, which provides detailed discussion on the following areas:

- Scientific Grand Challenges
  - Resolving the f-electron challenge to master the chemistry and physics of actinides and actinide-bearing materials
  - Developing a first-principles, multiscale description of material properties in complex materials under extreme conditions
  - Understanding and designing new molecular systems to gain unprecedented control of chemical selectivity during processing
- Priority Research Directions
  - Physics and chemistry of actinide-bearing materials and the f-electron challenge
  - Microstructure and property stability under extreme conditions
  - Mastering actinide and fission product chemistry under all chemical conditions
  - Exploiting organization to achieve selectivity at multiple length scales
  - Adaptive material-environment interfaces for extreme chemical conditions
  - Fundamental effects of radiation and radiolysis in chemical processes
  - Predictive multiscale modeling of materials and chemical phenomena in multi-component systems under extreme conditions
- Crosscutting Research Themes
  - Tailored nanostructures for radiation-resistant functional and structural materials
  - Solution and solid-state chemistry of 4f and 5f electron systems
  - Physics and chemistry at interfaces and in confined environments
  - Physical and chemical complexity in multi-component systems

A 2006 workshop co-sponsored by the Offices of Nuclear Energy and Advanced Scientific Computing Research of the U.S. Department of Energy focused on modeling and simulation related to advanced nuclear energy systems. The reader is directed to the workshop report, which identifies several key challenges and discusses potential issues and approaches for developing and implementing modeling and simulation tools useful for advancing nuclear technologies. The modeling and simulation challenges related to separations processes include the following:

- Plant-scale simulation
  - integrated toolset to enable full-scale simulation of a plant – chemistry, mass transport, energy input, and physical layout
  - dynamic plant models
- Computational fluid dynamics

    - multiple fluid phases, fully developed turbulence, non-Newtonian flows, interfacial phenomena, radical chemical processes due to the presence of ionizing radiation
- Predictive methods for thermodynamics and kinetics data as input to process simulators
    - extend currently limited thermodynamics data reliably into broader parameter ranges
    - incorporate limited experimental data and use computational chemistry
- Rational design of the separations system from first-principles physics and chemistry
    - predict what molecules will have the desired properties and can be synthesized
    - reliably predict the properties of liquids, solvation, and kinetics in solution
- Connecting/crossing time and length scales, with uncertainty quantification
    - access longer times without dramatic changes in theoretical and algorithmic approaches
    - span spatial regimes; critical regime is the mesoscale (1 nm–1 μm)
- Data management and visualization
    - capture, manage, integrate, and mine data from a wide range of sources to enable the optimal design and operation of separation processes
    - provide sufficient computer resources and access
    - export control issues

The Advisory Committee on Nuclear Waste and Materials of the Nuclear Regulatory Commission has identified the following research needs relevant to modeling and simulation of reprocessing systems:

- "Knowledge of the split of each chemical species in each process step in the plant (the separation factors), especially concerning tritium, iodine, technetium, neptunium, and radioactive material associated with the cladding
- Developing a model that simulates the interconnected equipment in a facility flowsheet using the separation factors to determine the radionuclide concentrations and inventory. Such models need to accommodate complexation, colloids, internal recycle streams, and important conditions in bulk fluids (e.g., temperature, acidity, radiolysis)
- Understanding stability of organic extractants, solvents, and ion exchange materials and the safety implications of degradation product"

Figure 1 provides a vision toward the possible hierarchy and integration of future development efforts in modeling and simulation to advance technologies and capabilities in nuclear separations and accompanying safeguards to address the challenges listed above. This figure presents four planes—a top plane that represents the physical reality of the interacting unit operations of an integrated recycling plant and three planes that represent different levels of modeling. A primary goal is the development of a plant model that allows dynamic simulations of the separations plant operations under various configurations and conditions, and integration of relevant analysis modules for specific tasks. Future codes will be developed on modern, expandable architecture with flexibility to explore and evaluate a wide range of process options. While the top-level models will initially incorporate relatively simple models for each process, the codes will be developed with the capability for bridging to subscale models to provide required fidelity in chemical and physical processes.

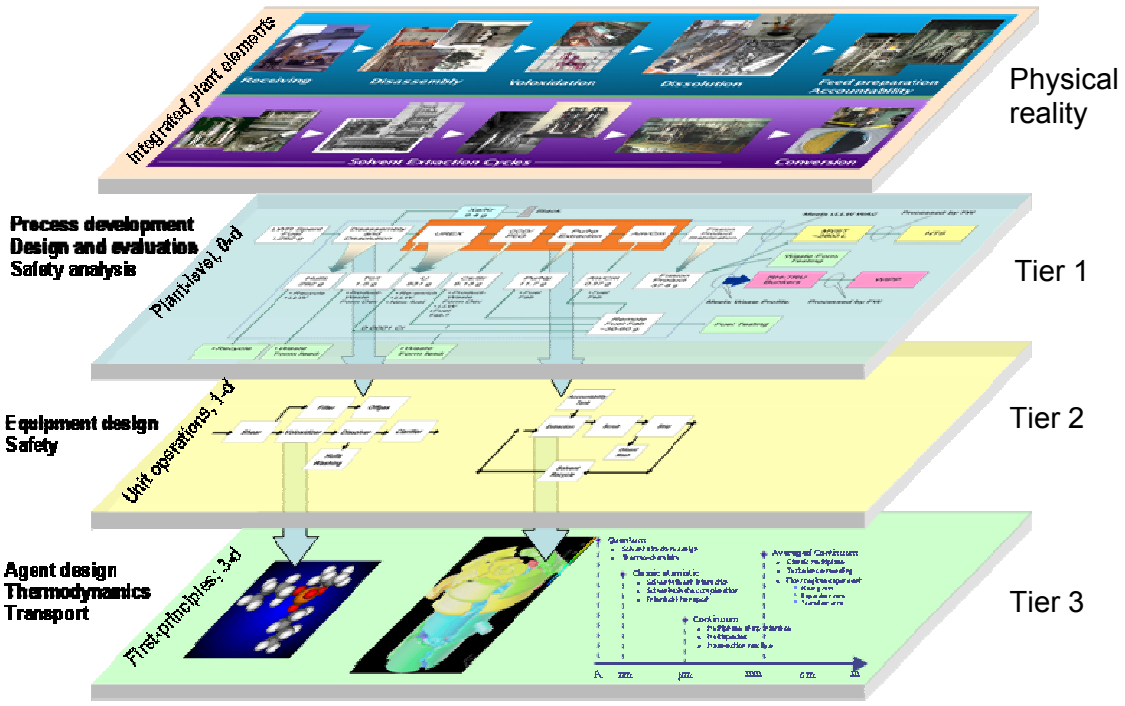

Figure 1. Vision of the hierarchy and integration of modeling and simulation efforts for separations and safeguards.

The top level of modeling in Figure 1 (Tier 1) is a network of units represented by discrete events providing throughput analysis, scheduling impacts, and output chemical compositions. Models at this level generally summarize significant sections of the separations plant lumped together or connected as individual units. These high-level models (i.e., zero-dimensional models for which elapsed time is the principal independent variable) interact through a simulation environment and are calibrated by a combination of experimental data and simulations at other levels of finer detail. Plant-level modeling and simulation is a key practical tool for designing, operating, and safeguarding a separations plant and should be able to follow thousands of streams and chemical species. The computational power requirements for this effort are relatively modest; a dedicated cluster of commodity workstations (e.g., hundreds of processors) is probably sufficient for keeping a live plant simulator running in real time.

The safeguards performance model described previously is an example of current work at the plant level. That work has shown that the process modeling tools currently available provide the capability for a user to define a system that constitutes all or part of a plant, perform simulations under a wide variety of operating conditions, track a large number of variables, and visualize the transient response of any variable in the process (see example in Figure 2). The current tools provide the flexibility to readily change the operations included in the process and their connectivity. The models used for each unit operation can be created with the desired level of sophistication; in addition, it is possible to link to other codes. With further development of transient process models for unit operations, powerful tools will be accessible for analysis of recycling plant design and operation.

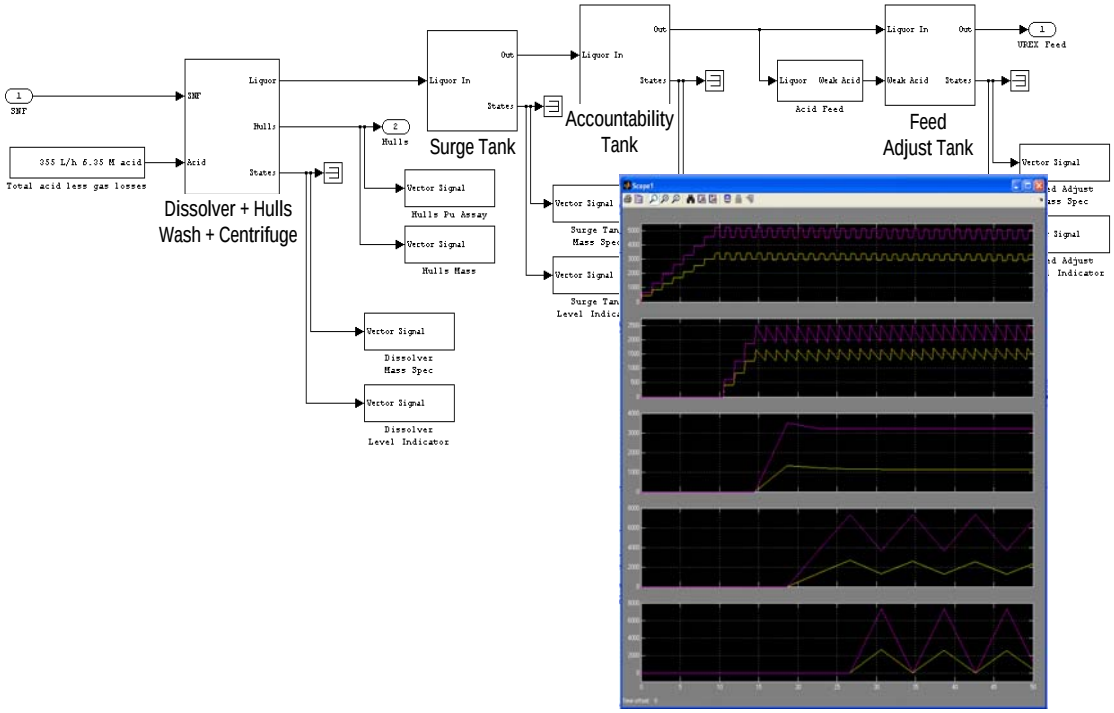

Figure 2. Example of dynamic process modeling made possible with easily adaptable current tools. This figure30 shows a portion of process and instrumentation connections for a model of the head-end section of a plant and sample visualization of transient response in the inventories of multiple tanks through a series of operational cycles.

In view of stringent specifications for the output of a separations plant, the calibration of the plant-level models needs to be improved. This prompts the development of a second hierarchical modeling and simulation level that focuses on any of the top-level units for which a more accurate model is needed. Models at this level (Tier 2 in Figure 1) are modular and may consist of several interchangeable models for the same separations plant section at varying levels of detail. This level includes accurate models of multi-phase flows, high-temperature chemistry, hydraulics, phase transformation, and/or transport, which must be developed for the design and optimization of many required plant operations. These models are necessary to simulate the performance of units under expected plant conditions and to ensure greater reliability in designs before they are physically installed and operated, addressing issues related to process scale-up. The models that currently exist for many unit operations can be further refined; however, for several key operations, even basic models do not currently exist. Unit operations where models can provide substantial immediate benefit to the design of a full-scale plant include voloxidation, solidification, and solvent extraction. In addition to the process chemistry, criticality, and radiation effects must be factored into rigorous designs for optimal operation. Efforts in model and code development must be combined with concurrent experimentation at the benchtop and engineering-scale testing for validation and verification. The computational power required for each of the models at this level is estimated as up to several dozen dedicated processors. Therefore it is envisioned that simulations at this level can take place in parallel to the plant-scale level on an on-demand basis, which could bring the estimate of computing power for both levels together to several hundreds of processors.

An example of current work at this level is fluid dynamics modeling of centrifugal contactors for solvent extraction. Recent efforts have illustrated the capability of continuum modeling to help provide better understanding of the complex flows within these devices. Figure 3 shows examples of simulations of the flow of a single liquid phase in the annular mixing zone of a contactor. These simulations have provided significant insight into system performance and may enable further advancements in design and operation of these units. Further expansion of this capability to provide realistic description of turbulent flows of multiple liquid phases in contactors will require model development and experimental validation; fruitful research in these areas is ongoing.

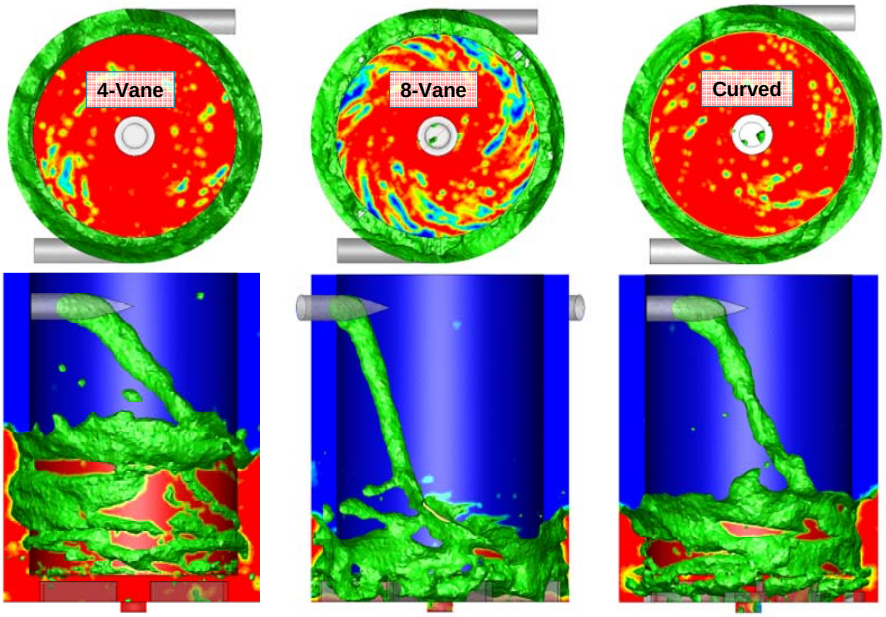

Figure 3. Example of fluid dynamics modeling of the complex free-surface flows in centrifugal contactors used for solvent extraction33. This figure compares the flows of a single liquid phase under identical conditions except for the configuration of the vanes at the bottom of the vessel. Surfaces are color-coded blue for gas-solid, red for liquid-solid, and green for liquid-gas.

At the most fundamental level are key topics for which greater complexity is warranted to deliver accurate predictions, including chemical thermodynamics, interfacial phenomena, reaction kinetics and equilibria, radiolytic and hydrolytic degradation, design of new separating agents, and multicomponent transport in three-dimensional, turbulent multiphase flows. At this root level (Tier 3 of Figure 1) of the modeling and simulation hierarchy resides the most computer-intensive calculations needed to accurately describe time-dependent, three-dimensional (or higher-dimensional) systems. For example, these models could include specific fluid-phase equilibrium and heat and mass transfer calculations, microstructured computational fluid flow, specialized data retrieval, detailed adsorption models, and could even include detail as fine as molecular dynamics, computational chemistry, and radiative transport, if it is necessary, to adequately model the system of interest. Computing power requirement at this level could be the highest available—to hundreds of thousands of processors. These fundamental modeling efforts will calibrate simpler models at the higher level and will form the basis for future advancements in long-term research and development. Fundamental model development projects at this level will necessarily be conducted in close connection with experiments for

validation. The fundamental nature of the work will actively engage academic research, providing educational opportunities, and will enable international cooperation.

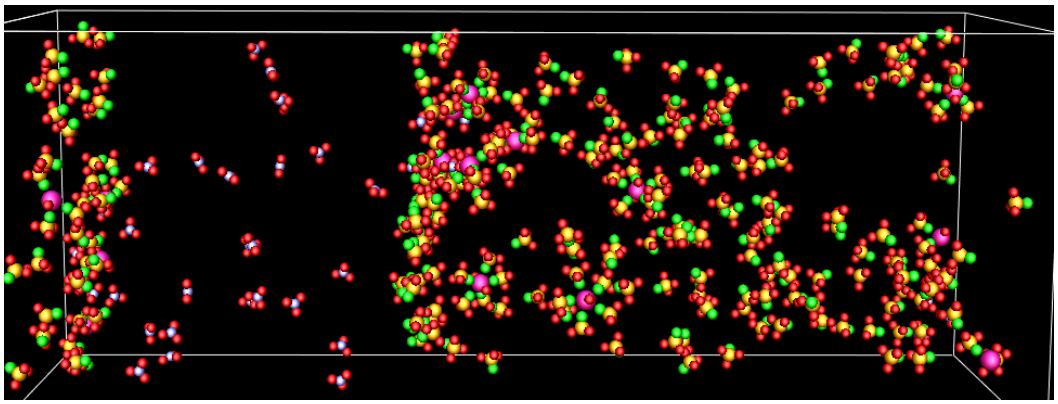

Figure 4. Snapshot of a molecular dynamics simulation of interfacial transport of uranyl nitrate extracted by tri-butyl phosphate (TBP) diluted in dodecane34. In this image, the water and dodecane molecules and the hydrocarbon tails of the TBP molecules are not shown. The uranium atoms of uranyl ions are depicted by magenta spheres, the nitrogen atoms of nitrate ions by blue spheres, the phosphorous atoms of TBP molecules by yellow spheres, and the electro-active oxygen of the TBP phosphoryl groups by green spheres. The complexation of uranyl by TBP is readily visualized by the clustering of green spheres around a magenta center.

Molecular-level modeling has advanced to a point where it can provide valuable contributions toward the development of separations systems. In addition to the agent-design example discussed above, the simulation of molecular-level transport processes near interfaces is an example of an important area where progress in modeling may translate into practical understanding on the performance of separations processes. An example of current work at this fundamental level is shown in Figure 4, a snapshot of a molecular dynamics simulation of uranyl nitrate extraction from an acidic aqueous phase into a tri-butyl phosphate/dodecane solvent. Recent simulations, which include a large number of atoms (>18,000), long-chain hydrocarbon diluents, and flexible molecules, have provided significant insight into the nature of interfacial transport. For example, surfactant behavior of TBP was exhibited, with a resultant heterogeneous distribution of diluent near the interface. This suggests a bridging mechanism for transporting complexed ions into the bulk phase of the solvent. These simulations indicate potential value in further experimental and computational study of molecular aggregation and transport at interfaces. For molecular-level simulations to provide useful data on transport processes for use in higher level models, further work is needed to improve force fields for description of molecules, including systematic experimental calibration.

The NEAMS Program

Motivated by the challenges and needs in nuclear energy systems that can be addressed by modeling and simulation, the Office of Nuclear Energy of the U.S. Department of Energy has articulated a vision for a Nuclear Energy Advanced Modeling and Simulation (NEAMS) program. The NEAMS vision is “To rapidly create, and deploy next generation, verified and validated nuclear energy modeling and simulation capabilities for the design, implementation, and operation of future nuclear energy systems to improve the U.S. energy security future.”

NEAMS is aimed toward building on the success of recent programs in advanced scientific computing, namely, ASCI and SciDAC, with a focus on very different challenges. These challenges include the need for nuclear energy systems to be licensed by regulators and moving advanced technologies out of the research environment and into the hands of the engineers who will design, build, and operate the new nuclear energy systems. NEAMS will provide a comprehensive solution and is organized into the following five elements:

- Integrated Performance and Safety Codes—End-to-end codes to understand the detailed, integrated performance of new nuclear systems including the following:
  - Nuclear Fuels
  - Reactor Core & Safety
  - Separations and Safeguards
  - Waste Forms and Near-Field Repositories
- Fundamental Methods and Models
- Verification, Validation, and Uncertainty Quantification
- Capability Transfer Enabling Computational Technologies

Summary

Modeling and simulation have provided useful input to the development of fuel cycle separations over the past several decades. With significant scientific advancements and vast increases in computational power, modeling and simulation can play an increasing role in solving the complex challenges to be overcome in developing advanced nuclear energy systems. In conjunction with experimental efforts, concurrent development of tools at three levels of detail—plant, unit operations, and fundamental—is needed to enable fruitful progress in the near, mid, and long term.

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