Development of Integrated Process Simulation System Model for Spent Fuel Treatment Facility (SFTF) Design

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Project title: Development of Integrated Process Simulation System Model for Spent Fuel Treatment Facility (SFTF) Design

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Abstract

The Advanced Fuel Cycle Initiative (AFCI) and Transmutation Research Program-University Participation Program (TRP-UPP) supported by Department of Energy of the United States have been developing many important technologies for the transmutation of nuclear waste to address long-term disposal issues. While successfully embedding AMUSE module into a dedicated System Engineering Model (TRPSEMPro), developed by the Nevada Center for Advanced Computational Methods (NCACM) at the University of Nevada-Las Vegas collaborating with Argonne National Laboratory (ANL), ANL is interested in further simulating the Light Water Reactor (LWR) Spent Fuel Treatment Facility (SFTF) combining commercial process simulation and analysis packages and core calculation of the AMUSE that derived for using with the UREX+ process. The designed SFTF will receive, temporarily store, and prepare spent nuclear fuel for leaching. The major objectives of this research proposal are to develop a framework for simulating the Spent Fuel Treatment Facility (SFTF) process using AMUSE code, commercial process package such as ASPEN-PLUS, HYSYS and PRO/II and system engineering model such as TRPSEMPro’s flexible parameter optimization modules, to develop a middleware package that can communicate between the AMUSE code and any selected commercial packages, to extend the existing system engineering model for optimization process that includes process simulation results, and to include a scenario-based database system that efficiently reports required information as chart output using web-based programming, and Microsoft Visual Basic (MS VB). In addition, the proposed project will lead to two M.S. theses from the participating graduate students in this three-year project and will also generate at least three journal papers and a few conference proceedings.

1. Overview

The Advanced Fuel Cycle Initiative (AFCI) and Transmutation Research Program-University Participation Program (TRP-UPP) supported by Department of Energy of the United States have been developing many important technologies for the transmutation of nuclear waste to address long-term disposal issues. Integrating and enhancing the Argonne National Laboratory’s AMUSE code that consists great part of chemical separations processing, are the major part of the previous 3-year project. Figure 1 shows a block diagram of the chemical process as envisioned by Argonne National Laboratory (ANL) researchers. Nearly all issues related to risks to future generations arising from long-term disposal of such spent nuclear fuel are attributable to ~1% of its content. This 1% is made up primarily of plutonium, neptunium, americium, and curium (the transuranic elements) and long-lived isotopes of iodine and technetium created as products from the fission process in power reactors. When transuranics are removed from discharged fuel destined for disposal, the toxic nature of the spent fuel drops below that of natural uranium ore (that was originally mined for nuclear fuel) within a period of several hundred years.
Figure 1 – Overall Chemical Separations System for AFCI Program.

While successfully embedding AMUSE module into a dedicated System Engineering Model (TRPSEMPPro), developed by the Nevada Center for Advanced Computational Methods (NCACM) at the University of Nevada-Las Vegas, Argonne National Laboratory is interested in further simulating the Light Water Reactor (LWR) Spent Fuel Treatment Facility (SFTF) combining commercial process simulation packages and core calculation of the AMUSE that derived for using with the UREX+ process. The designed SFTF will receive, temporarily store, and prepare spent nuclear fuel for leaching. The leached fuel will then be separated by solvent extraction to recover:

- Uranium, Technetium and Iodine for disposal as low-level waste (LLW)
- Cs/Sr for decay storage and eventual disposal as LLW while Pu/Np for production of mixed-oxide (MOX) fuel
- Am/Cm for short-term storage and eventual use as fast-reactor fuel
- Two raffinates from the UREX+ process containing all soluble fission products but Cs, Sr, Tc, I, and the rare earth elements will be converted to a solid for disposal in the repository.

The following technologies will be used for product stream solidification:

- Evaporation/denitration for Uranium
- Borohydride technetium reduction for Tc, producing Tc metal
- Continuous oxalate precipitation process for Pu/Np stream, producing Pu/Np oxide
- Evaporation/denitration for Am/Cm stream, producing Am/Cm oxide
• Steam reforming applies to Cs/Sr and raffinates, generating a mineralized solid waste forms for disposal.

Key process unit operations involved in SFTF modeling in Figure 2 are:
• Fuel dissolution, including mechanical chopping, voloxidation, and leaching processes
• Fuel separations, consisting of a series of five solvent-extraction flowsheets that defines the following operations:
  o Tc/U recovery (UREX),
  o Cs/Sr recovery (CDC-PEG),
  o Pu/Np recovery (NPEX),
  o Am/Cm and rare-earth fission products recovery (TRUEX), and
  o Am/Cm separation from the rare earths (CYANEX 301).
• Product solidification and waste forms, including evaporation, steam reforming, calcining, and distillation
• Off-gas treatment, including H-3 molecular sieves, I2 adsorption, and C-14 carbonate precipitation

Figure 2 not only shows a further simplified construction blocks but also clearly identify the proposed chemical separation processes, such UREX and TRUEX, as well as system processes, such as precipitation and denitrification. The chemical separation process is calculated by the Argonne Model for Universal Solvent Extraction (AMUSE) code, developed in the 1980s, that designs multi-stage countercurrent flowsheets for the TRUEX solvent extraction process. The GTM and AMUSE predicts chemical behavior in solvent extraction processes by calculating component distribution ratios. Further, the countercurrent mass balance algorithm contains terms for stage efficiency and other-phase-carryover for both the aqueous and organic phases. All five process segments of the UREX+ process were programmed into AMUSE code. The currently developed TRPSEMPro (Transmutation Research Project System Engineering Model PROgram) by the NCACM can be highly integrated with the AMUSE code and commercial packages to calculate the complex interactions between proposed process changes.
Figure 2. Definition of construction blocks for chemical separation and system process.

System simulation generated from the commercial package requires the ability to define actinides and fission product physical-chemical databases, the modules that can accurately predict aqueous electrolyte systems and the ability to create a thermodynamic package for phase diagrams and coupling of phase diagrams with kinetic process modeling.

A complete work frame for integrating three major parts together is shown in Figure 3. Core chemical separation calculation is carried out by the AMUSE code. Process simulation is generated by commercial package while a parameter optimization process is fine tuned by the TRPSEMPRO package. To simulate the SFTF, AMUSE will be used for all the solvent extraction processes. Each solvent extraction process is treated as one AMUSE unit operation. These unit operations will be integrated with the commercial process simulation package as shown in Figure 3. It is critical that the commercial process simulation package be able to easily communicate with AMUSE. All principal parameters within the AMUSE code can be systematically adjusted by the TRPSEMPro system and feed back into process simulation.
The development process is considered a multi-year task. Significant data communication and analysis work have been development from the previous 3-year project funded by the TRP-UPP/DOE program. A complete new research scope here will be heavily benefited from the well-established knowledge in communicating with AMUSE code as well as data manipulation and reporting.

Figure 3. Complete framework for the newly proposed research work.
2. Summary – System Engineering Model Development

2.1 System Engineering Model

2.1.1 Overview

Implementing system engineering model is an approachable solution to solve complex chemical separation process. To help user to build a model on a system level and perform system analysis, a software tool for building system models was created. In the NCACM’s software design, system engineering model includes analysis tools, such as parameter study, optimization, and Design of Experiments (DOE) study, especially designed for the chemical separation process. Schematically, the model development approach is shown in Figure 4.

![Figure 4. Schematic representation of the process for model development](image)

The beginning of any system study is Block 1 in Figure 4, System Identification and Familiarization. Often, to a simple system, identifying the essential components of the system that collectively undergo the cause and effect action associated with the system is obvious. Three center blocks in Figure 4 are the major development tasks for the project. The involved TRP chemical separation process can be systematically identified as a group of blocks that have specific separation functions: existing LWR spent fuel, separation processes, fuel fabrication, transmutation, disposal as a low-level waste (LLW), and the reprocessing of fuel after transmutation. One block’s effluent flows into another block as input. Each block has its process target. Task in ANL is to develop each individual model for these chemical separation processes. Most of the blocks (sub processes) are still under development or revision. Currently our research is focused on the Uranium Extraction process (UREX) block as shown in Figure 5.

The UREX process is for separation of TRU (Transuranic) elements from the dissolved spent fuel as shown in Figure 5. The process needs to report 95% of Tc and 99.9% of U to a separate effluent. The U/Tc–free TRU/Fp stream is then fed to PYRO-A process. The recovered Uranium is purified for low-level waste (LLW) disposal while recovered technetium is used for transmutation of targets. While chemical engineering systems are getting more complex, the process becomes more difficult to analyze mathematically. The development of Systems Engineering Model allows industries to model the process more quantitatively and to study the interactions between subsystems and performance of the model under the influence of various design parameters.
Systems engineering is a multidisciplinary function dedicated to controlling design so that all elements are integrated to provide an optimum, overall system. Developing a systems engineering model of the overall chemical separation process would be beneficial to analyzing complex interactions between proposed process changes. The model will evolve over several years to incorporate all process steps and to improve process modules as more knowledge is gained. To UREX process, the visual definition is performed in program module AMUSESimulator, and user can actively define inputs/outputs of this process module in graphic format. The definition of inputs/outputs is the key to a proper mass balance. Block diagrams, signal-flow graphs, and organizational diagrams, as graphical modeling tools have been developed in this project. The basic single-input, single-output model has been widely used and justified as an excellent beginning model for many systems.

In TRPSEMPro, design problems are specified, and simulation codes from multiple disciplines are coupled into a system model description file written in XML. After a description file is created, the user can use the application interface to set up, monitor, and analyze a design run. The designed system model includes four main parts as shown in Figure 6.

- **System Manager** – System Manager is the main application interface, from where a user can launch any of the application interfaces. The System Manager allows the user to set up and run a design problem.

- **Model Integration** – Model Integration enables user to couple simulation programs to system engineering model and specify their execution sequence. Model Integration provides a GUI that acts as a front end for creating a system engineering model description file written in XML language.

- **Study Plan** – Study Plan provides a convenient means to provide problem formulation information to specific design parameters, then allowing user to control information in a specific problem. Techniques such as Optimization and Design of Experiments (DOE) are available in Study Plan.

- **Solution Viewer** – Solution Viewer provides a visual means to monitor the optimization process as it moves through the design space. Solution Viewer provides several tables and graphs that can be used to view the runtime changes.
The role of TRPSEMPro is to automate the design-evaluate-redesign cycle, eliminate the human intervention bottleneck. The Study Plan (a single, self-contained design unit with its own parameters, optimization strategy) drives the automation of the cycle. By eliminating costly human intervention, TRPSEMPro lets the designer refocus valuable engineering time on design analysis and selective refinement of the design process. TRPSEMPro system can be harnessed to execute more than just computer simulation programs. TRPSEMPro can automate the execution of a sub model (independent models that are invoked by the main model) to support hierarchical, multilevel design problems.

2.1.2 Key Features for the System Engineering Model

1. SystemManager Initialization - When user starts the program TRPSEMPro, the main GUI of SystemManager is the default user interface, as shown in Figure 7, where user can access any three main modules - ModelIntegration, StudyPlan, and SolutionViewer.

2. ModelIntegration module shown in Figure 8 can add individual models into the system engineering model. These individual models can be the calculation model, the simcode model or the chemical separation models.
2.2 Integration Between the AMUSE Code and the System Engineering Model (TRPSEMPro)

2.2.1 Overview

Figure 9 shows the framework of AMUSESimulator. Initially, each of the different steps in the UREX process needs to be outlined on the screen and allows easy modification by the user. After all the modifications have been made, AMUSESimulator will write all the modifications into Export File which is the input file for AMUSE Macro, and triggers the execution of AMUSE Macro; at the end of AMUSE execution, a report file is generated. To expedite the data usage, AMUSESimulator provides user with the capability that all the Export File and Report File information can be saved to the Microsoft SQL Server 2000 and XML databases.

The main objects in UREX process can be identified in Figure 10. According to the functionality required for AMUSESimulator, the main domain classes and architecture of AMUSESimulator are then obtained. Database design for AMUSESimulator is shown in Figure 11. Figure 12 shows the database interface that displays all the saved testing sets from the previous test scenarios. The strength of the database management is to provide researchers with the capability to retrieve historical testing cases for further research.
Figure 9. Framework of AMUSESimulator

Figure 10. Identify Objects in UREX Process.

Figure 11. Diagrams for the Structure of Database Design.
2.2.2 Key Features for the AMUSESimulator

1. The Main GUI for AMUSESimulator - The main GUI for AMUSESimulator includes five parts as shown in Figure 13. Menu and toolbar, located at the top part of the interface, can invoke all the available commands. Flowsheet contents displayed in tree view format, located at the left part, can select different section, stream, while the middle part shows the drawing blocks where user can select different sections, streams by clicking mouse at corresponding area. Property list on the right displays all the properties of selected object, such as flowsheet, section, or stream. Status Window on the bottom shows detailed information of the selected object.

2. Flowsheet property dialog shown in Figure 14 provides property information for flowsheet, where the user can see and modify the properties of the flowsheet.

3. Figure 15 shows the case that a section is selected; both the property window (right part) and status window (lower part) show the information about this selected section. Figure 16 further shows the property dialog for the selected section, where user can see and modify the properties of the selected section.

4. Figure 17 shows the case that a stream is selected; both the property window (right part) and status window (lower part) show the information about this selected stream. Figure 18 shows the property dialog for stream, where user can see and modify the properties of the selected stream.
5. Database Viewer, shown in Figure 19, is a utility program that user can browse through entire historical data.

Figure 13. Whole Flowsheet is Selected by Default

Figure 14. Property Dialog for Flowsheet
Figure 15. Information display for the selected Section.

Figure 16. Property Dialog for Section
Figure 17. Interface for the selected stream.

Figure 18. Property Dialog for Stream.
3. Proposed Research – Overview

The major objectives can be described as below that will lead to a creation of framework that combines all the strengths of AMUSE’s complicates calculation, well-established commercial system process package such as ASPEN-PLUS, HYSYS and PRO/II and TRPSEMPro’s flexible parameter optimization modules. Development of the process simulation code can be done using the solvent extraction process experience at Argonne National Laboratory and in collaboration with the Nevada Center for Advanced Computational Methods (NCACM) at the University of Nevada-Las Vegas (UNLV).

- Develop a framework for simulating the Spent Fuel Treatment Facility (SFTF) process using AMUSE code, commercial process package, such as ASPEN-PLUS, and system engineering model.
- Develop a middleware package that can communicate between the AMUSE code and any selected commercial packages.
- Extend the existing system engineering model for optimization process that includes process simulation results.
- Include a scenario-based database system that efficiently reports required information as chart output using web-based programming, and Microsoft Visual Basic (MS VB).

A comprehensive study of commercial simulation packages has been performed by the Argonne National Laboratory. The three most promising packages identified were ASPEN-PLUS,
HYSYS and PRO/II, while all these packages require purchasing the aqueous electrolyte package, OLI that can incorporate AMUSE as the unit operation for solvent extraction. Since the ASPEN-PLUS provides a greater capability for solids handling, the major task here will create a communication tool between the AMUSE code and the ASPEN-PLUS. ASPEN-PLUS or an alternative, used for the SFTF process simulation, will be thoroughly studied for data parsing capability.

Due to the extremely importance of AMUSE calculation, more communication between AMUSE and any other process-related packages is expected. The NCACM team intends to develop a middleware package that can communicate between the AMUSE code and any selected commercial packages.

Although current SFTF process, prepared by the Washington Group International (WGI), utilized ASPEN-PLUS calculations contained in AMUSE, the single-scenario calculation is used. Such process can be further refined into multi-scenario simulation with optimization concept in mind while using the currently developed TRPSEMPro package.

An interface will be developed that incorporates AMUSE as the solvent extraction unit operation and any commercial package as the head-end and back-end processes.

4. Technical Impact

A comprehensive SFTF process simulation can greatly facilitate the evaluation of overall systems options. This capability will be become increasingly important as it becomes necessary to down-select reactor types, fuel types, and multi-recycle modes. Scenario-based database systems analysis will make it possible to present decision-makers with concise evaluations of system options and their characteristic features. Optimization tool associated with the system engineering model can further speed up the selection of proper chemical separation parameters.

At the end of the year three, the level of sophistication for such framework will conceivably to make relative comparisons of process options with regard to waste generation, proliferation resistance, throughput capabilities, facility requirements, and cost. With confidence in the models, the decision-making process can be given greater objectivity and technical credibility.

5. Research Approach

The proposed research activities have been broken down into several tasks. These tasks are outlined below.

Activity 1: Identify the parameter correlations and data manipulation channels between the AMUSE code and the selected process simulation software package

1. Identify AMUSE input/output (from AMUSE Macros)
   - Specify input streams associated with typical spent fuel.
   - Determine all inputs and outputs for each process block.
- Select key components to monitor throughout process.
- Document key individuals or organizations for each process block.

2. Identify AMUSE input/output (from TRPSEMPro package)
   - Specify input streams associated with typical spent fuel.
   - Determine all inputs and outputs for each process block.
   - Select key components to monitor throughout process.

3. Identify the communication protocols from the selected process simulation package
   - Specify the possible data manipulation routes.
   - Determine the most suitable channel for data transferring.
   - Streamline the discrepancies among AMUSE code, TRPSEMPro and the selected process package such as ASPEN-PLUS, HYSYS and PRO/II.

Activity 2: Develop middleware for data manipulation among the AMUSE code, system engineering model and the selected process simulation software package


2. Middleware might include a database design for temporary data repository.

3. XML data communication standard will be used for data manipulation. However, some of the process package might not support XML-enabled transfer and will be considered respectively.

4. Middleware implementation – use programming languages, such as MS VB and VB.net.

Activity 3: Integrate with the System Engineering Model (TRPSEMPro)

1. Review/Analyze Code Structure – Each year this review activity is carried out to make sure we are still on track and developing a useful product for the national laboratories. This work insures that all input data is properly stored within the existing AMUSE environment and that the output from AMUSE is easily accessed and analyzed.

2. Modify/Improve Software – The overall system model is evolving as the TRP Project continues. UNLV will work to incorporate changes to the overall process as time advances. Make changes to the software that allows it to be used as a part of the overall systems engineering model and that allows it to be used as a stand-alone process model. Determine what modifications would be necessary to allow the optimization of the process.

3. Systems Analysis – Use the software to analyze a variety of proposed configurations (input from Argonne National Laboratory Collaborators).
Dr. Chen’s efforts will center on initializing and designing the framework relationship from the process and chemical engineer’s viewpoint. He will also provide software architecture guidelines to establish the baseline SFTF process model from which modifications and improvements can be made.

Dr. Sean Hsieh will carry the work from design, implementation to the deployment stage. Further verification work will also considered and will work closely with student workers, staffs and the ANL researchers.

Student programmers will work on developing Visual Basic interface between process simulation package such as ASPEN-PLUS, HYSYS and PRO/II and the AMUSE-related calculation. Improvement made for the system engineering model will be responsible for both students and Dr. Hsieh.

6. Capabilities at UNLV and DOE Labs

Dr. Yitung Chen is Associate Professor of the Department of Mechanical Engineering and Associate Director of the Nevada Center for Advanced Computational Methods (NCACM) at the University of Nevada Las Vegas, and would serve as Principal Investigator. He received his B.S. degree in Chemical Engineering in 1983, and his M.S. and Ph.D. degrees in Mechanical Engineering in 1988 and 1991, respectively, from the University of Utah. He also has a minor degree in Nuclear Engineering. He was a consultant for several engineering companies from 1991 to 1993. Dr. Chen is an expert in experimental and computational aspects of momentum, heat, and mass transfer. His research interests include chemical kinetics modeling, high level radioactive waste repository design, atmospheric sciences, magnetohydrodynamics modeling, ground water transport, energy conservation, and biomedical engineering. He also has a strong background in organic chemistry, biochemistry, polymer chemistry, and physical chemistry. His research experience includes being PI and co-PI on projects involving the study of flow and heat transfer and species transport in unsaturated porous media funded by DOE, the Transmutation Research Program-University Participation Program funded by DOE, the high temperature heat exchanger design funded by DOE, the Solar Thermal Chemical Hydrogen (STCH) Generation funded by DOE, the burning of rocket motors under the Joint Demilitarization Technology (JDT) program funded by DOD, Radiography Stockpile Stewardship Program funded by DOE, ATLAS project funded by DOE, JASPER project funded by DOE, high-level radioactive waste material repository design funded by DOE, high performance computing project funded by NSF, and atmospheric modeling funded by the NOAA Cooperative Institute for Atmospheric Sciences and Terrestrial Applications. He is also co-PI on an EPA project dealing with environmental monitoring for public access funded by EPA and a groundwater modeling project funded by DOE.

Dr. Sean Hsieh is Assistant Research Professor of the Department of Mechanical Engineering and a member of the Nevada Center for Advanced Computational Methods at the University of Nevada, Las Vegas. He received his B.S. degree from the Department of Geology, National Taiwan University, Taiwan, in 1986, M.S. degree from the Department of Geological Science, University of Rochester, NY in 1992 and Ph.D. degrees from the Environmental Institute,
Oklahoma State University in 1997. During the Post-doc research period, Dr. Hsieh worked with
the Sandia National Laboratories, Albuquerque, NM on the Waste Isolation Pilot Plant (WIPP)
project for two years. Before employed by the NCACM, Dr. Hsieh has been working as an
independent consultant in information technology related fields for three years. Dr. Hsieh is
specialized in system integration, database design and implementation, digital image processing
and data mining. He has working with Argonne National Laboratory on “System Engineering
Model for spent fuel extraction” project. His current research interests also include atmospheric
science forecasting, groundwater contaminant transport, energy conservation and high
performance computing. His research experience includes being PI and co-PI on projects
involving air quality system integration, funded by State of Nevada and system engineering
modeling study, funded by DOE, respectively.

Researchers in the Chemical Technology Division of Argonne National Laboratory have been
involved in the development of chemical separations technologies for the nuclear industry since
formation of the division in 1948. Argonne is leading the development of chemical processing
technology for the AAA program and, along with the Savannah River Technology Center and
the Oak Ridge National Laboratory, is actively involved in the conception and experimental
confirmation of advanced separations processes for a variety of spent fuel types. Two senior
scientists with Argonne’s Chemical Technology Division, Drs. Laidler and Vandegrift, will
provide support to this project. Dr. Laidler is currently responsible for all U.S. work on the
development of chemical separations technology for the AAA program, and Dr. Vandegrift has
over two decades of experience in the development of solvent extraction processes for chemical
separations. Dr. Vandegrift is the developer of the GTM code that was recently renamed
AMUSE.

7. Project Timeline with Milestones and Deliverables

The proposed schedule for all tasks and significant meetings is shown on the following page.
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<td>1. Identify AMUSE parameters (from AMUSE)</td>
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<td>3. Identify communication protocols from the package</td>
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<td>4. Reporting Requirement</td>
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<td><strong>Develop middleware for data manipulation</strong></td>
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<tr>
<td>1. Define Framework Architecture</td>
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<td>2. Design/develop database repository with middleware</td>
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<td>3. Implement XML data communication standard</td>
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<td>4. Implement Middleware</td>
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<td>1. Review/Analyze Code Structure</td>
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<td>3. Systems Analysis</td>
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Notes: In addition to the proposed formal meetings, researchers from UNLV and ANL staffs will correspond as needed to insure program goals are being met.

Trip 1 Meeting with ANL – Discuss Progress and project refinement
Trip 2 Meeting with ANL – Discuss Progress

Figure 20 - Proposed Timeline for Research Tasks.

Work is assumed to commence on September 1, 2004. Additional travel may be necessary for interactions relating to other specific tasks.