

Systemic Analysis, Mapping, Modeling, and Simulation of the Advanced Accelerator Applications Program

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Abstract

Advanced chemical separations methods envisioned for use in the Department of Energy Advanced Accelerator Applications (AAA) program have been studied using the Systemic Analysis, Mapping, Modeling, and Simulation (SAMMS) method. This integrated and systematic method considers all aspects of the studied process as one dynamic and inter-dependent system. This particular study focuses on two subjects: the chemical separation processes for treating spent nuclear fuel, and the associated non-proliferation implications of such processing. Two levels of chemical separation models are developed: level 1 models treat the chemical process stages by groups; and level 2 models depict the details of each process stage. Models to estimate the proliferation risks based on proliferation barrier assessment are also developed. This paper describes the research conducted for the single-stratum design in the AAA program. Further research conducted for the multi-strata designs will be presented later. The method and models described in this paper can help in the design of optimized processes that fulfill the chemical separation process specifications and non-proliferation requirements.

I. INTRODUCTION

The Department of Energy's Advanced Accelerator Applications (AAA) program supports the long-term role of nuclear power in U.S. energy production. The proposed AAA system reduces by 95% the mass of high level waste to be disposed by separating out waste appropriate for low level storage and greatly reduces the radiotoxicity of nuclear waste by transmuting most of the remaining heavy metal as well as important long-lived fission products.

Two of the primary goals of the AAA program are (1) to reduce the amount of material sent to high-level waste repositories and (2) to eliminate from high-level waste certain radionuclides that dominate its radiotoxicity, by transmuting them with high energy neutrons in an accelerator-based transmutation system. By separating the uranium from the spent fuel before transmutation, nearly 95% of the mass can be disposed of as non-transuranic (TRU) low level waste. This not only permits

disposal of the large mass of uranium at a much lower cost than high-level waste, but also removes the material from the more advanced processing steps needed to prepare the AAA targets and fuel. By transmuting specific isotopes, significant reductions can be achieved in the potential dose rate to the general public from materials in the repositories. The key elements to be transmuted include technetium, iodine and the transuranic elements Np, Pu, Am and Cm.

In order to analyze and optimize the above chemical separation processes, the AAA processes have been studied using the SAMMS (Systemic Analysis, Mapping, Modeling and Simulation) method. This particular study focuses on two subjects: the chemical separation processes that treat and process spent nuclear fuel and the associated non-proliferation implications.

The purpose of this study is to develop a set of quantitative models of these subjects to simulate and evaluate the effect of spent fuel processing on reduction of proliferation risk and environmental impact. The dynamic model simulations can also provide valuable feedback to optimize the specifications of the chemical separation processes. The processes would then be designed and operated to meet total program goals, both short-term and long-term.

The current study, however, did not include the modeling of new fuel fabrication, reactor processes or transmutation, their entry and exit stream constituents, or the internal transformation of material. Therefore, important material flow consistency checks could not be included. Assurance of consistency was not possible between the streams entering fuel fabrication from chemical separation, on the one hand, and, on the other, the subsequent streams from reactor or transmutation facilities into the follow-on chemical separations.

The detail of this study is presented in a report¹. This paper provides a summary of the study.

II. BACKGROUND

Figure 1 shows a flowsheet for the baseline chemical separation process conceived for the single-tier transmutation system that was the subject of the original study of an accelerator-driven transmuter. Since the development of this approach, other concepts that include the use of fast-spectrum reactors have been introduced. As these concepts are developed, the methodology and models presented in this paper can be readily adapted to analyze those systems.

After an appropriate cooling time, spent commercial reactor fuel is sent to the separations facility. This fuel is typically uranium oxide enriched to a few percent in ²³⁵U that has been cooling in local storage facilities for a number of years. As indicated in Fig. 1, the fuel is removed from the subassemblies and individual fuel pins are chopped into segments and the oxide is dissolved. During this step, iodine is released and collected for later incorporation into

ATW target assemblies. The uranium in the fuel is separated from the rest of the material using the UREX process. This uranium is packaged for disposal at a low-level waste repository. During UREX processing, Tc is separated and collected for later inclusion in target assemblies. The cladding hulls are separated, cleaned, and sent to a process that consolidates them into a metal waste form suitable for disposal.

The remaining spent fuel components, the transuranic elements and the fission products, are then sent to the PYRO-A process. This pyrochemical process separates the majority of the fission products from the TRU elements. The fission products are sent to a processing station that produces waste forms acceptable for long-term storage at high-level waste repositories. The TRU material is fabricated into fuel elements for the subcritical assemblies that will be irradiated in the accelerator-driven transmutation system.

Once the transmuter units are in equilibrium operation, the fuel and target assemblies will be reprocessed until the desired level of burnup (i.e., isotope reduction) is achieved. The PYRO-B process will remove the TRU remaining in the transmuter fuel and route it back to the fuel element production station. Fission products are removed and sent to the same waste treatment and packaging steps that handle the fission products from the PYRO-A process.

III. APPROACH

The basic approach developed in this study is to track the spent fuel material breakdown components as they pass through each stage in the AAA system. The quantitative relationships between the components are captured and built into the simulation models. The co-existing and time-dependent relations associated with key elements are accounted for as well. The variations of the spent fuel material that occur in each stage have been systemically analyzed and considered. The phenomena shown in each stage are recorded and integrated according to the sequences into the models to perform the simulation of the entire process.

Two sets of models are developed, namely Chemical Separation Models and Proliferation Resistance Barrier Models, to analyze the chemical separation processes and the associated non-proliferation implications, respectively. They are then integrated together to form the simulations.

¹ Systemic Analysis, Mapping, Modeling, and Simulation Applied to the Advanced Accelerator Application Program, Developed for US DOE and ANL by Advanced Systems Technology and Management, Inc. May 2001

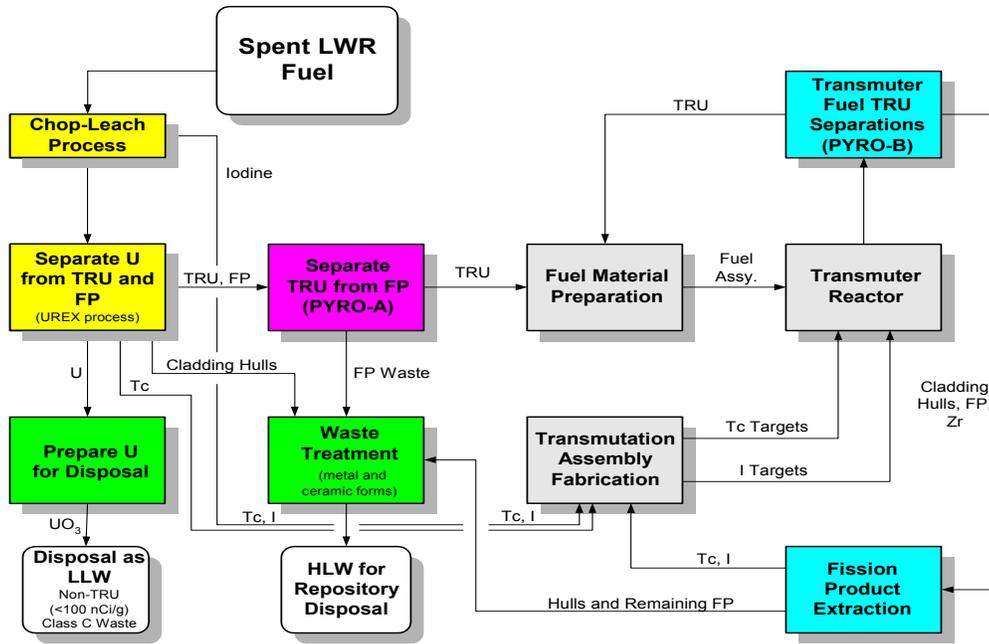


Figure 1. Baseline AAA chemical separations process flowsheet

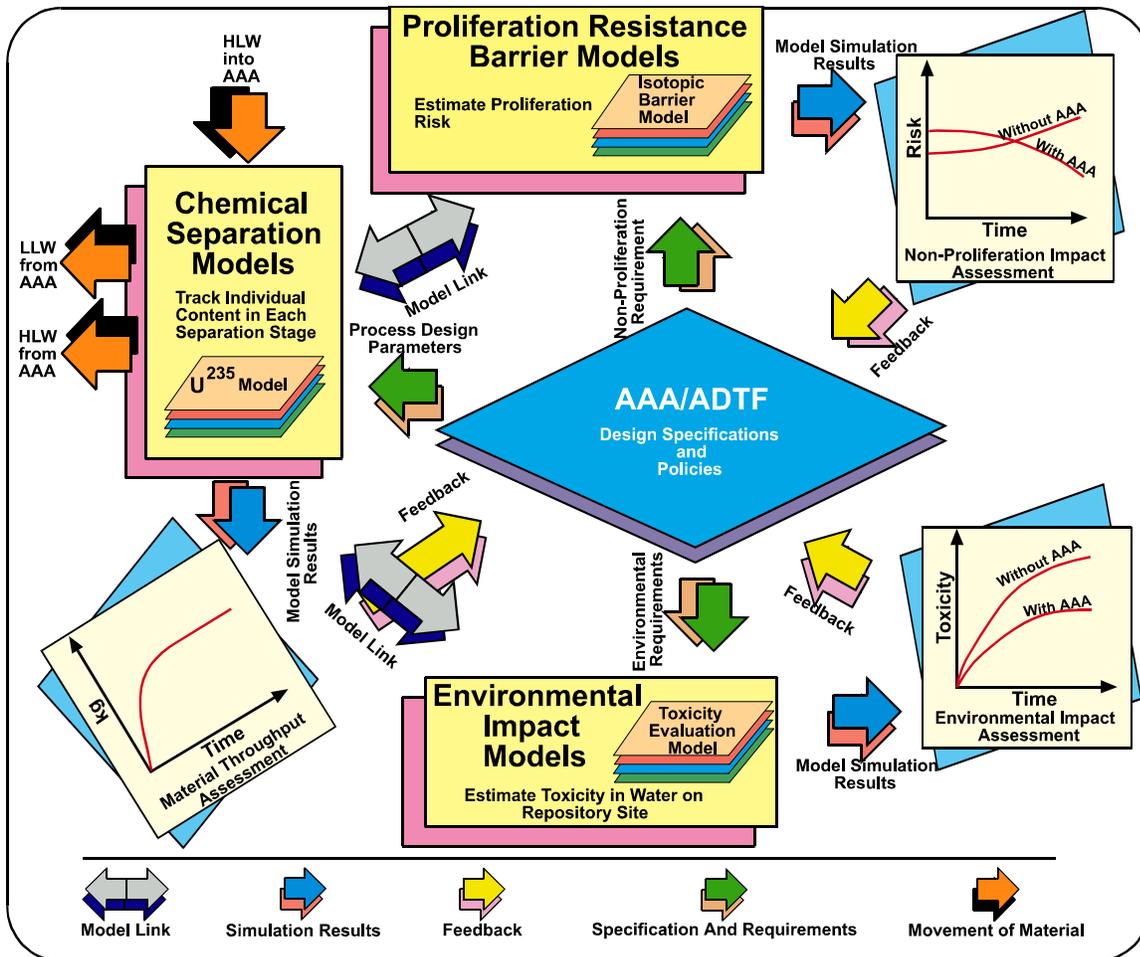


Figure 2. SAMMS Models and Purposes

In order to evaluate the AAA system and compare the results with direct spent fuel disposal, the study begins with the modeling of the global picture of spent nuclear fuel processing and storage. The model allows simulations of two scenarios: spent fuel storage with chemical separation processing and spent fuel storage without such processing.

In order to quantify the chemical processes and the associated non-proliferation assessment, individual material components are tracked through each process in the chemical separation models. Two levels of chemical separation models are needed and developed in order to facilitate different levels of decision making: level 1 models treat the chemical process stages by groups (UREX, PYRO-A, and PYRO-B) to provide global pictures and avoid the details; and level 2 models depict the details of each process stage. However, model simulations can be executed on either level to produce the throughput results for individual material components at each processing stage. The model is developed to be easily modified to include process design limits (e.g., a value to limit only a certain percent of fission products in a particular process stream). Thus, the overall chemical separation processes can be optimized to ensure that those limits are effectively maintained.

Models to estimate the proliferation risks based on proliferation barrier assessment are developed. The proliferation resistance barrier models are associated with individual material components and the processes they go through. Thus, the barrier models are dynamically linked with the chemical separation models. Currently, when the simulation is executed, and while the throughput of individual material components are calculated at each stage of the process, the proliferation risk is also calculated at each time step. Further development of the models will eventually allow feedback from the barrier models to the chemical separation models. If a barrier requirement is not met in the simulation, this information will be sent to the chemical separation models to alter chemical processes so that the total proliferation risks can be minimized.

For the models to provide a complete material inventory and process simulation, they would account fully for all feeds to and products from each of the processes. The chemical separation of the spent nuclear fuel will produce reusable new fuels for power reactors and transmutation facilities. Once the recycled fuel is irradiated, it may again go through follow-on chemical separation processing. Thus, the

complete life-cycle management of spent nuclear fuel to be modeled includes the initial spent fuel separation and fuel fabrication processes, the nuclear transformation inside the reactor and the accelerator transmutation facilities, and the follow-on separation and fuel fabrication processes.

Figure 2 shows how the SAMMS modeling and simulation relate to the emerging details of AAA policies and design specifications. The chemical separation models are dynamically linked with the proliferation resistance barrier models. The model simulation can be carried out for many years. It can simulate the AAA under various scenarios to determine the optimum chemical separation process designs and to demonstrate the impact of the AAA program on spent fuel management in respect to proliferation risk reduction and the reduction in environmental concerns. The model simulation outputs provide feedback to design specifications and policies.

The models have a built-in dynamic input link with Excel spreadsheets. Currently, all data inputs needed to execute level 1 models are linked to this Excel file. The level 1 models have already imported the current input data in the spreadsheets. These input data are the resident data in the model.

The model simulations can be executed with or without links to the Excel file. When the model is executed without the links, the resident data will be used in the simulation. When the models are executed with dynamic input links the input data can be changed or updated in the Excel spreadsheets so that a new scenario can be simulated.

IV. MODEL DESCRIPTION

IV.1 Spent Nuclear Fuel Processing and Storage Model

The global model of spent nuclear fuel processing and storage is shown in Figure 3. The model allows the simulation of two options in which spent fuel accumulated on nuclear power plant sites (designated "Acc Spent Fuel on NPP Site") is either sent into long-term storage, without processing, as high level waste (HLW) or is passed through AAA processing first. Within AAA, the material in the exit stream is separated into low level waste (LLW), cladding, and HLW. Processing chemicals are added

and in part recovered, designated as “CM” in the figure.

The overall AAA process map (named AAA Spent Fuel Process, and abbreviated SFPr in the designation of all its components) is shown in Figure 4. This map is similar to Figure 1 and the baseline process flow-sheet in ANL-99/15². ANL-99/15 was used in developing most of the chemical separation models in this report.

As shown in Figures 1 and 4, spent fuel is sent to a chop/leach process where iodine is extracted and the clarified dissolver solution is sent to the UREX process. Here, uranium, Tc, cladding hulls, and other heavy metals are separated into different product streams. The stream containing Pu and other fission products is sent to the PYRO-A process, where plutonium and the minor actinides are separated from the remaining fission products before being sent to fuel fabrication. In the single-tier system considered here, the new fuel is sent to a transmuter reactor and the resulting spent fuel from this reactor is sent to the PYRO-B process. In the PYRO-B process, iodine and Tc, cladding hulls, fission products, and other heavy metals are separated and sent to different streams. In this map it is assumed that the iodine and technetium are sent to target assembly fabrication for subsequent transmutation to stable xenon and ruthenium.

The UREX, PYRO-A, and PYRO-B processes have been modeled. The transmuter reactor and/or multi-tier power reactors, and the fuel/target fabrication processes are not yet included in the models.

IV.2 Material Breakdown

The focus of the models of the chemical separation processes is the material flowing through them. The material is broken down into components (individual isotopes, groups of isotopes, chemicals used in the processes) by mass. The volumes in the processes are also tracked. The fineness of the component breakdown is determined by:

1. Ability to capture the specific chemical processes;

2. Data requirements for estimating proliferation resistance barrier values;
3. Data requirements for evaluating the environmental impact.

At any point in the process, the components are recorded in a Material Breakdown (MB) structure array. An example material array in the model uses the following material breakdown structure:

1. Volume
2. Iodine
3. ²³³U
4. ²³⁵U
5. Other uranium
6. Technetium
7. Cladding
8. Other fission products
9. Neptunium
10. ²³⁹Pu
11. Other heavy metal
12. Chemicals

In the sample data for the present study, the neptunium components are set to 0, with neptunium masses included in “other heavy metal.” The “chemicals” component is intended to include elements such as the oxygen in UO₂ feed, as well as reagents added in chemical processes.

IV.3 Level 2 Chemical Separation Models

Level 1 models are broken down into the next level to reveal the details of the individual process stages. The modeling of the individual process stages produced the level 2 models. In level 2 models, the materials flowing to and from a particular process stage can be more precisely defined by the characteristics of the process stage. It is in this level that process refinement and optimizations can be accomplished through modeling and simulation.

Level 2 models were developed for UREX, PYRO A, and PYRO B. All material feed streams and exit streams in level 1 models are accounted for in level 2 models. Dynamic connections between level 1 and level 2 models are developed to allow dynamic data exchange between the two levels.

² A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology, DOE/RW-0519, 1999.

IV.4. Level 2 Model Input

The inputs to the level 2 model are the spent fuel feeds to UREX and PYRO-B, the chemical additions to each stage in the UREX, PYRO-A and PYRO-B processes, and the initial amount of materials in each of these level 2 process stages. These inputs are provided to the model according to the material breakdown structure.

In the level 2 model, a particular material breakdown component in an exit stream of a specific process is calculated by a factor times the total amount of the material within the process, calculated as in the level 1 model. The factors for each material breakdown exit stream for each process are inputs to the model.

IV.5. Proliferation Resistance Barrier Model

The proliferation resistance (PR) portion of the model derives a description of the PR attributes of a process stage from the MB array at any point in time. It attempts to automate the criteria defined in the October, 2000, document "Annex: Attributes of Proliferation Resistance for Civilian Nuclear Power Systems," referred to in this paper as simply "Annex." Figure 5 shows the structure for assigning PR attributes to a process stage, here the long-term HLW repository. The material contents are recorded in the box labeled "Repository HLW," arrayed by MB.

Each process in the level 1 chemical separation models carries a replica of the structure displayed in Figure 5. The target of this synthesis is the circle on the right labeled "PRR Barriers." It is an array, indexed by barrier category, of numbers from 0 to 4. The numbers represent barrier height characterizations from "Insignificant" to "Very High." The document "Annex" recommends using qualitative characterizations rather than numbers, but the model's programming environment does not permit non-numerical variables. Besides, as will be

seen below, many of the barrier variables summarize numerical computations.

The fan formation around "PRR Barriers" consists of "ghosts" in the terminology of this programming environment, meaning that the actual computation of an individual barrier is calculated elsewhere in the model. The individual barriers that were modeled are:

- Isotopic Barrier Model;
- Chemical Barrier Model;
- Radiological Barrier Model;
- Mass and Bulk Barrier Model;
- Time Barrier Model.

They are individually developed and integrated to form the Proliferation Resistance Barrier Model.

As shown in Figure 5, an overall proliferation risk value for the facility is computed by "PRR Prolif Risk" as a qualitative conversion, from the maximum of the various barrier heights, to a value between 0 and 1.

V. CONCLUSION

Application of the Systemic Analysis, Mapping, Modeling, and Simulation (SAMMS) methodology to the AAA program is being evaluated. The chemical separations processes for spent fuel partitioning and transmutation have been modeled and simulated. A proliferation resistance barrier model has been developed as well. This study gives reason to believe that the SAMMS method can provide a powerful dynamic simulation means with the potential to be further implemented to model other aspects of the AAA program.

