

Flowsheet and Facility Design to Support Safeguards and Security by Design (SSBD) for Future Nuclear Fuel Cycle Facilities

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Abstract

Chemical process models have been developed for the prediction of material distribution and additional process characteristics in an electrochemical reprocessing facility for the treatment of used oxide fuel and its key unit operations. These modeling capabilities are part of a toolkit assembled by the Materials Protection, Accounting, and Control Technologies (MPACT) campaign, within the U.S. Department of Energy Office of Nuclear Energy, for the demonstration of an advanced Safeguards and Security by Design (SSBD) approach applied to an advanced nuclear fuel cycle facility. These high-fidelity models developed for the MPACT 2020 Milestone demonstration have incorporated the fundamental chemical theory and available data into a predictive capability that relates operational parameters and process observables to material compositions and quantities. A baseline flowsheet has been defined for a pilot-scale facility using existing reference technologies at lab and engineering scales. The implications of flowsheet options on the applied safeguards and security system are discussed.

Introduction

The Materials Protection, Accounting, and Control Technologies (MPACT) campaign, of the U.S. Department of Energy Office of Nuclear Energy, brings together modeling capabilities with experimental testbeds to address next-generation safeguards challenges.¹ Modeling capabilities have been developed representing electrochemical processing as a demonstration of an advanced Safeguards and Security by Design (SSBD) approach, although the same approach can be applied to other facilities and unit operations.² This example demonstrates how the approach can practically be implemented for a new facility. The ability to predict the in-process inventory and related observables in a fuel cycle facility is an important component of an integrated approach to safeguards and supports the design of modern, efficient safeguards and security systems for new fuel cycle facilities.

Flowsheet modeling is a means to establish the initial facility design, which can iteratively be refined as safeguards and security concerns are taken into account.³⁻⁶ Chemical process models of individual unit operations predict key metrics including material flows, separation factors, and inventories based on process operation parameters. Flowsheet and facility models incorporate unit operation models to predict overall material flows and inventories, identify areas of material holdup, and elucidate subtle interdependencies that are not immediately apparent from the unit operation perspective. Analyses using unit operation and flowsheet models can inform higher-level systems models for safeguards, security, and other targeted analyses.

Chemical process model development has proceeded on two distinct levels. A facility or flowsheet model has been developed to evaluate overall mass balance, throughput, inventories, interdependencies and timing, and to aid in defining a reference flowsheet. Unit operations models offer technical basis for empirical relationships used in higher level models and identify characteristics that require further investigation to resolve, particularly if those characteristics could interfere with or pose an issue for safety, safeguards, or security. Greater emphasis has been placed on developing models for unit operations that affect key separations and accountable material inventories. Chemical process models at the unit and facility level can validate that design assumptions are reasonable and have a firm technical basis. Dynamic models are important for predicting system behavior beyond the steady state or average characteristics of the plant because a complete facility design must meet the range of normal conditions to be supported. This paper describes the general capabilities and activities to support the MPACT program's Milestone 2020 efforts. Specific details and data generated by these models under the Milestone 2020 activity contain Sensitive Nuclear Technology and are therefore excluded from this paper.

Reference Flowsheet Description

Electrochemical processing facilities use electrochemical separation processes conducted in molten salt media to remove fission products from used nuclear fuel and recover valuable actinide materials for recycle or storage.⁷ Key drivers for performing this separation include the high activity and/or long-term radiotoxicity of fission products and their negative effect on the performance of recycled fuel. An electrolyzer (ER) is the central piece of process equipment. Additional unit operations support it to provide an appropriate feed and manage products, wastes, and process fluids.

A reference flowsheet was developed to represent a generic electrochemical processing facility for the treatment of used oxide fuel, shown in Fig. 1, and used to support the demonstration of an integrated safeguards and security approach to this example of an advanced fuel cycle facility. While pyroprocessing technology is suited to treatment of various fuel types, including both oxide fuels from commercial light water reactors and metal fuels from fast reactors, a flowsheet for processing oxide fuels utilizes all of the major process equipment necessary for these typical fuel types. Furthermore, the selection of an oxide fuel treatment facility as the reference leveraged additional test bed opportunities and integration of new measurement technologies targeting the oxide reduction (OR) process. While the flowsheet may appear complex, process equipment can be grouped according to four basic functions:

- Head-end operations to prepare fuel assemblies for electrorefining,
- Electrorefining (electrochemical separation),
- Packaging of products and wastes, and
- Process salt management.

A pilot-scale facility with an annual throughput of 100 metric tons heavy metal per year represents a significant increase beyond existing facilities and is near enough to production scale for a meaningful comparison of traditional and advanced safeguards approaches. Detailed process equipment requirements and assumptions for this scale were identified and a baseline flowsheet defined.⁸ A commercial pressurized water reactor fuel assembly, a Westinghouse 17 x 17 array,⁹ serves as a reference fuel input, with composition for a burnup of 50,000 MWd/MTIHM (~5 at% burnup) and 10 year cooling time calculated with the ORIGEN code. The flowsheet provided here is a baseline design, and several processing options exist that would affect facility design. These variations and their potential impacts of the implementation of a safeguards and security system are discussed in a separate section below. A general discussion of unit operations in the baseline flowsheet is presented here.

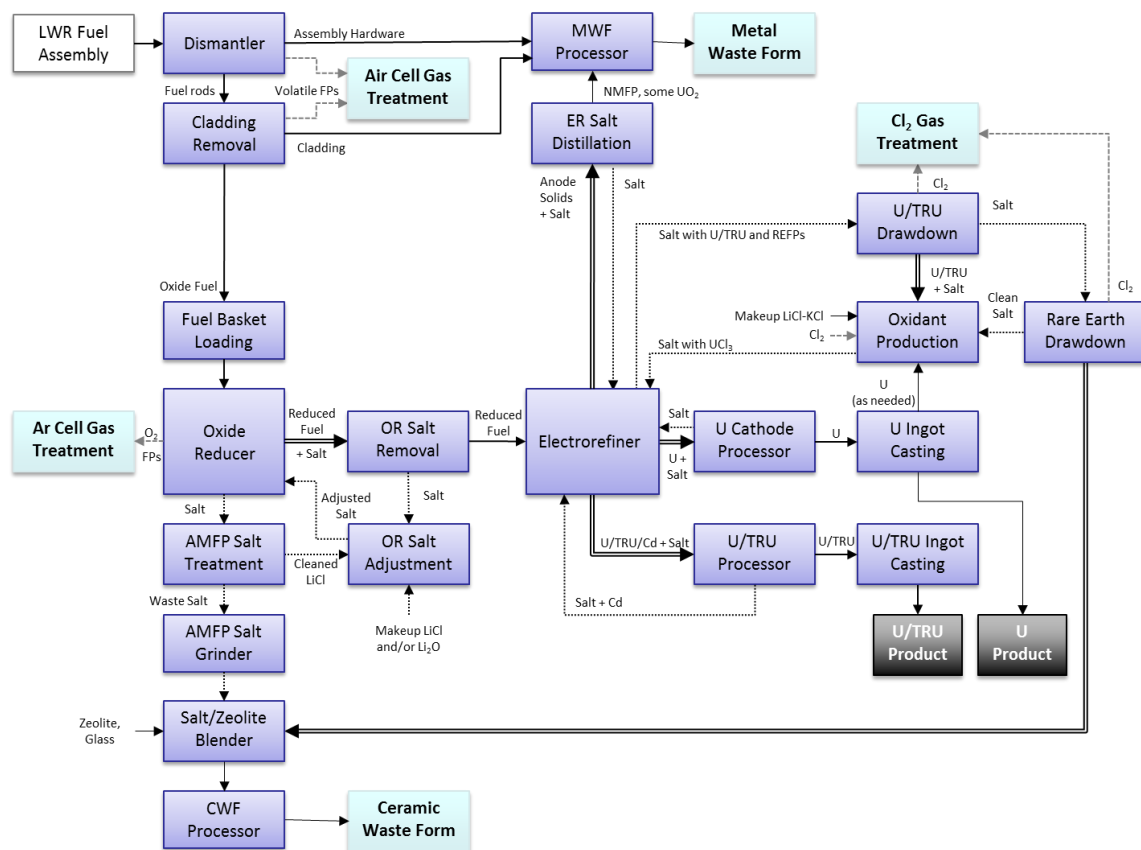


Figure 1. Reference flowsheet

Head-end processing equipment must be capable of receiving a used fuel assembly and segregating the fuel from assembly hardware and cladding. Typically, this is achieved by dismantling the fuel

assembly to separate it into fuel pins and fuel assembly hardware. Fuel pins are then processed to remove fuel material from the cladding by mechanical methods. Thermal treatment may also be applied, to improve fuel/cladding separation, homogenize the fuel, and simplify fission gas handling. The fuel is loaded into baskets for processing with OR and electrorefining equipment. Assembly hardware, cladding and fission gases are collected for waste treatment. These operations may be conducted in an air atmosphere cell and are similar to head-end processing steps in existing aqueous reprocessing facilities.

The primary function of OR equipment is to reduce the used oxide fuel to metal in preparation for electrorefining. OR is conducted in a molten lithium chloride-lithium oxide electrolyte at 650°C. In this step, actinides in the fuel are reduced to base metals and are retained in the basket. Rare earth fission products are also partially reduced from oxides in the fuel to metals. Noble metal fission products are assumed to exist as metals in the fuel and are retained with the actinides and rare earths. Alkali and alkaline earth fission products, generally referred to as active metal fission products (AMFPs), are more stable as chlorides and partition to the molten salt. Oxygen produced by the operation and any remaining fission gases are collected for waste treatment.

Some salt management and waste handling process equipment is associated with the OR. In the reference flowsheet, this includes a salt management unit operation to recover salt adhering to the baskets between the OR and ER steps. A unit to manage the accumulation of alkali and alkaline earth fission products in the OR salt is also needed. For this flowsheet, vaporization is the process used to recover salt from the baskets. Several options exist for the removal of AMFPs from the OR salt. Recently developed melt crystallization techniques have been selected as a reference over historical ion exchange methods. The method has been shown to be effective at separating Cs, Sr, and Ba and has the benefit of not requiring additional chemicals or consumables that add to the facility waste volume.^{10, 11} It is expected to be effective for other components that may accumulate in the salt, such as chalcogens and halogens, although additional study of the behaviors of these species is needed.

Because a much larger amount of available data has been published for the LiCl-KCl eutectic system, an ER utilizing this process salt was selected for the reference flowsheet. The ER is the central piece of process equipment in the pyroprocessing facility. In it, two major separations occur: (1) uranium, the bulk of the fuel, is recovered in high purity on a solid cathode, and (2) an actinide (U/TRU) product containing both uranium and the transuranics is collected using a separate, smaller cathode. Various configurations exist for U/TRU product collection in terms of cathode materials and operating conditions. To achieve high purity product collection, a minimum concentration of uranium chloride must be maintained in the ER salt during operation. Therefore, an oxidant production unit is required to regenerate uranium chloride needed by the ER. A salt distillation unit is also required for recovery of actinide-bearing salts from the noble metal-containing fuel baskets after their removal from the ER.

Product processing operations handle consolidation of the uranium and U/TRU deposits collected from the ER. Equipment for product processing must accomplish separation of ER salt from the collected metal and conversion of the metal to a product ingot. For the uranium product, vaporization of the salt is required and would be followed by a high temperature casting operation. The vaporization and casting could potentially be combined into a single piece of process

equipment. For the reference flowsheet, they are represented as separate units. Similarly, U/TRU product processing achieves separation of residual salt from the product material and conversion to an appropriate form for storage and transport. In the case of a liquid metal cathode such as cadmium, cathode metal must also be separated from the product material. For this flowsheet, units that perform vaporization to remove cadmium and salt and casting of U/TRU product ingots meet the necessary processing requirements.

When a salt recovery step is inserted between the OR and ER, minimal accumulation of alkali and alkaline earth fission products in the ER salt is expected to occur. Therefore, treatment of salt to remove these impurities is not explicitly included for the ER salt as it is for the OR salt. Salt treatment for the ER salt is required, however, for the removal of rare earth fission products and can be achieved by electrolysis of a portion of the ER process salt in a two-step operation. The first step is an actinide drawdown that recovers the U/TRU present in the salt for return to the ER. This is necessary to prevent loss of U/TRU to the rare earth waste stream. After the U/TRU drawdown, rare earth drawdown then collects the rare earth fission products as waste. Because both U/TRU drawdown and rare earth drawdown are performed on the same salt batch, it is possible for these operations to be conducted in a single process vessel despite being represented as separate unit operations in the flowsheet. Treated salt would be passed from drawdown to the oxidant production process prior to return to the ER.

The remaining process equipment comprises those units involved in conditioning and packaging waste from the facility. Three types of waste are produced during pyroprocessing of used fuel. Metal waste types include assembly hardware, cladding, and noble metal fission product waste, with separate zirconium-rich and iron-rich waste forms utilized to achieve the requisite durability. Gaseous waste types include fission product gases (ex. Kr, Xe) and gases generated during OR (O_2) and drawdown (Cl_2). Finally, a ceramic waste type is used for the disposal of alkali, alkaline earth, and rare earth fission products. For the most part, preparation of metal and ceramic waste forms and separation of Kr and Xe can be achieved using established processes already proven effective in commercial aqueous facilities.

Unit Operations Model Development & Applications

In order to predict separation efficiencies and understand their relation to normal and off-normal operating modes, detailed unit operations models have been developed as high-fidelity tools in a broader SSBD toolkit. Detailed models are necessary to evaluate the dynamics of multicomponent systems and validate empirical assumptions used in higher level models. An emphasis was placed on developing models for operations that have an important impact on the distribution of accountable material in the overall process.

Electrochemical Operations

Electrorefining,¹²⁻¹⁴ electrolysis (or electrowinning),¹⁵ and electrolytic OR¹⁶⁻¹⁸ (Fig. 2) are all processes involving similar physicochemical phenomena in a molten salt phase and electrochemical reactions at salt/electrode interfaces. A dynamic ER model (DyER) has been developed to represent this process chemistry. The model was originally developed to track the dynamic evolution of anode, cathode, and salt inventories during an electrorefining process.

Electrolysis, the process used for U/TRU drawdown and rare earth drawdown operations, is treated as a special operating mode of the ER model in which chlorine gas is evolved at the anode in place of the oxidation of impure metal feed. Both processes involve deposition of oxidized species in the salt as metals on the cathode. The model was further extended to predict electrode and salt inventory changes due to exchange reactions that occur when these phases are in contact at temperature in an open circuit configuration.

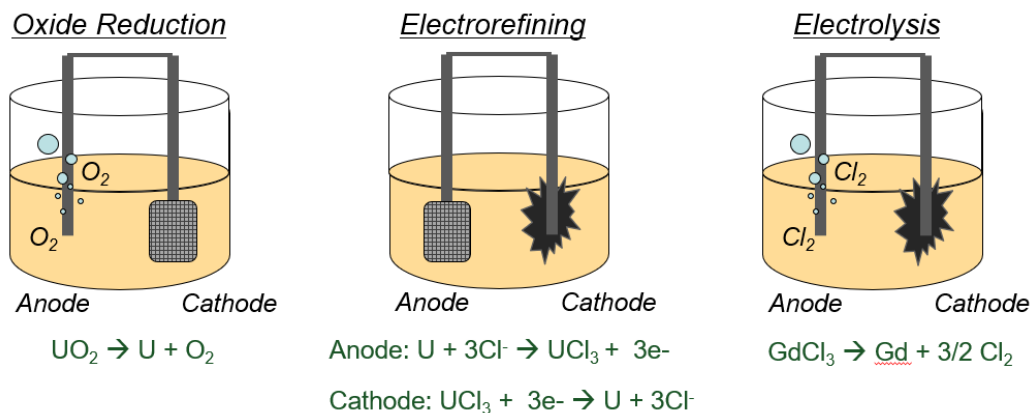


Figure 2. Schematic of electrochemical unit operations

Various modeling efforts have been made that address features of electrochemical operations.¹⁹⁻³⁴ The DyER code has been developed in Mathworks MATLAB® from fundamental theory. Component behavior is modeled using the Butler-Volmer theory of electrode kinetics, thermodynamic equilibria at the electrode surface governed by the Nernst equation, and mass transfer effects for one-dimensional diffusion-limited transport.³⁵ With the constraint that partial currents for all species must total the applied current, these relationships are used to predict dissolution and deposition rates for individual components in the salt during controlled-current (galvanostatic) or controlled-voltage (potentiostatic) operating modes. From these rates, material inventories in the salt and in the cathode deposit can be predicted at any point during the operation of the unit. Depending on operating mode, the induced current or electrode potentials are also predicted.

Fifty-four elements are tracked in oxidized and reduced forms and values for the relevant thermodynamic, kinetic and physicochemical properties of these components have been aggregated from the literature for the LiCl-KCl salt system. Aggregation of data was a key effort in order to capture the unique behavior of individual elements, rather than relying on empirical assumptions about chemical behavior of groups of elements (transuranics, rare earth elements). Temperature and concentration dependencies have been incorporated where available although concentration dependencies have not been evaluated, for the most part. The majority of studies focus on ternary systems, that is, LiCl-KCl eutectic plus a single component of interest, at low concentrations. The effects of multicomponent interactions and concentration effects are largely unexplored. Certain components, notably U and Pu, and some of the rare earths, have been studied extensively for molten chloride salt systems. However, the behavior of other fuel components is not well-characterized, or the published data are inconsistent. Estimates based on similar species have been used to fill gaps in the data. Several thermodynamic, kinetic, and physical parameters are employed in the model, such as apparent standard potential, activity coefficient, diffusion

coefficient, charge transfer coefficient, standard rate constant, solubility limits, and densities. Although data for the LiCl-KCl eutectic system has been implemented thus far, the code is structured such that data for other salt systems could be easily incorporated. As applicable, parameters have been collected with reference to both inert solid electrodes (e.g. W, Mo) and the liquid cadmium electrode.

The code has been validated for limited combinations of components and operating conditions using literature data. Recent validation studies indicate that model results for uranium, a component typically present in significant concentrations, were generally within 10% of experimental values for mass and concentration, but did differ by as much as 15–40% for some experimental data points. Larger discrepancies tend to correlate with small measured values. Considering that model input parameters (initial salt volumes, cell dimensions) were in some cases estimated or inferred from the source documents and that errors in the measurements themselves were not necessarily reported, this is considered good agreement. Digitization of the original data plots likely introduced some error as well, especially for small values. When a model of this type is applied to a larger-scale system with well-known input parameters (such as a facility operator would have), the error between predicted and measured values is expected to be significantly reduced. With sufficient refinement, modeling could then be used for materials accountancy declarations with periodic verification from measurements.

Process Salt Recovery and Removal (Distillation)

Another process common to multiple points in the flowsheet is one in which process salt must be recovered or removed from other material, typically a metal or oxide phase. This separation is most often accomplished by vaporization or distillation of the molten salt, and can also be applied to removal of the cathode material of a liquid metal electrode (e.g. cadmium).³⁶⁻⁴¹ In a related phenomenon, evaporation of process fluids during electrochemical operations has been studied⁴² and various approaches to modeling vaporization behavior have been reported.⁴³⁻⁴⁷ Due to differences in chemical behavior of individual components, this type of process has an important impact on the distribution of accountable material in the overall flowsheet. For this reason, a detailed chemical process model was developed to elucidate the effects of operating parameters on material distribution.

The model applied to molten salt distillation incorporates diffusion-driven mass transport of the vaporized species away from the gas-liquid interface. An approach described by Hur et al. for evaporation of salt during electrolytic reduction⁴² was adapted to model the distillation process and extended to the full set of components tracked in the DyER code. The rate of vaporization depends on the concentration gradient with mass transport defined by Fick's law. The species of interest is assumed to traverse a one-directional concentration gradient, perpendicular to the liquid surface, through a stagnant gas volume. A schematic of this is depicted in Figure 3. A rate relation can be derived for the vaporization of species in the molten salt. The evaporative flux depends upon the dimensions of the gas-liquid interface and gas space, system pressure and temperature, and properties of each component, including partial pressure and diffusion coefficients in the gas phase. Where diffusion coefficient data for individual components were unavailable, values were calculated using Chapman-Enskog theory.^{48, 49} Lennard-Jones parameters for the collision diameter and the collision integral were obtained from tabulated values or calculated with an appropriate correlation.^{49, 50, 51}

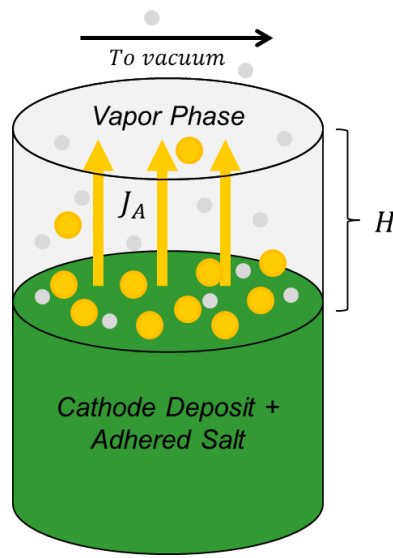


Figure 3. Schematic of Distillation Diffusion Process (J_A = evaporative flux, H = height of stagnant gas volume)

Equations based on fundamental theory were implemented in a distillation unit operation model in MATLAB®. The same species are tracked in this model as in DyER, facilitating interface between the models and integration into the facility-level model. The distillation model allows for flexibility in operation modes, accepting operating temperature and pressure parameters as specific set points with constant ramp rates and hold periods, or as a variable profile versus time. The model predicts independent distillation rates for each component, which are summed to yield an overall salt vaporization rate. Validation of the model with published experimental data confirmed the predictive capability of the model for distillation of molten salt and cadmium.

Head End Operations

At the head end, used fuel must be converted from complete fuel assemblies to a form suitable for the OR process. Compared to unit operation models for electrochemical processes, models of head-end processing equipment are very simple. A dismantler unit must be capable of receiving a used fuel assembly and performing mechanical actions such as cutting, grasping, and lifting to segregate assembly hardware from fuel pins. A basic MATLAB® model for the dismantler unit operation has been created for use with the facility-level model that reads in user-specified fuel assembly data and populates variables with compositional information for the fuel, cladding, endcaps, and other hardware such as nozzles and spacer grids. Outputs of the dismantler model are an inventory of fuel pins with cladding intact, the removed hardware, and a fission gas stream. Removal of a least one set of endcaps is assumed necessary to separate the fuel pins from the assembly hardware. This would release some gaseous fission products and the remainder of those gases would be retained in pores in the fuel at this point in processing.

Fuel rods must be size reduced to appropriate dimensions for further processing. Mechanical operations such as chopping or shredding have been proposed, since it is technically feasible to perform OR and ER operations on chopped fuel segments without first removing the cladding. However, carrying the cladding through these process steps would significantly affect the sizing and operation of the electrochemical process equipment, pose a challenge to process throughput goals, and increase cost. For these process optimization reasons, some type of mechanical and/or chemical decladding process will most likely be necessary. This presents a simultaneous opportunity to insert some type of input accountancy measurement with minimal impact on the process design. Mechanical decladding can be achieved through conventional methods and various equipment concepts exist.⁵²⁻⁵⁷ Chemical decladding typically involves a voloxidation process, where the fuel is heated in air to oxidize it, producing fine fuel fragments that release from the cladding.^{58, 59} A larger fraction of gaseous and volatile fission products is also released, which may have the benefit of easing the gas handling requirements for the argon atmosphere process cell. To facilitate input accountancy, decladding equipment may need to be coupled with diminution equipment that homogenizes the size and composition of the fuel feed and produces representative samples for analysis.^{60, 61} All of these operations have been modeled, but with limited detail.

Salt Treatment

During pyrochemical processing operations, alkali and alkaline earth fission products from the fuel accumulate in the process salt due to their thermodynamic stability as chlorides. Treatment of salt to remove these impurities is necessary to maintain the salt composition within operational requirements. Calculations with the flowsheet model have shown that periodic and relatively infrequent treatment operations can be sufficient to meet operational requirements. The quantity of process salt treated can be a fraction of the total in use, thereby allowing parallel operation of these processes. The treatment process also does not need to be highly efficient in removing impurities. Incomplete removal can be tolerated provided that the average rate of impurity removal via the treatment process is enough to balance the rate at which these fission products are introduced to the system via the fuel. The timing and sizing of the treatment process can be optimized as a function of allowable concentration of the impurities in the process salt and fraction of total salt treated.

Melt crystallization has been shown to be effective in separating Cs, Sr, and Ba in lab-scale experiments, and has the benefit of not requiring additional chemicals or consumables that increase facility waste volumes.^{10, 11} It is expected to be effective for other components that may accumulate in the salt, such as chalcogens and halogens. Using the data reported by Cho, a basic model was developed to calculate salt separations via melt crystallization. The model applies the correlation between the initial and final impurity concentrations in the salt. This model is limited to the type of system for which the correlation was developed, but provides sufficient detail for processes that do not involve actinide-bearing material streams.

Waste Handling

Three primary types of wastes are produced from the facility. Metal waste comprises assembly hardware, cladding, and noble metal fission products that concentrate in the anode heel of the electrolyzer.^{62, 63} Separate zirconium-rich and iron-rich waste forms can be utilized to achieve

the requisite durability in ultimate disposal.⁶⁴ Gaseous waste types include fission product gases (ex. Kr, Xe) and gases generated during OR (O₂) and drawdown (Cl₂). A ceramic waste type is used for the disposal of alkali, alkaline earth, and rare earth fission products, with zeolite and glass added to prepare the final consolidated waste form.^{65, 66}

All of these operations are modeled as material consolidation and repackaging steps and the mass of non-radioactive material is tracked with each unit cycle. For the ceramic waste processing model, this includes the mass of zeolite and glass added and for metal waste processing, the mass of alloying material. As the amount of accountable material in these waste handling units is negligible, much less detail has been incorporated into these models.

Flowsheet Model Development

Concurrent with the development of unit operation models, a facility-level model was also developed. This code, the Argonne Model for Pyrochemical Recycling (AMPYRE) calculates a time-dependent mass balance across the set of unit operations handling material in an electrochemical reprocessing facility. It predicts the dynamic evolution of material compositions and inventories as used nuclear fuel is processed. Products, wastes, and in-process material are all accounted for. The code follows the major fuel and salt components and has been configured such that all major material movements are captured, including recycle of salt recovered from materials extracted from process vessels. As a comprehensive tool, the code can be used to design flowsheets or to study operational effects on process observables. The AMPYRE output is being interfaced with a 3D plant simulation using the Blender suite (<https://www.blender.org/>) with the goal of imparting both dimensionality and mobility to the various process vessels and material streams to better assess the interoperability of unit operations, and to further aid placement and selection of sensors.

The pyrochemical recycling process primarily utilizes electrochemical and mechanical operations for separation of products and waste through various unit operations. The AMPYRE code has been developed on the MATLAB® platform to track the movement of material throughout the facility through prediction of the evolution of the salt composition, and its effect on the compositions of products and waste forms. This result is accomplished by integrating chemistry-based mass-balance models of individual plant functions, including those discussed above. The resulting facility model converts user-defined process and operating parameters into an elemental mass balance across principal plant operations, allowing changes in compositions to be tracked as a function of the batches of fuel processed. The configuration of unit operations in the modeled flowsheet is implemented in a modular way. The code is therefore flexible and has been easily adapted to the addition of new unit operations and rearrangements of the flowsheet. Because of this structure, it could also be applied to other fuel cycle facilities (aqueous reprocessing, fuel fabrication, etc.) or other processes that consist of interconnected unit operations.

The AMPYRE code currently tracks fifty-four elements in oxidized and reduced forms. Species tracked include fuel components (uranium, TRU, rare earth, active metal, and noble metal fission products), sodium, which may be present as a metal fuel bond layer, along with lithium and potassium, the primary cationic components of the molten salt.

A key feature, that has recently been incorporated in the code to facilitate the assessment of real-world systems, is independent unit operation timing. An example of unit operation timing resulting from an AMPYRE simulation is shown in Figure 4. Variables to account for unique cycle times for each piece of process equipment have been added. Variables to account for pre- and post-cycle equipment operations such as heating and cooling times have also been added, to extend beyond the baseline timing of the equipment being “in-use”, e.g. held at temperature or with applied current. The AMPYRE code dynamically manages plant inventories and process equipment availability to trigger unit operation as an integrated system. These revisions to the code have allowed for iterative improvements to flowsheet design. Areas of material holdup and potential process backlogs have been identified and corrected using this capability.

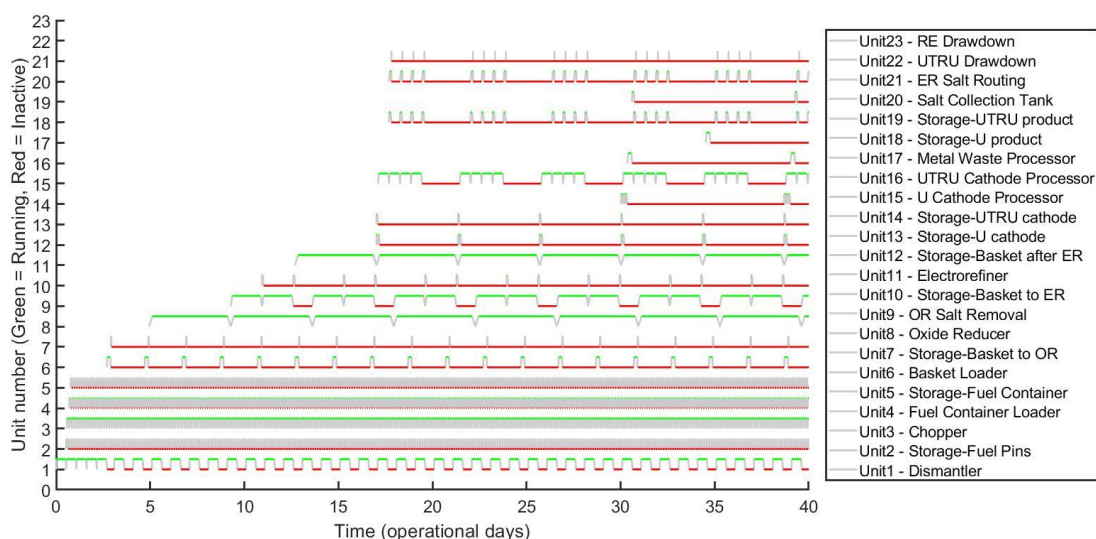


Figure 4. Unit operation timing generated using AMPYRE

3D Visualization

Visualization of plant data in multidimensional renderings aids in identifying certain features that may necessitate changes to layout and design but are not readily apparent when assessing the data in other formats. Potential software options were evaluated for multidimensional visualization of pyroprocessing plant data generated by AMPYRE. The free and open source software suite Blender was selected based on its availability and features. Blender is a 3D computer graphics design environment that supports a broad range of computer graphics development activities, including 3D modeling and animation. It features an interactive graphical user interface (GUI) and uses the programming language Python. The Blender tool has also been used as part of security modeling⁶⁷ which requires 3D facility layouts, and future work could better integrate the layout.

A multidimensional process equipment visualization (MPEV) tool to visualize and manipulate data output from AMPYRE was then created using Blender. The flow of data between the modeling capabilities is illustrated in Figure 5. Input parameters that define a pyroprocessing facility and its individual unit operations are set by the user. These inputs include material feeds,

operating conditions, and facility layout parameters. The input data are fed into AMPYRE, which makes calls to various unit operation models to simulate material flow through the plant and calculate a time-dependent mass balance and other observables. Results generated by AMPYRE are passed to the MPEV tool to render the data in 3D views or a 4D animation.

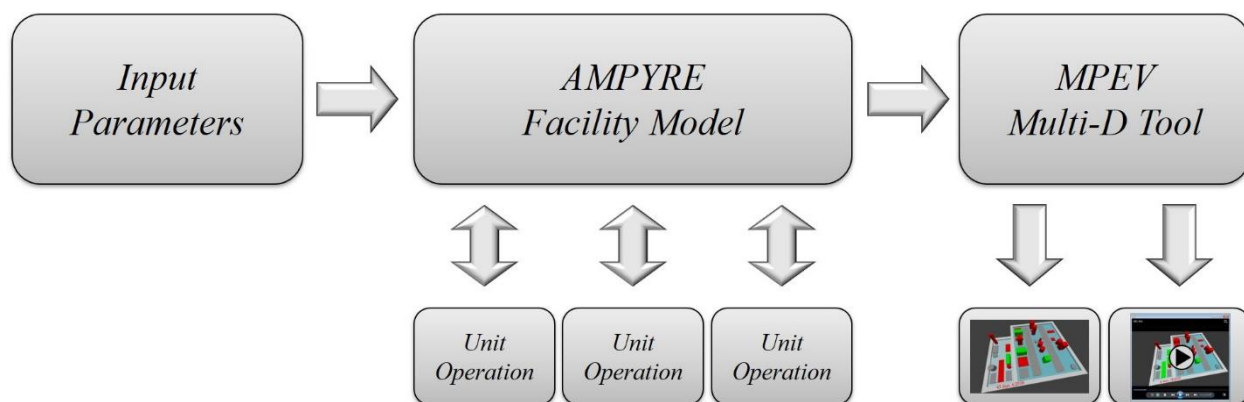


Figure 5. Information flow between modeling capabilities

For an initial rendering, the facility floor and process equipment are represented using simple abstractions. Code structure is modular such that these can be replaced with more detailed renderings. As a proof-of-concept, data for the cycling of individual unit operations was passed from AMPYRE to the MPEV and successfully rendered as both 3D static views and with animation of the unit operation timing.

Flowsheet Variations

Work on flowsheet modeling has identified many variations for the electrochemical flowsheet that are possible. The particular choice of technologies and processing parameters in an actual facility will depend on the complex balance of several factors – a country’s or operator’s needs, regulatory requirements, operational constraints, engineering issues, and safeguards/security impacts. Some notable processing options and their impact on the design of a safeguards and security system are reviewed here. Implications of some of these options have been evaluated using the developed electrochemical reprocessing facility and unit operations models (AMPYRE, DyER, etc.). These examples reinforce the importance of the SSBD approach, which seeks to identify and evaluate safeguards implications early in the design process of the advanced fuel cycle facility.

Head End Options

Decladding and voloxidation are two examples of design options to evaluate at the head end of the facility. The baseline flowsheet includes mechanical decladding in order to route cladding to a metal waste stream early in the flowsheet. This prevents cladding from being sent with the fuel through the OR and ER processes. If cladding is not removed up front, it would be largely unaffected by OR and ER processes and would follow the noble metal fission products through these steps, ultimately being transferred from the used ER anode basket to a metal waste processing

unit. Zirconium-containing cladding (e.g. zircalloy) may impose some operating constraints in order to avoid oxidation of Zr into the ER salt. However, omission of an initial decladding step would have the effect of decreasing the packing density of fuel material in the fuel baskets, thus lowering plant throughput or necessitating increased basket volumes. A basket volume increase would, in turn, require an increase in vessel sizes for both the OR and ER units. The safeguards system would need to accommodate the larger holdup of accountable material in the ER salt and higher background during accountancy measurements.

Mechanical decladding is therefore an alternative to help reduce holdup and background activity in the process cell. One potential tradeoff is that mechanical decladding may leave more residual fuel on the cladding than if it was carried through the ER, thereby not achieving the desired or required separation for a final waste form. An added processing step to recover fuel residues from cladding material may be required.

Voloxidation, a high-temperature thermal decladding process that oxidizes UO_2 in the fuel to U_3O_8 , typically follows shearing or chopping of the fuel pins or of the fuel assembly itself. This may simplify the flowsheet if disassembly can be omitted. Voloxidation of the fuel may be required if removal of more of the residual fuel material is desired. Advantages of using voloxidation include reduced particle size, improved fuel separation from the cladding, and volatile fission product removal. Decladding by some means might be necessary in order to homogenize fuel for input accountancy.

Primary Process Systems

In the main process cell, the choice of salt for the OR and ER vessels is an early design consideration. The baseline flowsheet assumes different process salts, LiCl -1 wt% Li_2O in the OR and LiCl - KCl eutectic in the ER, with a distillation process after OR to recover process salt from the reduced fuel baskets. This helps to keep most of the active metal fission products in the OR salt, while rare earth fission products remain in the ER salt. These are by far the most common salt systems studied but not the only design option. A common salt, i.e. electrolysis in LiCl , could be used to justify elimination of the OR distillation step. A higher operating temperature (650°C vs 500°C) is required which affects separation behavior and materials choices. A small amount of Li_2O carryover with each basket would slightly reduce efficiency through undesirable reactions with uranium or other metal species. In this instance, a detailed comparison between models of both systems could provide objective data to inform the choice of system. This design choice may also depend on lessons learned from larger-scale operation. Some carryover of active metal fission products into the ER salt is possible even with distillation. If both salts ultimately need both active metal and rare earth fission product removal steps, it might make more sense (from an engineering point of view) to keep the salts the same. On the other hand, accumulation of these components may take place over a very long timescale. Suppression of active metal fission products, and the accompanying gamma signature, also makes it easier to use gamma measurements of ER salts and products for safeguards measurements.

The distillation operation between the OR and ER units could potentially be omitted, even if the LiCl - KCl salt system is used. In such a configuration, the buildup of alkali and alkaline earth fission products in the reducer salt could be managed by the drag-out of salt to the ER without the need for process equipment to treat the reducer salt. Regular additions of potassium chloride would be required to maintain the eutectic composition. Additions of fresh salt to the reducer would also

be required to compensate for salt carryover to the ER. However, any flowsheet that omits salt removal between the reducer and ER would still require process equipment to remove alkali and alkaline earth elements, but this equipment would more likely process the ER salt rather than OR salt. Creative alternatives to vaporization, such as salt removal by centrifugal force,⁶⁸ have been investigated. This could be a mechanism to suppress salt carryover between vessels but requires further testing before it could be practically implemented.

Salt Handling and Waste Options

Drawdown is another area for potential variation. The baseline flowsheet periodically draws down a fraction of the ER process salt to maintain the rare earth content within an acceptable range. However, higher rare earth concentrations in the process salt results in higher rare earth contamination in the U/TRU product. Since some rare earths are neutron poisons, this can affect performance of fuels fabricated from the electrorefining product. There may or may not be a need to suppress the rare earth content depending on the end use of the ER products, since some advanced reactor designs are more forgiving in this respect. This is somewhat of an engineering issue to determine an appropriate level within the constraints of an identified product use.

Operations to prepare the final waste forms are also subject to variation due to different methods of isolating of fission products. Some methods, like drawdown, pull the waste material directly from the salt to be recycled. Others, like melt crystallization, concentrate waste materials in the liquid salt phase. There are different options on if or how the two fission product groups can be combined that consider their chemical stability in a particular waste form and contribution to other waste characteristics (radiotoxicity, surface and centerline temperatures, etc.). Waste minimization goals may influence the decision to process and recycle chlorine gas in the facility, or to allow it to be a once-through stream. These engineering options would have to consider the effect of any additional process equipment.

Finally, continuous collection of uranium product requires a minimum level of UCl_3 content in the ER salt as an oxidant for more electroactive species in the fuel and an electrotransport species. The baseline flowsheet reuses a fraction of the U product to create the oxidant UCl_3 that is recycled back into the ER. This could alternatively be prepared from an external source of uranium (e.g. depleted uranium) as well.

Summary

As these examples demonstrate, facility design work can involve a large number of considerations. Adopting a SSBD approach can help to address these early in the process, before substantial resources have been committed, and thereby avoiding costly retrofits and non-optimized solutions.

Conclusion

Flowsheet and unit operation modeling capabilities have been developed to support application of a safeguards and security by design (SSBD) methodology using an electrochemical processing facility as an example. The Argonne Model for PYrochemical REcycling (AMPYRE) can inform

both the design and implementation of process monitoring and material accountability systems for a pyrochemical recycling facility. Modular time-dependent models of the individual unit operations that comprise the facility have been developed and integrated with AMPYRE to provide higher fidelity representations of material flows and inventories in the overall process. A facility layout and relative sizing of process equipment were successfully visualized using the multidimensional process equipment visualization (MPEV) tool. These capabilities are applicable and adaptable to the evaluation of next generation fuel cycle facilities.

A baseline flowsheet was defined for a pilot-scale electrochemical processing facility with consideration of existing reference technologies at lab and engineering scales. Implications of various flowsheet options on the applied safeguards and security system have been reviewed, and a safeguards and security by design approach is recommended to help address these considerations early in the facility design process.

Keywords

Electrochemical; Pyroprocessing; Modeling; Safeguards and Security by Design.

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