Pyre: A Cyclus Pyroprocessing Facility Archetype

Gregory T. Westphal* and Kathryn D. Huff

Dept. of Nuclear, Plasma and Radiological Engineering, University of Illinois at Urbana-Champaign
**gtw2@illinois.edu*

ABSTRACT

Quantifying potential signatures and observables of material diverted from a pyroprocessing facility presents a modeling challenge. This work assesses system parameters that influence separation efficiency and throughput. We leverage these parameters to implement a customizable pyroprocessing facility archetype for use with the Cyclus framework. This generic facility model will allow simulations to quantify signatures and observables associated with various operational modes and material throughputs for a variety of facility designs. Such quantification can timely detection of material diversion. This paper describes the Pyre facility archetype design, pyroprocessing flowsheets captured by the model, and simulation capabilities it enables. To analyze data retrieved from the model, we propose a class for tracking and observing signatures and observables which will be extensible for other facility archetypes in the future.

INTRODUCTION

The diversion of significant quantities of special nuclear material from the nuclear fuel cycle is major non-proliferation concern [1]. These diversions must be detected in a timely manner using signatures and observables in order to properly safegaurd the fuel cycle. Pyroprocessing is a used nuclear fuel separations technology capable of both converting current generation waste into sodium fast reactor fuel, and reprocessing next generation molten salt fuel types. With a new reprocessing technology comes new signatures and observables which in turn necessitate new diversion detection methods. The goal of this research is to identify potential signs of material diversion in a pyroprocessing facility and implement models of these processes into a detailed pyroprocessing facility archetype to the modular, agent-based, fuel cycle simulator, Cyclus [2]. This facility archetype will equip users of the Cyclus fuel cycle simulator to investigate the detection timeliness enabled by quantifying signatures and observables in various fuel cycle scenarios.

BACKGROUND: CYCLUS

Cyclus models the flow of material through user-defined nuclear fuel cycle scenarios. Facilities in nuclear fuel cycles vary, requiring a diverse collection of pre-designed facility process models, known as *archetypes*. Cycamore, the CYClus Additional MOdules REpository, provides common facility archetypes (separations, enrichment, reactor, etc.) [3]. Archetypes are predetermined facility layouts with known parameters corresponding to an output for the Cyclus framework. Simulations run in discrete time steps and allow exact isotopes to be dynamically tracked between facilities [2]. This work seeks to add signature and observable tacking capabilities to Cyclus. Potentially trackable signatures and observables include truck deliveries, power draw, and steam production [4, 5]. Cyclus' discrete time allows users to investigate diversion and flag the time and location diversion occurs.

PYROPROCESSING

Pyroprocessing is an electrochemical separation process used to recycle spent fuel into metallic fuel for use in advanced reactors. With the capability of processing various forms of waste, efficiencies will differ according to design. There are four major systems within pyroprocessing with observable waste: voloxidation, electroreduction, electrorefining, and electrowinning [6].

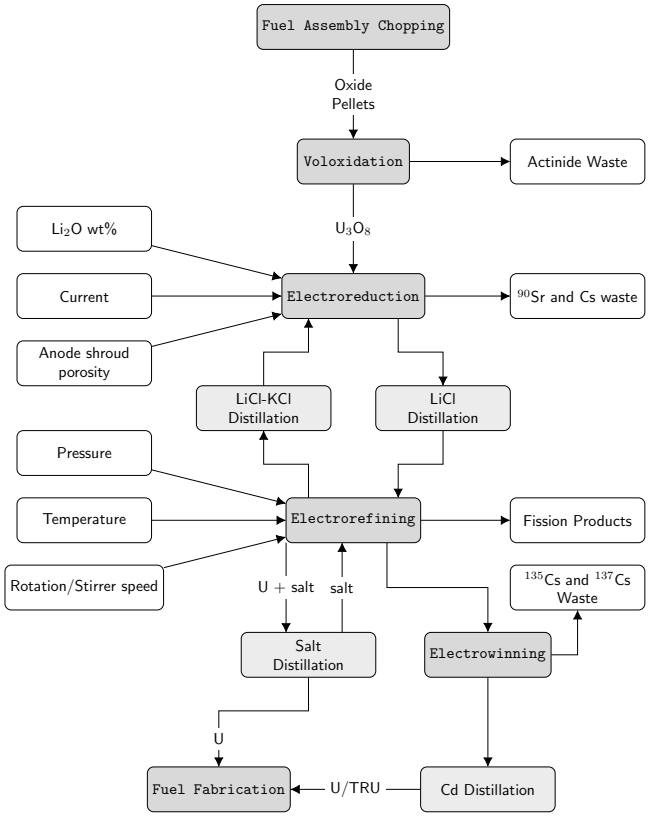

Fig. 1. An archetype design flowchart of pyroprocessing facilities including observable outputs and Cyclus variables.

Figure 1 demonstrates the primary separations steps involved in a general pyroprocessing facility. The main process parameters along the left side of the flowsheet, each applied to processes most significantly impacted. The boxes on the right

side of the processes contain the observable waste produced by each step that CYCLUS can track. Each of main processes are described below regarding information to be used by CYCLUS.

Voloxidation

For LWR fuel, the fuel must be initially treated and separated before proceeding with electrolytic processes. Heated under 500°C, noble gases, carbon, and tritium are collected to decay in storage, and uranium dioxide is converted to U_3O_8. Actinides are also converted to their stable oxide forms and a majority are removed [7, 8]. Voloxidation throughput can be increased by heating the uranium dioxide to temperatures in excess of 800°C. Other methods to improve the U_3O_8 reaction rate are to use alternative oxidants such as cycling between H_2 and air [8].

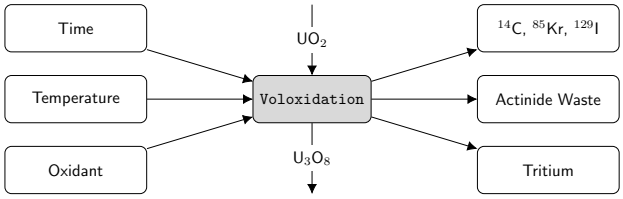

Fig. 2. A material balance over the voloxidation sub-process, including observables.

Electroreduction

Oxide pellets, created in voloxidation, enter the cathode, a negatively charged metal basket. Current density between 100 and 500 mA/cm^2 is applied to the anode in a molten LiCl salt. The electrolytic reduction process primarily results in the diffusion of Cs, Ba and Sr, along with the reduction and conversion of zirconium into metallic form [9, 7]. Electroreduction can further improve its throughput by the addition of Li_2O as a catalyst and to prevent dissolution of the anode [9]. Since Li_2O is used to speed up the reaction, it is important to note that for signatures and observables the operators could add more oxide than reported to IAEA. More frequent shipments of lithium oxide can be tracked as an observable to match records.

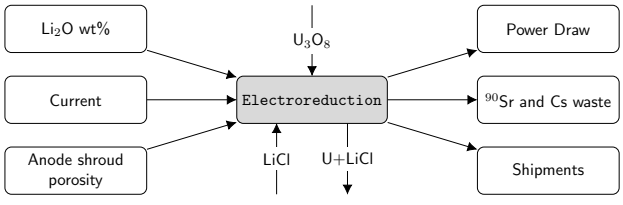

Fig. 3. An enlarged material balance over the electroreduction process with signatures and observables.

Electroreduction produces the majority of Sr and Cs waste with a considerable decay signature proportional to the efficiency and size of the feed batch [6, 7].

Electrorefining

Recoverable waste from reduction is fed into an anode basket suspended in a graphite cathode. A LiCl-KCl eutectic is used as an electrolyte above 500°C [7, 10]. The uranium dissolves at the anode to recombine at the cathode as metallic uranium. The waste transuranics (TRUs) and lathanides are in a soluble chloride form while fission products and cladding remain in the anode basket. Finally, actinides and fission products are removed from the cladding electrochemically [10].

Lee et al. [11] show that the addition of vacuum pressure to the system improves removal efficiency. Temperature, however, exhibits the opposite effect: as temperature decreases so does salt removal. This comes into effect particularly depending on material choice of instrumentation and containment [11]. The most limiting material choice is iron because a eutectic forms between Fe and U at 725°C [12]. In facilities where iron equipment is present temperatures are limited to 700°C, and efficiency is significantly hindered. Cathode arrangement and anode rotation speed also affect the collection of uranium dendrites [11]. The addition of a central stirrer allows mixing of uranium dendrites stuck on the bottom of the vessel, improving separation efficiency and increasing throughput.

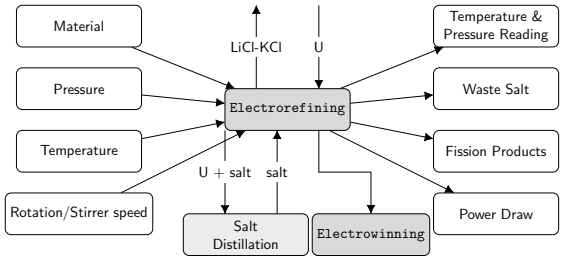

Fig. 4. A material balance over the electrorefining sub-process, including observables.

The electrorefining process also produces the fission product waste stream which requires monitoring. The following products are produced and tracked in this step: Tc, Ag, Pd, Rh, Ru, Mo and Zr [7].

Electrowinning

The molten salt contains TRUs from electrorefining that are separated through electrowinning with trace uranium quantities. With a temperature of 500°C there is approximately 99 wt% reduction in actinides and lanthanides [7]. Throughput is also dependent on material choice for the inert electrodes whose operating voltages vary, impacting separation efficiency [13]. A shroud surrounds the anode to provide a path for O^{2-} ions to the anode and prevent Cl_2 from corroding the anode [14, 9]. Optimum operating current depends on the material choice for the anode shroud since a nonporous shroud limits ion pathways to the points of contact with the anode. Higher porosity corresponds to free ion paths and a higher current. With a higher current the separation time for reduction and winning are reduced [9].

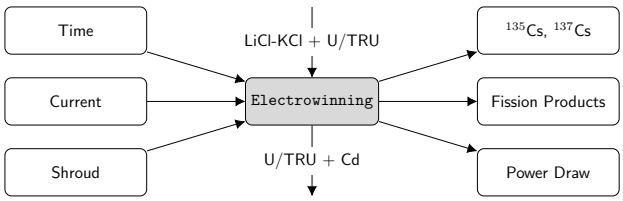

Fig. 5. A material balance over the electrowinning sub-process, including observables.

Figure 5 shows that electrowinning produces Cs waste similar to electroreduction except in reduced quantities. Any fission products remaining in the salt from electrorefining are also removed here. Outside of physically tracked quantities, facility power can also be monitored to observe the use of current to increase throughput.

METHOD: CYCLUS SIMULATION

The separations facility provided by the Cycamore library is used as an initial model of a simple PRIDE facility. The user provides the separations archetype with a feed stream and facility efficiencies. Each waste stream requires a material balance over voloxidation, electroreduction, electrorefining and electrowinning. The main waste streams are metallic waste, ceramic waste from electrowinning and electroreduction, and vitrified waste. Vitrified waste contains the majority of TRUs, Sr, and rare-earth elements. The elemental separation efficiencies are determined through theoretical material balance determined by the NEA and Hermann et al [7, 15]. The simple simulation using a separations facility was run to verify the table of efficiencies input to Cyclus. A scenario was created of one separations facility with a feed of five year cooled spent LWR fuel at a burn-up of 45GWd/MTIHM to match results seen in [7].

A pyroprocessing facility can be modeled with the separations archetype at low fidelity by a dedicated archetype. The goal for Pyre is to include facility configuration parameters and their respective effects on the efficiency table. Efficiencies also vary according to the feed stream resulting in different waste streams for LWR and FR fuels, for example. Multiple material choices exist for anodes and cathodes as well as other design choices that require consideration.

Parameters

It's important to consider facility configuration parameters in the construction of a pyroprocessing archetype. Facility designs vary in multiple aspects which affect the throughput and efficiencies of different waste streams. Variables have been described above through material balance of each sub-process and their system parameters. Table I consists of the inputs derived from the previously mentioned process parameters and material flow.

The signatures and observables contain quantities that are measured directly inside the facility and indirect characteristics observables at distance. A broader category for the facility as a whole is also described for larger parameters such as throughput and batch size. Since throughput is a facility parameter, it is seen in a majority of the sub-processes. Reduction is limited by batch size therefore reducing the throughput of proceeding steps to the electrochemical process. The number of shipments and cars in the parking lot also serve as an indicator to excess work being done. Thermal imaging, further, can determine the operational status of the facility.

TABLE I: Archetype inputs and signatures & observables at each sub-process.

Sub-process	Parameters	S & O	Refs
Voloxidation	Volume	Tritium	[8]
	Oxidant	^{14}C	[7]
	Flow Rate	^{129}I	
	Temperature	^{85}Kr	
	Time	Actinides	
Electroreduction	Volume	^{90}Sr	[6]
	Batch Size	^{135}Cs	[7]
	Li_2O wt%	^{137}Cs	[9]
	Current	Power Draw	[10]
	Porosity	Shipments	[16]
	Distillation Speed		
Electrorefining	Volume	Fission Products	[11]
	Throughput	Power Draw	[10]
	Material	Waste Salt	[7]
	Anode Rotation	Vacuum Pressure	[13]
	Stirrer Speed	Temperature	[14]
	Pressure		
	Temperature		
Electrowinning	Current	Power Draw	[7]
	Shroud Material	Cadmium Waste	[10]
	Time	Fission Products	[6]
	Flow Rate	Lanthanides	
		^{135}Cs	
		^{137}Cs	
Facility	Throughput	Shipments	
	Batch Size	Parking Lot Thermal Image	

DISCUSSION

Using a material balance area over electrorefining and electroreduction yields the majority of detectable waste from the electrochemical processes. Material balances over the remaining processes are used to verify diversion did not occur. Fuel fabrication is of high risk since finished product can be diverted with no additional processing steps, therefore a material balance must also be taken here.

To determine the most significant places of diversion and where to observe them, multiple scenarios must be considered. Each facility parameter must be varied to observe their effects, as well as using a limited number of material balance areas. Scenarios will be run that include various monitoring points with the goal of determining if excess material was produced and what parameter was altered. For example, an increase in Cs production points to electroreduction and electrowinning. If both increase similarly then current is likely affected as these processes share an increase in efficiency with increased current. Further in this scenario, if Cs production increases while Sr does not, electrowinning must be the point at which parameters are altered. A set of these scenarios will be used for sensitivity and importance analysis on the generic pyroprocessing facility.

CONCLUSIONS

This analysis demonstrates the variability in commercialized pyroprocessing facilities and their affects on potential signatures and observables that will be tracked through a detailed archetype. CYCLUS also is outlined as tool in detecting shadow fuel cycles through its agent-based simulation and modular facilities, allowing for variations in plant design. Modeling and data collection of shadow fuel cycles will be performed in the CYCLUS environment after creation of a library specific to the unique needs of electrochemical refinement. Data from these simulations with additional signatures and observables will inform detector placements and measurement points leading to more reliable diversion detection.

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