

## Discrete Event System Based Pyroprocessing Modeling and Simulation: Oxide Reduction

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### 1. Introduction

Material balance for a newly developed process is mainly studied in a flowsheet [1]. However, this is nothing more than an accumulated amount of material transported through in and out streams during a specific period, in other words, equilibrium material balance. Thus, dynamic changes according to the batch operation cannot be predicted in an equilibrium material flow. This study began to build a dynamic material balance model based on the previously developed pyroprocessing flowsheet [2]. As a mid- and long-term research, an integrated pyroprocessing simulator [3] is being developed at the Korea Atomic Energy Research Institute (KAERI) to cope with a review on the technical feasibility, safeguards assessment, conceptual design of facility, and economic feasibility evaluation. The most fundamental thing in such a simulator development is to establish the dynamic material flow framework. This study focused on the operation modeling of pyroprocessing to implement a dynamic material flow. As a case study, oxide reduction was investigated in terms of a dynamic material flow.

### 2. Oxide Reduction

#### 2.1 Pyroprocessing

As shown in Fig. 1, pyroprocessing includes many processes and complex recycling flows. Since pyroprocessing mostly consists of dozens of batch-type processes, a discrete event system is preferred to model this system. It is still developing technology, and is not matured. A lot of effort has been placed into an investigation of its principle. Since the current experimental study focuses on unit process technology, not an integrated process, it is difficult to predict the overall behavior and mutual influence. However, modeling and simulation can make it possible to see unforeseeable results.

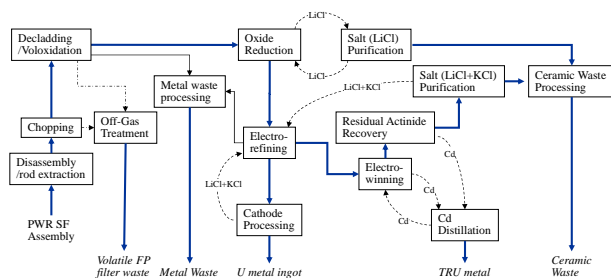


Fig. 1. Simplified pyroprocessing material flow diagram.

Each box in Fig. 1 indicates a grouped process and the number of unit processes is actually more than in Fig. 1. The arrows represent a material flow direction. Pyroprocessing produces not recyclable products from spent nuclear fuel (SNF) but also wastes to be disposed of. The final products of pyroprocessing are a U metal ingot and TRU metal, and final wastes are the filter, metal, and ceramic wastes.

Pyroprocessing features complicated the batch type operation, tangled material flow logic, and numerous SNF elements to be tracked. Thus, the material balance must be calculated whenever events such as feed arrival and product departure occur. Otherwise, a dynamic material flow cannot be tracked. The basic understanding of the whole process is well fulfilled by a flowsheet study, which represents an equilibrium material balance at a specific time.

#### 2.2 Oxide Reduction

The oxide reduction process receives oxide SNF feed material of porous pellet or fragment generated from the headend process. The oxide SNF is converted into metallic form in a LiCl molten salt bath. During the electrolytic reduction process, the oxide powder is reduced into a metal form, which normally contains most of the transition elements, all of the actinides and a certain fraction of rare earth elements. The reduced metal is sent to cathode processing to distill residual salt entrained in metal and oxide mixture, and then transferred to the next process, electro-refining. The remaining LiCl salt in an electrolytic reduction bath after several process operations contains most of the fission products with a high heat load, such as Cs, Sr and Ba, which are separated from the metallic powder. The LiCl salt is sent to a LiCl salt purification process to recycle it by separating LiCl residue concentrated with Cs, Sr, and Ba from pure LiCl. Fig. 2 illustrates three unit process and product streams regarding oxide reduction.

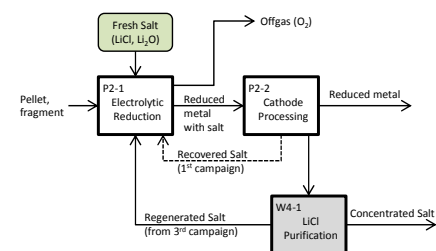


Fig. 2. Material flow diagram for oxide reduction.

### 3. Modeling

#### 3.1 Operation Procedure

The pyroprocessing flowsheet study represents equilibrium mass balance, i.e., accumulated amount of material transported through in and out streams during a specific period (one year is mostly used). It does not provide detailed information regarding the batch operation. Thus, the batch operation procedure was investigated based on the process currently under development. The electrolytic reduction (P2-1) has 50kgHM/batch and 400kg-salt/batch. It receives recovered salts after distillation in cathode processing (P2-2) every other batch operation during the 1<sup>st</sup> campaign (1<sup>st</sup> through 40<sup>th</sup> batch operations). Since P2-1 at the 1<sup>st</sup> batch operation cannot receive recovered salt from P2-2, the 3<sup>rd</sup>, 5<sup>th</sup>, ... , 39<sup>th</sup> batch operations are reasonably practicable to receive the recovered salts. Process P2-1 does not receive any salt from P2-2 during the 2<sup>nd</sup> campaign (41<sup>st</sup> through 80<sup>th</sup> batch operation) but receives fresh salt as much as insufficient amount. It receives regenerated salts for every other batch operation from the LiCl purification process (W4-1) since the 3<sup>rd</sup> campaign, i.e., the 81<sup>st</sup> batch operation. If the regenerated salt is not enough to facilitate the process of P2-1, fresh salt can be added. As the number of P2-1's campaign changes, the direction of material flow changes in process P2-2, the recovered salts is transferred to P2-1 during the 1<sup>st</sup> campaign but to W4-1 since the 2<sup>nd</sup> campaign.

The above operation procedure is changed according to the batch operation number. Consequently, the amount of generated work in process (WIP) and its direction of flow are affected by the operation procedure. Such transient behavior must be well described in a model in terms of the inventory management, nuclear material accountability, and productivity of the integrated process.

#### 3.2 Material Balance

Dynamic material balance describes the amount of feed, hold-up, and product in any process for every batch operation. If accumulating the amounts of received and departed material for a specific period, an exact equilibrium material balance can be obtained. Because the above operation procedure is difficult to implement, an equilibrium material balance tends to simplify the complicated operation to the averaged one, i.e., every batch operation is assumed to be the same. However, if we can build an exact model through appropriate tools or methodologies to reflect such tangled operation requirement, the assumed equilibrium material balance obtained from a flowsheet study [2] can be replaced with an exact material balance in an exact model.

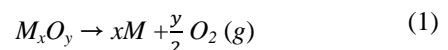
#### 3.3 Equilibrium and Dynamic Material Balance

For comparison between equilibrium and dynamic material balance, it is assumed that the oxide reduction treats 10tHM per year, which corresponds to 200 batch operations of process P2-1. An equilibrium mass balance in process P2-1 is shown in Table I.

Table I: Equilibrium material balance in P2-1.

Material via stream	type	SNF mass(kg)	Salt (LiCl, Li <sub>2</sub> O)
new salt	feed	-	824
pellet/fragment	feed	11,331	-
recovered salt	feed	5	350
regenerated salt	feed	6	1,146
Sum of inputs		11,341	2,320
cathode product	WIP	9,997	1,935
O <sub>2</sub>	product	1,331	-
Sum of outputs		11,328	1,935
remaining salt	hold-up	13	385
Sum of hold-up		13	385

Since the equilibrium material balance shows accumulated results over numerous batch operations, the difference of each batch is ignored. Process P2-1 has a total of 4 inputs and 2 outputs. The sums of the inputs and outputs are not the same because process P2-1 can hold a small amount of SNF in its bath. Therefore, the sum of inputs exactly matches the sum of outputs and hold-up. We cannot predict from equilibrium material balance any transient behavior affected by operation procedure described in section 3.1. Tables II and III show dynamic SNF and salt balances, respectively, obtained from a discrete event system (DES) model of oxide reduction. It shows different results from every batch operation: amount of inputs, outputs, and hold-up in process P2-1 for every batch operation. In Table II, every batch operation of P2-1 receives 50kgHM/batch fragment or pellet from the previous process excluding O<sub>2</sub> weight. The 2<sup>nd</sup> column in Table II represents the minimum weight of oxide form of a fragment or pellet. Excluding oxide, only the SNF element weight becomes 50kgHM/batch. The weight of oxide form can be more than the sum of 50kg and O<sub>2</sub> weight measured at the output stream because reduction yield ratios are not 100% about all SNF oxide elements. The reduction yield ratio is one of the parameters that significantly influence the material balance at the out stream. For example, actinide elements are almost reduced to metal form such that 99.5% of those oxides convert into metal form but lanthanide elements are rarely reduced such that only 30% of those oxides convert into metal form. Generally, the overall reaction for oxide reduction of an arbitrary metal oxide can be described as follows [4]:



In the case of actinide, 99.5% of  $M_xO_y$  converts into metal by electrolytic reduction but 0.5% of  $M_xO_y$  still keeps its original form. Some elements (Se, Rb, Cs, Sr, Ba, Eu, and Te) are dissolved and transferred to a salt bath to become chloride forms. Therefore, cathode

product contains three types of product forms: metal, oxide, and chloride form. The chloride form is entrained in a cathode product along with LiCl salt when the cathode product is transferred to cathode processing P2-2. The 4<sup>th</sup> column represents only the element weight of chloride form in salt. The 5<sup>th</sup> column in Table II represents the sum of the three types. The 3<sup>rd</sup> and 4<sup>th</sup> column indicate only element weights of chloride forms contained in salt.

Table II: SNF material balance in P2-1.

batch #	inputs			hold-up remaining salt(kg)	outputs	
	fragment/ pellet(kg)	recovered salt(kg)	regenerate d salt(kg)		cathode product(kg )	O <sub>2</sub> (kg)
1	56.67	-	-	0.28	49.72	6.67
2	56.67	-	-	0.54	49.73	6.67
3	56.59	0.02	-	0.83	49.74	6.59
4	56.67	-	-	1.08	49.75	6.67
5	56.59	-	-	1.33	49.75	6.59
6	56.67	-	-	1.57	49.76	6.67
7	56.59	0.05	-	1.85	49.77	6.59
8	56.67	-	-	2.08	49.77	6.67
9	56.59	0.08	-	2.38	49.78	6.59
...	...	...	...	...	...	...
41	56.59	-	-	10.25	49.97	6.59
42	56.67	-	-	10.27	49.98	6.67
43	56.59	-	-	10.30	49.97	6.59
44	56.67	-	-	10.32	49.98	6.67
45	56.59	-	-	10.35	49.97	6.59
46	56.67	-	-	10.37	49.98	6.67
47	56.59	-	-	10.40	49.97	6.59
...	...	...	...	...	...	...
81	56.59	-	0.08	11.03	49.99	6.59
82	56.67	-	-	11.04	50.00	6.67
83	56.59	-	0.08	11.13	49.99	6.59
84	56.67	-	-	11.13	50.00	6.67
85	56.59	-	0.08	11.21	49.99	6.59
86	56.67	-	-	11.22	50.00	6.67
87	56.67	-	0.08	11.30	49.99	6.67
...	...	...	...	...	...	...
194	56.67	-	-	13.30	50.05	6.67
195	56.67	-	0.11	13.37	50.04	6.67
196	56.67	-	-	13.32	50.05	6.67
197	56.67	-	0.12	13.39	50.04	6.67
198	56.67	-	-	13.34	50.05	6.67
199	56.67	-	0.12	13.41	50.04	6.67
200	56.67	-	-	13.36	50.05	6.67
total	11,331.06	4.55	5.66	13.36	9,996.85	1,331.06

Table III: Salt material balance in P2-1.

batch #	inputs			hold-up remaining salt(kg)	outputs	
	fresh salt(kg)	recovered salt(kg)	regenerate d salt(kg)		cathode product(kg)	O <sub>2</sub> (kg)
1	404.00	-	-	394.06	9.94	-
2	-	-	-	384.13	9.93	-
3	-	19.67	-	393.88	9.92	-
4	-	-	-	383.96	9.92	-
5	-	-	-	374.06	9.91	-
6	-	-	-	364.15	9.90	-
7	-	19.64	-	373.90	9.89	-
8	-	-	-	364.01	9.89	-
9	-	19.61	-	373.74	9.88	-
...	...	...	...	...	...	...
41	42.26	-	-	394.31	9.69	-
42	-	-	-	384.62	9.68	-
43	19.38	-	-	394.31	9.69	-
44	-	-	-	384.63	9.68	-
45	19.37	-	-	394.31	9.69	-
46	-	-	-	384.63	9.68	-
47	19.37	-	-	394.31	9.69	-
...	...	...	...	...	...	...
81	0.17	-	19.17	394.33	9.67	-
82	-	-	-	384.66	9.67	-
83	0.17	-	19.17	394.33	9.67	-
84	-	-	-	384.67	9.66	-
85	0.17	-	19.16	394.33	9.67	-
86	-	-	-	384.67	9.66	-
87	0.17	-	19.16	394.33	9.67	-

...	...	...	...	...	...	...
194	-	-	-	384.77	9.61	-
195	0.17	-	19.06	394.38	9.62	-
196	-	-	-	384.77	9.61	-
197	0.17	-	19.06	394.38	9.62	-
198	-	-	-	384.77	9.61	-
199	0.17	-	19.06	394.38	9.62	-
200	-	-	-	384.77	9.61	-
total	824.24	349.83	1,146.07	384.77	1,935.37	-

### 3.4 DES based Operation Model

The material flow of oxide reduction as shown in Fig. 2 was modeled as shown in Fig. 3 by using multi-system modeling software, ExtendSim [5]. Process P2-1 has four input connectors to receive pellet/fragment, fresh salt (LiCl and Li<sub>2</sub>O), recovered salt, and regenerated salt, and it also has three output connectors to transfer cathode product and O<sub>2</sub> to the next processes such as P2-2 and W3-1, as shown in Fig. 3.

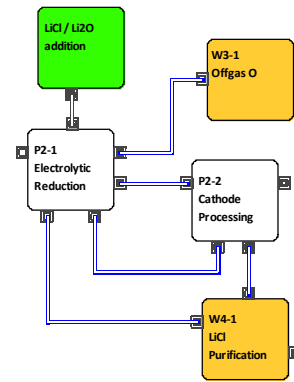


Fig. 3. Operation model of oxide reduction.

Each box in Fig. 3 is a hierarchical block, which contains many blocks and complicated flows to implement functions inherent to the process: feed material receipt, hold-up calculation, product material calculation, and so on. If any box is double-clicked in the model, detailed models as shown in Figs. 4 and 6 pop up.

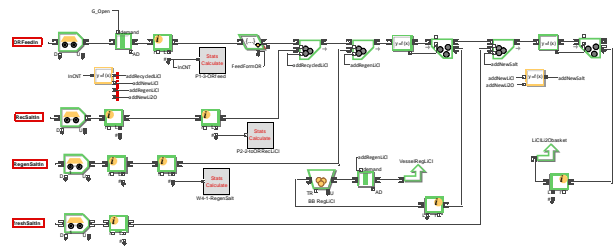


Fig. 4. A selected part in P2-1 for receipt of feed materials.

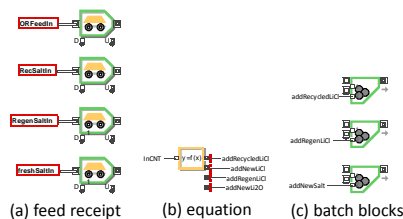


Fig. 5. Selected blocks needed for operation logic in Fig. 4.

Fig. 4 shows the first part of P2-1 representing the receipt of feed materials. An item flow in the DES model is controlled by transport blocks, equation blocks and batch blocks, as shown in Fig. 5. An oxide reduction model begins with transport blocks that describe the receipt of four types of feed materials (fragment/pellet, recovered salt, regenerated salt, and fresh salts). The feed of fragment/pellet is always needed for every batch operation. However, the recovered salt, and regenerated and fresh salts, can be received or not according to the batch operation procedure. The complicated operation logic of section 3.1 was perfectly built in Figs 4 and 5.

The materials generated in process P2-1 are remaining salt in bath as a hold-up, and cathode product and  $O_2$  as products. The total amount and elemental composition for the three types of material are calculated in the later part of Fig. 4, as shown in Fig. 6. Fig. 7 shows some important blocks to calculate them and time delay blocks to describe the operation time. Fig. 7(a) calculates the remaining salt and fission products (FPs) dissolved in the salt. Two equation blocks in Fig. 7(c) calculates the amount and composition of cathode product and  $O_2$ .

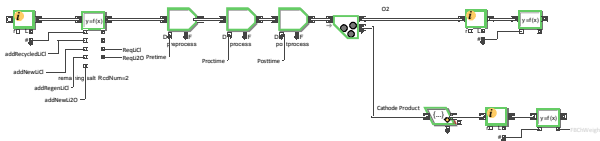


Fig. 6. A selected part in P2-1 for calculation of mass compositions of hold-up and products (cathode product and  $O_2$ ).

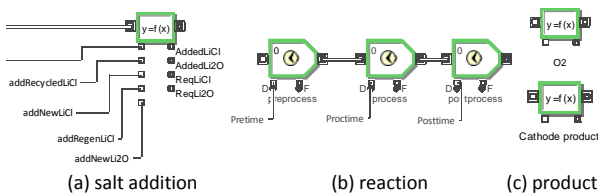


Fig. 7. Blocks for calculation of hold-up, process operation delay, and product compositions.

Every calculation results are written in internal database tables of ExtendSim by equation blocks in Fig. 7(a) and 7(c), so that a further detailed analysis can be performed after simulation by an investigation of the recorded data during simulation.

## 4. Simulation

### 4.1 Default Scenario

The basic input parameters resulted in Table II and III are as follows: The capacities of process P2-1, P2-2 and W4-1 are 20 kgHM/batch, 100 kgHM/batch and 50 kgHM/batch, respectively, and operation times taken by

the processes are 20 day/batch, 44 h/batch, and 165 hours/batch, respectively. One piece of equipment is deployed for each process. Chemically, 20% salt over the weight of the cathode product is carried to the next process P2-2. Actinide oxide and noble metal oxide has a 99.5% reduction yield ratio, but a rare earth oxide has a 30% reduction yield ratio. The process P2-2 recovers 99.9% salt by distillation and 0.1% salt transfers to the next process. This is the default scenario to run the simulation. To reflect the experimental results in the future or to analyze various alternative operations, these parameters can be changed.

### 4.2 Experimental Results

Simulation is performed based on the above default scenario for 200 batch operations of process P2-1. The 200 batch operations end within 250 days. Figs. 6 and 7, and Figs. 8 and 9 show process P2-1's material balance for the SNF element, and salt ( $LiCl+Li_2O$ ), respectively. Figs. 6 and 8 show the material balance to mean the amount of material that is received from the input and sent to output each batch. On the other hand, Figs. 7 and 9 show the material balance for the amount accumulated from the first batch. The amounts at the end batch operation in Figs. 7 and 9 mean the equilibrium mass balance. In Fig. 6, the fragment/pellet includes 50kgHM plus oxide weight. Therefore, after the process, the cathode product losses as much weight as the oxygen generated during the reduction. The bottom figure of Fig. 6 is magnified to properly investigate small amounts such as FPs contained in the hold-up, recovered salt, and regenerated salt. In the bottom figure of Fig. 7, FPs increase in hold-up because recycled salts containing small amounts of FPs are constantly used except for the 2<sup>nd</sup> campaign. Since the 1<sup>st</sup> campaign uses distilled salt directly without purification, the concentration of FPs steeply increases, but since the 3<sup>rd</sup> campaign uses regenerated salt containing very small amounts of FPs, the concentration gradually increases.

Fig. 8 shows the salt material balance such that the sum of fresh salt, recovered salt, and regenerated salt exactly match the sum of entrained salt carried with the cathode product and hold-up increment for every batch operation. Fresh salt addition occurs during only the 2<sup>nd</sup> campaign and the 1<sup>st</sup> batch of 2<sup>nd</sup> campaign needs more fresh salt than the others to supply the accumulation of 0.1% loss amount during the 1<sup>st</sup> campaign because cathode processing recovers only 99.9% salt. An insufficient amount of salt during the 1<sup>st</sup> campaign is supplemented through a recycling of recovered salt but is through the recycling of regenerated salt during the 3<sup>rd</sup> campaign. Fig. 9 shows an accumulation of batch mass balance. The hold-up in Fig. 9 indicates the amount of remaining salt in bath right after transfer of the cathode product, i.e., accumulation of hold-up. Since the 3<sup>rd</sup> campaign, the remaining salt approximately retains 400kg. The amount of salt carried

with the cathode product gradually decreases because the FPs in proportion to the entrained salt increases.

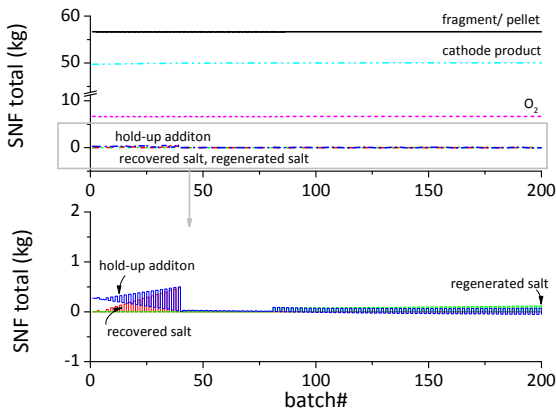


Fig. 6. Material (a total sum of 52 SNF elements) balance every batch in P2-1.

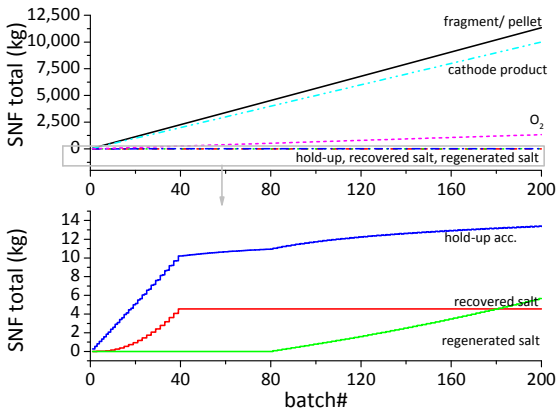


Fig. 7. Accumulated material (a total sum of 52 SNF elements) balance in P2-1.

### 5. Conclusions

DES based modeling was applied to build a pyroprocessing operation model. A dynamic material flow as the basic framework for an integrated pyroprocessing was successfully implemented through ExtendSim's internal database and item blocks. Complex operation logic behavior was verified, for example, an oxide reduction process in terms of dynamic material flow. Compared to the equilibrium material flow, a model-based dynamic material flow provides such detailed information that a careful analysis of every batch is necessary to confirm the dynamic material balance results. With the default scenario of oxide reduction, the batch mass balance was verified in comparison with a one-year equilibrium mass balance.

This study is still under progress with a mid-and long-term goal, the development of a multi-purpose pyroprocessing simulator that is able to cope with safeguards assessment, economic feasibility, technical evaluation, conceptual design, and support of licensing for a future pyroprocessing facility.

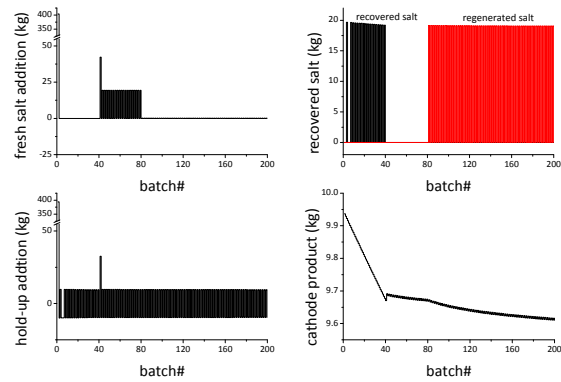


Fig. 8. Material (salt: LiCl and Li<sub>2</sub>O) balance every batch in P2-1.

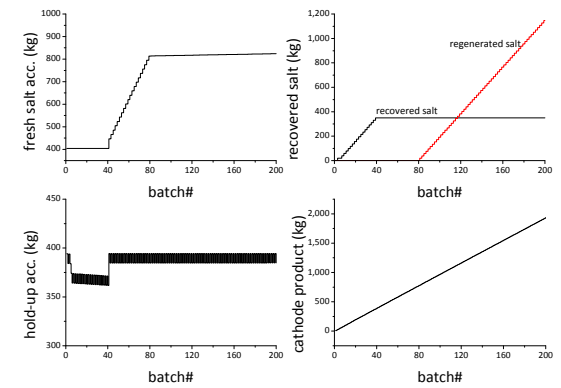


Fig. 9. Accumulated material (salt: LiCl and Li<sub>2</sub>O) balance in P2-1.

### ACKNOWLEDGEMENTS

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