Modeling and Simulation of Pyroprocessing Oxide Reduction

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Abstract: Pyroprocessing operation modeling features complicated batch type operation, tangled material flow logic, handling many numbers of unit processes. Discrete event system (DES) modeling was applied to build an integrated operation model of which simulation showed that dynamic material flow was accomplished. In the model simulation, the amount of material transported through upstream and downstream in a process satisfied the mass balance equation for every batch operation. This study also analysed in detail an oxide reduction process and showed that every stream's material flow could be exactly tracked under DES modeling environment.

1 INTRODUCTION

Material balance for a newly developed process is mainly studied in a flowsheet (Piet et al., 2011). 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 (Lee^a et al., 2013). As a mid- and long-term research, an integrated pyroprocessing simulator (Lee^b et al., 2013) 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. Therefore, 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.

There are some recent interesting works similar to this study, the US devoted to developing a spent nuclear fuel (SNF) reprocessing plant level toolkit named RPTk (Reprocessing Plant Toolkit) (McCaskey et al., 2011). Japan developed an analysis code (Okamura and Sato, 2002) for an estimation of the material balance for the system design of the pyrochemical reprocessing plants consisting of batch processes. As a preliminary study, Korea also developed DES based model to implement simplified dynamic material flow for pyroprocessing (Lee et al., 2011).

2 OXIDE REDUCTION

2.1 Pyroprocessing

As shown in Figure 1, pyroprocessing includes many processes and complex recycling flows. 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. Since pyroprocessing mostly consists of dozens of batch-type processes, a discrete event system is preferred to model this system if main concerns are not the chemical reaction within one batch operation.

Each box in Figure 1 indicates a grouped process and the number of unit processes is actually more than in Figure 1. The arrows represent a material flow direction. Pyroprocessing produces not

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recyclable products from SNF but also wastes to be disposed of. The final products of pyroprocessing are a uranium (U) metal ingot and transuranic (TRU) metal, and final wastes are the filter, metal, and ceramic wastes.

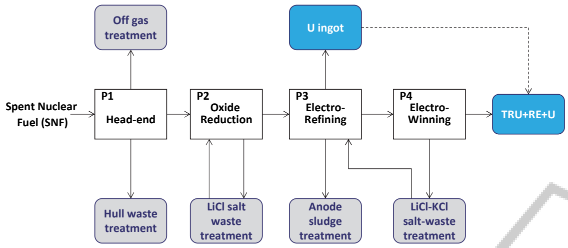


Figure 1: Simplified pyroprocessing material flow diagram.

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 reduced metal deposit 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. Figure 2 illustrates three unit process and product streams regarding oxide reduction.

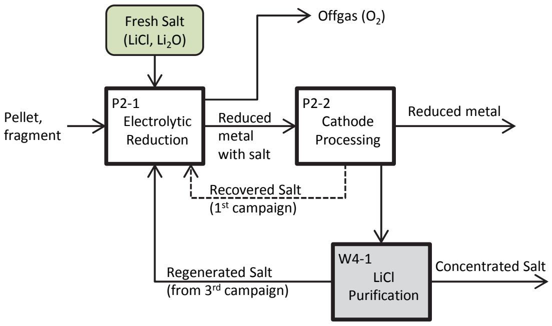


Figure 2: Material flow diagram for oxide reduction.

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 1st campaign (1st through 40th batch operations). Since P2-1 at the 1st batch operation cannot receive recovered salt from P2-2, the 3rd, 5th, … , 39th batch operations are reasonably practicable to receive the recovered salts. Process P2-1 does not receive any salt from P2-2 during the 2nd campaign (41st through 80th 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 3rd campaign, i.e., the 81st 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 1st campaign but to W4-1 since the 2nd 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 accountancy, 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 can be replaced with an exact material balance in an exact model.

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 1.

Table 1: Equilibrium material balance in P2-1.

Material via stream	type	SNF mass(kg)	Salt ($LiCl$, Li_2O)
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	product	9,997	1,935
O_2	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 2 and 3 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 2, every batch operation of P2-1 receives 50kgHM/batch fragment or pellet from the previous process excluding O_2 weight. The 2nd column in Table 2 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 O2 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.

Table 2: SNF material balance in P2-1.

	inputs			hold-up	outputs	
batch #	fragment/ pellet(kg)	recovered salt(kg)	regenerated salt(kg)	remaining salt(kg)	cathode product (kg)	O2(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

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 (Phongikarron et al., 2011):

$$M_xO_y \rightarrow xM + \frac{y}{2}\,O_2\,(g) \qquad (1)$$

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 4th column represents only the element weight of chloride form in salt. The 5th column in Table 2 represents the sum of the three types. The 3rd and 4th column indicate only element weights of chloride forms contained in salt.

Table 3: Salt material balance in P2-1.

batch #	inputs			hold-up	outputs	
	Fresh salt(kg)	recovered salt(kg)	regenerated salt(kg)	remaining salt(kg)	cathode product(kg)	O2(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	-

Every unit process model must satisfy dynamic material balance equation for every batch operation.

$$\sum_{i=1}^{m} I_{i,k} = \sum_{i=1}^{n} O_{i,k} + (H_k - H_{k-1}) \qquad (2)$$

where m is the number of inputs; n the number of outputs; k the current number of batch operation; $I_{i,k}$ the k-th input amount of mass transported through the i-th upstream, $O_{i,k}$ the k-th output amount of mass transported through the i-th downstream; H_k hold-up until k-th batch.

When the equilibrium material balance equation is considered instead of dynamic mass balance equation, the equation (2) can be modified into:

$$\sum_{i=1}^{m} {}^{acc}I_{i,k} = \sum_{i=1}^{n} {}^{acc}O_{i,k} + H_k \qquad (3)$$

where ${}^{acc}I_{i,k}$ is the k-th accumulated input amount of mass transported through the i-th upstream so far; ${}^{acc}O_{i,k}$ the k-th accumulated output amount of mass transported through the i-th downstream so far.

3.3 Operation Model

3.3.1 DES Modeling

The material flow of oxide reduction as shown in Figure 2 was modeled as shown in Figure 3 by using multi-purpose system modeling software, ExtendSim. Process P2-1 has four input connectors to receive pellet/fragment, fresh salt (LiCl and Li_2O), recovered salt, and regenerated salt, and it also has three output connectors to transfer cathode product and O_2 to the next processes such as P2-2 and W3-1, as shown in Figure 3.

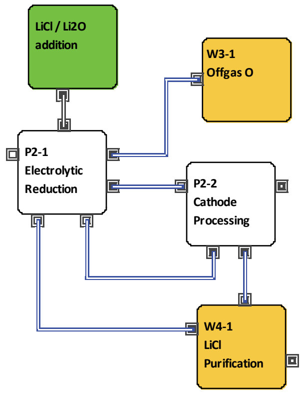


Figure 3: Operation model of oxide reduction.

Each box in Figure 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 Figures 4 and 6 pop up.

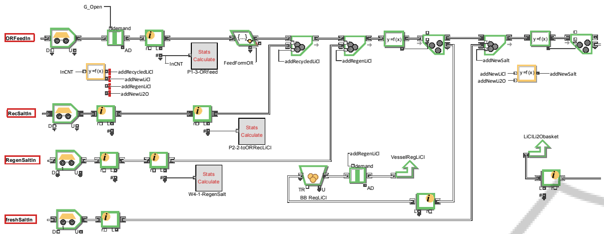
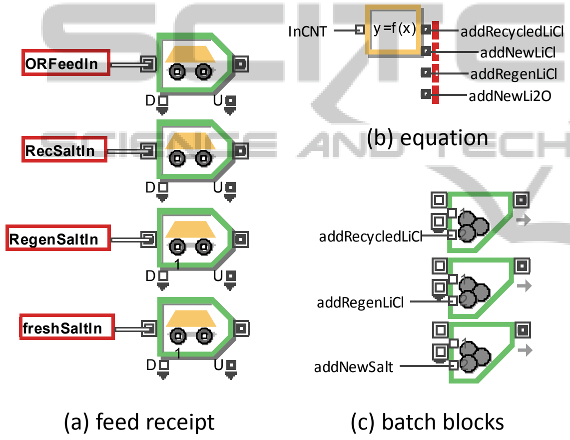


Figure 4: A selected part in P2-1 for receipt of feed materials.



(a) feed receipt

(b) equation

(c) batch blocks

Figure 5: Selected blocks needed for operation logic in Figure 4.

Figure 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 Figure 5. Transport blocks move items from the start of a path to the end based on distance and speed information, so they are used here for simulating feed material's movements that consume time. 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 (ORFeedIn in Figure 5(a)) is always needed for every batch operation. However, the recovered salt (RecSaltIn in Figure 5(a)), and regenerated and fresh salts (RegenSaltIn and freshSaltIn in Figure 5(a)), can be received or not according to the batch operation procedure. The complicated operation logic of section 3.1 was perfectly built in equation block in Figure 5. Input value connector (InCnt) in Figure 5(b) indicates the number of receipt of fragment/pellet (ORFeedIn) item. In other words, it indicates the number of batch operation of P2-1 because fragment/pellet (ORFeedIn) is always needed for every batch operation. The equation block judges if RecSaltIn, RegenSaltIn, and freshSaltIn feeds need to be received for current batch, and sends out 1 or 0 output signals through its output value variables (addRecycleedLiCl, addRegenLiCl, and addNewLiCl) to the batch blocks' second input value connectors in Figure5(c) to determine the corresponding feed is added to fragment/pellet feed.

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 Figure 4, as shown in Figure 6. Figure 7 shows some important blocks to calculate the amounts of hold-up and products, and to describe the operation time. The equation blocks in Figure 7(a) calculates hold-up, i.e., the remaining salt and fission products (FPs) dissolved from fragment/pellet feed in the salt before electro-chemical reaction occurs. In Figure 6, the equation block is located right before three activity blocks representing pre-process, main process, and post-process of the electro-chemical reaction. Three sequential activity blocks in Figure 7(b) merely play a role of consuming the corresponding process time. Actually, the electro-chemical reaction should be simulated with a continuous system modeling methodology if the main concerns are the changes according to time by electro-chemical reaction within a batch operation. However, this model calculates only the final result after the electro-chemical reaction because our main concern is how consecutive batch operations affect the material balance throughout the whole process.

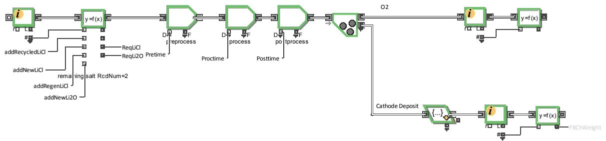
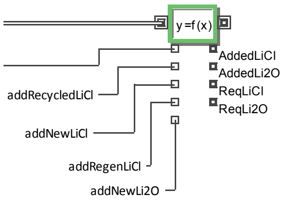
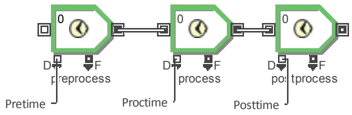


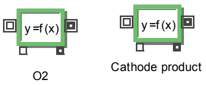
Figure 6: A selected part in P2-1 for calculation of mass compositions of hold-up and products (cathode product and O_2).



(a) salt addition



(b) reaction



(c) product

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

Two equation blocks in Figure 7(c) calculates the mass composition of cathode product and O_2. These blocks are located in downstream right after three activity blocks representing the electro-chemical reaction. Every calculation results are written in internal database of ExtendSim by equation blocks in Figure 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 2 and 3 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. Figures 8 and 9, and Figures 10 and 11 show process P2-1's material balance for the SNF element, and salt ($LiCl+Li_2O$), respectively. Figures 8 and 10 show the material balance to indicate the amount of material that is received from the input and sent to output each batch. On the other hand, Figures 9 and 11 show the material balance for the amount accumulated from the first batch. The amounts at the end batch operation in Figures 9 and 10 mean the equilibrium mass balance. In Figure 8, 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 top figure of Figure indicates sum of inputs (fragment/pellet, recovered salt and regenerated salt) are equal to sum of outputs (cathode product and O_2) and hold-up addition for every batch operation. The bottom figure of Figure 8 is magnified to properly investigate small amounts such as FPs contained in the hold-up, recovered salt, and regenerated salt.

In the first batch operation, SNF addition in hold-up (i.e., remaining salt) occurs owing to dissolution of some of PFs of fragment/pellet feed. Its contribution approximately amounts to 0.28kg per batch operation by first row in Table 2. Without any other contribution to SNF addition in hold-up, the amount of 0.28kg is the same over the batch operation. However, the SNF addition in remaining salt until 40th batch operations fluctuates more or less severely.

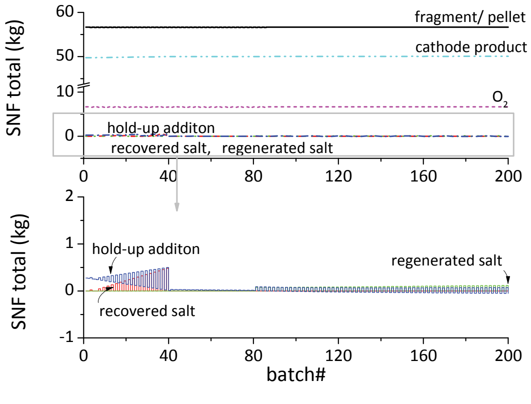


Figure 8: Material (a total SNF elements) balance every batch in P2-1.

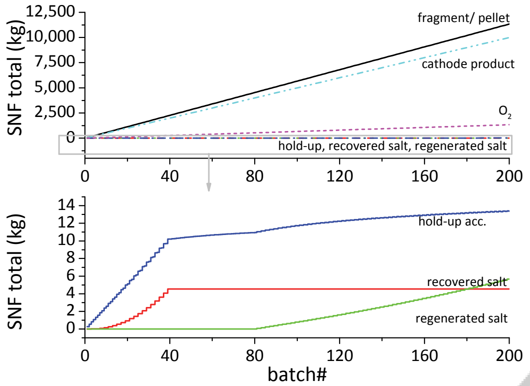


Figure 9: Accumulated material (a total of SNF elements) balance in P2-1.

When the hold-up addition is more than 0.28kg is when recovered salt is recycled. In such batch operation, PFs (which plays + contribution) accompanied with the recovered salt is more than PFs (which plays - contribution) accompanied with the entrained salt with cathode product. On the other hand, when the hold-up addition is less than 0.28kg is when the recovered salt is not recycled. In such batch operation, PFs accompanied with the entrained salt with cathode product goes out of the remaining salt. From the 2nd to the 40th operation, hold-up addition is fluctuated according to whether recovered salt is added or not. Since the recovered salt is the salt recovered from the salt entrained in cathode product, it contains the same FPs concentrations as the remaining salt in salt batch of P2-1. Hold-up addition increases every other batch since the recovered salt is added every other batch by operational procedure in section 3.1. During 2nd campaign (41~80 batch operations), neither recovered salt with high concentration of FPs nor regenerated salt with very low concentration of FPs is received. Only fresh salt without FPs is added to supplement a shortage of LiCl. It does not cause any abrupt change of hold-up addition compared to the 1st campaign. Approximately, 0.03kg of SNF is added in remaining salt for every batch operation during 2nd campaign.

Compared to Figure 8, the Figure 9 represents the accumulated in- and out-mass of P2-1. If the Figure 8 is simply integrated over batch operation number, it becomes Figure 9. In the bottom figure of Figure 9, FPs increase in remaining salt (see the line marked 'hold-up acc.' of the bottom of Figure 9) because recycled salts containing high concentration of FPs are constantly re-used without purification. In steep increase during 1st campaign, supplementary fresh salt addition during 2nd campaign does not bring about a significant increase in remaining salt. The gradual increase in remaining salt during 2nd campaign is only due to fragment/pellet's dissolution. Since 3rd campaign, the accumulation of SNF in remaining salt increases gradually but more steeply than 2nd campaign due to contribution of both the dissolution of some FPs of fragment/pellet and the regenerated salt addition with very low concentration of FPs. The SNF input accumulation by the recovered salt addition exponentially increases during 1st campaign, however, the accumulation stops increasing since 2nd campaign.

Figure 10 shows not the SNF material balance but the salt material balance such that the sum of fresh salt, recovered salt, and regenerated salt exactly match the sum of salt entrained with the cathode product and hold-up increment for every batch operation. Fresh salt addition occurs during only the 2nd campaign and the 1st batch of 2nd campaign needs more fresh salt than the others to supply the accumulation of 0.1% loss amount during the 1st campaign because cathode processing recovers only 99.9% salt. An insufficient amount of salt during the 1st campaign is supplemented through a recycling of recovered salt but is through the recycling of regenerated salt during the 3rd campaign. Figure 11 shows an accumulation of batch mass balance. The hold-up in Figure 11 indicates the amount of remaining salt in bath right after transfer of the cathode product, i.e., accumulation of hold-up. Since the 3rd 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.

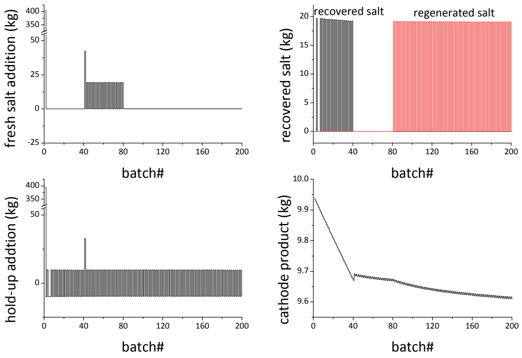


Figure 10: Material (salt: LiCl and Li_2O) balance every batch in P2-1.

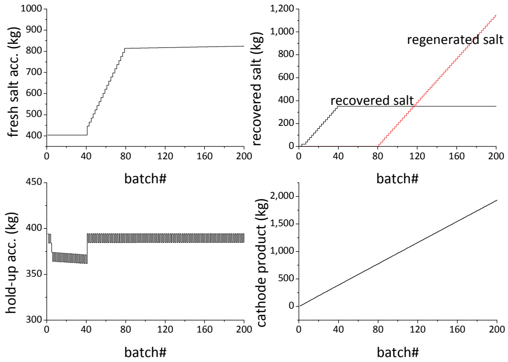


Figure 11: Accumulated material (salt: LiCl and Li_2O) balance in P2-1.

Validation of dynamic material flow was performed in every level of details for guaranteeing completeness of the model. Especially, dynamic mass balance using equation (2) was carefully checked for every batch operation and showed that SNF and salt mass balance is always satisfied.

5 CONCLUSIONS

DES based modeling was applied to build a pyroprocessing operation model, specifically, oxide reduction 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, dynamic material balance was verified for every batch operation.

This study is a meaningful step to confirm a part of an integrated pyroprocessing simulator in terms of dynamic material flow and its implementation under DES environment. The development of a multi-purpose integrated pyroprocessing simulator is still under progress with a mid-and long-term goal to cope with safeguards assessment, economic feasibility, technical evaluation, conceptual design, and support of licensing for a future pyroprocessing facility.

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