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Modeling of oxide reduction in repeated-batch pyroprocessing

Hyo Jik Lee *, Hun Suk Im, Geun Il Park

Department of Nuclear Fuel Cycle System Development, Korea Atomic Energy Research Institute, 111, Daekdaero 989 beon-gil, Yuseong-gu, Daejeon 305-353, Republic of Korea

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ABSTRACT

Pyroprocessing is a complicated batch-type operation, involving a highly complex material flow logic with a huge number of unit processes. Discrete event system modeling was used to create an integrated operation model for which simulation showed that dynamic material flow could be accomplished to provide considerable insight into the process operation. In the model simulation, the amount of material transported upstream and downstream in the process satisfies a mass balance equation while considering the hold-up incurred by every batch operation. This study also simulated, in detail, an oxide reduction group process embracing electrolytic reduction, cathode processing, and salt purification. Based on the default operation scenario, it showed that complex material flows could be precisely simulated in terms of the mass balance. Specifically, the amount of high-heat elements remaining in the molten salt bath is analyzed to evaluate the operation scenario.

1. Introduction

The material balance for pyroprocessing was first studied using a flowsheet (Piet et al., 2011). However, this was nothing more than a record of the amount of material transported through the incoming and outgoing streams during a specific period, in other words, the integral material balance. Thus, dynamic changes according to the batch operation cannot be predicted in an integral material flow. This study set out to build a dynamic material balance model based on the previously developed pyroprocessing flowsheet (Lee et al., 2013a). As a mid- to long-term goal, an integrated pyroprocessing simulator (Lee et al., 2013b) is being developed at the Korea Atomic Energy Research Institute (KAERI) for application to a review of the evaluation of the technical feasibility, safeguards, conceptual design, and economic feasibility. The most fundamental aspect of the development of such a simulator is the establishment of a 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.

Some interesting studies similar to this have been published recently. Researchers in the US set out to develop 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 estimating a material balance for the system design of pyrochemical reprocessing plants for performing batch processes. As a preliminary study, Korea also developed a DES-based model to implement a simplified dynamic material flow for pyroprocessing (Lee et al., 2011).

2. Oxide reduction

2.1. Pyroprocessing

The pyrometallurgical processing (pyroprocessing) of spent nuclear fuel (SNF) is now regarded as being one of the most promising options for future nuclear cycles in Korea (Kim, 2006). The Korea Atomic Energy Research Institute (KAERI) has been developing pyroprocessing technologies, which can reduce the increasing amounts of spent nuclear fuel and thus dramatically decrease the disposal load, through the recycling and destruction of the toxic waste (such as long-life fission products) in spent nuclear fuel (You et al., 2007). Pyroprocessing is still a developing technology and far from being mature. Considerable effort has been put into the investigation of the basic principles. Since the current study focuses on the individual processes, not on the overall process, it is difficult to predict the overall behavior of the system and the mutual influences of the processes. However,

Abbreviations: ASCI, advanced strategic computing initiative; CS, continuous system; Cs, cesium; Sr, strontium; Br, barium; Rb, rubidium; U, uranium; TRU, transuranic; DES, discrete event system; KAERI, Korea Atomic Energy Research Institute; Pu, plutonium; RE, rare earth; SNF, spent nuclear fuel; PRIDE, pyroprocessing integrated inactive demonstration facility.

* Corresponding author. Tel.: +82 42 868 4866; fax: +82 42 868 8679.
E-mail address: hyojik@kaeri.re.kr (H.J. Lee).

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modeling and simulation make it possible to predict unforeseeable behavior.

As shown in Fig. 1, pyroprocessing includes many processes and complex recycling flows. Since it consists almost entirely of batch-type processes, even though some resemble continuous processes, the use of a discrete event system for modeling is preferred if the main concerns are not the electro-chemical reactions within a single batch operation.

Each box in Fig. 1 indicates a grouped process. The number of unit processes is actually much greater than that shown in this figure. The arrows represent the material flow direction. Pyroprocessing produces not only recyclable products from SNF but also waste requiring disposal. The final products of pyroprocessing are uranium (U) metal ingots and transuranic (TRU) metal. The final waste consists of filter waste, metal waste, and ceramic waste.

Pyroprocessing features complicated batch-type operations, a complex material flow logic, and numerous SNF elements, all of which must be tracked. Thus, the material balance must be calculated whenever events such as feed arrival and product departure occur. Otherwise, the dynamic material flow cannot be tracked. A basic understanding of the overall process can be attained by the application of a flowsheet study, which captures the integral material balance at a specific time.

2.2. Oxide reduction

Although there are many unit processes involved in pyroprocessing, this study addressed the oxide reduction process. This process receives oxide SNF feed material in the form of porous pellets or fragments generated from the headend process. The oxide SNF is converted into a metallic form in a bath of molten lithium chloride (LiCl). During the electrolytic reduction process, the oxide pellets/fragments are reduced to a metal, 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 a cathode processing process to distill any residual salt entrained in the reduced metal, after which the metal is transferred to the next process, namely, electro-refining. The remaining LiCl salt in the electrolytic reduction bath after several process operations contains most of the fission products and presents a high heat load. These fission products include cesium (Cs), strontium (Sr), and barium (Ba), which have been separated from the reduced metal. The LiCl salt that is separated in the cathode processing process is then sent to a LiCl salt purification process to recycle it by separating the concentrated LiCl residue containing Cs, Sr, and Ba from pure LiCl. Fig. 2 illustrates the three unit process and product streams for oxide reduction.

The oxide reduction model deals with the 52 elements listed in Table 1, as well as two chemical compounds, namely, lithium chloride (LiCl) and lithium oxide (Li_2O). The volatile or semi-volatile elements such as tritium (H^3), carbon-14 (C^{14}), krypton (Kr), xenon (Xe), bromine (Br), and iodine (I) will have already been separated in the headend process such that the oxide reduction process does not have to deal with those elements. Alkali metal (AM) and alkali earth (AE) elements generate significant amounts of heat and have a half-life of approximately 30 years. Those elements are separated by electrolytic reduction (P2-1) into the molten salt bath and are then transferred to decay storage to allow the temperature to fall, at which point they are classified as lower- and intermediate-level waste. Therefore, it must be possible to track these elements in the model. One of the critical elements affecting proliferation-resistance is europium (Eu), given its high gamma dose rate.

3. Modeling

3.1. Operation

The pyroprocessing flowsheet study represents an integral mass balance, i.e., the total amount of material transported through the in- and out-streams during a specific period (typically one year). It does not provide detailed information regarding batch operations. To implement dynamic material flow, the batch operation procedure should be investigated, including the batch capacity and time.

The electrolytic reduction process (P2-1) uses a 50-kg heavy metal (HM)/batch and a 400-kg salt/batch. The electro-chemical reaction consumes 20 h/batch, and the pre-processing and post-processing times change according to the number of batches. The first batch of every campaign (one campaign is equivalent to 40 batch operations) requires 96 h for the pre-processing which is much more than the two hours required for the other batch operations. The last batch of every campaign requires 96 h of post-processing, which again is much more than two hours required for the other batch operations. Fig. 3 illustrates how pre- and post-operation times are assigned according to the batch operation number.

The electrolytic reduction receives recovered salt distilled in cathode processing (P2-2) of every other batch operation during the 1st campaign (1st through 40th batch operations). P2-1 of the first batch operation cannot receive the recovered salt from P2-2 because P2-2 is not running at that time. Therefore, in the 3rd, 5th... 39th batch operations, but excluding the 1st, P2-1 receives the recovered salts. Process P2-1 does not receive any salts from P2-2 during the 2nd campaign (41st through 80th batch operations) but instead receives an amount of fresh salt equal to the

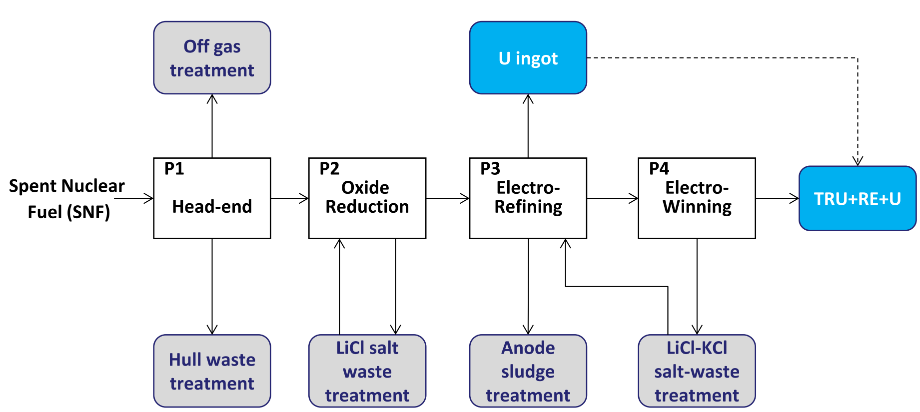

Fig. 1. Simplified material flow of pyroprocessing.