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AN OBJECT-ORIENTED SYSTEMS ENGINEERING MODEL DESIGN FOR INTEGRATING SPENT FUEL TREATMENT FACILITY AND CHEMICAL SEPARATION PROCESSES

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ABSTRACT

The mission of the Transmutation Research Program (TRP) at University of Nevada, Las Vegas (UNLV) is to establish a nuclear engineering test bed that can carry out effective transmutation and advanced reactor research and development effort. TRPSEMPro package, developed from previous project period, integrated a chemical separation code from the Argonne National Laboratories (ANL). Current research focus has two folds: development of simulation system processes applied to Spent Fuel Treatment Facility (SFTF) using ASPEN-plus and further interaction of ASPEN+ program from TRPSEMPro interface. More details will be discussed below.

ANL has identified three processes simulations using their separation technologies. The first process is to separate aqueous acid streams of acetic acid, nitric acid, water and a variety of fission product nitric salts. Distillation separation method is used to remove the desired components from the streams. The second simulation is to convert plutonium nitrate to plutonium metal. Steps used for the process simulation are precipitation, calcinations, fluorination and reduction. The third process currently under development is vitrification of fission product of raffinate streams. During the process, various waste streams from the plant are mixed and fed to a process that converts them to a solid state glass phase. The vitrification process used by the Hanford and Savannah River facilities was selected as a guideline to develop the prototype simulation process using ASPEN-Plus. Current research is focusing on identifying unit operations required to perform the

vitrification of the waste streams. The first two processes are near completion stage.

Microsoft Visual Basic (MS VB) has been used to develop the entire system engineering model package, TRPSEMPro. Currently a user friendly interface is under development to facilitate direct execution of ASPEN-plus within TRPSEMPro. The major purpose for the implementation is to create iterative interaction among system engineering modeling, ANL separation model and ASPEN-Plus process that outputs optimized separation/process simulation results. The ASPEN-plus access interface from TRPSEMPro allows users to modify and execute process parameters derived from the ASPEN Plus simulations without navigating through ASPEN-Plus. All ASPEN-plus simulation results can be also accessible by the interface. Such integration provide a single interaction gateway for researchers interested in SFTF process simulation without struggling with complicate data manipulation and juggling among various software packages.

INTRODUCTION

In 2003, the Department of Energy (DOE) launched the Advanced Fuel Cycle Initiative (AFCI) to address nuclear issues facing the United States. Amongst the major issues as identified by the AFCI were energy and waste management concerns. There are several types of nuclear waste, each classified by their origin and toxicity such as spent nuclear fuel (SNF) and transuranic (TRU) wastes. SNF waste is fuel that has been discharged from a nuclear reactor after being used for

at least one cycle or a reactor operation. TRU wastes are those that contain alpha-bearing radionuclides with atomic numbers greater than uranium (greater than 92 protons). Currently, the U.S. plans to store its SNF and high-level waste (HLW) in a deep geologic disposal repository located at Yucca Mountain, Nevada.

There are many questions regarding safety issues about the adverse health effects of long term storage of nuclear waste. Yucca Mountain has a finite capacity to which it can store spent nuclear waste. The limiting factor for determining this capacity is the temperature rise of the rock cause by the decay of the SNF. The majority of this heat comes from the transuranic elements. If these elements are removed from the waste, the storage capacity of the mountain would increase five times before the temperature would again become a problem. Specifically, the removal of cesium-137 (137Cs) and strontium-90 (90Sr) from the waste streams would allow for a one hundred fold increase in storage capacity.

Nearly all issues related to risks to future generations arising from long-term disposal of spent nuclear fuel is attributable to approximately one percent of its content. Chemical species responsible for the high toxicity can be broken down into two main groups: the transuranic elements - plutonium (Pu), neptunium (Np), americium (Am), and curium (Cm); and the long-lived isotope products from the fission process during power reaction. Searching for feasible technologies for nuclear transformation or transmutation that changes the contents of the nucleus (protons and/or neutrons) is the main mission for the Transmutation Research Program (TRP) at University of Nevada, Las Vegas (UNLV). Such program attempts to establish a nuclear engineering test bed for carrying out effective transmutation and advanced reactor research and development effort.

Figure 1 outlines a current process envisioned by the Argonne National Laboratory (ANL) researchers [1], [2], [3]. A continuous effort between the UNLV and ANL has created a system engineering model package that interfaces the ANL's chemical separation model, Argonne Model for Universal Solvent Extraction, (AMUSE) with database management system and optimization tools. By treating each separation process as a group of connected blocks with specific separation functions, an object-oriented systems engineering model software, TRPSEMPro (Transmutation Research Program System Engineering Model Project), was designed and developed. Since a complete AFCI request involves system process analysis utilizing both the AMUSE simulation and process analysis.

The second phase of the research is to integrate the process analysis tool, ASPEN-plus with the chemical separation code (AMUSE) under the TRPSEMPro package. Figure 2 outlines the draft relationship among the ASPEN-plus, AMUSE code,

and systems engineering model. Since complete SFTF contains many operations and processes for separating chemical species from the spent fuel, this paper mainly demonstrates two key processes, acid and plutonium metal production separations processes, and their integration with the system engineering model from previous research.

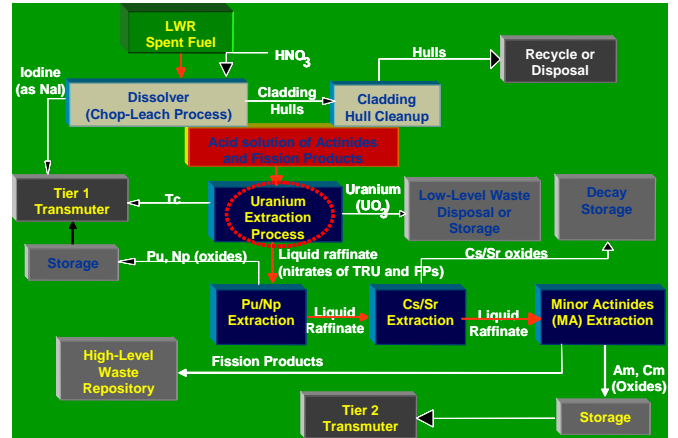


Figure 1 Chemical Separations System for TRP Program.

METHODOLOGY

This research is a further integration from the UNLV's previously developed system engineering model for solving complicate chemical separation process [4]. Fundamental architecture and concepts are the same while new middleware development is required for communicating with the already object-oriented designed ASPEN-plus software package.

System Engineering

System Engineering Model concerns with defining and implementing an approach to solving problems, while managing complexity and communicating over the entire lifetime of a project [5]. Such model can be generally divided into two parts [6]: modeling and optimization. The model usually performs requirements definition, conceptual design, subsystem partitioning, and system validation.

Software Architecture Design

A high-level architecture design deals with more on sufficiency and flexibility than on robustness and efficiency. Robustness and efficiency are usually better handled at lower levels of design. More details can be found on previous research [4]. To incorporate the ASPEN-plus software package into the systems engineering model, ASPENSimulator program is introduced as a communication layer that allows the user to pass data derived from AMUSE simulation into ASPEN-plus package.

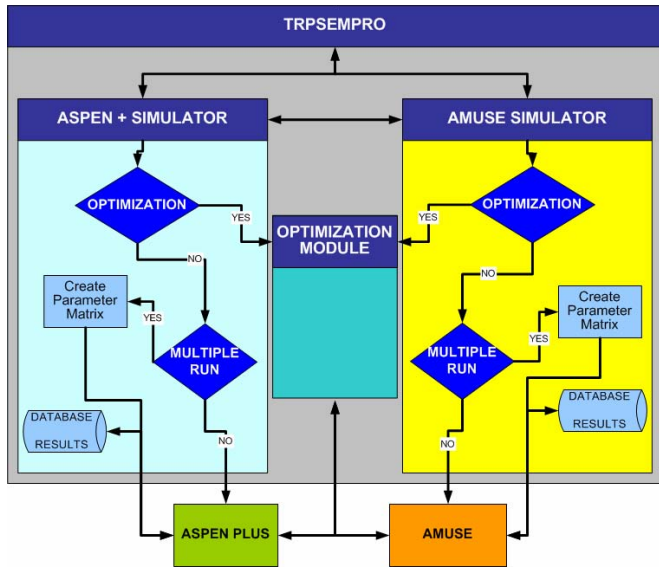


Figure 2 Module relationships among ASPEN-Plus, AMUSE-simulator and Systems Engineering Model.

SYSTEM PROCESS

The ASPEN-plus is a commercial software package with OOP modules available for end users. The possible scenario for long-term waste disposal risks is from approximately 1% of the spent fuel with the composition of transuranic group and long-lived isotopes of Iodine and Tc. While removing transuranic species from the fuel, the toxicity of the waste requires several hundred years to drop below natural uranium detection limits. Possible chemical separation simulations using ASPEN-plus are extractions of U/Tc, Cs/Sr, and Pu/Np, and Am/Cm Separation.

Nitric Acid Recycle

A key concept in the SFTF plant design is the recycle of nitric acid. The purpose of the nitric acid recycle system is to concentrate the spent nitric acid to a form that can be recycled back to the process users. The spent nitric acid streams from many processes are collected and sent to a distillation column where they can be separated from the impurities collected in the various separation processes. The feed to the separation column contains acetic acid as well as nitric acid. Figure 3 depicts one process in which nitric acid in the separations processes.

Plutonium Metal Production

The NPEX process is used by ANL scientists to remove plutonium and neptunium from spent fuel. The overall NPEX process can be seen in the Figure 4. In the NPEX process, tributyl-phosphate (TBP) in n-dodecane is used to extract the plutonium/neptunium from the cesium/strontium solvent

extraction raffinate. A slight nitric acid scrub is used to remove the fission products, americium/curium and the lanthanides. These species are collected in the NPEX raffinate stream and fed to the vitrification process elsewhere in the plant (beyond the scope of this thesis). Once the Pu/Np has been removed from the spent fuel, it needs to be further separated and eventually processed into pure plutonium metal. The work contained in this project is the simulation of the process following the removal of the plutonium/neptunium strip product (shown in red in the above figure). There are a variety of methods of producing plutonium metal each with their own individual strengths and weaknesses.

Developed at ANL and Handford National Laboratory (HNL), plutonium metal production by direct denitration is the simplest, most straightforward process. There are relatively few processes required for this option, resulting in a simple equipment operation. However, this process has been shown to have high corrosion rates due to nitric acid fumes; which results in extensive clean-up steps. Furthermore, direct denitration provides no decontamination from impurities. Oxide production via this method is severely limited and it is not known if the plutonium oxide would meet the required fuel specifications [7].

The plutonium peroxide precipitation method was originally developed at LANL but has been practiced at the SRP as well as at RFP. Advantages of this method include excellent decontamination from impurities as well as feasible methods of remote operation and maintenance. Disadvantages of this option include a presence of excess peroxide in the filtrate as well as a surge in pressure caused by the potential decomposition of hydrogen peroxide [8]. Precipitation of Pu (III) oxalate is the main process by which LANL scientists use for concentrating plutonium during recovery operations. Advantages of this process are numerous. The plutonium precipitate is easily enough handled and filtered in addition to having low decontamination from impurities. Furthermore, losses of plutonium to the filtrate are low.

The Sol-gel process developed by ORNL takes the plutonium nitrate feed and reacts it with n-hexanol to produce a sol-gel (colloidal suspension of silica particles). The major drawback of this option is that the process is quite complex and has not been demonstrated on an engineering scale [9].

The most successfully proven option is that of Pu (IV) oxalate precipitation. In general, this process provides a good yield of easily enough filterable precipitate which can be calcined to an oxide powder which meets product specifications. The work contained in this thesis uses the Pu (IV) oxalate precipitation method as the backbone for plutonium metal production.

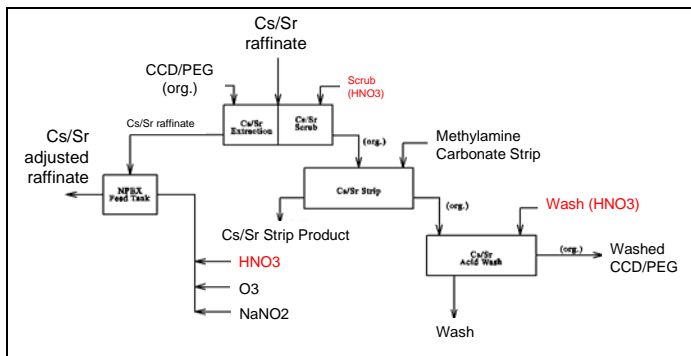


Figure 3 Flowsheet of the Nitric acid separation.

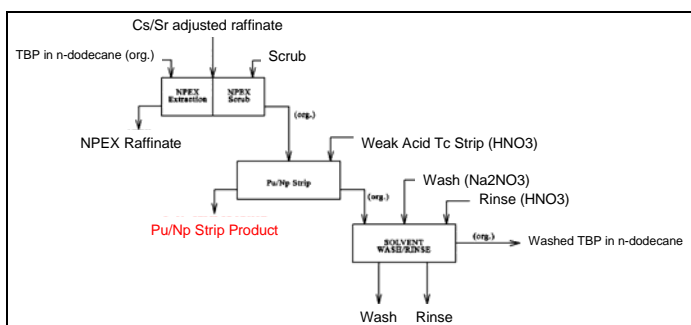


Figure 4 Flowsheet of the NPEX Process.

This plutonium metal production process using precipitation-calcination-fluorination reduction that is the motivation for the simulation is that which is in operation at the Department of Energy's Hanford Site. This process follows the commonly used method to produce plutonium metal, namely precipitation-calcination-fluorination-reduction. In this research, precipitation, calcination and fluorination are to be simulated by ASPEN Plus. The reduction operations are beyond the scope of this project. The plutonium nitrate product stream from the NPEX process is received in product receiver (PR) cans. The PR cans are manually loaded into staging tanks where they are blended and sampled. The solution batches are vacuum transferred to a preparation tank. In this preparation tank, the solution acidity, plutonium concentration, and valence of the nitrate feed are operator adjusted in an effort to guarantee optimal performance for the conversion of nitrate-to-oxalate in the first reactor. Concentrated solutions of 12M and 2M nitric acid are used to adjust the acidity and overall plutonium concentration in the PR. Hydrogen peroxide is used to adjust the plutonium valence via reduction-oxidation. Samples of the solution are taken after the adjustments have been made in an effort to ensure the correct feed conditions.

The first operation in the plutonium metal production process is the reaction of the plutonium nitrate feed with oxalic acid to produce brown plutonium oxalate solids.

The oxalate solids are then converted to plutonium oxide in a screw calciner. This reaction is performed through countercurrent contact with an air stream of near 450 degree C. After the plutonium oxide has been produced, it enters a fluorinator where it becomes plutonium tetrafluoride by reaction with hydrogen fluoride and oxygen at 525 degree C. The conversion of plutonium to the fluoride compound is the final step in the simulation. From here, the plutonium tetrafluoride is collected and reduced to plutonium metal (beyond the scope of this project). This project takes the feed stream of plutonium nitrate and follows the above process to produce plutonium tetrafluoride. This flowsheet can be seen in Figure 5.

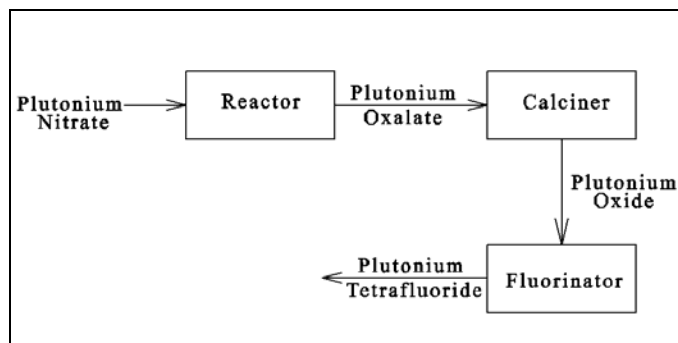


Figure 5 Plutonium Production Flowsheet.

ASPEN PLUS SIMULATION FLOWSHEETS

Nitric Acid Recycle

The spent nitric acid streams from processes are collected and sent to a distillation column where they can be separated from the impurities collected as indicated in Figure 6. The feed to the separation column contains acetic acid as well as nitric acid.

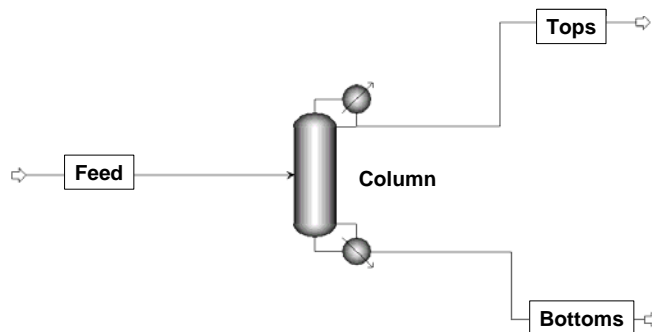


Figure 6 ASPEN Plus simulation plot for the nitric acid recycle.

Plutonium Metal Production

The plutonium metal production process is simulated by ASPEN Plus with a variety of assumptions. The first of these

assumptions is that we are only concerned with Pu (IV). For that reason, only the Pu (IV) ion is present in the feed. The process can be separated into three distinct unit operations. The first unit is the reaction of plutonium nitrate with oxalic acid in the reactor to produce plutonium oxalate while the second unit operation is the calcination of plutonium oxalate into plutonium oxide. In the calciner, the plutonium oxalate is converted to plutonium oxide (green powder). The oxalate is reacted with air in a countercurrent fashion at around 450 degree C. The calciner has a length of 7 feet, an outside diameter of 10 inches and has a pitch of 1 inch. ASPEN Plus does not have a block dedicated to a calciner, therefore the process will be simulated as a reactor. The final step in the plutonium metal production process is the fluorination of plutonium oxide. The plutonium oxide from the calciner is fed to the fluorinator and allowed to react with a gaseous mixture of hydrogen fluoride (HF) and oxygen at 525 degree C. In the fluorinator, the countercurrent stream of HF and O₂ reacts with the PuO₂ to form plutonium tetrafluoride (PuF₄) in accordance with the following reaction.

The temperature in the fluorinator is kept at 525 degree C. The fluorinator is about five feet long and has six heating elements for which to keep the internal temperature at the desired warmth. The PuF₄ product is collected in powder pans and transferred to the process responsible for reduction.

It should be noted that Figure 7 is a simplified version of the plutonium reduction process and the icons used are simply used as visual reference. Eventually more unit operations will be included in an effort to accurately represent the reduction process later based on more inputs from the ANL.

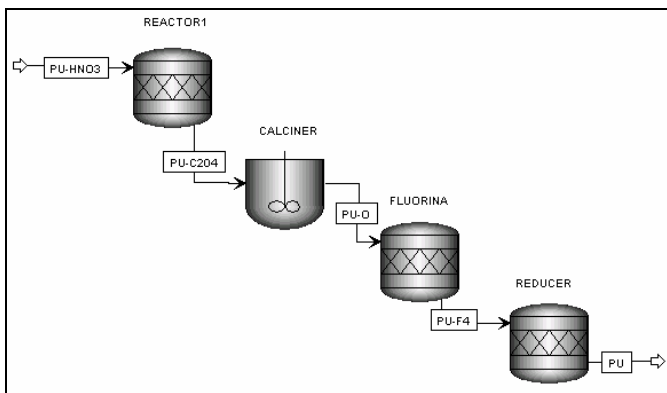


Figure 7 Pu extraction schema using ASPEN-Plus simulation.

SIMULATION RESULTS

Some preliminary results from nitric acid reduction are reported here. The feed stream consists of 100 lbmol/hr of nitric acid, 10 lbmol/hr of water and 10 lbmol/hr of acetic acid. The temperature of the stream is set to 78 degree F under 1 atm

(14.7 psi) pressure. The distillate rate is 25 lbmol/hr with 15 stages. Figure 8 shows the molar flow rate result for the bottom unit while varying reflux ratio. From the graphs it can be seen that an increase in reflux ratio leads to an increase in the molar flow of both acetic acid and water in the bottoms stream. Conversely, the amount of acetic acid present in the bottoms decreases. The opposite is true for the tops stream. It should be noted that the changes in molar flowrates are small. Changing the reflux ratio from 5 to 100, results in minimal variation of outlet flowrate. Therefore, varying reflux ratio has a minimal effect on the changing the outlet molar flowrate.

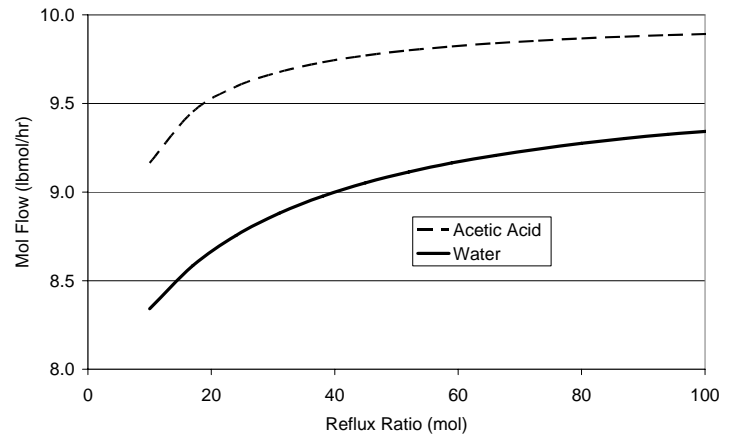


Figure 8 Molar flow rate results for the bottom unit of nitric acid reduction.

Figure 9 shows the molar flowrate results for the nitric acids with the variation of stage numbers. The physical conditions are held the same as the previous scenario. The distillate rate is 25 lbmol/hr and the reflux ratio is 15. The results show that the maximum required number of stages for the separation occurs at around 10. When the number of stages is more than 10, there was no measurable notice of an increase of molar flowrates. From 15 to 100 stages, the flowrates in both the tops and bottoms streams remained constant. In the bottoms streams, the molar flowrates of acetic acid and water increased with increasing number of stages; while the flow of nitric acid decreased with the same increase. The opposite holds true for the tops stream. This study shows that it is not necessary to run more than 10 stages in the column.

Figure 10 compares molar flowrate results with the variation of distillate rate. Increasing the distillate rate causes a drastic decrease in the molar flowrate of nitric acid in the bottoms stream (and thus causes an increase in the tops flowrate). Similarly, increasing the distillate causes a decrease in acetic acid water. The extra cost of adding an additional tower to further separate the tops product stream (of large quantity produced nitric acid) might not be justified when it might be more economically feasible to produce a less acceptable tops stream at a lower cost.

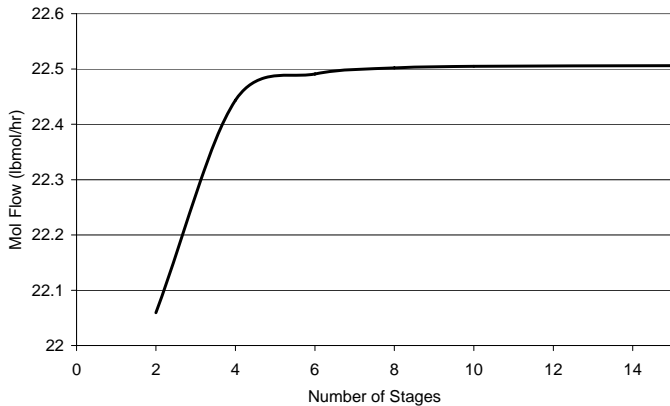


Figure 9 Molar flow rate results for the top unit of nitric acid reduction while varying number of stage.

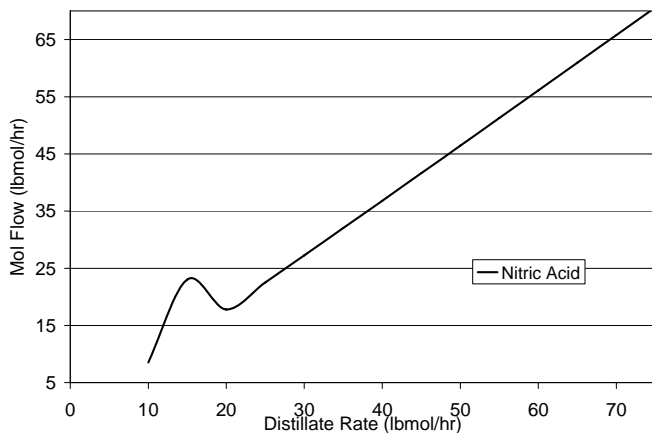


Figure 10 Molar flow rate results for the top unit of nitric acid reduction while varying distillate rate.

CONCLUSIONS

The entire chemical separation process in Figure 1 is complex to the point that definitely requires certain level of systematic coordination. To perform smoothly and meet the target extraction rates and cost-effective processes, this research proposes an interface to communicate with ASPEN-plus and to interact with the AMUSE simulation results. This research considers design concepts from requirements definition and conceptual design to system partitioning, and finally system validation. Lengthy pre-coding of the design process and recursive modification provides a system package with high degree of flexibility and robustness.

The system model integrates AMUSE code from ANL and the commercially available ASPEN Plus package. TRPSEMPro can interact with both identities and perform optimization process in the future. The created system allows industries to

model their process quantitatively and to study the interactions between subsystems and performance of the model under the influence of various design parameters. This research reports several ASPEN Plus simulation results that demonstrate the capability of using ASPEN Plus to simulate interested processes.

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