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Separations and Safeguards Model Integration

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Separations and Safeguards Model Integration

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Abstract

Research and development of advanced reprocessing plant designs can greatly benefit from the development of a reprocessing plant model capable of transient solvent extraction chemistry. This type of model can be used to optimize the operations of a plant as well as the designs for safeguards, security, and safety. Previous work has integrated a transient solvent extraction simulation module, based on the Solvent Extraction Process Having Interaction Solutes (SEPHIS) code developed at Oak Ridge National Laboratory, with the Separations and Safeguards Performance Model (SSPM) developed at Sandia National Laboratory, as a first step toward creating a more versatile design and evaluation tool. The goal of this work was to strengthen the integration by linking more variables between the two codes. The results from this integrated model show expected operational performance through plant transients. Additionally, ORIGEN source term files were integrated into the SSPM to provide concentrations, radioactivity, neutron emission rate, and thermal power data for various spent fuels. This data was used to generate measurement blocks that can determine the radioactivity, neutron emission rate, or thermal power of any stream or vessel in the plant model. This work examined how the code could be expanded to integrate other separation steps and benchmark the results to other data. Recommendations for future work will be presented.

Acknowledgement

The authors would like to acknowledge the help of Valmor de Almeida at Oak Ridge National Laboratory for his contribution in the original integration of SEPHIS with the SSPM, and J.D. Smith for running the ORIGEN source term files.

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Acronyms

AMUSE	Argonne Model for Universal Solvent Extraction
CCD-PEG	Cesium/Strontium Extraction
FCRD	Fuel Cycle Research and Development
GWD	Gigawatt-Day
MBA	Material Balance Area
MPACT	Material Protection, Accounting, and Control Technologies
MT	Metric Ton
NDA	Non-Destructive Analysis
NEAMS	Nuclear Energy Advanced Modeling and Simulation
ORIGEN	Oak Ridge Isotope Generation and Depletion Code
PUREX	Plutonium and Uranium Extraction
PWR	Pressurized Water Reactor
SEPHIS	Solvent Extraction Process Having Interaction Solvents
SSPM	Separations & Safeguards Performance Model
TALSPEAK	Rare Earth Fission Product Extraction
TRUEX	Transuranics Extraction
UREX	Uranium Extraction
V&V	Verification and Validation

1.0 Introduction

The Fuel Cycle Research and Development (FCRD) program performs the long-term research needed for the next generation of nuclear fuel cycle facilities. Reprocessing plants in particular are challenging facilities due to high costs and licensing concerns as well as safeguards, safety, and security requirements. A transient plant model can provide a useful platform for the design and evaluation of these systems without requiring costly experimental facilities. The purpose of this work was to develop such a reprocessing plant model with realistic solvent extraction and a versatile source term library.

The Separations and Safeguards Performance Model (SSPM) is a transient plant model that was originally built for the design of safeguards systems at Sandia National Laboratories [1]. It models material flow rates through a reprocessing plant and simulates measurements that would be used for material protection, accounting, and control. The original model did not include solvent extraction chemistry, and instead used assumptions to control the separations processes.

The Solvent Extraction Process Having Interaction Solutes (SEPHIS) model was developed at Oak Ridge National Laboratory to model transient solvent extraction in the PUREX process [2,3]. This model was integrated into the SSPM code to provide solvent extraction chemistry in a more versatile plant model. The initial integration was originally funded through the National Nuclear Security Administration [4].

In addition, a versatile source term was desired to allow the users of this tool to have a variety of different fuels to choose from. The type of fuel being reprocessed can impact the plant design as heat loads and radiation levels change. Radiation levels can also impact the design of measurement instrumentation. Another goal of this work was to introduce a variety of fuel types and also allow tracking of radioactivity and heat load at any point in the plant.

A future reprocessing plant design will use input from all of the various working groups within the FCRD program. This project overlaps with the Separations, MPACT (Material Protection Accounting and Control Technologies), and the NEAMS (Nuclear Energy Advanced Modeling and Simulation) working groups. A further goal of this work was to serve as an integration point between the three working groups to help design the integrated model into a tool that will be useful for both separations and safeguards experts. Although this tool can be used for near term research, the results provided here can also form the basis of more detailed plant-level simulation codes being developed in the NEAMS campaign.

2.0 Background

2.1 Separations and Safeguards Performance Model (SSPM)

The Separations and Safeguards Performance Model (SSPM) [1] is a high-level materials tracking model of a reprocessing plant developed at Sandia National Laboratories for materials accountancy and process monitoring analysis. The SSPM is constructed in Matlab Simulink and tracks cold chemicals, bulk fluid flow, solids, and mass flow rates of elements 1-99 on the periodic table. However, since separations modeling was not the goal of the SSPM, the chemical processes were described with simplified models, and all separation efficiencies were originally assumed with static values. The original purpose of the model was to simulate materials accountancy and process monitoring measurements. This data is used to simulate inventory difference calculations and examine the instrumentation response to material loss scenarios.

Figure 1 shows the front end of the SSPM in the Simulink environment. The processing vessels are shown as the black blocks and contain functionality that models their operation. Each signal connecting the blocks contains a 101-element array that keeps track of the mass flow rates of elements 1-99, the total liquid flow rate, and the total solids flow rate.

The blue blocks, which may be connected to either process streams or vessel inventories, are used to simulate accountancy or process monitoring measurements. For example, the "Acc MS" block above the accountability tank simulates a plutonium concentration measurement from a sample taken once every 8 hours. Each measurement block is customized for the particular measurement. Diversion blocks can also be added throughout the model to determine the instrumentation response to material loss. Elsewhere in the model (not shown), all of the data from the measurements are used to determine an overall inventory difference.

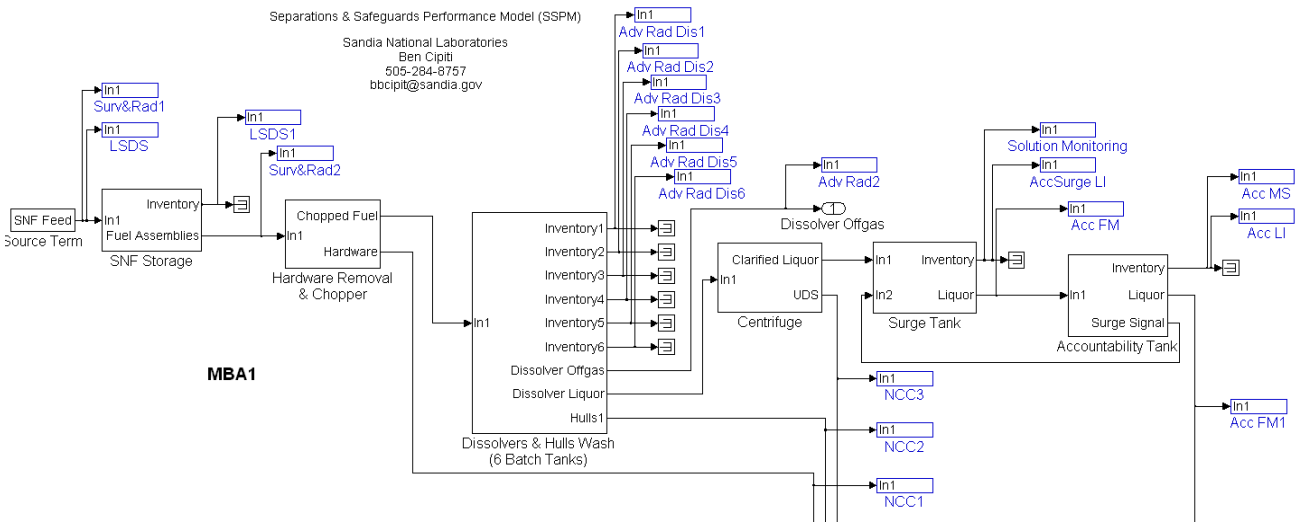

Figure 1: Front End (MBA1)

Figure 2 shows the separations portion of the model for a PUREX reprocessing plant (which separates U and Pu only), used for this study. However, a UREX+ version of the model exists which includes other extraction steps such as CCD-PEG (for Cs/Sr), TRUEX (transuranic and rare earth fission product extraction), and TALSPEAK (rare earth fission product removal). The PUREX Contactors block shown in Figure 2 was modified in this work to provide a more accurate model of separations.

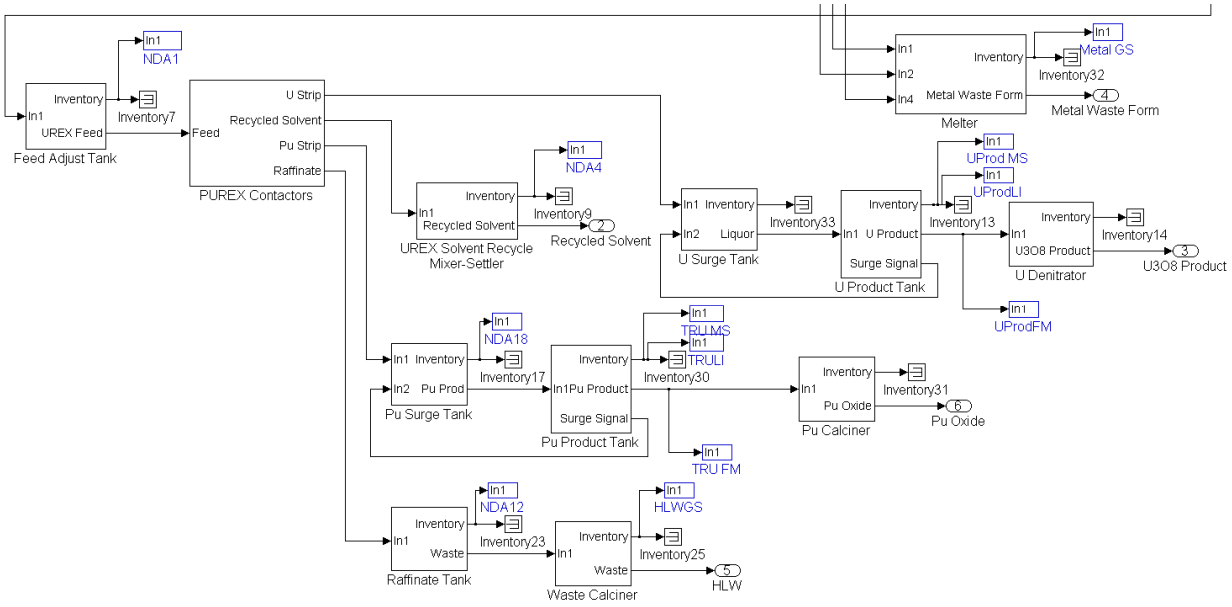

Figure 2: Separations - PUREX (MBA2)

2.2 Solvent Extraction Process Having Interaction Solutes (SEPHIS)

SEPHIS (Solvent Extraction Process Having Interacting Solutes) is an existing process model that predicts solute concentration profiles in aqueous and organic phases throughout a solvent extraction process as a function of time. The previous year's work used a mathematical model based on the SEPHIS code to develop a solvent extraction module for integration with the SSPM. The model is described in reference 4 but will be repeated in this section for background.

The advantage of SEPHIS is that it provides transient results. The SEPHIS model was originally designed from a simulation for the PUREX process based on an idealized model for mixer-settlers [5]. However, other extraction steps have been modeled in the years since its development. SEPHIS has been used as the basis for PUREX flowsheet designs at Y-12, SRNL and Hanford.

The model considered a number of solutes for the PUREX process, including nitric acid, uranium, plutonium (IV), plutonium (III), a plutonium reductant, and inextractable nitrates. The flow of solutes throughout the extraction process is modeled via ordinary differential equations. The distribution of solutes between the aqueous and organic phases is determined by subroutines

that estimate distribution coefficients based on experimental data. Reduction reactions between solutes are also controlled in program subroutines.

The model can be potentially applied to other contactors such as pulsed columns or centrifugal contactors through appropriate selection of parameters. The solutes enter each mixer through an aqueous or organic feed stream, the aqueous stream from the previous stage, and the organic stream from the next stage. Perfect mixing and equilibrium is assumed stage-wise; therefore, the aqueous and organic streams containing the corresponding compositions are defined by the distribution coefficients and are separated as they enter a respective settler. Upon leaving the settler, these streams can either exit the system or continue as interstage flow.

SEPHIS is designed to model transients in a bank of interconnected solvent extraction stages. The basic equation in SEPHIS is an unsteady-state mass balance of mixer contents. Between stage mixers are settlers, whose concentrations must also be solved. The settlers propagate outgoing concentrations from the mixer to the next stage or to a product stream; additional reactions may be taking place in the settlers such as plutonium reduction. The dynamics of transport through the system is governed by the residence time in each stage, directly related to the mixer and settler volumes and the fluid flow rates.

SEPHIS provides a knowledge base upon which to develop transient process models. The main program block performs process-generic mass balance calculations and iterations, while process-specific calculations (extraction coefficients, density changes, temperature effects) are performed in other code blocks. These coefficient values are, in all instances, calibrated by experimental data. Future needed work with SEPHIS includes improved treatment of kinetic processes and improved predictions of partitioning. However, the key relevant feature of SEPHIS as a solvent extraction process model is its ability to model the build up of solutes in a bank of ideal contactors as the transfer process approaches steady state and the transient effects of process upsets.

2.3 Integration of SSPM and SEPHIS (Past Work)

One of the advantages of using Simulink for the modeling platform is that the Matlab workspace can be used to calculate more advanced functions. Matlab embedded functions and m-files can be used to perform calculations on the signals in Simulink. This was the method for integrating the SEPHIS-based solvent extraction module into the SSPM.

An embedded function block was used in the Simulink model to convert the signals going into the PUREX contactor block into variables in the Matlab workspace. This block is called periodically (not on every time step) to save on computational time. A calculation every 15 minutes appeared to be adequate for this model. Originally, the only input into this embedded function block was the feed flow rate and composition. A separate m-file was called to populate an input file and run the solvent extraction unit module.

After the solvent extraction unit m-file ran, the embedded code used the results and converted them back into Simulink outputs. These outputs were used to calculate the uranium and

plutonium mass flow rates in each of the output blocks. Much more detail about the embedded function and solvent extraction unit file are given in Reference 4.

3.0 Modifications to the Integrated Code

The goal of this past year was to strengthen the integration by linking the cold chemicals from the SSPM model as additional drivers for the solvent extraction unit file. The cold chemicals include the organic solvent and the various scrub and strip solutions. These flow rates were originally assumed and hard coded into the solvent extraction unit file. By allowing these flow rates to change as the model changes, it provides more realism for actual input conditions. In addition to improving plant operations modeling, it also provides a capability for examining diversion scenarios (for safeguards) that may be due to process upsets or intentional altering of the cold chemical flows.

Figure 3 shows the embedded function block which calls the SEPHIS executable. The Feed stream is the dissolved fuel going into the contactor train. The cold chemicals are also shown as inputs on the left driving the enabled subsystem. Both the feed and cold chemical data are used to populate the input file for the solvent extraction code. After running this modified version of SEPHIS, the output data is used to determine the flow streams out of this block.

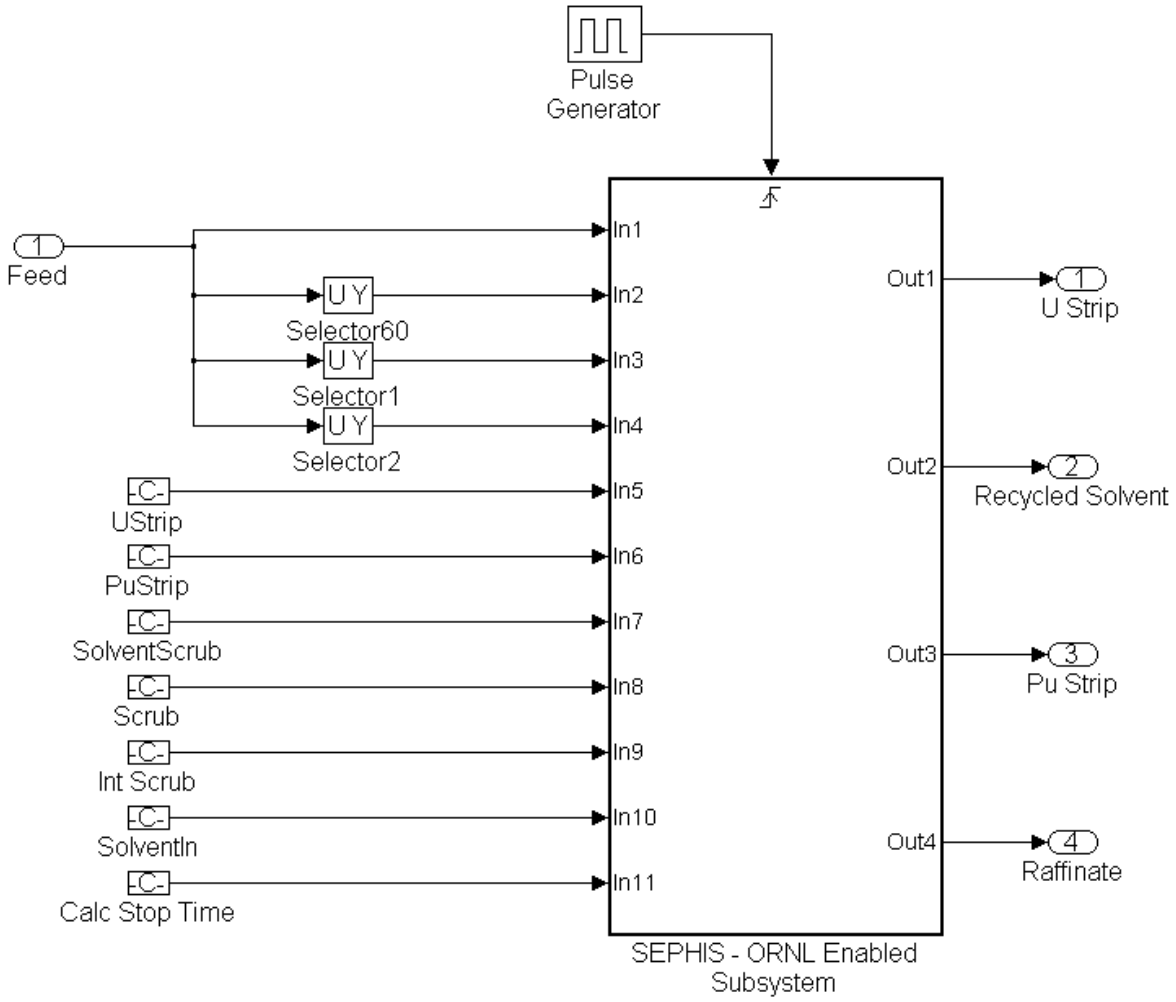

Figure 3: Embedded SEPHIS Function Block

3.1 Results

A series of runs were completed once these modifications were completed to test the operation of the model. These runs looked at the uranium and plutonium mass flow rates in the PUREX feed, uranium product, and plutonium product streams.

Figure 4 shows the results during normal operation. For this run, it is assumed that a plant starts without nuclear material present, and the feed to the contactor train turns on at hour 27. At this point, the feed is mostly constant, but the operation of the plant does lead to slight flow differences with time. Figures 4a and 4b show the uranium and plutonium flow rates (respectively) in the feed stream. Figure 4c shows the uranium mass flow rate in the uranium product stream, and as expected it contains most of the uranium. The plutonium flow rate in the uranium product (Figure 4d) is zero. Figure 4f shows the plutonium flow rate in the plutonium product stream, and figure 4e shows that a small amount of uranium is also present.

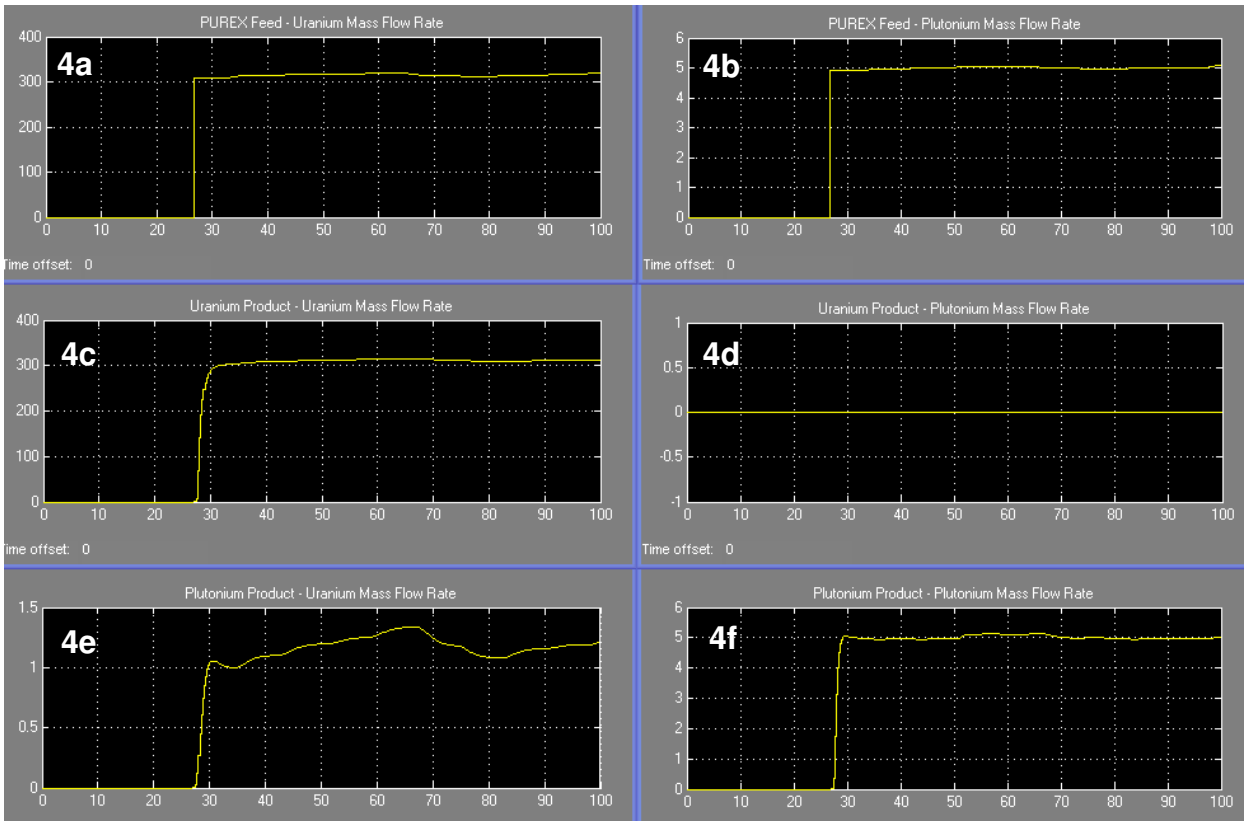

Figure 4: SEPHIS-SSPM Integration, Normal Operation

In all cases the transient nature of the SEPHIS calculation is shown since the product stream is not a step function. It takes several hours for steady-state to be reached from a startup condition, and this trend is shown in the figures.

Figure 5 shows the results with a transient. In this transient, the fuel feed into the plant changed midway through the run to a higher plutonium concentration. This change is shown in the feed

in Figure 5b. Figure 5f shows that this small transient is dealt with quickly, and the operation appears to be normal.

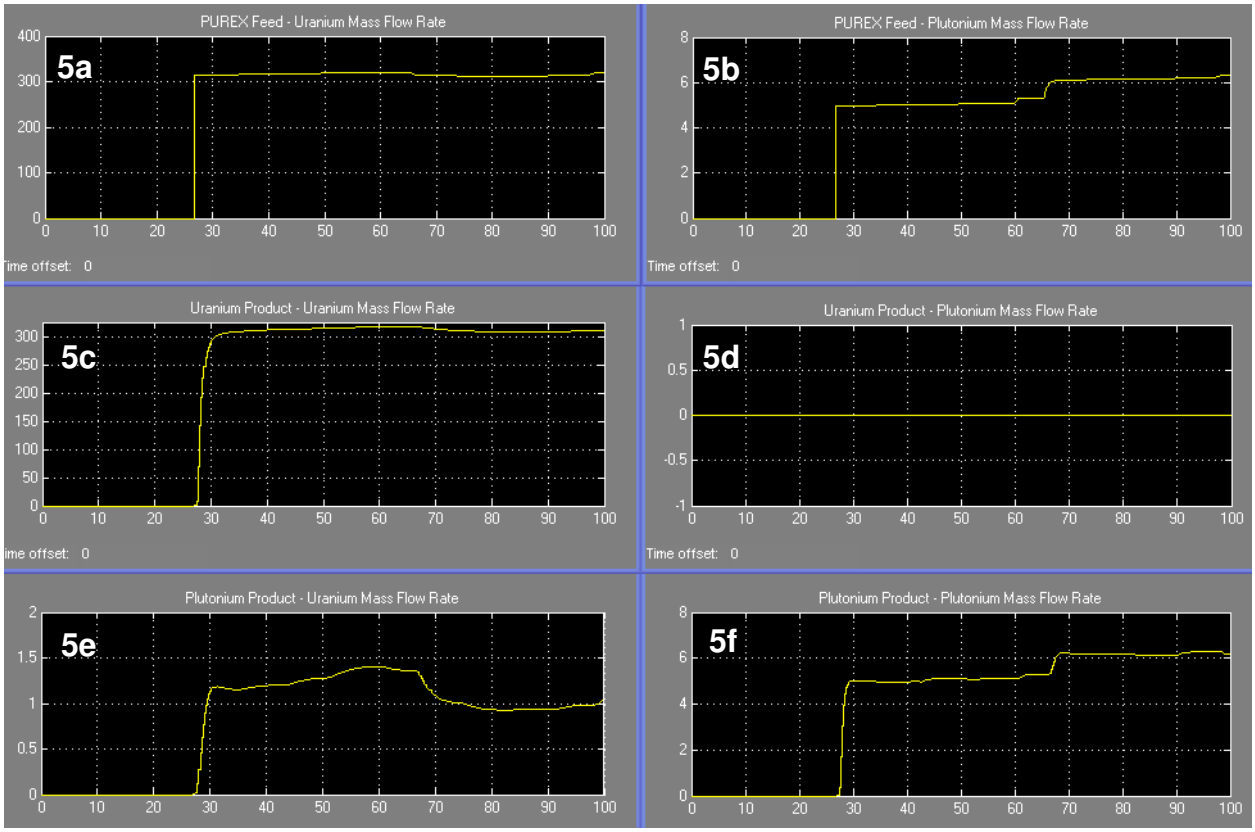

Figure 5: SEPHIS-SSPM Integration, Transient

3.2 Discussion

Given the nature of contactor operation, the results in the previous two figures appear to be reasonable. The integrated model results were compared to the SEPHIS stand-alone results in other tests and were found to be the same. This verified that the integration did not change the data in any way. However, it would be useful to benchmark or validate the model in a more robust way. Future work may examine validation in more detail.

One of the disadvantages of this integration is that it is based on PUREX, whereas future plants may use different extraction processes (UREX+). Past work at ORNL has modified SEPHIS for UREX+ extraction processes [6], but more work would be required in order to integrate these models into the SSPM. This path forward would be very useful to provide a more complete transient plant since UREX+ is a likely design choice for the future.

4.0 Integration of ORIGEN Data

The second goal of this work was to provide a versatile source term to run the SSPM model with different fuel types. The integration of the ORIGEN code [7] with the SSPM was expressed as an interest in order to generate various source terms. However, after careful consideration, it was decided that integration of the code was unnecessary, and that the same functionality could be created simply by integrating ORIGEN output. This approach also considerably conserves computational expense.

A set of source term data tables were created to represent a range of spent light water reactor fuel. ORIGEN was used to generate 9 source terms for fuel with the following burnups and enrichments:

33 GWD/MT 2.60% enrichment	33 GWD/MT 3.30% enrichment	33 GWD/MT 4.00% enrichment
45 GWD/MT 3.30% enrichment	45 GWD/MT 4.00% enrichment	45 GWD/MT 4.70% enrichment
60 GWD/MT 4.03% enrichment	60 GWD/MT 4.73% enrichment	60 GWD/MT 5.43% enrichment

Table 1: Nine ORIGEN Source Terms for SSPM Input

For each of these 9 runs, the data was plotted at 1, 5, 10, 25, and 50 years after discharge. Thus, this set of data provides 45 different options of burnup, enrichment, and decay time that can be used as the input for the model.

The data generated for the runs included concentration (g/MT), radioactivity (Ci/MT), thermal power (W/MT), alpha radioactivity (Ci/MT), and neutron emission (n/s/MT). This data was extensively reorganized into excel files that could be easily read into data arrays by Matlab.

A Matlab script was generated to allow the user of the SSPM to enter the desired fuel source term. Based on those inputs, the appropriate data tables are read into data arrays. The concentration values are used to directly determine the mass flow rates of the spent fuel as it enters the model. The other data in the tables were converted into ratios based on the concentration. For example, the radioactivity values per element were converted into Ci/g of the element present. This was done to make it easier to use the data.

With this setup in the Matlab/Simulink environment, Simulink blocks were created that could be used to determine the total radioactivity, thermal power, alpha radioactivity, or neutron emission of any stream or vessel in the plant (see Figure 6). The advantage of this setup is that the data is not carried through the entire model (which would take up unnecessary computational time). Rather, the information is calculated from the original data tables only when needed.

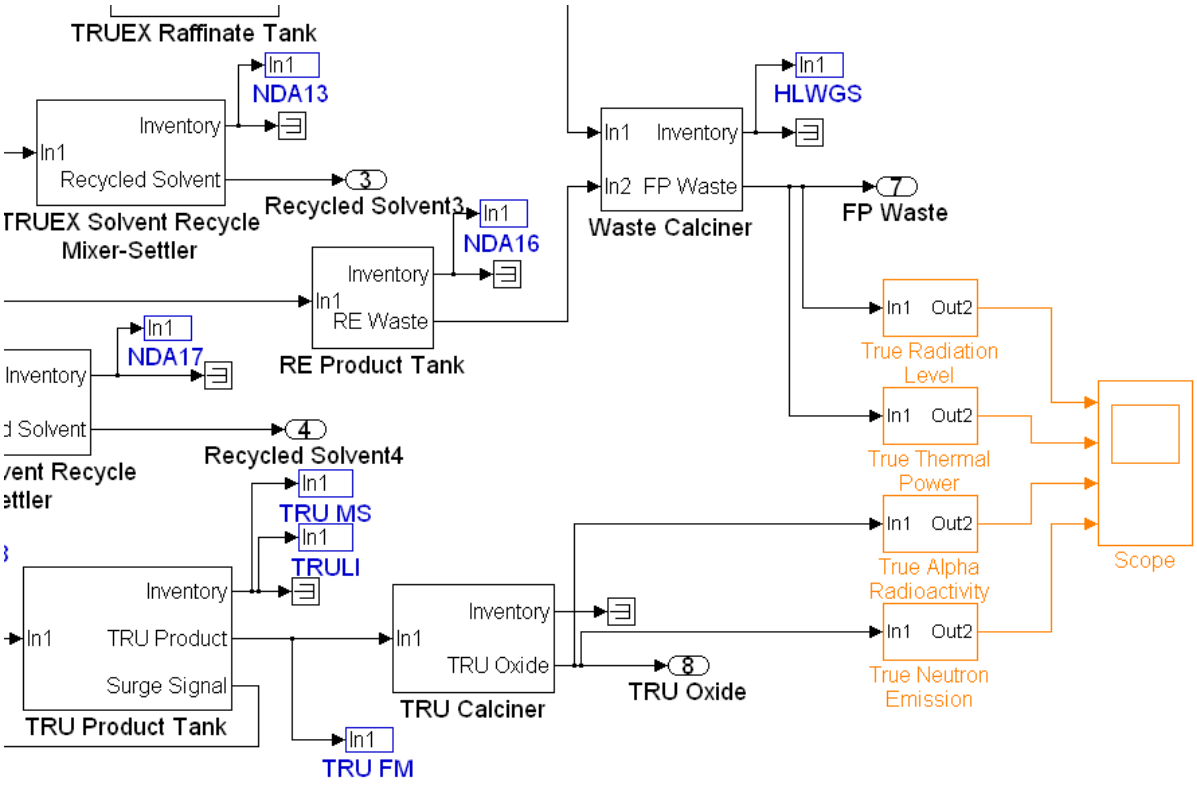

Figure 6: Radioactivity, Thermal Power, Alpha, and Neutron Emission Blocks

4.1 Testing the Emission Blocks

The four new emission blocks were tested by hooking them up to the key inputs and output streams in the separations portion of the UREX+ model. To avoid confusion, it should be noted that this model did not include the SEPHIS integration as described in the previous section. For consistency, the blocks were attached to steady-flow streams in order to make a direct comparison. The input stream used was the feed into the UREX contactors. The four outputs used were the U/Tc product, TRUEX raffinate, rare earth product, and the TRU product streams. Correct implementation was indicated if the sum of the outputs equaled the input stream value. (In all cases, the fuel input assumed was 60 GWD/MT, 4.03% initial enrichment, and 10 year cooled fuel.)

Figure 7 shows the results from the five streams with the Total Radioactivity block connected to each. The top line shows the total radioactivity of the UREX feed stream given as Ci/hr—it reached steady-state at 306,600 Ci/hr. The blue line shows the radioactivity of the TRUEX raffinate with a steady-state value at 210,800 Ci/hr. This makes sense since the Cs/Sr are part of this stream. The green line is the TRU product with a steady-state value at 79,170 Ci/hr. The red line is the rare earth product at 16,400 Ci/hr steady-state. Finally, the lowest line on the plot is the U/Tc product at 300 Ci/hr steady-state. Combined, the four product streams add up to the input value.

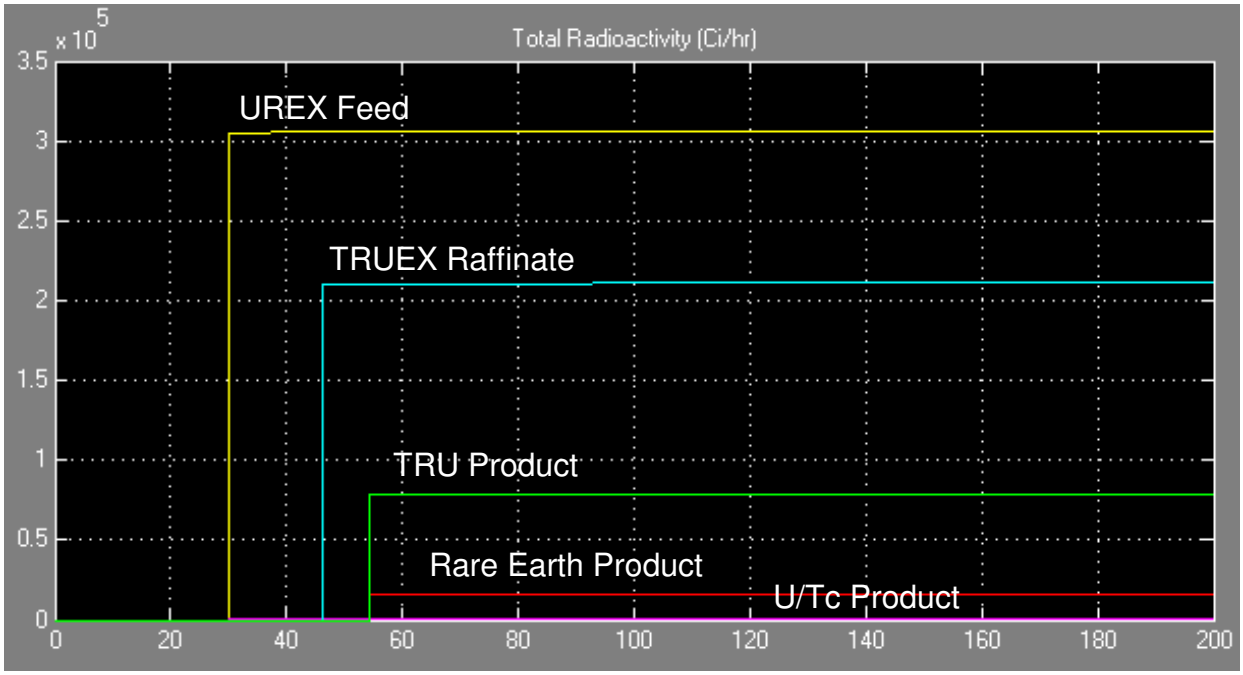

Figure 7: Total Radioactivity Balance Test

Figure 8 shows the results from the five streams with the Total Heat Load block connected to each. The trends are similar to those in Figure 7. The top line shows the total radioactivity of the UREX feed stream given as W/hr—it reached steady-state at 983 W/hr. The blue line shows the radioactivity of the TRUEX raffinate with a steady-state value at 517 W/hr. The green line is the TRU product with a steady-state value at 424 W/hr. The red line is the rare earth product at 42 W/hr steady-state. Finally, the lowest line on the plot is the U/Tc product at 1 W/hr steady-state. Again, the four product streams add up to the input value.

Figure 9 shows the results from the five streams with the Total Neutron Emission block connected to each. In this case, the neutron emission is due to the actinides, so the rest of the streams only have small values due to actinide impurities. The top line shows the total neutron emission of the UREX feed stream given as n/s/hr—it reached steady-state at 7.75e8 n/s/hr. The green line is the TRU product with a steady-state value at 7.73e8 n/s/hr. The rest of the lines are not distinguishable on the plot, and all fall below 1e6 n/s/hr. Again, the four product streams add up to the input value.

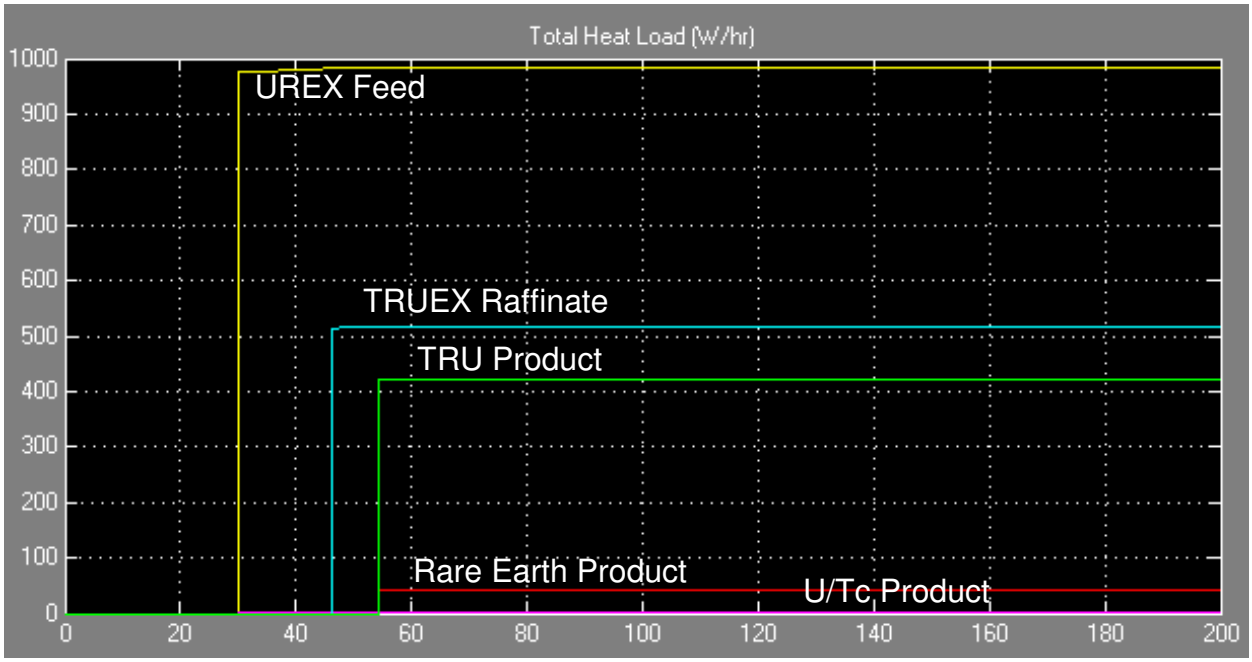

Figure 8: Total Heat Load Balance Test

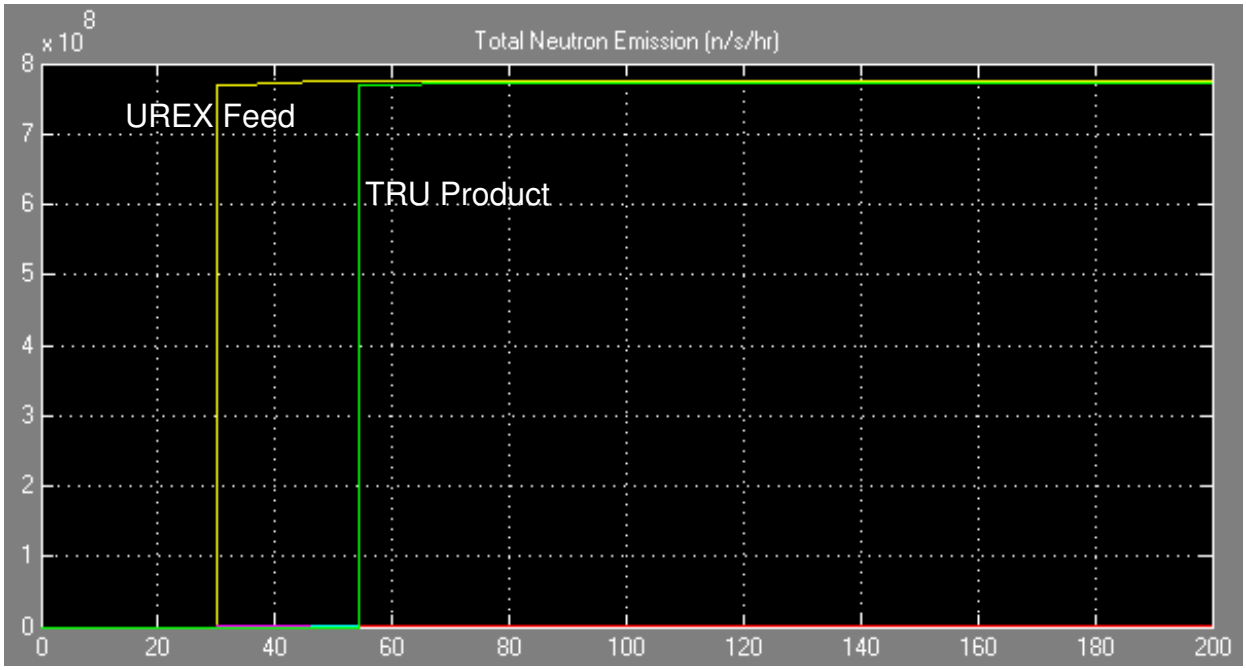

Figure 9: Total Neutron Emission Balance Test

4.2 Discussion

This modification provides a capability that will be useful in the design of the plant, including the design of the safeguards system. The ability to measure the thermal power is useful for design of cooling systems. The radioactivity measurements help to design shielding requirements and may be used to assist in the design of NDA measurements.

The ORIGEN data that was utilized also included gamma emission spectra that could be used in the model as well. This emission data is divided into energy bins, and would be more difficult to convert for use in the model. The determination of emission by bins would be useful for shielding considerations, but provides limited value for design of safeguards instrumentation. For that goal, specific gamma spectra would be required.

One potential way to achieve this goal is to integrate the ORIGEN data with a gamma spectra code such as RADSource [8], GADRAS, [9], or SYNTH [10]. These codes can simulate gamma spectra for a particular detector geometry given the isotopic mixture and geometry. In general, the gamma spectra in most of the reprocessing plant streams gets swamped by a few key isotopes such as Cs/Sr, so the value of integrating one of these codes may be limited. However, there may be areas of the plant where the radiation signature is such that more detail can be seen. Future work may need to look into this in more detail.

5.0 Conclusion

The modifications to the SSPM described above provide new capabilities for this software tool. The integration of SEPHIS provides a transient solvent extraction model, and the Simulink model puts SEPHIS results into a much more visual output. The higher detail transient modeling provides a more realistic basis for evaluating diversion scenarios.

It would be useful to develop similar embedded functions for the UREX+ type extraction steps as well. Future work may examine the work at ORNL in this area. It would also be useful to fold in the Argonne Model for Universal Solvent Extraction (AMUSE) data to this effort since AMUSE provides steady-state separations chemistry for the UREX+ separations.

The addition of the ORIGEN source term tables now provide 45 different source terms that the user can use to run the model. These tables span the range of PWR spent fuel, and future efforts could create source terms for spent mixed oxide fuel as well. The ability to determine the radioactivity, heat load, alpha, and neutron emission at any point in the plant may prove to be a valuable design feature both for plant operations and for the design of safeguards and safety systems.

A challenge for this area of work is verification and validation (V&V). No plan for V&V currently exists, but it will be required if the SSPM starts to be used for facility design. Validation has been performed in only a very limited case to roughly determine if the results match up to independent SEPHIS runs. Future efforts will need to outline a plan for comparing the results to actual data.

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