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Transformative Monitoring Approaches for Reprocessing

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Abstract

The future of reprocessing in the United States is strongly driven by plant economics. With increasing safeguards, security, and safety requirements, future plant monitoring systems must be able to demonstrate more efficient operations while improving the current state of the art. The goal of this work was to design and examine the incorporation of advanced plant monitoring technologies into safeguards systems with attention to the burden on the operator. The technologies examined include micro-fluidic sampling for more rapid analytical measurements and spectroscopy-based techniques for on-line process monitoring. The Separations and Safeguards Performance Model was used to design the layout and test the effect of adding these technologies to reprocessing. The results here show that both technologies fill key gaps in existing materials accountability that provide detection of diversion events that may not be detected in a timely manner in existing plants. The plant architecture and results under diversion scenarios are described. As a tangent to this work, both the AMUSE and SEPHIS solvent extraction codes were examined for integration in the model to improve the reality of diversion scenarios. The AMUSE integration was found to be the most successful and provided useful results. The SEPHIS integration is still a work in progress and may provide an alternative option.

Acknowledgement

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Acronyms

AMUSE	Argonne Model for Universal Solvent Extraction
CuSum ID	Cumulative Sum of the Inventory Difference
MBA	Material Balance Area
MPACT	Material Protection, Accounting, and Control Technologies
NDA	Non-Destructive Analysis
NRTA	Near Real Time Accountability
PUREX	Plutonium and Uranium Extraction
SASPE	Stagewise Algorithm for Speciation and Partitioning Equilibrium
SASSE	Spreadsheet Algorithm for Stagewise Solvent Extraction
SEPHIS	Solvent Extraction Process Having Interaction Solvents
SSPM	Separations & Safeguards Performance Model
TALSPEAK	Rare Earth Fission Product Extraction
TBP	Tributyl Phosphate
TRU	Transuranics
TRUEX	Transuranics Extraction
UREX	Uranium Extraction
UV-Vis-NIR	Ultraviolet-Visible-Near Infrared

1.0 Introduction

Nuclear fuel reprocessing plants face economic constraints due to their need for stringent safety, safeguards, security, and environmental requirements. As such, one of the drivers of the Fuel Cycle Technologies program is to develop designs that can significantly reduce the cost of various systems in a future reprocessing plant. While these systems need to be efficient and cost-effective, they also may face tighter requirements than those of existing plants in operation around the world.

The goal of this work is to examine transformative plant monitoring approaches that use plant data more efficiently, reduce the burden of accountancy systems on the operator, and reach a new regime of complete plant awareness in near real time. This work utilizes a systems approach to design and evaluate advanced plant monitoring systems. New technologies developed in both the Separations and MPACT (Material Protection Accounting and Control Technologies) working groups were used as the basis for this study.

The design and analysis was completed using the Separations and Safeguards Performance Model (SSPM) which was developed at Sandia National Laboratories. The SSPM is a transient aqueous reprocessing plant model that tracks mass flow rates through the various unit operations. Materials accountancy and process monitoring measurements are modeled throughout the plant to generate simulated data. This data is then used to calculate inventory differences to determine how well the instrumentation responds to diversion events.

Parallel work in the MPACT working group has developed the architecture for the integration of bulk process monitoring data with traditional materials accountancy [1]. In this work, the technologies being developed in the Separations working group were also integrated. The UV-Vis-NIR spectroscopic technique provides a method for continuous on-line monitoring of the chemical signature of a processing solution [2]. The SSPM was used to determine the places within the plant where this technology can make the greatest impact. In addition, current work at Idaho National Laboratory is examining micro-fluidic sampling and more automation in the analytical lab to significantly speed up precision measurements of actinides and other elements in reprocessing samples [3,4]. The SSPM was used to determine how this technology can have the greatest impact as well. The design and analysis will be presented.

As a tangent to this work, two solvent extraction codes were examined for integration into the SSPM. Both the SEPHIS (Solvent Extraction Process Having Interaction Solutes) code [5] at Oak Ridge National Laboratory and the AMUSE (Argonne Model for Universal Solvent Extraction) code [6] at Argonne National Laboratory were evaluated for integration. Either of these codes can provide a higher degree of detail in the plant model for more realistic diversion scenario results. The initial results of the integration will be described.

2.0 Background

2.1 Separations and Safeguards Performance Model (SSPM)

The Separations and Safeguards Performance Model (SSPM) [1,7,8] is a transient reprocessing plant model 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. The number of tracked species can be expanded to include cold chemicals or specific isotopes as needed.

The SSPM contains a number of capabilities including:

- Spent fuel source term library for user-defined runs with varying initial enrichment, burnup, and cooling time.
- Heat load and radioactivity tracking at any location in the plant.
- Customizable measurement points with user-defined errors.
- Automated calculation of inventory difference and error propagation in real-time, including statistical modules for determining alarm conditions.
- User-defined diversion scenarios.
- Integration of process monitoring data, material measurements, material accountancy procedures, and physical security data for complete plant monitoring.

The model is used for design and analysis of advanced plant monitoring systems. Recent work has evaluated the integration of bulk process monitoring data (level measurements, mass flow rates, volumetric flow rates, etc.) with traditional accounting [1]. The SSPM provides a platform for testing new measurement technologies and evaluating the overall improvement to safeguards. In general improvement may be measured as high sensitivity to diversion events or more timely detection of diversion. The SSPM has also been evaluated for integration of safeguards and security.

Figure 1 shows the model during operation. The underlying model is shown in the background. Black blocks represent key unit functions, blue blocks are measurement points, and red blocks are potential areas for setting up diversion scenarios. The scope shows an example of an alarm condition being surpassed during a diversion scenario. The model is setup to automatically calculate inventory differences and look for plant abnormalities that might represent material loss.

Various versions of the model have been created. The baseline model is a UREX+ version that includes the UREX, TRUEX, and TALSPEAK extraction steps. An alternative PUREX model also exists. The UREX+ version is the most up-to-date and was used for the analyses presented here.

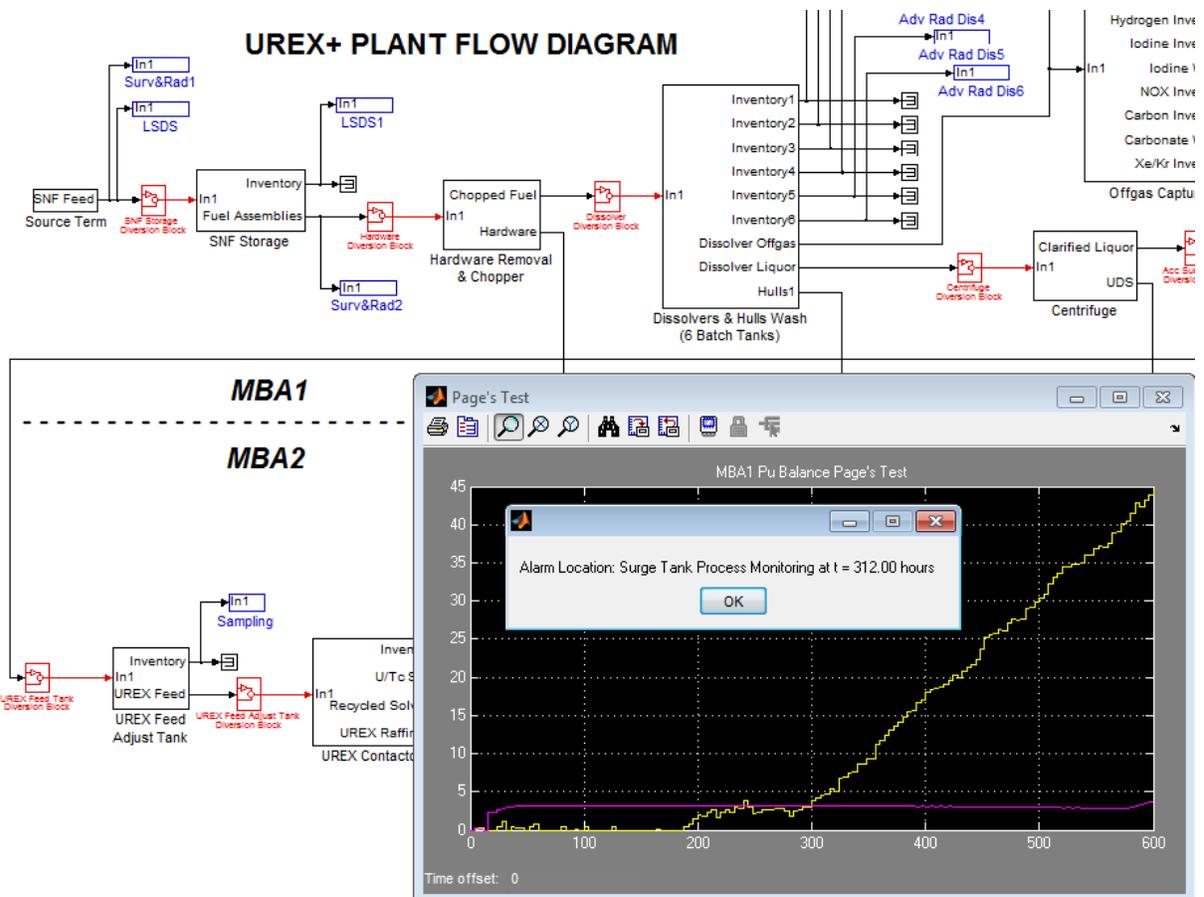


Figure 1: SSPM operation under a diversion scenario

2.2 Process Monitoring Technologies Evaluated

The Separations Working Group is evaluating two key technologies that may provide significant benefit to both process control and materials accountancy. UV-Vis-NIR (ultraviolet-visible-near infrared) and Raman spectroscopy are being evaluated to continuously monitor chemical compositions in process streams including actinides, fission products, or cold chemicals. Fiber optic probes easily allow for this measurement in the process cell. Using Raman spectroscopy, uranium concentration can be measured with an uncertainty around 0.8%. Using Vis-NIR spectroscopy, the concentration of tetravalent plutonium can be measured with an uncertainty around 1.7%. This technology is robust in the wide range of elements and chemicals that can be monitored [2].

Micro-fluidic sampling is also being evaluated in the program to improve turn-around-time, reduce cost, and reduce waste of sampling and analytical measurements for reprocessing accountancy [3,4]. This concept utilizes a microchip capillary-based sampling system and more automated interface to a mass spectrometer. Micro-liter samples significantly reduce the waste and exposure to lab workers and provide more samples for later reference. A more automated

interface will speed up the measurements to reduce overall cost and can provide the potential to sample in more places in the reprocessing plant. In addition, a more rapid turn-around-time can reduce the size of accountability tanks which saves capital cost for the plant.

The goal of this work was to determine where these technologies could make the greatest impact in the reprocessing environment. Spectroscopic techniques may not have the low uncertainty needed at accountability tanks, but they can provide tremendous value at other points in the process. Micro-fluidic sampling enables more sampling for precise analytical measurements, but its use will not make sense everywhere due to the impact on operations. The SSPM was used to optimize how these technologies fit into the overall safeguards system.

2.3 Solvent Extraction Codes

The development of advanced plant monitoring systems requires a model that is realistic of actual process conditions in a reprocessing plant. While a significant amount of detail exists in the SSPM for a majority of the process units, the separations modules originally were too simplistic. Fortunately, other solvent extraction codes can be added to the SSPM to improve the modeling of the contactor banks.

SEPHIS 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. It has been developed at Oak Ridge National Laboratory. The advantage of SEPHIS is that it provides transient results. The SEPHIS model was originally designed as 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 [9]. SEPHIS has been used as the basis for PUREX flowsheet designs at Y-12, SRNL and Hanford.

A rudimentary integration of SEPHIS into the SSPM was completed in past work, but this work utilized a PUREX model [8,10]. A first start at the coupling of the two codes was completed and provided transient plutonium and uranium flow rates that appeared consistent with what would be expected. However, the coupling was not very elegant and was not designed to model UREX, TRUEX, TALSPEAK or other extraction steps. One of the goals of this work was to examine if a more seamless integration with the UREX+ version was possible.

AMUSE (Argonne Model for Universal Solvent Extraction) is a more recent code that originally was developed for steady-state flow sheet design for the UREX+ suite of extraction steps [6]. The code has progressed and current work is developing a transient version. As an alternative to the SEPHIS, AMUSE may provide another route to including solvent extraction in the SSPM. The goal of this work was to integrate AMUSE to the SSPM and compare which code can be more easily integrated directly into the model.

3.0 Modeling of Advanced Measurement Technologies

Current and past work under the MPACT (Material Protection Accounting and Control Technologies) campaign has examined the move toward near real time accountability (NRTA) of actinides in reprocessing. NRTA requires actinide measurements at all points within a plant as well as at the accountability tanks at the beginning and ends of a material balance area (MBA). NRTA provides much more timely detection of material loss, so a plant operator could respond more rapidly to any plant anomaly (which may include an intentional diversion or a leaking vessel).

The NRTA approach is different for the two different MBAs that have been examined. MBA1 either contains spent fuel in solid form or in solution. Since no extractions have taken place yet, every area of the front end has roughly equal importance to the MBA1 balance. In other words, each area in MBA1 is processing large quantities of plutonium, so precision measurements are needed throughout. These measurements depend highly on a plutonium measurement in spent fuel, as it could be used to estimate plutonium content in most of the rest of MBA1. Sampling and spectroscopic techniques are not appropriate here, so MBA1 will be ignored for the rest of this section.

In MBA2, most of the areas in the plant are processing liquids, so sampling and spectroscopy techniques may be applicable in many locations. A previous sensitivity study found which areas within this MBA contribute the most to overall uncertainty [1]. Figure 2 shows the areas that process large amounts of plutonium in yellow—these key tanks would need to be sampled and measured with low uncertainty using analytical techniques for NRTA. The micro-fluidic sampling technology could be useful in these locations since NRTA requires more measurements as compared to traditional accounting. The areas shown in green are areas that process small quantities of plutonium. This may be because either the plutonium has been separated or because the amount of material in process at any one time is very small. For these areas, higher measurement uncertainties (10-20%) are adequate. These areas may be ideal locations for spectroscopic techniques.

The SSPM was used to test this overall safeguards architecture for MBA2. The goal was to parametrically determine how these technology improvements would improve the overall system. In the model, the areas shown in yellow were assumed to use micro-fluidic sampling and analytical measurements to determine plutonium concentration to σ_r (random error) = σ_s (systematic error) = 0.2%. The areas shown in green were assumed to utilize a spectroscopic or other NDA technique to estimate total plutonium content to $\sigma_r = \sigma_s = 2\%$. In reality, both analytical sampling and spectroscopic techniques do not determine total plutonium content, but rather they can determine concentration. Tank level measurements are used in parallel to determine total plutonium content. The integration of the bulk process monitoring data will be discussed in more detail later.

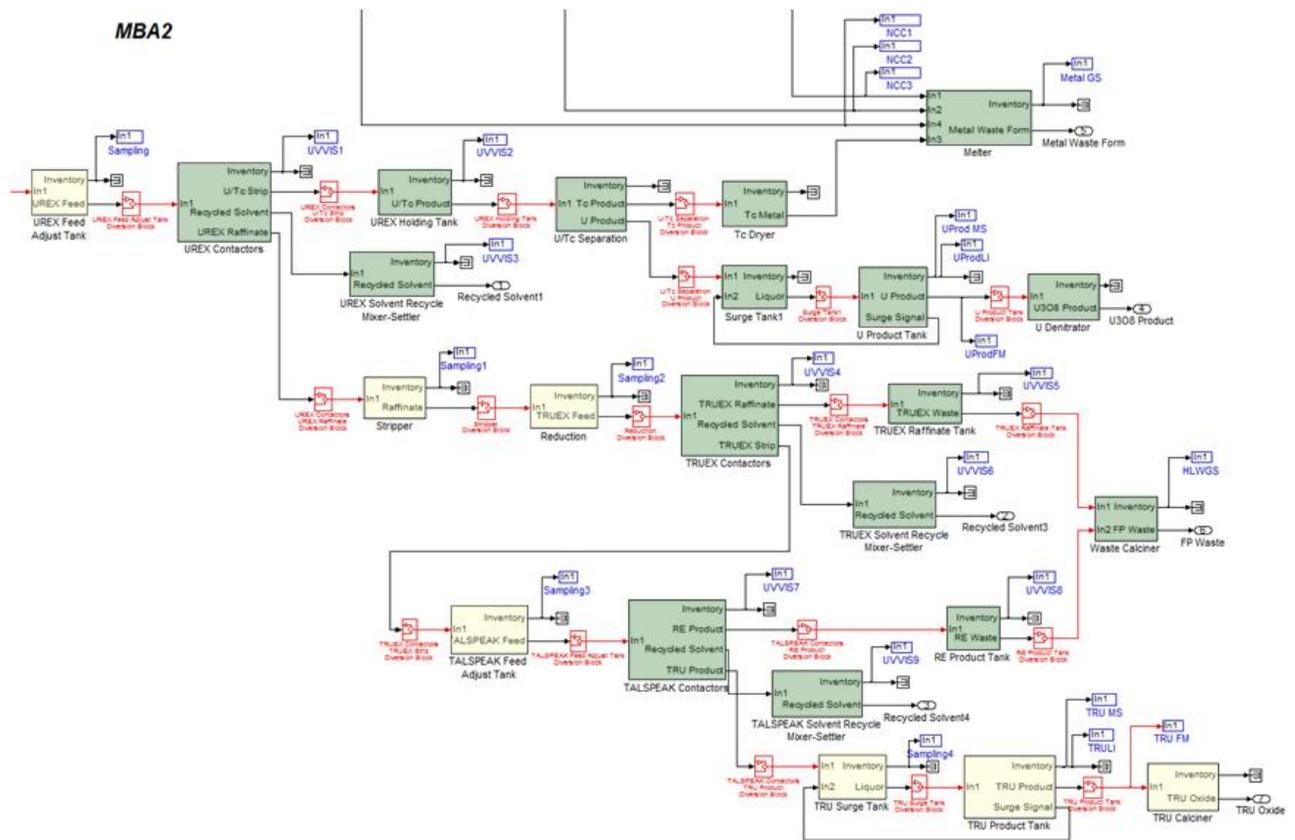


Figure 2: MBA2 with areas processing small (green) and large (yellow) quantities of plutonium

The effects of the above changes needed to be compared to a baseline that is representative of what is done in existing plants. This was somewhat difficult to implement since existing reprocessing plants use PUREX, and MBA2 is simpler than that shown here. Discussions with an expert in the implementation of safeguards on Rokkasho were used to estimate how the existing plants work [11]. The baseline assumed sampling and analytical measurements at the accountability tank and the three key product tanks in MBA2 (uranium product, fission product waste, and TRU product tanks). These areas were assumed to achieve plutonium concentration measurements with $\sigma_r = \sigma_s = 0.2\%$. The rest of the plant can only use bulk process data to estimate plutonium content, but does not measure specific actinides. Actinide content can only be estimated from measurements in the accountability tank. In addition, this estimating leads to an uncertainty that was modeled as a 10% random error.

Four diversion scenarios were evaluated for both the baseline and advanced monitoring strategies. Both abrupt and protracted diversions were modeled, as well as both direct and substitution diversions. An abrupt diversion is an immediate removal of material (usually over a day or less), while protracted diversions involve a gradual removal of material over weeks or months. Direct diversion involves directly removing the material or processing solution; substitution diversions involve removing material and replacing it with a surrogate (such as nitric acid solution). The following section describes the results of the diversion scenarios.

3.1 Diversion Scenario Results

A total of 4 diversion scenarios were examined for both the baseline and advanced NRTA system. These included the following:

- Abrupt, direct diversion – Removal from the Stripper Tank before the TRUEX extraction starting at hour 300 and ending at hour 324 for a total removal of 8 kg of plutonium.
- Abrupt, substitution diversion – Same as the previous, except the removed material was replaced with clean solution.
- Protracted, direct diversion - Removal from the Stripper Tank before the TRUEX extraction starting at hour 300 and ending at hour 1900 for a total removal of 8 kg of plutonium.
- Protracted, substitution diversion – Same as the previous, except the removed material was replaced with clean solution.

The baseline system utilized the process monitoring balances and the Page's Test to look for bulk material loss in the Stripper Tank. On the other hand, the plutonium balance was not able to use the Page's Test due to the higher uncertainty of the baseline system. (This was not a limitation of the test, but rather of the way the test was implemented in the model. In order to run the Page's Test as the model runs, a bias correction was first applied. With the higher measurement uncertainties of the baseline system, the bias correction did not work well.) Instead, the cumulative sum of the inventory difference (CuSum ID) was used to determine if the diversion could be detected.

Figure 3 shows the results of the four diversion scenarios under the baseline system. In all cases, the plutonium CuSum ID was not able to detect material loss. The scatter in the data due to the uncertainty was high, and 8 kg of plutonium could easily be hidden. The error bars in the CuSum ID plots grew very quickly due to the 10% uncertainty on the estimation. Even if a more formal Page's Test were applied to this data, the errors are too high to provide any level of detectability. For the direct diversion scenarios, the process monitoring system was able to detect both the abrupt and protracted diversions (as indicated by the alarm message windows), but as expected, the substitution diversions were not detected. Therefore, the process monitoring balances do provide some level of protection.

Figure 4 shows the results of the same four diversion scenarios under the advanced NRTA system. With the much lower inventory measurement uncertainty, the Page's Test on the plutonium balance was able to detect all four diversion scenarios. Detection occurred before one half of a significant quantity was removed, so this system would provide adequate time for a security force to respond. Again, the process monitoring system was only able to detect direct material loss, but the detection was quicker than for the plutonium balance for these two cases. Combined, this new system is able to detect all four classes of diversion scenarios in a timely manner. There will be a limit to the detectability of very long protracted diversions, but this system is a significant improvement over current facilities.

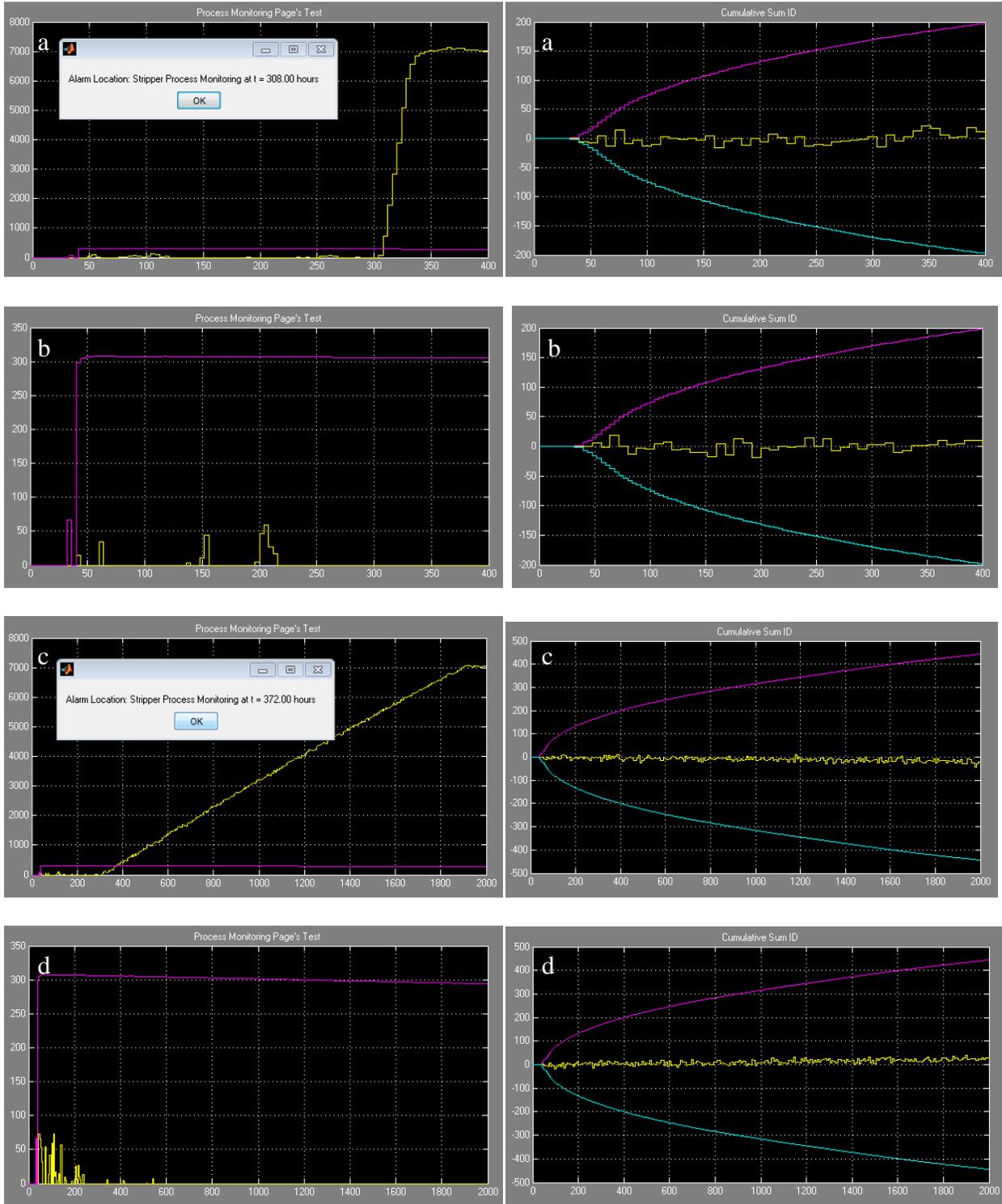


Figure 3: Baseline system results (a) abrupt direct diversion, (b) abrupt substitution diversion, (c) protracted direct diversion, (d) protracted substitution diversion

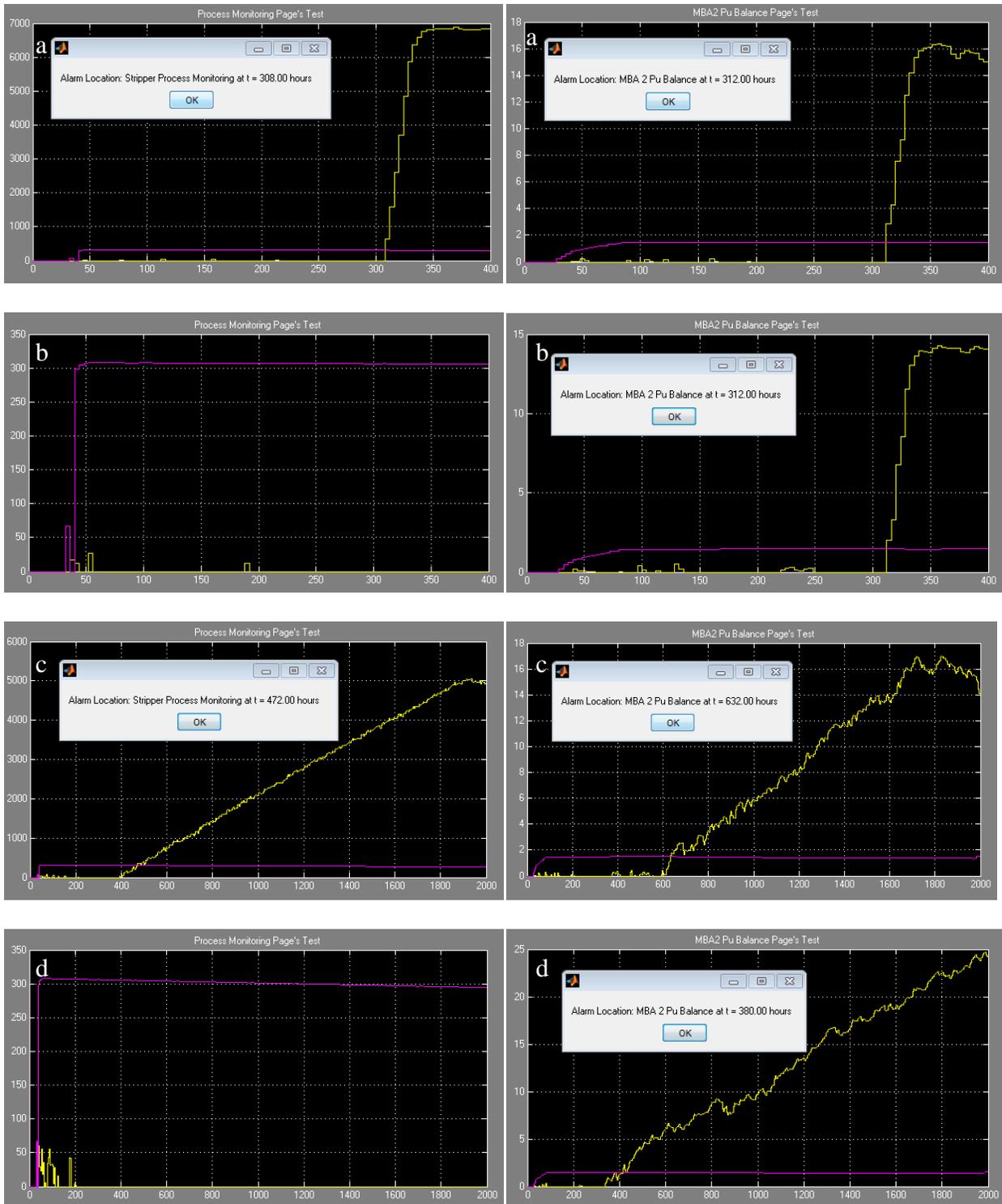


Figure 4: Advanced NRTA system results (a) abrupt direct diversion, (b) abrupt substitution diversion, (c) protracted direct diversion, (d) protracted substitution diversion

3.2 Diversion Scenario Discussion

The traditional accounting approach was not able to detect substitution diversions at all. Because the traditional approach only uses bulk process data for all of the internal tanks/vessels along with an assumption of actinide content from the accountability tank, a substitution diversion is not realized. In reality, a substitution diversion may be detected at plant flushout when the sum total of the inputs is balanced against the sum total of the outputs. However, material could be long gone by that point. The 10% estimation error makes it very difficult to detect any type of diversion using the plutonium balance alone.

The NRTA system is superior to the baseline system both because of the improvement in measurement uncertainty at the key plutonium tanks internal to MBA2, and because the measurements throughout the internal tanks provides protection against substitution diversions. The micro-fluidic technique, or some similar technology, will be required to allow for rapid sampling and analysis of these additional tanks. The UV-Vis-NIR technique, or similar, will be required to provide protection against substitution diversion scenarios in the areas processing smaller quantities of actinides.

In reality, the UV-Vis-NIR system cannot detect total plutonium, but rather the concentration in the process stream. For direct material loss, UV-Vis-NIR will not show any change to the concentration, but the bulk process monitoring balance will indicate the material loss. For a substitution diversion, the bulk material balance will not show any change, but UV-Vis-NIR will show a drop off in concentration. The two systems must be integrated for full protection. It also provides an advantage in that it tells the operator whether the material loss was abrupt or substituted. This system can also be used to determine other plant anomalies such as problems with the extraction steps or potential leaks in the system.

4.0 Integration of Solvent Extraction Codes

Past work examined the integration of the ORNL SEPHIS code with the SSPM in order to provide more realism of the contactor blocks in the plant model. However, this integration was limited to PUREX and was a rough coupling between the codes. Part of the goal of the effort this year was to examine if an alternative approach could be used. A SEPHIS version that was designed for UREX, TRUEX, and TALSPEAK was examined for more efficient integration. Also, the ANL AMUSE code was examined for integration.

4.1 SEPHIS

The past integration of a PUREX version of SEPHIS in the SSPM was limited in the number of elements tracked. It also required many layers of integration which led to insights for future improvements. The main drawback was that the code was written as an executable that had to be called outside of MATLAB. Direct coding into MATLAB m-files or as embedded functions is more ideal.

A recent version of SEPHIS was modified to include the UREX, FPEX, TRUEX, and TALSPEAK extraction steps [9]. It was also converted into Fortran-90 to make a more current code. The Fortran-90 source files (as developed at ORNL) were used as the starting point for integration into MATLAB. An open-source matlab code [12] was used to convert the Fortran-90 files to MATLAB m-files.

The conversion was not completely straight-forward and led to a number of errors. These were corrected by hand first with the goal to get the code to run. A larger effort was required to examine the m-files to make sure the code was producing useful results. Unfortunately, due to the limited amount of time spent on this conversion process, the analysis was not completed by the end of the year. The code did compile and run in MATLAB, but it will take more time to ensure that the code is producing correct results.

One of the insights of the past work with SEPHIS was that it is based on mixer-settler tanks. These tanks are much larger by volume than contactors, and past plant designs may have used lower flow rates. Therefore, the equilibrium times were on the order of hours. SEPHIS does take into account time dependence, which is a key benefit, but the volumes and flow rates will need to be examined for applicability to contactor banks. Since contactors have low volumes and since the plant design of the SSPM has high flow rates, the equilibrium time is likely to be on the order of minutes. More success was achieved with the AMUSE integration as described in the next section.

4.2 AMUSE

The AMUSE model for solvent extraction uses two components. SASPE (Stagewise Algorithm for Speciation and Partitioning Equilibrium) performs the calculation for distribution ratios and SASSE (Spreadsheet Algorithm for Stagewise Solvent Extraction) performs the mass balance [13]. Since the SSPM already performs a mass balance for each species, only the SASPE code

was needed for integration. Researchers at ANL developed a mex file (which is compatible with MATLAB) for SASPE in a version that is universal for a number of aqueous extraction processes. The extraction type, the important species and their concentrations, and the extractants need to be specified.

SASPE determines the distribution ratio for the aqueous and organic outputs for a particular centrifugal contactor. A contactor bank requires running SASPE a number of times for each contactor and continuously updating the concentration profiles between each stage. A mass balance is calculated for each stage as well.

The integration of SASPE in the Simulink model required the use of an embedded MATLAB code block that pulls the concentration data from the input streams into a contactor bank. The embedded code sets up the input variables needed for SASPE for that particular contactor bank. The inputs for the SASPE calculation change for the three different sections of the contactor bank, so three call functions are used. The distribution values are calculated, and then the mass balance across each contactor is used to calculate the output concentrations. The calculation takes into account the counter-flow of the aqueous and organic streams. This embedded code along with the three SASPE call function m-files are shown in the appendix.

Working in the time dependence was one of the more tricky parts of the integration with SASPE. SASPE calculates steady-state results and does not include any time dependence. In order to estimate time dependence, the calculation was performed in discrete time increments based on the contactor filling time. Initially during a startup condition, the concentrations of the species of interest are all zero between all stages. Once the dissolver solution reaches the eighth contactor (since the aqueous solution travels backwards from contactor 8 to 1), the first SASPE calculation will only produce an output for contactor 8. The next calculation will produce outputs for 8 and 7, and so on until an output comes out of the first contactor. The contactor volume was simply divided by the feed rate to determine the time increment.

This approximation for time dependence is not ideal, but probably a good approximation for this model. The time to steady-state is on the order of a few minutes due to the small contactor volumes and high flow rates. From the standpoint of safeguards, this equilibrium time is almost negligible, so as long as the calculation is in the right order of magnitude, it satisfies the needs of the SSPM. ANL is currently working on a time-dependent AMUSE model. In the future, it might be best to directly integrate this version instead.

4.2.1 AMUSE Results

The SASPE integration was set up for the UREX contactor bank to test the results. Uranium, technetium, and plutonium were tracked through the calculations. A 24 stage contactor bank was assumed. The aqueous feed from the adjustment tank enters stage 8 and leaves stage 1 as the raffinate. The scrub enters stage 14, is added to the aqueous feed at stage 8 and leaves stage 1. The strip enters stage 24 and leaves stage 15 as the U/Tc product. The organic TBP solvent flows countercurrent and enters stage 1 and leaves stage 24 as cleaned solvent for recycle.

The inputs into the SASPE calculation from the model included the aqueous flow, scrub flow, strip flow, and solvent flow rates; and the concentrations of uranium, technetium, and plutonium. The calculation is set up so that these values can change from the model, but the results will be directly calculated with a time step of 0.01 hours.

The following 4 figures show the results. All of the following figures are zoomed in the x-axis to the startup condition to readily show the transient nature of the response. It takes about 30 hours for material to make it to the UREX contactors in the SSPM. Figure 5 shows the uranium mass flow rates for the feed, raffinate, and strip product. As expected, the uranium ends up in the strip solution, indicating that the SASPE model and calculation are behaving as expected. The transient nature shows the buildup in the contactor bank with time—since this is mainly driven by the contactor volumes, steady-state is reached on the order of minutes.

Figure 6 shows the plutonium mass flow rates. In this case, the plutonium ends up in the raffinate stream. Figure 7 shows the technetium mass flow rate. Similarly to the uranium, the technetium ends up in the strip product. In all cases, the output from the contactor banks balances completely with the input after steady-state is achieved, which validates the mass balance calculations. Figure 8 shows the uranium, plutonium, and technetium mass flow rates coming out with the solvent, and these values are very small as expected.

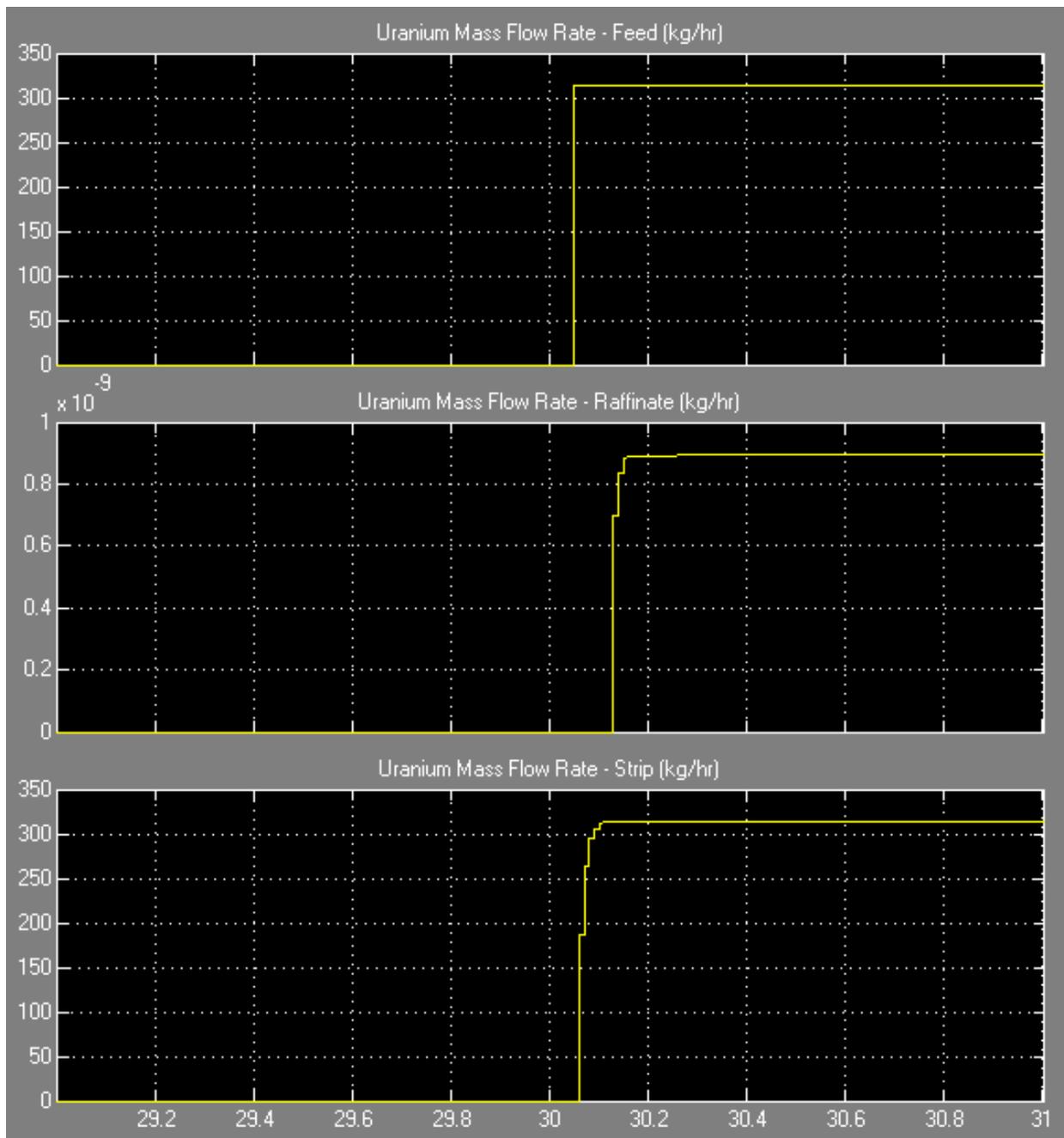


Figure 5: Uranium separation in UREX contactors using SASPE

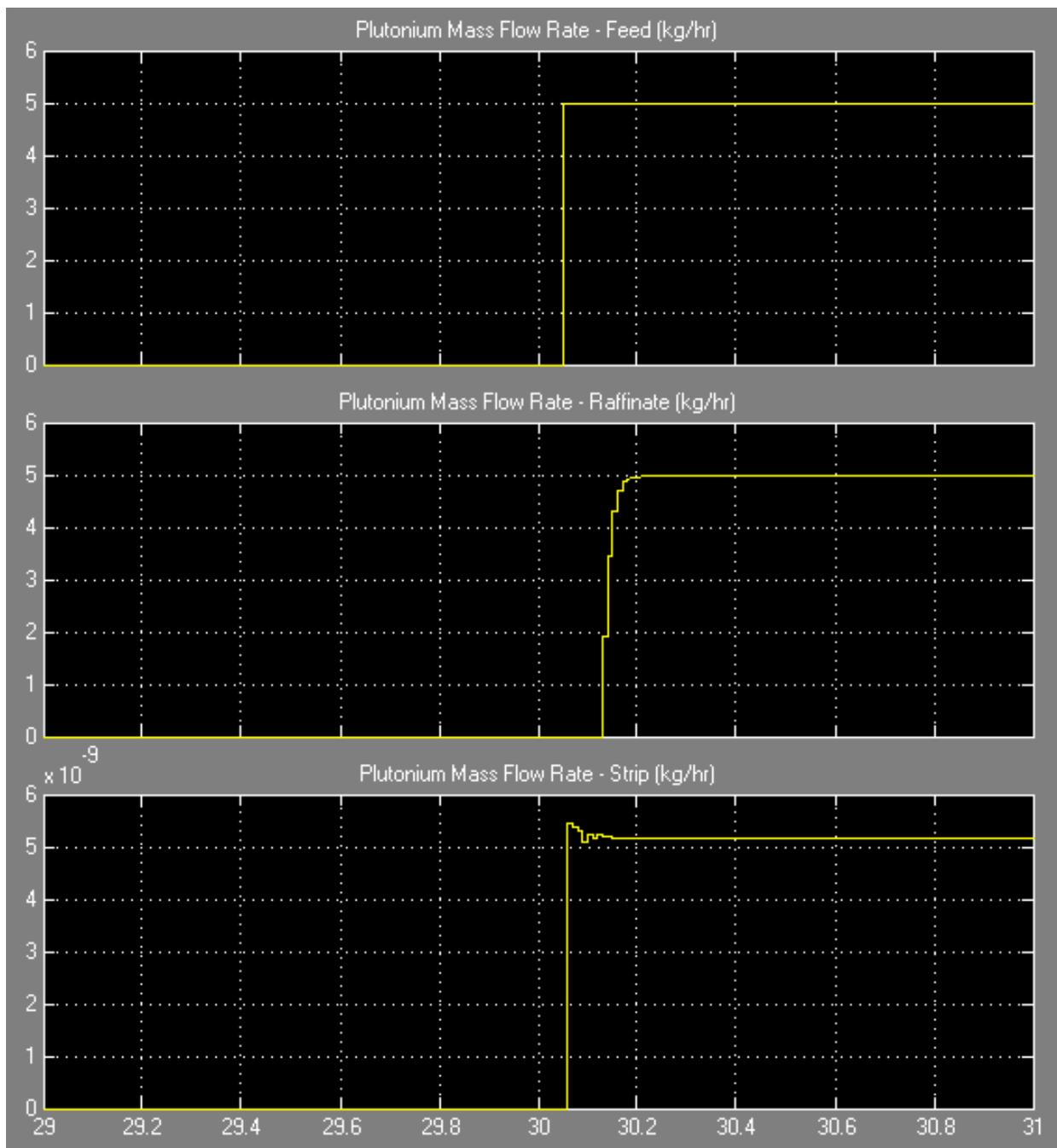


Figure 6: Plutonium separation in UREX contactors using SASPE

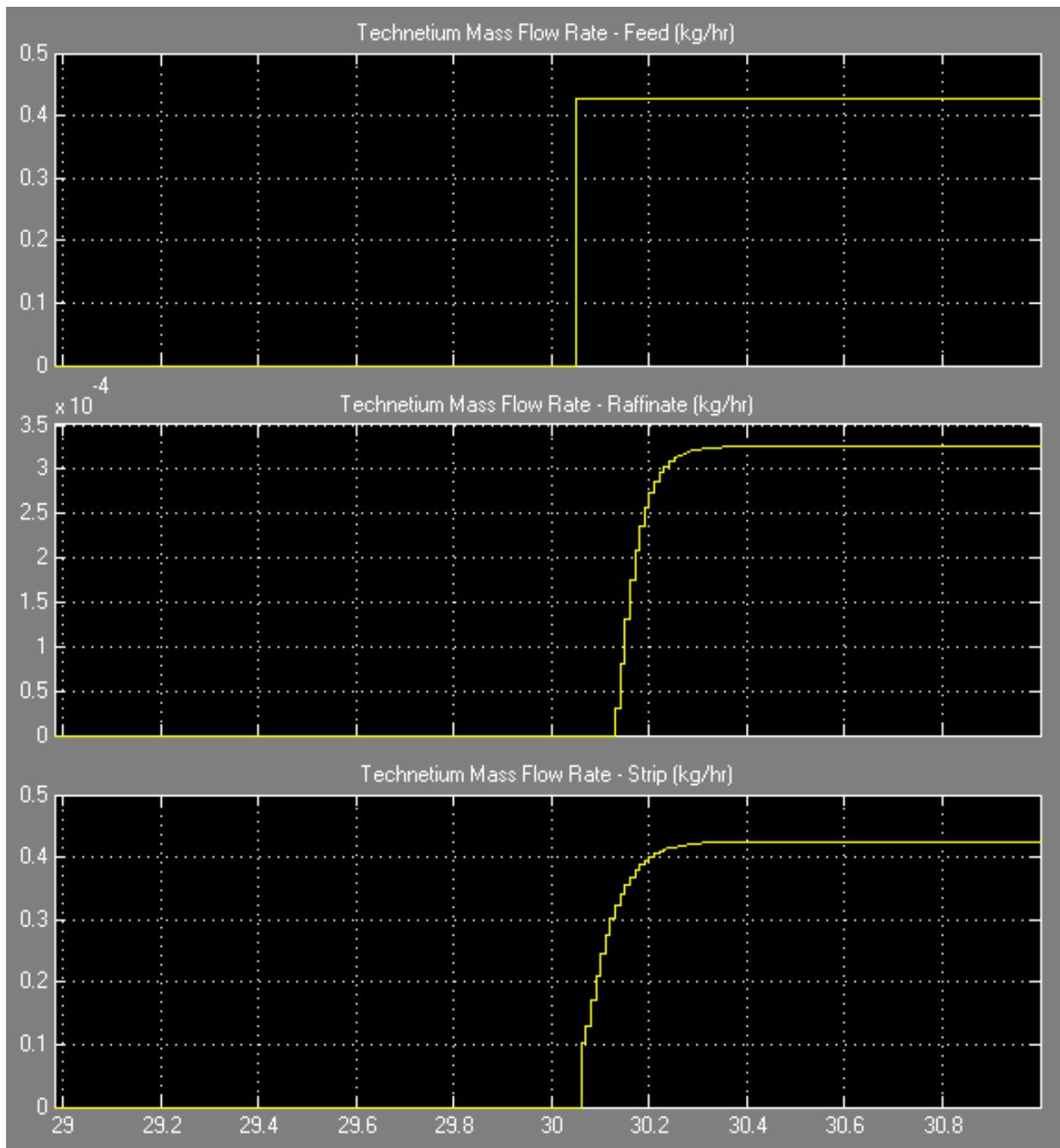


Figure 7: Technetium separation in UREX contactors using SASPE

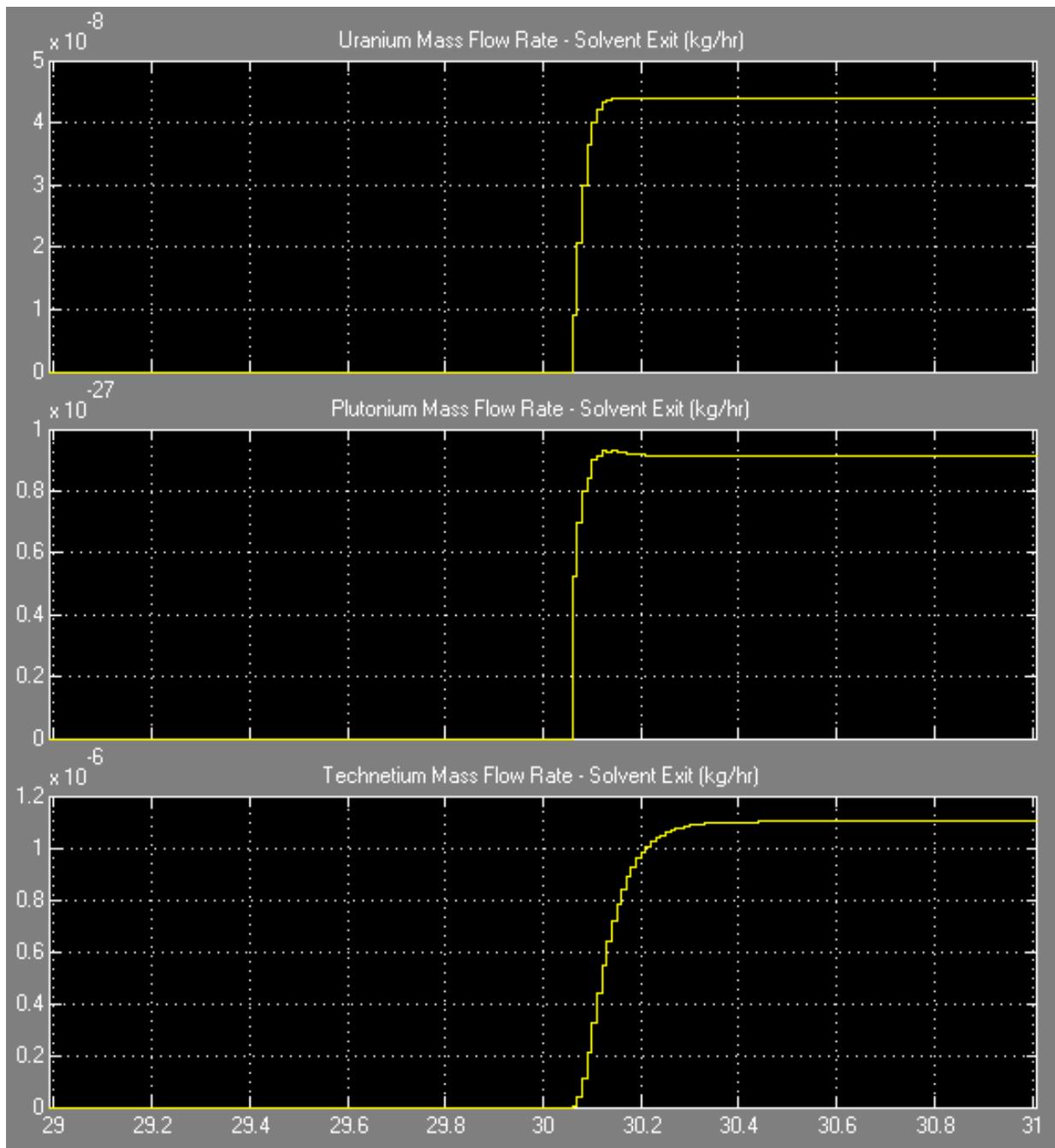


Figure 8: Solvent output mass flow rates using SASPE

4.2.2 AMUSE Discussion

The AMUSE integration was successful with the one uncertainty about time dependence. One of the key advantages of the SASPE mex file is that it can be used for any extraction process in the SSPM, so integration with the other extraction steps should be straight-forward. The modeling allows the user to know how the actinides are distributed in the contactor bank.

A disadvantage of the SASPE integration is that it slowed the model down considerably. Since the time dependence was tied directly to the calculation update time, changing it to make the model run more quickly will create a false time dependence. On the other hand, this work showed that the equilibrium times for the contactor banks were small (on the order of minutes) due to the small contactor volumes and large flow rates. From the safeguards perspective, these transient times are almost negligible in the model. A work-around would be to calculate the equilibrium directly without time dependence and run SASPE less often.

5.0 Conclusion

NRTA can be achieved for a vast majority of an aqueous reprocessing plant by utilizing additional sampling at key plutonium tanks and by utilizing simple spectroscopic or other NDA measurements at areas processing small quantities of plutonium. The micro-fluidic sampling work at INL can speed up the measurement time, making it possible to sample in more locations for the same operating cost. The UV-Vis-NIR spectroscopic techniques being investigated at PNNL provide a simple and effective method for measuring areas processing small actinide quantities. Coupled with process monitoring measurements that must be present in the plant anyway, such an integrated system is unlikely to burden the operator more than with existing monitoring systems. Diversion scenarios have shown that such a system is able to detect various diversion events before half of a significant quantity is removed, as opposed to many months later during a plant flushout.

The integration of solvent extraction codes was examined to add robustness to the model. The AMUSE integration (through integration with SASPE) was completed with a modest amount of time. Initial results show expected separation fractions, but time dependence is not inherent in the model. A rough workaround was used for time-dependence. However, it was found that equilibrium times were on the order of minutes, so transients are unlikely to affect diversion scenarios which are typically on longer time scales. The integration with SEPHIS will take more time to complete, and may be worthwhile to include time dependence. The initial results with SASPE were more encouraging and easier to replicate throughout the plant for a number of species.

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Appendix

Embedded MATLAB function for calling SASPE:

```
function
[raffuout, raffpuout, rafftcout, stripuout, strippuout, striptcout, tbpuout, tbppuou
t, tbptcout] = fcn(aflow, oflow, uconc, puconc, tcconc)
% This block supports the Embedded MATLAB subset.
% The purpose of this embedded function is to call the Solvent Extraction
Unit m-file.
% This block was specifically written for UREX, but it does not include the
% scrub step

eml.extrinsic('callSASPE');
eml.extrinsic('callSASPE2');
eml.extrinsic('callSASPE3');
eml.extrinsic('disp');

dVals=[0;0;0;0;0]; %must be initialized first

persistent aquconc;
persistent aqpuconc;
persistent aqtcconc;
persistent oruconc;
persistent orpuconc;
persistent ortcconc;
persistent utemp;
persistent putemp;
persistent tctemp;
persistent tem9u;
persistent tem9pu;
persistent tem9tc;

if isempty(oruconc)
    aquconc=zeros(1,25);
    aqpuconc=zeros(1,25);
    aqtcconc=zeros(1,25);
    oruconc=zeros(1,25);
    orpuconc=zeros(1,25);
    ortcconc=zeros(1,25);
    utemp=zeros(1,24);
    putemp=zeros(1,24);
    tctemp=zeros(1,24);
    tem9u=0;
    tem9pu=0;
    tem9tc=0;
end

aquconc(9)=uconc/1.5+tem9u/3; %adds in the aq stream coming from scrub
aqpuconc(9)=puconc/1.5+tem9pu/3;
aqtcconc(9)=tcconc/1.5+tem9tc/3;
aquconc(15)=0; %reset to zero since strip is pulled out here
aqpuconc(15)=0;
aqtcconc(15)=0;
```

```

aquconcc(25)=0;
aqqpuconcc(25)=0;
aqtccconcc(25)=0;

for i=1:8 % three scripts for the 3 different sections of the bank
[dVals] = callSASPE(aquconcc(i+1), aqqpuconcc(i+1), aqtccconcc(i+1), oruconcc(i),
orpuconcc(i), ortccconcc(i));
utemp(i)=dVals(2);
putemp(i)=dVals(3);
tctemp(i)=dVals(4);
end

for i=9:14
[dVals] = callSASPE2(aquconcc(i+1), aqqpuconcc(i+1), aqtccconcc(i+1), oruconcc(i),
orpuconcc(i), ortccconcc(i));
utemp(i)=dVals(2);
putemp(i)=dVals(3);
tctemp(i)=dVals(4);
end

for i=15:24
[dVals] = callSASPE3(aquconcc(i+1), aqqpuconcc(i+1), aqtccconcc(i+1), oruconcc(i),
orpuconcc(i), ortccconcc(i));
utemp(i)=dVals(2);
putemp(i)=dVals(3);
tctemp(i)=dVals(4);
end

% Convention is that conc(i) is the concentration between contactors
% i-1 and i. Also aqueous flows backwards so output of contactor 7 is
% aqueous index 7 and organic index 8.

for i=1:8
aquconcc(i)=(aflow*1.5*aquconcc(i+1)+oflow*oruconcc(i))/(aflow*1.5+utemp(i)*oflo
w);
oruconcc(i+1)=utemp(i)*aquconcc(i);
aqqpuconcc(i)=(aflow*1.5*aqqpuconcc(i+1)+oflow*orpuconcc(i))/(aflow*1.5+putemp(i)*
oflow);
orpuconcc(i+1)=putemp(i)*aqqpuconcc(i);
aqtccconcc(i)=(aflow*1.5*aqtccconcc(i+1)+oflow*ortccconcc(i))/(aflow*1.5+tctemp(i)*
oflow);
ortccconcc(i+1)=tctemp(i)*aqtccconcc(i);
end

for i=9:14
aquconcc(i)=(aflow/2*aquconcc(i+1)+oflow*oruconcc(i))/(aflow/2+utemp(i)*oflow);
oruconcc(i+1)=utemp(i)*aquconcc(i);
aqqpuconcc(i)=(aflow/2*aqqpuconcc(i+1)+oflow*orpuconcc(i))/(aflow/2+putemp(i)*oflo
w);
orpuconcc(i+1)=putemp(i)*aqqpuconcc(i);
aqtccconcc(i)=(aflow/2*aqtccconcc(i+1)+oflow*ortccconcc(i))/(aflow/2+tctemp(i)*oflo
w);
ortccconcc(i+1)=tctemp(i)*aqtccconcc(i);
end

```

```

for i=15:24
aquconc(i)=(aflow*10*aquconc(i+1)+oflow*oruconc(i))/(aflow*10+utemp(i)*oflow)
;
oruconc(i+1)=utemp(i)*aquconc(i);
aqpucconc(i)=(aflow*10*aqpucconc(i+1)+oflow*orpuconc(i))/(aflow*10+putemp(i)*oflow);
orpuconc(i+1)=putemp(i)*aqpucconc(i);
aqtccconc(i)=(aflow*10*aqtccconc(i+1)+oflow*ortccconc(i))/(aflow*10+tctemp(i)*oflow);
ortccconc(i+1)=tctemp(i)*aqtccconc(i);
end

tem9u=aquconc(9); % required for adding above
tem9pu=aqpucconc(9);
tem9tc=aqtccconc(9);

raffuout=aquconc(1);
raffpuout=aqpucconc(1);
rafftcout=aqtccconc(1);
stripuout=aquconc(15);
strippuout=aqpucconc(15);
striptcout=aqtccconc(15);
tbpuout=oruconc(25);
tbppuout=orpuconc(25);
tbptcout=ortccconc(25);

end

```

callSASPE.m file (calculates D-ratios for stages 1-8):

```

function [dVals] = callSASPE(auc, apuc, atcc, ouc, opuc, otcc)

comps = {'H_(1)', 'UO2_(2)', 'Pu_(4)', 'TcO4_(-1)', 'CH3CONHOH_(0)'};
proc = 'UREX';
aConc = [0.87, auc, apuc, atcc, 0.33];
oConc = [0.0, ouc, opuc, otcc, 0.0];
vRatio = 2.0;
temp = 25.0;
exTBP = 1.3;
exCMPO = 0.0;
exCCD = 0.0;
exPEG = 0.0;
exCROWN = 0.0;

[dVals] =
SASPE_mx_140(comps, proc, aConc, oConc, vRatio, temp, exTBP, exCMPO, exCCD, exPEG, exCROWN);

end

```

callSASPE2.m file (calculates D-ratios for stages 9-14):

```
function [dVals] = callSASPE2(auc, apuc, atcc, ouc, opuc, otcc)

comps    = {'H_(1)', 'UO2_(2)', 'Pu_(4)', 'TcO4_(-1)', 'CH3CONHOH_(0)'};
proc     = 'UREX';
aConc    = [0.3, auc, apuc, atcc, 0.5];
oConc    = [0.0, ouc, opuc, otcc, 0.0];
vRatio   = 6.2;
temp     = 25.0;
exTBP    = 1.3;
exCMPO   = 0.0;
exCCD    = 0.0;
exPEG    = 0.0;
exCROWN  = 0.0;

[dVals] =
SASPE_mx_140(comps,proc,aConc,oConc,vRatio,temp,exTBP,exCMPO,exCCD,exPEG,exCR
OWN);

End
```

callSASPE3.m file (calculates D-ratios for stages 15-24):

```
function [dVals] = callSASPE3(auc, apuc, atcc, ouc, opuc, otcc)

comps    = {'H_(1)', 'UO2_(2)', 'Pu_(4)', 'TcO4_(-1)', 'CH3CONHOH_(0)'};
proc     = 'UREX';
aConc    = [0.1, auc, apuc, atcc, 0.0];
oConc    = [0.0, ouc, opuc, otcc, 0.0];
vRatio   = 0.31;
temp     = 25.0;
exTBP    = 1.3;
exCMPO   = 0.0;
exCCD    = 0.0;
exPEG    = 0.0;
exCROWN  = 0.0;

[dVals] =
SASPE_mx_140(comps,proc,aConc,oConc,vRatio,temp,exTBP,exCMPO,exCCD,exPEG,exCR
OWN);

end
```

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