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The Effect of Phase Disengagement on Criticality Safety Calculations for Aqueous Separations

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INTRODUCTION

Counter current two-phase solvent extraction, for nuclear or non-nuclear applications, relies on the ability of the two phases to rapidly disengage after the extraction step. This facilitates the process moving on to the next extraction step with the two phases flowing in different directions. In the HM-process used at the H-Canyon facility at the Savannah River Site, the separation phases are an aqueous, higher density solution of uranyl nitrate in nitric acid and an organic lower density solution of tributyl phosphate in normal paraffin diluent. These two phases flow in opposite directions through banks of mixer settler stages. They are mechanically mixed in the mixing section and then expected to separate in the long settling section such that at the end organic may exit near the top of the settling section and aqueous near the bottom.

Previous criticality safety analyses (Ref. 1-3) analyzed the chemical composition of each individual mixing and settling section. These analyses modeled the settling section as two distinct compositions directly adjacent to the mixing section composition, i.e. instantaneous phase disengagement was assumed. This assumption is believed to be conservative for criticality safety analyses, resulting in a higher calculated neutron multiplication factor. That assumption is scrutinized in this work through the simulation of delayed phase disengagement.

SEPHIS-ACM is to predict the chemistry composition of the aqueous and organic phases in each mixing and each settling section. KENO-VI in SCALE 6.1 is used to simulate the neutron multiplication conditions.

DESCRIPTION OF THE WORK

Process Description

The Savannah River Site H-Canyon facility currently operates a uranium separation and purification process known as HM-Process which is derived from Purex process chemistry and utilizes a low TBP concentration due to the processing of highly enriched uranium (Ref. 1,4). Two cycles of processing are used: the first cycle separates uranium from fission products, transuranics, and the fuel matrix. The second cycle purifies the uranium. The first cycle is modeled for this work consists of a 16-stage mixer settler (A-Bank), a 16-stage mixer settler (B-Bank), and a 12-stage mixer settler (C-Bank). Fig. 1 illustrates the concept of phase disengagement within a stage.

Upon exiting the mixing section the two phases, which are nearly immiscible by nature (there is trace amount of carryover from one phase to the other) begin to settle and separate or disengage. The time it takes for this to occur depends upon the density, viscosity, and surface tension of the two phases. These parameters are affected not only by the feed solution that is sent to the process, but also by the degree of degradation of the solvent. Ref. 5 experimentally quantified the disengaging time for HM-Process solution in both fresh solvent and in solvent taken from the process hold tanks which had been degraded by the exposure to fission products via both chemical contaminants and by radiolytic decomposition of the TBP. The fresh solvent disengaged from the aqueous phase in 29 seconds (without a reductant in the aqueous phase) and the degraded solvent in 43 seconds (without a reductant in the aqueous phase). Presence of a ferrous sulfamate reductant in the aqueous phase increased disengaging time by 7 to 16 seconds. The cycle analyzed in this work did not have a reductant added.

Flow rates of the two phases are one of the parameters controlled in the process and are known values. The depth to which a slug of mixed phases penetrates into the settling section can be approximated by using the disengaging time, the cross-sectional area of the settling section, the density of the combined phases, and the flow rate. The density is available from SEPHIS-ACM calculations and the cross-sectional area from engineering drawings.

Modeling Phase Disengagement

Four degrees of phase disengagement are simulated in this work (Fig. 2). Instantaneous phase disengagement is simulated as was done in referenced work (Ref. 1-3) for consistency. Phase disengagement with clean and degraded solvent disengaging times was also simulated. Finally, incomplete disengagement was simulated wherein the mixed phases extended to the end of the settling section.

Geometrically, the mixed phase is modeled as a triangular wedge penetrating into the settling section. The base of the wedge is the height of the exit of the mixing section, which is approximately the height of the portion with the impeller (Fig. 1). The aqueous inlet portion does not exit to the settling section. Mixed solution exits the mixing section through a set of baffles (not simulated) and that opening is approximately the entire height of the mixing section. The width spans the width of the settling section, this is also approximately the exit width of the mixing section. The length of the wedge varied based on disengaging time. The length of the wedge is determined by the following formula:

$$L = \frac{V_{org} + V_{aq}}{A} t \tag{1}$$

Where L is the length of the wedge to be modeled, and t is t disengaging time. V_{org} is the volumetric flow rate of the organic stream and V_{aq} is the volumetric flowrate of the aqueous stream both in cubic centimeters per second. A is the area of the outlet of the mixing section, i.e. the width times the height of the mixing section. A is 258.1 cm^2 for the 16-stage mixer-settler design and 465.4 cm^2 for the 12-stage mixer-settler design. For the clean and degraded disengaging times 29 and 43 seconds were used as t. For the incomplete disengagement model, t was adjusted for each stage such that the length of the wedge was equal to the length of the settling section. Depending on the bank and stage in question, this value of t was between 52 and 207 seconds.

Equation 1 represents a very simplified model of the interspersed phases. More complex models using computational fluid dynamics, drop-rise simulation, etc. could be made. However, as will be shown later in this work, the impact on neutron multiplication indicated such time-consuming efforts were not warranted.

Chemically, the mixed phase is modeled as the same mixture that is present in the mixing section. By nature, it is effectively at a density less than that of the aqueous phase but greater than that of the organic phase. The total fissile mass of the system is preserved in all cases regardless of the degree of phase disengagement.

Selection of Process Conditions to Simulate

Two of the existing process upsets postulated to result in a criticality event was chosen for this work. Of the process upsets available (stream flow rates, uranium concentration, acid concentration, TBP concentration, and temperature) acid concentration was chosen. Concentration or dilution of acid entering the process is expected to cause the most significant and most rapid upset conditions.

The First Uranium Cycle is chosen for this work. This is the partitioning part of the process. A clarified nitric acid solution of uranium, aluminum, fission products, and trace amounts of transuranics is fed to a solvent extraction process. The process consists of two 16-stage mixer-settlers (A and B banks) and one 12-stage mixer-settler (C-Bank). Dissolved fission products are rejected to waste in the A-Bank and neptunium and plutonium are rejected in the B-Bank. The product stream of uranyl nitrate solution leaves the C-Bank destined for a second cycle of purification before storage and downblend to low enriched uranium.

The upsets chosen are those in the acid streams feeding the A-Bank and B-Bank. Table I indicates the fixed process parameters and the ranges of the two streams upset. Acid is given in molarity of nitric acid and the solvent is a 7.5 vol.% tributyl phosphate in a normal-paraffin diluent. The upsets are analyzed separately pursuant to the Double Contingency Principle; multiple concurrent upsets are not analyzed. The process is controlled by operator actions and by active engineered interlocks. The normal condition is the setpoints and specified chemistry of the streams. The credible abnormal is the terminal points of the allowable range. Once a process parameter is outside the range it must either be immediately corrected or the process will shut down by either operator action or engineered safety interlocks. Chemicals found out of specification before the run would not be used at all. Clarification, the process would not run for example if the feed uranium concentration was too high. The values in Table I simulate a flowsheet allowing up to 73 wt.% enriched uranium, thus the uranium content will be modeled as 73 wt.% U-235 and 27 wt.% U-238. U-234 and U-236 content are neglected. Trace Np is modeled as 99.9 wt.% Np-237, 0.1 wt.% Np-236 and trace Pu is modeled as 93.6 wt.% Pu-239, 6 wt.% Pu-240, and 0.4 wt.% Pu-241.

Simulation Tools

Process chemistry conditions are simulated for each stage of each bank using the SEPHIS-ACM model (Ref. 6). SEPHIS-ACM is a recent update to the SEPHIS program which has been used for flowsheet development of Purex type processes for more than 40 years. Among the ACM version improvements include tracking of 36 unique chemical species (up from 6 ions in older versions), a detailed graphical user interface, improved chemistry simulation, neptunium distribution correlation, iron and aluminum distribution correlation, and solvent recycle simulation. The ACM version also provides stage-wise aqueous and organic stream composition in atom density format which can be directly used in SCALE.

Neutron multiplication simulation is carried out in the KENO-VI model of SCALE 6.1 using the ENDF-VII 238-group cross section library. All chemical species tracked in SEPHIS-ACM are specifically modeled in SCALE, broken down into isotopics defined by natural abundance or assumed distribution. The SCALE model simulates three compositions per stage – a mixed solution in the mixer section and interspersed region, an organic phase in the settling section, and an aqueous phase in the settling section. The stainless steel plates that make up the mixer settler are explicitly modeled, however the baffles, flow weirs, impeller, and flow inlets are not. This simplification conservatively adds more fissile solution to the system than would be present due to the space occupied by these components. The bank is surrounded by an air pocket indicative of actual configuration and is then given a reflective boundary condition beyond that.

Determination of k_{SAFE}

An in-house validation for the SCALE 6.1 KENO-VI code for HEU solution systems was performed. A conservative k_{SAFE} assumed for this work was 0.9564.

RESULTS AND FUTURE WORK

Perturbation of the A-Bank acid stream had significant impacts on the uranium and acid distributions in A-Bank, somewhat muted impacts in B-Bank, and negligible changes in C-Bank. Perturbation of the B-Bank acid stream had significant impacts on the uranium and acid distributions in the B-Bank, and appreciable changes to the C-Bank only in the extreme of the perturbation range.

Table II and Table III show the bank eigenvalues for the analyzed cases for the A-Bank and B-Bank acid upsets, respectively. For practical application (i.e. to two significant figures), the bank multiplication factor is unchanged by phase disengagement delays. Beyond the second significant figure, the bank multiplication appears to be reduced by the presence of an interspersed phase. In the B-Bank upset (Table III), in many cases the reduction is greater than the uncertainty in the calculation, implying a real effect.

The one exception appears to be the extreme low acid case in the A-Bank upset. Generally, the solutions are highly over moderated. However, in the 0.2M case for A-Bank, uranium concentration in the aqueous phase is such that in several stages the aqueous composition is closer to optimal moderation, which is why multiplication jumps from ~0.6 to ~0.9 between 0.5M and 0.2M acid cases, respectively. The organic phase is still well over moderated but, when mixed with the aqueous phase, the interspersed phases are also nearer to optimal moderation than in other higher acid cases. The result is unique to this case and the condition is not credible for the A-Bank in the HM-Process. Sampling would detect 0.2 M acid before it is fed to the system and there is insufficient head tank volume to dilute acceptable molar acid to 0.2 M acid prior to operation. There are also engineered interlocks that shut down the system if the A-Bank acid drops below 3.1 M during operation.

For practical considerations, the impact of delays in phase disengagement appear typically to have a negligible effect to one of mildly reducing the multiplication. Therefore, assuming instantaneous phase disengagement appears to be acceptable, and arguably mildly conservative.

Future work will examine other process upsets in the First and Second Uranium Cycles to determine if criticality is or is not credible given the chemistry conditions. However, in light of the results of this work, further phase disengagement studies do not appear necessary from a criticality safety aspect. The instantaneous disengagement assumption is sufficient for practical analysis.

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Table I. Process Parameters

Parameter	**Description**
Process feed stream	6.1 L/min, 4.5 g U/L, 5.00 M
A-Bank acid stream	1.4 L/min, nominally 3.70 M, modeled from 0.20 to 8.00 M
A-Bank solvent stream	10.7 L/min
B-Bank acid stream	6.3 L/min, nominally 1.55 M, modeled from 0.20 to 4.50 M
B-Bank solvent stream	18.7 L/min
C-Bank acid stream	7.6 L/min, 0.02 M

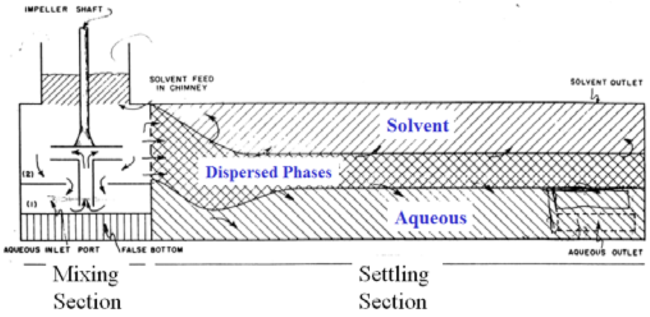

Fig. 1. Conceptual illustration of phase disengagement, lateral view of a mixer settler stage.

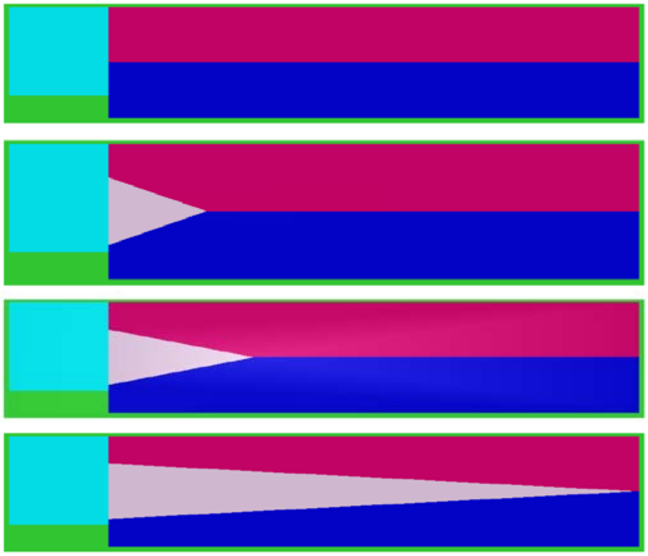

Fig. 2. Simulated phase disengagements in Stage 1 of the A-Bank. From top to bottom: instant, clean solvent, degraded solvent, and incomplete disengagement.

Table II. Bank Multiplication Factors for an Upset in A-Bank Acid Concentration

Acidity (M)	Instantaneous		Clean Solvent (29 second)		Degraded Solvent (43 second)		Incomplete	
	k-eff	Uncertainty	k-eff	Uncertainty	k-eff	Uncertainty	k-eff	Uncertainty
0.20	0.907480	0.000520	0.910480	0.000730	0.910300	0.000650	0.922520	0.000630
0.50	0.609790	0.000430	0.609770	0.000380	0.609150	0.000460	0.603800	0.000440
1.00	0.262170	0.000140	0.262510	0.000180	0.262160	0.000170	0.260150	0.000150
1.50	0.161529	0.000095	0.161081	0.000094	0.160982	0.000098	0.160679	0.000090
2.00	0.135439	0.000098	0.135168	0.000084	0.134869	0.000077	0.132207	0.000078
2.50	0.127149	0.000076	0.126571	0.000083	0.126199	0.000086	0.122386	0.000079
3.10	0.123266	0.000071	0.122659	0.000079	0.122031	0.000078	0.117537	0.000072
3.70	0.121394	0.000086	0.120782	0.000085	0.120314	0.000079	0.115501	0.000078
4.07	0.120930	0.000085	0.120397	0.000076	0.119809	0.000085	0.114888	0.000073
4.50	0.120651	0.000086	0.119965	0.000084	0.119746	0.000086	0.114689	0.000075
5.00	0.120779	0.000094	0.120179	0.000087	0.119690	0.000080	0.114652	0.000076
5.50	0.121224	0.000077	0.120316	0.000085	0.120022	0.000070	0.114797	0.000071
6.00	0.121800	0.000079	0.121050	0.000071	0.120624	0.000077	0.115627	0.000088
7.00	0.123660	0.000081	0.123000	0.000073	0.122479	0.000076	0.117428	0.000068
8.00	0.125975	0.000090	0.125388	0.000082	0.125036	0.000086	0.120072	0.000069

Table III. Bank Multiplication Factors for an Upset in B-Bank Acid Concentration

Acidity (M)	Instantaneous		Clean Solvent (29 second)		Degraded Solvent (43 second)		Incomplete	
	k-eff	Uncertainty	k-eff	Uncertainty	k-eff	Uncertainty	k-eff	Uncertainty
0.20	0.463640	0.000310	0.460080	0.000290	0.456750	0.000340	0.453440	0.000270
0.40	0.652870	0.000380	0.652560	0.000540	0.652550	0.000390	0.649510	0.000400
0.60	0.408440	0.000250	0.407070	0.000270	0.405010	0.000290	0.400210	0.000270
0.80	0.216180	0.000160	0.214570	0.000130	0.213840	0.000150	0.211410	0.000120
1.00	0.132635	0.000073	0.132273	0.000091	0.131841	0.000079	0.131089	0.000081
1.25	0.086348	0.000050	0.086468	0.000049	0.086403	0.000048	0.086271	0.000046
1.50	0.067392	0.000035	0.067341	0.000036	0.067300	0.000036	0.067228	0.000054
1.55	0.065210	0.000039	0.065008	0.000046	0.064956	0.000035	0.064762	0.000040
1.60	0.063299	0.000039	0.063059	0.000033	0.062943	0.000040	0.062685	0.000032
1.80	0.057662	0.000031	0.057273	0.000041	0.057071	0.000030	0.056551	0.000030
2.00	0.054194	0.000036	0.053812	0.000029	0.053536	0.000030	0.052664	0.000036
2.50	0.049957	0.000037	0.049431	0.000033	0.048850	0.000036	0.047750	0.000027
3.00	0.048129	0.000034	0.047499	0.000046	0.046906	0.000030	0.045609	0.000026
3.50	0.047277	0.000031	0.046551	0.000034	0.045955	0.000028	0.044569	0.000030
4.00	0.046817	0.000031	0.046173	0.000032	0.045401	0.000033	0.044094	0.000029
4.50	0.046691	0.000029	0.045960	0.000030	0.045365	0.000025	0.043867	0.000025