ANL-LTR-NGCSSX-001

Initial Flowsheet for the Next Generation CSSX Process in the MCU. Report for FY2010

Chemical Sciences and Engineering Division

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The Next Generation Caustic Side Solvent Extraction (NG-CSSX) flowsheet is being developed in a collaboration among Oak Ridge National Laboratory (ORNL), Argonne National Laboratory (ANL), and Savannah River National Laboratory (SRNL) for the Modular CSSX Unit (MCU) facility at the Savannah River Site (SRS). This work is being done in cooperation with two groups at SRS (my principal contacts there are Mark Geeting, SRR, and Sam Fink, SRNL) and with one group at Oak Ridge National Laboratory (my principal contact there is Bruce Moyer). The evaluation of the various NG-CSSX solvent candidates was done using the SASSE (Spreadsheet Algorithm for Stagewise Solvent Extraction) program with cesium distribution coefficient (D_{Cs}) values supplied by ORNL. The final optimized solvent composition has 50 mM MaxCalix as the extractant. Using the SASSE spreadsheet to calculate the Cs decontamination factor (D.F.) as well as ROTOR program to evaluate contactor rotor throughputs, an initial flowsheet was developed for the optimized NG-CSSX solvent. The current CSSX flowsheet for the MCU at its maximum salt feed flow rate is shown in Fig. 1. The initial NG-CSSX flowsheet for the MCU at its targeted maximum salt feed flow rate is shown in Fig. 2. Comparing these two flowsheets, one can see that, for the NG-CSSX flowsheet, the maximum flow rate of the salt feed (DF) is increased 41%, from 8.5 gpm to 12.0 gpm. At the same time, because of the high concentration of the MaxCalix extractant in the NG-CSSX solvent, the Cs decontamination factor (D.F.) for the aqueous effluent from the extraction section (DW, the aqueous raffinate) is increased from 12 to 40,000. As required, the Cs concentration factor (C.F.) for the aqueous effluent from the strip section (EW) is maintained at 15.

Because the development of the optimum NG-CSSX solvent has been ongoing throughout the second half of FY10, some physical properties including solvent density,

effect of temperature on D_{Cs} and the dimensionless dispersion number (N_{Di}) were estimated based on results from the earlier solvents characterized during the development of the CSSX and NG-CSSX processes. When the complete set of physical properties for the optimum NG-CSSX solvent are measured at ORNL, an optimum NG-CSSX flowsheet will be developed at ANL. Based on the data that has been obtained at ORNL for the earlier NG-CSSX solvent candidates, it is expected that the NG-CSSX flowsheet shown in Fig. 2 will be close to the optimum flowsheet.

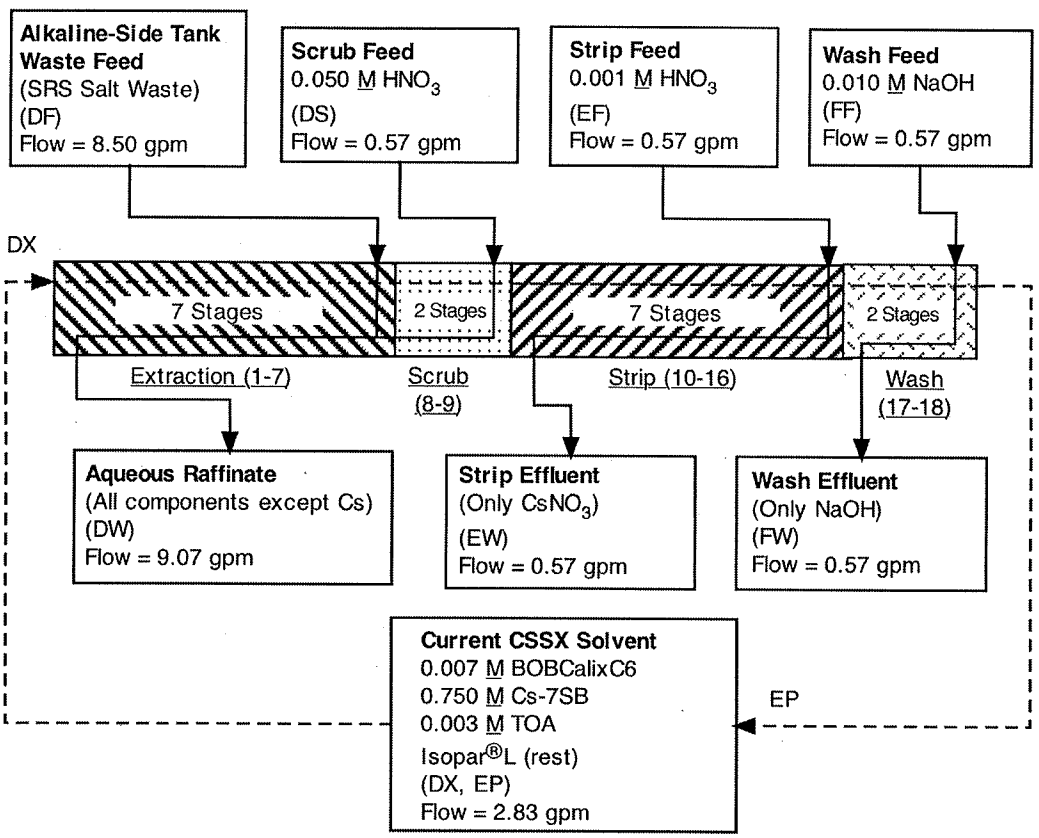

Fig. 1. Current CSSX Flowsheet for MCU Showing Maximum Flow Rates

The NG-CSSX flowsheet shown in Fig. 2 would have the same O/A flow ratios as the current CSSX flowsheet shown in Fig. 1 if the solvent (DX) flow rate were 4 gpm. However, this solvent flow rate is too high in the CINC V-05 (12.5-cm) contactors, that is, the scrub, strip, and wash sections, so the solvent flow rate was lowered to 3 gpm to decrease the total throughput there. Because the V-05 contactors are small relative to the CINC V-10 contactors (25-cm contactors) used in the extraction section of the MCU, total throughput

in the MCU is limited by the capacity of the V-05 units. Contactor rotor hydraulics were evaluated using N_{Di} and the ROTOR workbook in Excel. With a typical N_{Di} value of 0.0008 or better, hydraulic operation at a solvent (DX) flow rate of 3.00 gpm will be acceptable. However, earlier N_{Di} measurements showed that it could be as low as 0.0006. If this is the case, then the solvent (DX) flow rate must be dropped to 2.42 gpm to achieve acceptable hydraulic performance. Since the aqueous flow rates do not change, the NG-CSSX flowsheet will continue to give a maximum salt feed processing rate of 12 gpm. In this way, the solvent (DX) flow rate can be used to improve hydraulic performance in the MCU

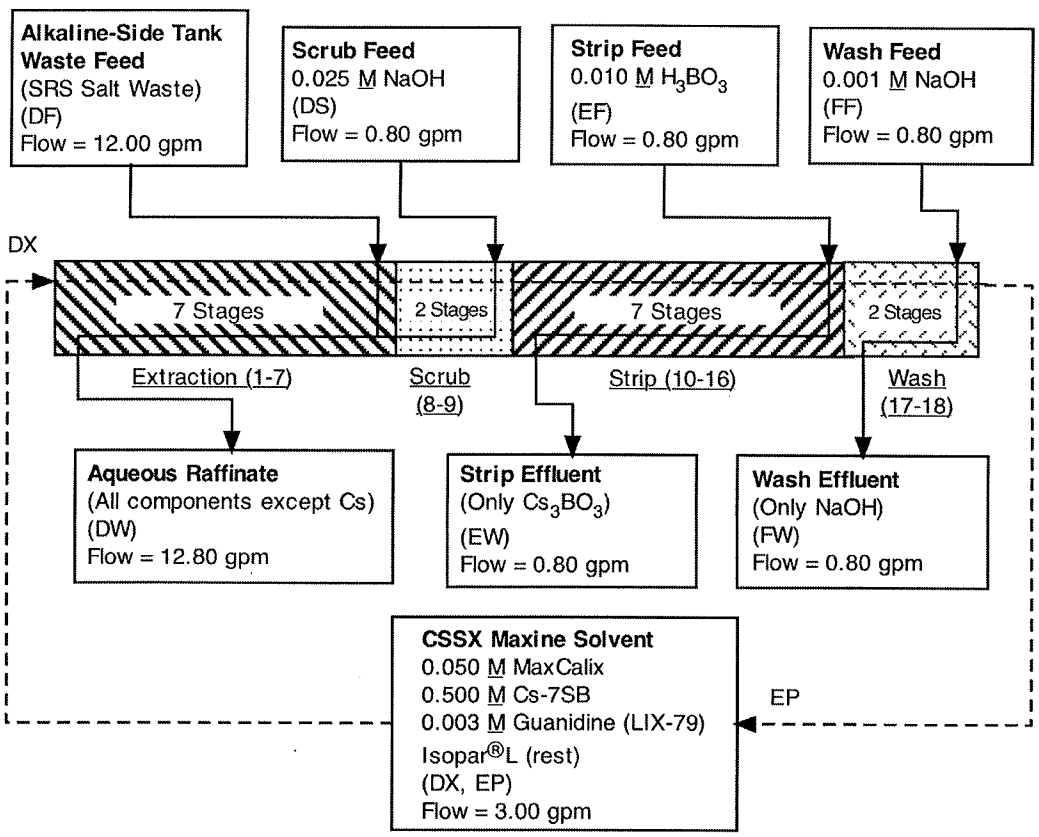

Fig. 2. Next-Generation CSSX Flowsheet for MCU Showing Maximum Flow Rates

The trade off in lowering the solvent (DX) flow rate is that process robustness will also be lowered. The robustness factor is defined as the calculated D.F. divided by 40,000, the desired D.F. The D.F. is calculated using the SASSE spreadsheet. The robustness values are shown in the first column of Table 1 for the flowsheet in Fig. 2 for a composite SRS feed with either 15 mM K or 45 mM K. If N_{Di} is 0.0006 so that a solvent (DX) flow rate 0f 2.42

gpm is required for good hydraulic performance, the process robustness will be lower as shown in the second column of Table 2. However, even for this lower solvent (DX) flow rate, the robustness is still be greater than 1.0 over the range of expected K concentrations.

Table 1. Robustness of Next-Generation CSSX Flowsheet at Maximum Aqueous Flow rates as a Function of Potassium Concentration in the Salt Feed and the Solvent Flow Rate

Concentration of K in SRS Salt (DF) Feed, mM	**Robustness (Calculated D.F./40,000)**	
	Solvent (DX) Flow Rate = 3.00 gpm ($N_{Di} \geq 0.0008$)	**Solvent (DX) Flow Rate = 2.42 gpm** ($N_{Di} \geq 0.0006$)
15	5.28	2.60
45	3.75	1.52

There is some additional robustness built into the SASSE calculations. The other-phase carryover used in the calculations was 0.5%. It is observed that other-phase carryover is typically 0.05% or less. If this lower value is used, the robustness of 1.52 becomes 2.07. In the calculations, the Cs distribution coefficients were the values determined at 25°C. In the MCU the extraction section is operated at 23±3°C and the strip section is operated at 33±3°C. Both of these temperature changes would give additional process robustness. If 23°C is used for the extraction section, then the robustness of 1.52 becomes 2.32. When both factors are combined, the robustness of 1.52 becomes 2.99, a very reasonable robustness value. In addition, if the MCU were operated at a temperature below 23°C for this new flowsheet, process robustness could be improved even more. The temperature of the extraction section thus becomes another possible control variable for the CSSX process. In every case, the stripping of the Cs is even better than the D.F. obtained for the extraction section.

In the SASSE calculations to get robustness, an average multistage efficiency of 90% was assumed. This is based on experimental measurements at SRS (Ref. 2) that shows single-stage efficiencies of 98% for the conditions (rotor speed of 2100 rpm and salt feed flow rate of 8.5 gpm or greater) in the V-10 contactors in use at the MCU. At lower salt feed rates and lower rotor speeds, single-stage efficiencies are lower. From work on multistage 2-cm contactors (Ref. 3), it was shown that multistage operation lowers the average stage

efficiency. This is due to the flow variations from stage to stage. These are quite severe in the 2-cm contactor. Thus, as the O/A flow ratio moves away from 1.0, either higher or lower, the apparent stage efficiency drops. At the O/A flow ratios of 0.23 and 0.19 for the 3.00 and 2.42 gpm solvent flow rate cases shown in Table 1, the apparent stage efficiency for a contactor with measured single-stage efficiency of 98% or greater will be about 90%. This value was used in the SASSE calculations. In larger contactors with higher flow rates, flow rates appear to be much steadier. Thus, in the V10 contactors, the average apparent multistage efficiency is probably higher than 90%. I would expect it could be 95% or greater. However, this has never been measured. If the average stage efficiency were increased even 1%, from 90 to 91%, the robustness of 1.52 in Table 1 would become 2.07. If the average stage efficiency were increased 5%, from 90 to 95%, the robustness of 1.52 in Table 1 would become 8.11.

While N(Di) values for the optimized NG-CSSX solvent were not available for calculating this initial flowsheet, N(Di) values with earlier NG-CSSX solvent candidates and from Ref. 1 show that lowering the solvent modifier, Cs-7SB, from 0.75 M to 0.50 M, gives rise to higher N(Di) values. In general, N(Di) values were increased by 38% or greater. This will improve contactor hydraulics and lead to a further increase in process robustness since the solvent (DX) flow rate could be increased.

The new NG-CSSX solvent has a lower density than the current CSSX solvent being used in the MCU. The current solvent is close to 855 g/L while the new solvent will be close to 820 g/L. Because of this, the ROTOR calculations show that the next larger upper (more-dense phase) weir will be required for the eleven CINC V-05 contactors in the MCU to be able to operate the flowsheet shown in Fig. 2. While increasing the upper (more-dense phase) weir size in the CINC V-10 contactors would also increase their maximum throughput with the new solvent, it is not necessary as the current weirs provide plenty of capacity for this flowsheet.

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