

Solvent Carryover Characterization and Recovery for a 10-inch Single Stage Centrifugal Contactor

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ABSTRACT

A test program has been performed to characterize the organic solvent carryover and recovery from centrifugal contactors in the Caustic-side Solvent Extraction (CSSX) process. CSSX is the baseline design for removing cesium from salt solutions for Department of Energy (DOE) Savannah River Site's Salt Waste Processing Facility. CSSX uses a custom solvent to extract cesium from the salt solution in a series of single stage centrifugal contactors. Meeting the Waste Acceptance Criteria at the Defense Waste Processing Facility and Saltstone, as well as plant economics, dictate that solvent loss should be kept to a minimum. Solvent droplet size distribution in the aqueous outlet streams of the CSSX contactors is of particular importance to the design of solvent recovery equipment. Because insufficient solvent droplet size data existed to form a basis for the recovery system design, DOE funded the CSSX Solvent Carryover Characterization and Recovery Test (SCCRT). This paper presents the droplet size distribution of solvent and concentration in the contactor aqueous outlet streams as a function of rotor speed, bottom plate type, and flow rate. It also presents the performance data of a prototype coalescer.

INTRODUCTION

CSSX is a continuous flow process utilizing 36 contactor stages for extraction, scrubbing, stripping, and washing of aqueous and organic streams. Cesium (Cs) is captured by contacting the Cs-bearing aqueous salt waste with a solvent (organic phase) in the extraction stage

contactors at a aqueous flow rate to solvent flow rate ratio (A:O) of 16:5. A simulated salt waste was used for SCCRT consisting primarily of sodium hydroxide, sodium nitrate, aluminum nitrate and sodium nitrite [1]. The solvent used in the CSSX process is primarily Isopar®L, with a specialty extractant (BOBCalixC6) a modifier (Cs-7SB), and a suppressant (tri-n-octylamine) [2]. The Cs-laden solvent exiting the extraction stage is scrubbed to remove Na and K impurities. The Cs is then stripped from the solvent. In the scrub and strip contactors, the A:O is 1:5. Both the scrub and strip phases utilize dilute nitric acid solutions. The stripped solvent is then washed with dilute caustic to remove any extracted impurities and solvent degradation products before it is recycled to the extraction system.

Each contactor stage mixes the aqueous and organic phases together, then separates them and discharges them separately. The separation is not absolute. A small amount of the aqueous phase remains entrained in the organic effluent, and a small amount of organic remains entrained in the aqueous effluent. While the entrained aqueous is of little consequence, the organic entrained in the aqueous phase is a major operational cost if left unrecovered. In addition, the aqueous product streams must meet Waste Acceptance Criteria at downstream waste immobilization facilities (the Defense Waste Processing Facility and Saltstone). To mitigate this cost and to meet downstream requirements, the SWPF design includes solvent recovery equipment on the aqueous effluents from the CSSX system.

Solvent recovery from processed aqueous feed in the CSSX process is one parameter in the design of the SWPF process and estimation of plant operating costs. The solvent droplet-size distribution in the aqueous outlet streams of the CSSX contactors is of particular importance to the design of solvent recovery equipment, in particular solvent coalescers. Because there was insufficient solvent droplet-size distribution and concentration data [3], the CSSX SCCRT was developed to address this particular data gap.

The primary objectives of the SCCRT included characterization of the droplet size distribution of solvent in the contactor aqueous outlet streams as a function of rotor speed, bottom plate type, and throughput; the test also demonstrated effective solvent recovery from the aqueous effluent utilizing a prototype coalescer. Solvent recovery was characterized as a function of flow rate to allow scale-up to the full-scale CSSX System and the selection of the optimal coalescer media type. The lengths of the coalescer media and the separation-zone were verified and passive interface control (i.e., through the use of weirs) for continuous removal of organic and aqueous phases from the coalescer was demonstrated.

EQUIPMENT DESCRIPTION

The CSSX SCCRT was conducted at the General Atomics (GA) facilities in San Diego, California. Fig. 1 illustrates a basic process flow diagram of the SCCRT test system. The SCCRT system comprised a CINC V-10 contactor, a coalescer, a flow splitter, droplet-size analyzer, fluid supply systems including pumps, heat exchangers, chiller, instrumentation and controls, and interconnecting piping and wiring to support continuous operation. The single contactor was configurable as either an extraction-stage or strip-stage contactor. Access ports were located to directly grab samples and a small slipstream was provided to the droplet-size analyzer. This configuration enabled evaluation of solvent droplet-size distribution in the contactor aqueous effluent as well as coalescer solvent-recovery performance. Sample ports were installed at upstream, mid-length, outlet, and downstream of coalescer. A 210L salt/strip solution (aqueous) feed tank was used to hold a solution volume that provided an one-minute residence time in the

tank. This configuration provides minimal settling time for the aqueous stream and thus provides a more conservative challenge to the SCCRT solvent recovery system.

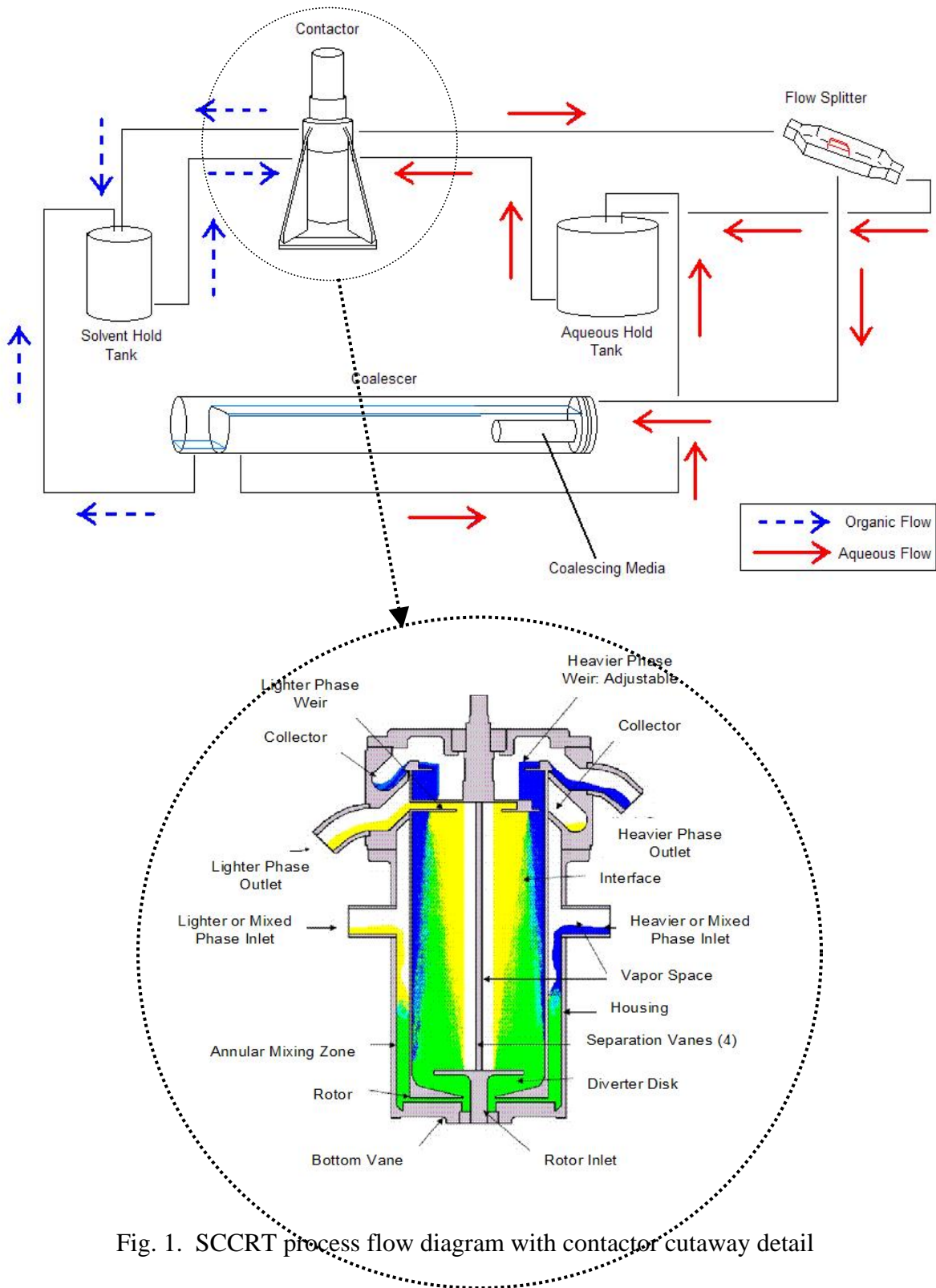


Fig. 1. SCCRT process flow diagram with contactor cutaway detail

Contactactor

Fig. 1 shows a centrifugal contactor cutaway schematic. A single-stage CINC Model V-10 contactor was used for SCCRT. The contactor is a centrifugal separator, in which immiscible liquids of different densities are mixed in the annulus between the stationary housing and spinning rotor, and then separated inside the spinning rotor by centrifugal force. The unit comprised a steel housing support, two inlets and outlets, rotor with drive motor, and variable frequency drive for control of rotor speed. In the contactor, the inlets and outlets are placed tangential to the rotor, with the fluid entering and exiting in the same direction as the swirling liquid. The salt simulant (aqueous) and extraction solvent solutions are introduced into the annular mixing zone through separate inlets. These process fluids are mixed in the annular zone and then pulled into the bottom vanes of the self-priming rotor. The rotor spins the fluids axially upward, along the rotor wall. The two liquid phases are separated by centrifugal force as they move axially up the wall, and then directed by the rotor and weir designs to flow out through separate outlets.

Fluid flows during extraction were nearly 3.5 times greater than flows during strip testing. The liquid height in the contactor mix zone would be reduced (or eliminated) at the substantially lower strip flow rates. Thus, a low-flow sleeve was installed inside the contactor prior to strip testing to ensure proper mixing in the contactor. The low flow sleeve decreased the cross-sectional area in the annulus proportionally to the reduced flow rate to ensure liquid height is maintained in the annulus.

Coalescer

The coalescer was a 20cm diameter by 305cm long cylindrical vessel that provided gas-liquid separation. The diameter was sized for two gpm liquid flow rate. Liquid residence time was approximately five minutes. The coalescer consisted of a fluid coalescing media, mist eliminator, and disengagement zone. Clear polyvinyl chloride was selected for the material of construction to allow visual observation. The coalescer was modular to facilitate rapid changes in media and media bed lengths, separation zone lengths, and interface controls (active versus passive). Two different stainless steel mesh coalescing media were tested; Amistco LL-430W 100 μ m mesh and Franken TPS III/500-M-F 8 μ m mesh.

Flow Splitter

Since the fabricated coalescer was sized for the strip flow rates and not for the extraction flow rates, a proportional cross-section of the incoming extraction raffinate flow had to be acquired. Obtaining representative samples for carryover analyses using conventional sampling techniques is problematic due to the two-phase nature of the contactor effluent and the relatively large aqueous flow rates. Sampling from a small slip stream (via a sample valve) could preferentially sample either the aqueous or organic phases depending on the location of the sample port. The required coalescer feed was approximately 10% of the aqueous stream from the contactor discharge during extraction conditions. For the same reasons described above, obtaining a representative slip stream to the coalescer is problematic using conventional methods. Therefore, a custom flow splitter was utilized to attain a cross-section of the aqueous flow rate for use in the coalescer and obtaining representative samples for carryover analyses. The flow splitter works by spreading and streamlining the flow over a wide area. A mesh flow distributor screen was used to promote uniform flow. A small portion of the streamlined flow passes into a port

between a fixed vane and an adjustable vane. By taking a vertical slice of the contactor effluent, a stream is produced that contains representative quantities of the both the organic and aqueous phases.

MEASUREMENT TECHNIQUE

A Jorin Visual Process Analyzer (ViPA) was used to analyze solvent droplet size and organic carryover concentrations in samples taken from the aqueous effluent. The ViPA comprised an optical unit, a peristaltic pump, a control computer, and software. The optical unit used a video microscope with lens, camera, and light source to examine the sample stream. The sample stream flowed between a pair of transparent sapphire windows inside the ViPA's optical unit. The camera looked through the liquid at the light source providing a backlit view of solid particles, liquid droplets, or gas bubbles. The ViPA was capable of measuring droplets ranging from 2.6 to 250 μ m in diameter and solvent concentrations from 0 to 2,500 parts per million (ppm).

A peristaltic pump was used to draw the sample fluids through the ViPA optical unit containing a digital video microscope. The ViPA operated by freezing a single frame of the video image and analyzing the objects present. A database of information was built by rapidly acquiring and analyzing sequences of these still images. The different objects or phases present in the frame were differentiated by using 17 measured and/or calculated parameters including area, size, aspect ratio, shape factor, curvature, and optical density. The ViPA measured dispersed-phase visible concentration, based on a known volume of liquid for each frame analyzed. The measured concentration was reported as visible ppm, because only those objects seen were measured and included in the concentration calculation. The ViPA reported changes in concentration over time.

RESULTS

The primary objectives of the SCCRT were to:

1. Characterize the solvent droplet-size distribution during extraction conditions,
2. Characterize the solvent droplet-size distribution during strip conditions, and
3. Demonstrate acceptable solvent recovery from both extraction and strip aqueous effluents.

Carryover Characterization Results

Meeting the first two objectives required characterization of contactor performance over the range of expected contactor operating conditions. Variable contactor operating parameters included contactor weir size, fluid flow rates, and rotor speed. The contactor performance characterizations were achieved by changing heavy-phase weir inner diameter and then studying the effects of different flows and rotational speeds for that weir diameter. Characterizations were done for extraction and strip conditions, using both straight- and curved-vane contactor bottom plates.

To illustrate the relationships observed between the contactor operational variables, interaction plots are provided from each of the 4 contactor test configurations. These were: 1) extraction conditions using a straight-vane lower plate, 2) extraction conditions using a curved-vane lower plate, 3) strip conditions using a straight-vane lower plate, and 4) strip conditions using a curved-vane lower plate. The interaction plots show general trends observed over the range of operating conditions tested. The JMP software used to analyze the data builds the interaction plots after generating linear regression models and performing F-tests on the data. Interaction plots are statistically accurate for the data as a whole; however they are not an appropriate tool for predicting performance under specific operating conditions. They are best suited to identifying general trends.

Fig. 2, containing four interaction plots, graphically illustrates how the three main variables in the tests interacted with each other to affect the solvent carryover concentration for extraction and strip conditions using a straight-vane and curved-vane lower plate. Fig. 2a shows that for extraction conditions with the straight vane bottom plate the solvent carryover is generally lower at higher rotor speeds. This can be determined by observing that the curves above the rotor speed square have negative slopes. The plot also shows that for a given rotor speed, the weir size and aqueous flow rate do not significantly effect solvent carryover concentration. This conclusion can be drawn by seeing that the curves to the left of the rotor speed square are generally flat. This plot also indicates that the effects of aqueous flow on solvent concentration may be weir dependant. This can be seen by observing that the curves above and to the left of the aqueous flow rate square, cross and have much different slopes. Fig. 2b graphically shows that the carryover is lower with the 13.5cm weir over the entire range of operating conditions. It also shows that for the 13.7cm weir, concentration is nearly constant throughout the rpm range tested. Fig. 2c is an interaction plot for strip conditions using a straight-vane bottom plate that graphically illustrates how the three main variables in these tests interacted with each other to affect the solvent carryover concentration. The plot shows that solvent carryover is lower at lower rotational speeds and that weir size has a fairly significant effect at high rotor speed. It also shows that changes in flow rate have little effect on carryover. Lastly, Fig. 2d is an interaction plot for strip conditions using a straight-vane bottom plate. Fig. 2d shows that droplet diameter increases with decreasing rotational speed, while weir size and flow rate have little effect.

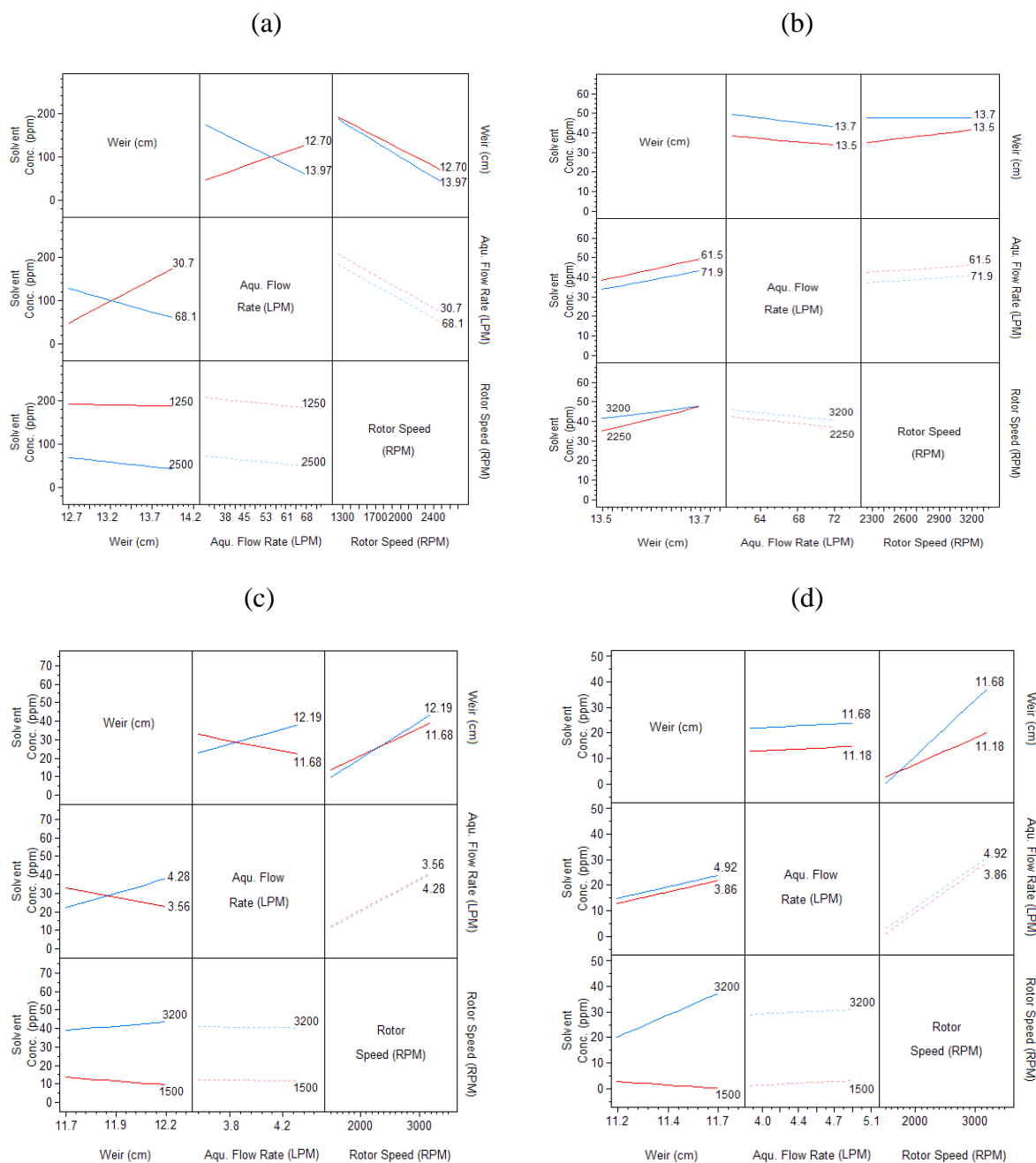


Fig. 2. Solvent carryover concentration variable interaction and response using (a) straight vane contactor configuration under extraction conditions. (b) curved vane contactor configuration under extraction conditions. (c) straight vane contactor configuration under strip conditions. (d) curved vane contactor configuration under strip conditions.

Droplet size and droplet size distributions were measured by the ViPA. Approximately 5000 measurements of size distributions were captured during testing. Fig. 3, containing four additional interaction plots, graphically illustrates how the three main variables in these tests interacted with each other to affect the solvent droplet diameter for the straight-vane and curved-

vane configuration under extraction and strip conditions. Droplet size distributions were essentially insensitive to the test variables and vane type during extraction testing, as illustrated by interaction plots in Fig. 3a and Fig. 3b. Fig. 3c and Fig. 3d show that droplet diameter increases with decreasing rotational speed and that there is again an interaction between flow and weir size during strip conditions.

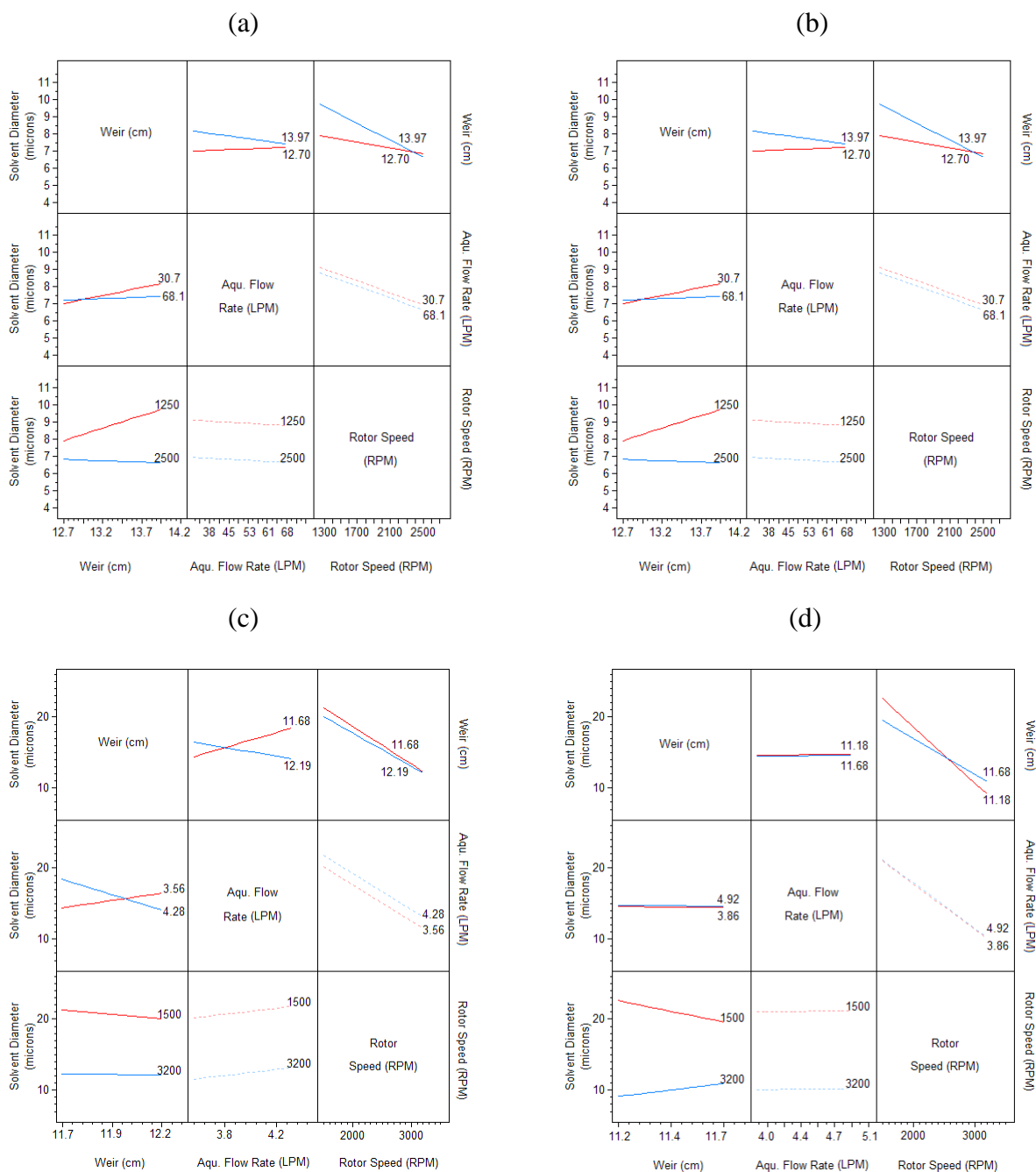


Fig. 3. Solvent droplet diameter variable interaction and response using (a) straight vane contactor configuration under extraction conditions. (b) curved vane contactor configuration under extraction conditions. (c) straight vane contactor configuration under strip conditions. (d) curved vane contactor configuration under strip conditions.

Droplet size and droplet size distributions were measured by the ViPA. Typical droplet size distribution data is shown in Fig. 4. As can be seen from the interaction plots above, the mean droplet size for extraction conditions shown in Fig. 4a was remarkably stable, generally hovering around $7\mu\text{m}$ with the largest mean size being $<10\mu\text{m}$ and the smallest mean size being $>6\mu\text{m}$. Fig. 4a illustrates that the distribution of the log of diameter is well characterized as a normal distribution as evidenced by the linear nature of the distribution data when plotted as cumulative volume vs. diameter using a log scale for diameter. The previous interaction plots show that mean droplet size during strip conditions was insensitive to all variables except rotor speed. Rotor speed showed a strong effect on mean droplet size, with droplet size decreasing with increasing rotor speed under strip conditions. As shown in Fig. 4b, the nature of the distribution is slightly different from the nearly perfect normal distribution displayed during extraction conditions. The distributions associated with strip conditions show small populations of droplets at the high and low extremes that are above the counts predicted by a normal distribution. In general, the distribution is wider than for extraction conditions, with a greater range of droplet sizes present. The mean droplet size under strip conditions ranged from $\sim 12\mu\text{m}$ to $22\mu\text{m}$.

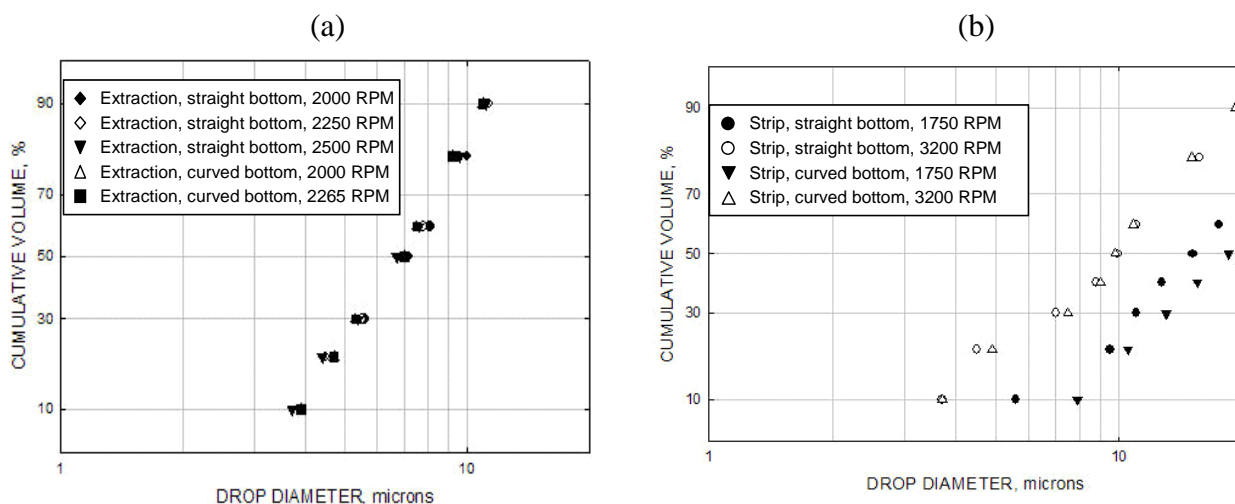


Fig. 4. (a) droplet size distributions under extraction conditions. (b) droplet size distributions under strip conditions

Solvent Recovery Results

The third primary objective of the test, a demonstration of acceptable solvent recovery from both extraction and strip aqueous effluents was accomplished using the SCCRT system coalescer. Two different media types were tested: one manufactured by Amistco and one by Franken.

The Amistco media was a conventional axial flow stainless-steel mesh style media. The Amistco media was tested at coalescer flow rates of 1.9 and 11.4 L/min under extraction conditions. No further testing was performed with the Amistco mesh because it was found to be ineffective at coalescing solvent from the aqueous stream at either flow rate. Given the size distribution of the solvent droplets and the large fiber diameters, this result was not unexpected.

The Franken media was a radial flow stainless-steel mesh media. With extraction conditions and Franken media the incoming concentrations of 67 to 104 ppm were reduced to less than 16 ppm

at the coalescer outlet for coalescer flow rates under 7.6 L/min. Flow rates ranging from 0.8 to 11.4 L/min were examined. As expected, recovery improved with decreasing flow. The coalescer effluent solvent concentration remained relatively flat at ~15 ppm over the flow range of 1.9 to 6.4 L/min.

It should be noted that the SCCRT coalescer was understood to be undersized for extraction conditions, where only a fraction of the total aqueous contactor effluent was diverted to the coalescer. The data obtained related to coalescer solvent outlet concentration versus flow rate will be used to scale the coalescer to typical SWPF flow rates. The scale-up of the vessel from SCCRT to full-scale is done by maintaining the superficial velocity profile in the vessel and increasing the coalescer media surface proportionally with flow rate. The coalescer flow rate can readily be converted to superficial velocity (assuming constant area) by dividing by the coalescer liquid cross-sectional area. Using simple geometry and selecting a full-scale flow rate basis, a plot of average solvent concentration as a function of vessel diameters can be generated. This scaling methodology predicts that an extraction coalescer diameter of ~91cm will be required for the SWPF.

Due to the recovery results from extraction conditions, only the Franken media was tested during strip conditions. Flow rates ranging from 0.8 to 4.5 L/min were examined. Under strip conditions with the Franken media incoming concentrations of 10 to 21 ppm were reduced to less than 0.5 ppm at the coalescer outlet. It should be noted that the coalescer used operating at flows typical for the SWPF strip conditions. Its performance should be representative of the SWPF strip coalescer.

SUMMARY

The following summarizes the results applicable to the SWPF CSSX design.

1. Results for tests performed under the extraction conditions tested:
 - The solvent-droplet mean diameter is consistently between 6 and 10 μ m. The droplet size can be increased by lowering the rotor speed, but unaffected by the bottom-plate type, weir size, or flow rate.
 - Solvent carryover concentration using the straight vane bottom plate decreases as rotational speed increases. Carryover using the curved-vane bottom plate is affected only slightly by rotor speed.
 - With the straight-vane bottom plate, the effects of aqueous flow rate on solvent carryover are weir dependant. Solvent carryover decreases with increasing flow rate for weir sizes larger than 13.2cm. Solvent carryover increases with flow rate for weir sizes 13.2cm and smaller.
 - With the curved-vane bottom plate, solvent carryover increases with increasing weir size.
 - With the curved vane bottom plate, aqueous carryover is minimized between 2600 and 3000 rpm, within the aqueous flow rate range of 49 to 68 L/min.
2. Results for tests performed under the strip conditions tested:

- The solvent-droplet mean diameter is consistently between 8 and 25 μ m. The droplet size increases with decreasing rotor speed, but is not significantly affected by bottom-plate type, weir size, or flow rate.
 - Use of a curved-vane contactor-bottom-plate produces results similar to that of the straight vane.
 - Solvent carryover increases with rotor speed
 - Weir size does not a significant effect on aqueous carryover.
 - With the straight-vane bottom plate, there are no identifiable trends between aqueous carryover, flow rate and rotor speed.
 - With the curved-vane bottom plate, there is no identifiable trend between aqueous carryover and flow rate.
 - With the curved-vane bottom plate, aqueous carryover decreases with increasing rotor speed.
3. Results for coalescer tests performed under the conditions tested are:
- The Franken TPS III/500-M-F 8 μ m mesh proved to be superior to the Amistco LL-430W 100 μ m mesh for all conditions.
 - Under extraction conditions, the Franken media showed clear evidence that the solvent concentration and mean droplet size in the aqueous phase were reduced. The incoming solvent concentrations of 67 to 104 ppm were reduced to less than 16 ppm at the coalescer outlet for coalescer flow rates under 7.6 L/min (the nominal maximum flow rate for the specified coalescer).
 - Under strip conditions, the solvent concentration in the aqueous phase was reduced by the Franken media. The incoming solvent concentrations of 10 to 21 ppm were reduced to less than 0.5 ppm at the coalescer outlet.
 - The Amistco mesh media did not show definitive results for reducing the solvent concentration or droplets size.
 - With the Franken coalescer media, solvent recovery increases as flow rate decreases.
 - Passive control of phase interface is a viable option for solvent recovery.

CONCLUSION

The SCCRT satisfied the defined test objectives. The following considerations will be noted for the upcoming full-scale CSSX test, which will embody a more integrated, scaled-up version of the system built for the SCCRT test program:

1. The curved-vane contactor-bottom-plate is recommended over the straight-vane bottom plate because less carryover is produced. Final selection of bottom-vane type will be confirmed during CSSX full-scale testing where measurements of stage efficiency will be performed.

2. Franken coalescer media is recommended for use in both the strip and DSS coalescers.
3. Outside lab analysis is recommended to verify the ViPA concentration data.

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