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Review of Benzene Stripping Alternatives for the Small Tank Precipitation Facility

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SUMMARY

Packed columns provide a proven technology for stripping benzene from salt solution. With continuous Small Tank Precipitation process the stripping load is reduced by a factor of four vs. former ITP cycling, and process continuity is maintained through to the Saltstone transfer tank. Lower stripping capacity allows new design options, including coarser packing and possibly reduced foaming packing.

Nitrogen sparging is a cost effective method for stripping benzene from dilute salt solutions. In theory, the sparging gas, dispersed in fine droplets in the solution, is rapidly saturated with benzene. Under Late Wash conditions, sparging was efficient down to 5 mg/L. Stripping to lower concentrations required additional time and stripping under more concentrated Small Tank conditions was not investigated. A key concern is foaming introduced by the gas bubbles, necessitating mechanical foam breakup equipment. Foaming in concentrated salt solutions is particularly problematic since this can lead to "solid stationary foam" difficult to move or disperse. A sparging operation would likely require two tanks, a feed tank and a sparge tank, and would result in a separate batch operation following continuous Small Tank Precipitation. Existing ITP packed columns and Late Wash tanks could be utilized for stripping and sparging.

The opposite of sparging (bubbles of gas in a liquid) would be a shower of liquid droplets in a gas to minimize foaming. This could be achieved in a static column, or dynamically using a vortex scrubbing device.

RECOMMENDATIONS

1. Consult with industry and equipment manufacturers to identify proven technology for processing concentrated salt solutions and minimizing foaming, including non-foaming column packing, shower columns, and vortex scrubbing.
2. Test selected components for operability and foaming under Small Tank Precipitation processing conditions.

3. Evaluate feasibility of raising Saltstone benzene acceptance limit from 3 mg/L to 5 mg/L.

BACKGROUND

Small Tank Precipitation (STP) process is one of the alternatives for High Level Waste (HLW) salt solution decontamination. As with the original In-Tank Precipitation (ITP) process, sodium tetraphenylborate (NaTPB) and monosodium titanate (MST) would be used to precipitate radioactive cesium-137 and strontium. However the new process would run continuously, in small tanks, and at a controlled temperature, 25 °C, to minimize TPB decomposition and benzene generation. A preliminary flow sheet and material balance prepared by R. A. Jacobs of HLW Process Engineering is shown in Figure 1. The decontaminated salt solution (Figure 1, Stream 9) is assumed to be saturated with benzene at 215 mg/L at 23 °C (Ref. 1) and requires stripping to meet 3 mg/L acceptance limits for Saltstone operations.

Benzene stripping in a packed column, as used in ITP, and nitrogen sparging, as planned for Late Wash, need re-evaluation for the STP facility, particularly in consideration of a continuous precipitation process and smaller continuous feed rate of decontaminated salt solution, 22 gpm vs. 114 gpm design for batch ITP. The existing smaller ITP stripping column has a design rate of 22 gpm.

PACKED COLUMN OPERATIONS

Operation of packed columns has been demonstrated in ITP and in extensive tests at Koch Engineering (Ref. 2). Benzene is sparingly soluble in concentrated salt solution, about 215 mg/L in 4.7 M Na+ (Ref. 1) and readily strippable to air or nitrogen. In the STP process the benzene stripper would follow the concentrate cross-flow filter, Figure 2A, Stream 9, maintaining a continuous flow through to the Saltstone transfer tank. Since the stripper would be in continuous use the decontaminated salt solution feed rate would be only 22 gpm vs. 114 gpm for ITP which was based on cyclic campaign operation. For ITP equivalent performance the stripper column diameter could be reduced from 30 to 13 in., approximately the size of the smaller, 16 in. diameter, ITP column. Extensive tests at Koch Engineering (Ref. 2) have shown that 100 to 1000 mg/L benzene in feed can be stripped to under 2 mg/L with existing ITP design: liquid flow 25 gpm/ft^2, nitrogen flow 90 SCFM/ft^2.

Key considerations in stripping are foaming and resultant increased column pressure drop which sets a limit on the gas flow. Tests at Koch Engineering (Ref. 2) have shown that the pressure drop can be controlled by addition of antifoam agents, with minimal effect on stripping efficiency.

New design for the STP process offers additional options to assure effective benzene stripping:

1. Increased column diameter to reduce pressure drop.
2. Coarser packing to reduce pressure drop.
3. Specialized packing to provide laminar or columnar liquid flow, minimizing bubble formation (foaming) when the gas phase is forced to penetrate the liquid phase.

In addition, an in-line continuous stripper would be simpler to operate as part of continuous STP processing.

IN-TANK SPARGING

Tests on 1/200 scale Late Wash sparging have shown that benzene can be readily removed from low sodium concentration salt solution by batch sparging in an agitated vessel (Ref. 3). Late wash test data showed that benzene concentration during sparging of salt solution falls off exponentially with purge time, down to about 5 mg/L , indicating vapor phase diffusion or liquid/vapor interface control (Figure 3). However , below 5 mg/L the stripping efficiency falls off rapidly, indicating slower liquid phase diffusion control. The effectiveness of benzene stripping from high sodium salt solutions has not been investigated. Assuming Late Wash data, Small Tank sparging time can be estimated down to about 5 mg/L as calculated in Table 1 and shown in Figure 4.

A major concern in sparging is foaming. A test designed for evaluating foaming in Late Wash was abandoned due to uncontrolled foaming in an 8 in. un-agitated column (Ref. 5). In earlier Late Wash filtration testing (Ref. 9), foaming was introduced by simulated ITP slurry recirculation and Surfynol® had to be used to control foaming. Stationary ITP foam formed around the edges of the recirculation container. The foam solidified with time as the solution concentrated by evaporation. The same could occur by channeling in sparging, allowing stationary foam to evaporate and solidify.

Sparging by definition, is making bubbles in the solution. This in turn leads to foaming. Controlled agitation and/or foam break up equipment could be needed to permit reliable sparging.

The STP sparging process would require two tanks: a decontaminated salt solution receipt tank, and a batch sparging tank (Figure 2B). Existing tanks in Late Wash facility could be used. Both tanks could be designed for sparging. Using Late Wash data, a one day production of decontaminated salt solution, 32K gal, could be stripped to 1 mg/L in about six hours (Appendix A, Figure 3), assuming 50% saturation of nitrogen with benzene, no drop off in efficiency below 5 mg/L, and no foaming.

ALTERNATE TECHNOLOGIES

Since foaming is known to occur during solution and gas mixing, the goal is to identify processes which eliminate or minimize foaming. An example would be a wetted wall column where the liquid flows down the wall and the gas flows up the center without breaking through the liquid. This type of stripping is good for studying mass transfer, but the process is inefficient.

A practical approach may be to identify non-foaming packing which generates columnar liquid streams or droplets, allowing the gas to flow by without breaking through liquid sheets which leads to foaming.

Another option is engineered power scrubbing, such as AEA Technology "Vortex Scrubber" (Ref. 6). This scrubber forms a planar stream of droplets by impinging two opposing streams against each other. The scrubbing gas is fed tangentially and exits along the axis of the scrubber (Figure 5). This device is compact, has no moving parts, and requires no demister. This is particularly important in handling concentrated salt solutions.

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Table 1. Small Tank Sparging Calculations

D:\app\BZStrip\[Small Tank Strip.xls]Table1

	Eqn Synb	ASSUME		CALCULATE	
Decon Solution Volume		**32342**	gal		
Decon Solution Volume	V			4324	CF
Tank Sparge Diameter		== Hight		17.66	FT
Tank Sparge Area				244.87	SF
Purge Rate		**0.30**	SCFM/SF		
Nitrogen Flow	S			73.46	SCFM
Efficiency	E	**50%**			
Sodium Conc, Na+ Molar		**4.7**			
Temperature		**23**	deg C		
Henry's Constant	H	**1.80**	(mg/L in Vapor)/(mg/L in Solution)		
Starting Benzene Conc		100	120	215	500

Time		Benzene Concentration at Time			
MIN	HR	**100 mg/L**	**120 mg/L**	**215 mg/L**	**500 mg/L**
0	0	100.00	120.00	215.00	500.00
30	0.5	63.21	75.85	135.90	316.04
60	1	39.95	47.94	85.90	199.77
90	1.5	25.25	30.30	54.30	126.27
120	2	15.96	19.16	34.32	79.81
150	2.5	10.09	12.11	21.69	50.45
180	3	6.38	7.65	13.71	31.89
210	3.5	4.03	4.84	8.67	20.16
240	4	2.55	3.06	5.48	12.74
270	4.5	1.61	1.93	3.46	8.05
300	5	1.02	1.22	2.19	5.09
330	5.5	0.64	0.77	1.38	3.22
360	6	0.41	0.49	0.87	2.03
390	6.5	0.26	0.31	0.55	1.29
420	7	0.16	0.20	0.35	0.81
450	7.5	0.10	0.12	0.22	0.51
480	8	0.06	0.08	0.14	0.32

Washing endpoint =	0.010	M nitrite
Wash ratio =	43.64	
Wash water vol =	3.776	gal/gal

Figure 1 Small Tank Precipitation Material Balance

Strm No.	1	2	3	4	5	6	7	8	9	10	10a	11	11a	12	12a	12b	12c	13	13a	13b
Strm Name	Salt soln xfer	Salt soln feed	NaTPB	MST	Dilution water	Recycle wash water	CSTR #1 Prod	CSTR #2 Prod	Decon salt soln	Conc'd precip	Conc'd precip	Wash water	Wash water	Washed precip	Washed precip	Post-wash conc'n	Post-wash conc'n	Post wash filtrate	Spent wash water	Spent wash water
[B]atch or [C]ont	B	C	C	C	C	C	C	C	C	B	avg	inst	avg	B	avg	B	avg	avg	avg	B
Flow - gal/min	100	16.5	0.52	0.072	0.36	6.70	24.27	24.27	22.46	–	1.81	15.4	6.14	–	1.72	–	1.17	0.56	6.70	–
Flow - lbs/hr	255734	10656	294	39.6	182	3574	14744	14744	13645	–	1099	7687	3075	–	879	–	599	280	3574	–
Aqueous flow - gal/min		16.5				6.70	24.08	24.08	22.46	–	1.63	15.4	6.14	–	1.63	–	1.07	0.56	6.70	–
Cycle time - hours	4.0	–	–	–	–	–	–	–	–	60	–	24	–	60	–	60	–	–	–	60
Batch size - gal	23760	–	–	–	–	–	–	–	–	6521	–	22120	–	6208	–	4206	–	–	–	24122
Temp - °C	amb	amb	25	amb	amb	25	25	25	25	25	25	amb	amb	25	25	25	25	25	25	25
Na+ - M	6.4	6.4	0.65			1.17	4.70	4.70	4.70	4.70	4.70			0.02	0.02	0.02	0.02	~ 0.02	1.168	1.17
Cs-137 - Ci/gal	1.38	1.38					0.95	0.95	<1.6E-04	14.0	14.0			14.0	14.0	21.3	21.3	?	?	?
Sg aqueous - g/mL	1.290	1.290	1.12		1.0	1.057	1.214	1.214	1.214	1.214	1.214	1.0	1.0	1.006	1.006	1.006	1.006	1.006	1.057	1.057
Sg - g/mL	1.290	1.290		1.10			1.214	1.214		1.212	1.212			1.019	1.019	1.025	1.025			
Wt% insoluble solids	0.039	0.039		11.4		0	?	0.75	0	10.0	10.0			6.82	6.82	10.0	10.0	0.0	0.0	0.0
Flows - lbs/hr water	169793	7075	243	35.1	182	3269	10804	10804	10074	43801	730	7687	3075	48788	813		535	278	3269	196168
Na salts	77615	3234	1.8			231	3466	3466	3232	14053	234			322	5.37		3.5	1.8	231	13841
KOH	190	7.93					?	~0	~0	~0	~0			0	0	0	0	0		0
CsOH	4.76	0.198					?	~0	~0	~0	~0			0	0	0	0	0		0
NaNO2	8131	339				24.2	363	363	338	1471	25			34	0.56		0.37	0.19	24	1449
sludge	98.5	4.1					4.1	4.1	0.0	246	4.1			246	4.1	246	4.1	0		0
MST				4.5			4.5	4.5	0.0	270	4.5			270	4.5	270	4.5	0		0
NaTPB (soluble)			49.4			50.0	0.59	0.59	0.55	2.4	0.04			0	0	0	0	0	50.0	3000
NaTPB (insoluble)							?	50.0	0.0	2998	50.0			0	0	0	0	0		0
KTPB (insoluble)							?	50.7	0.0	3042	50.7			3042	50.7	3042	50.7	0		0
CsTPB (insoluble)							?	0.60	0.0	35.8	0.60			35.8	0.60	35.8	0.60	0		0

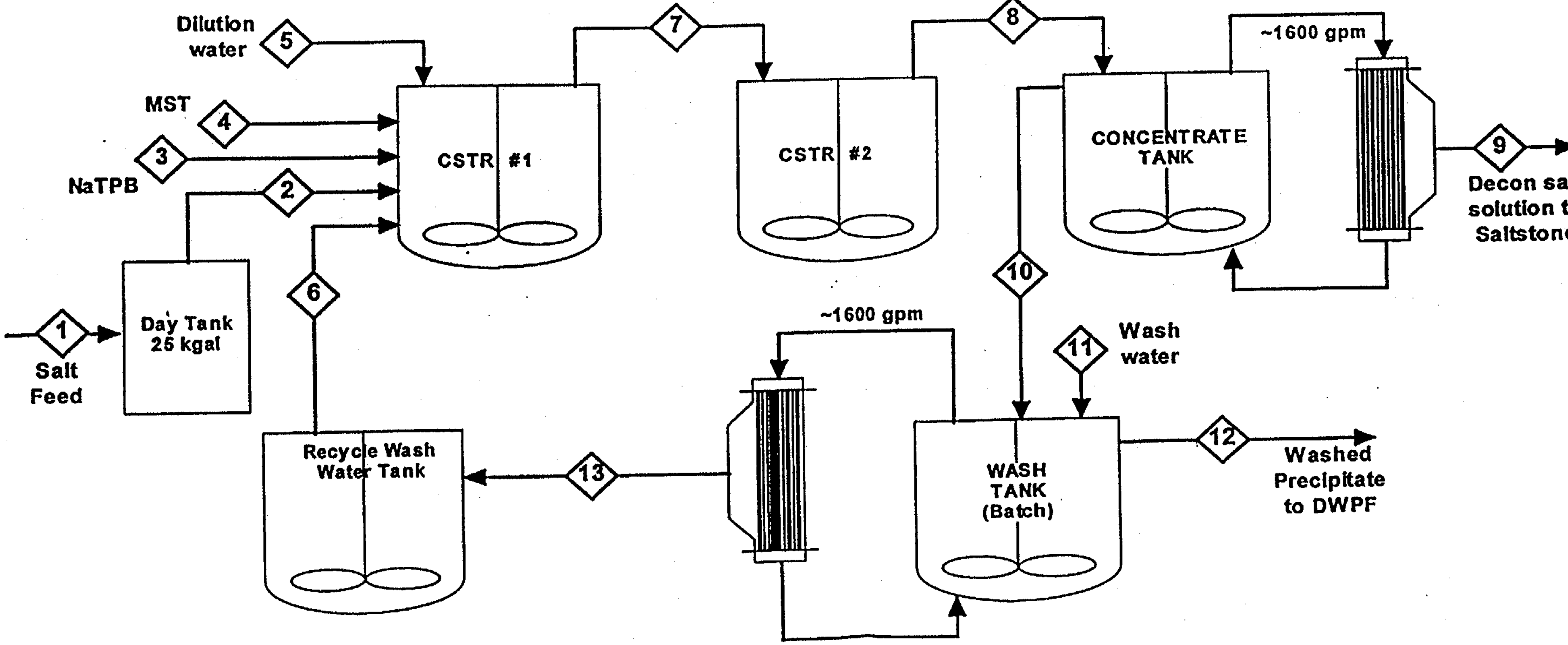


Figure 2
Small Tank Precipitation Process
Benzene Stripping Alternatives

Figure 2A Continuous Stripping in Packed Column –

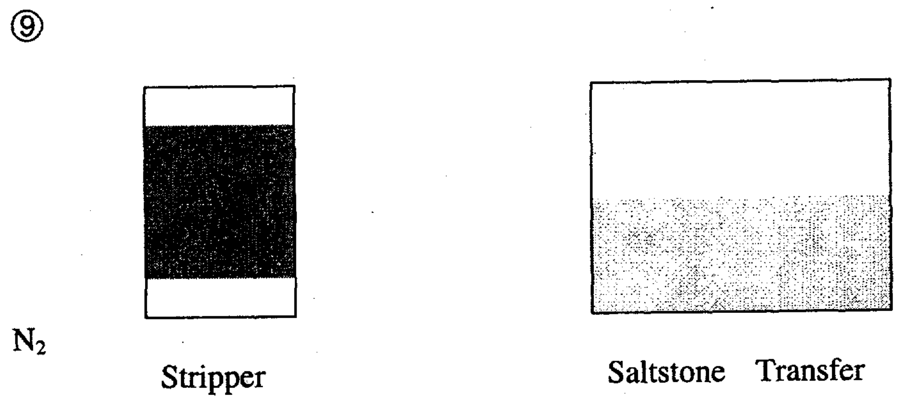


Figure 2B Batch Sparging of Benzene

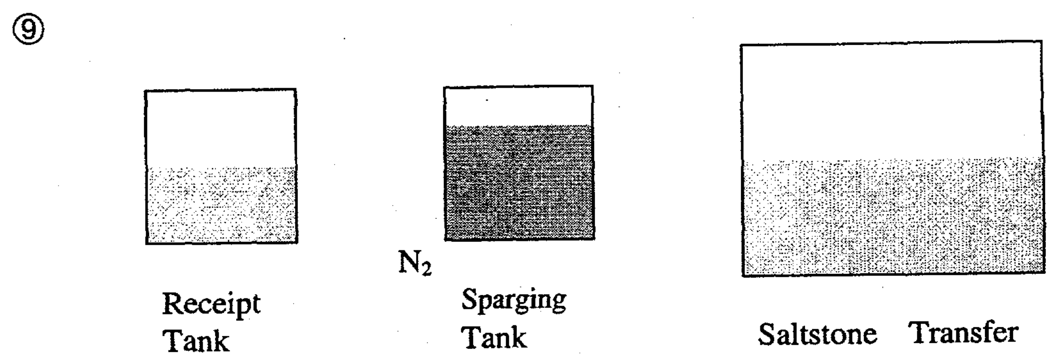


Figure 3 Late Wash Test Data, Run #3.

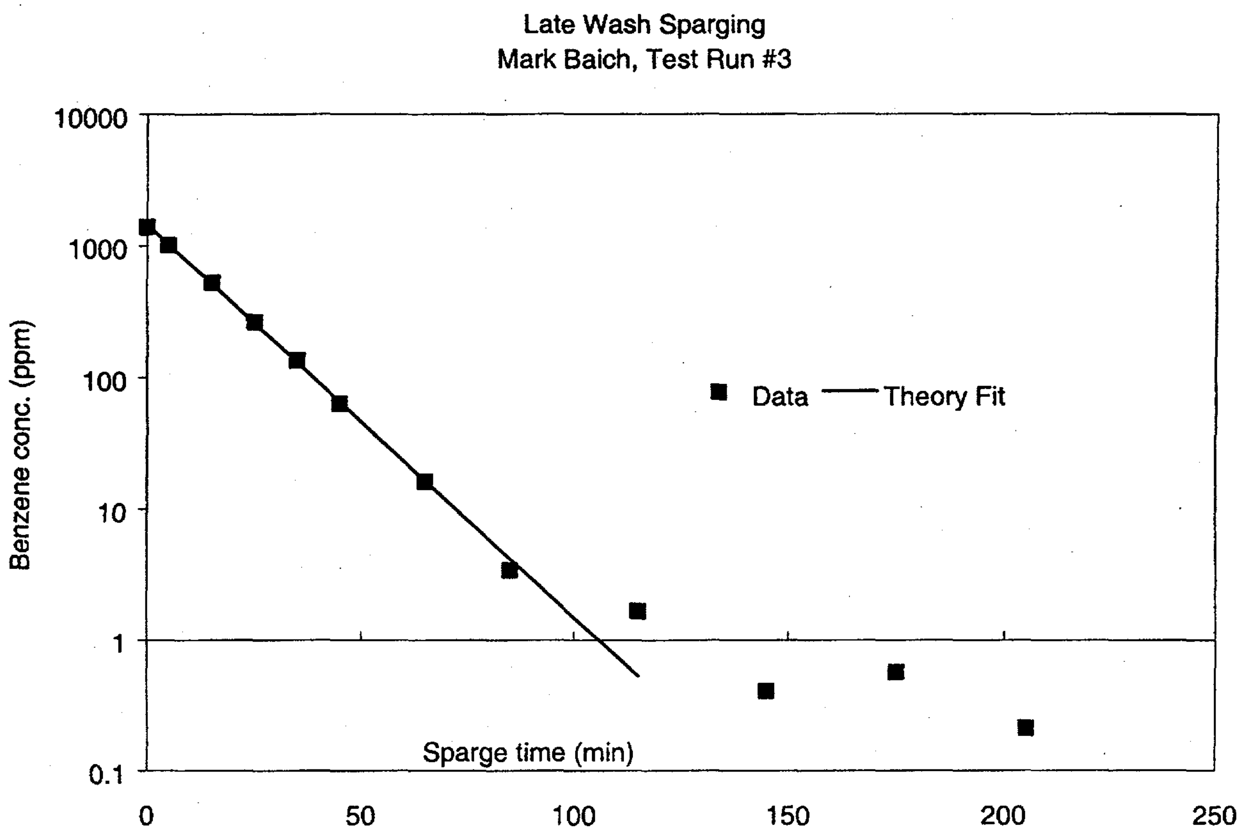


Figure 4 Small Tank TPB Process Sparging

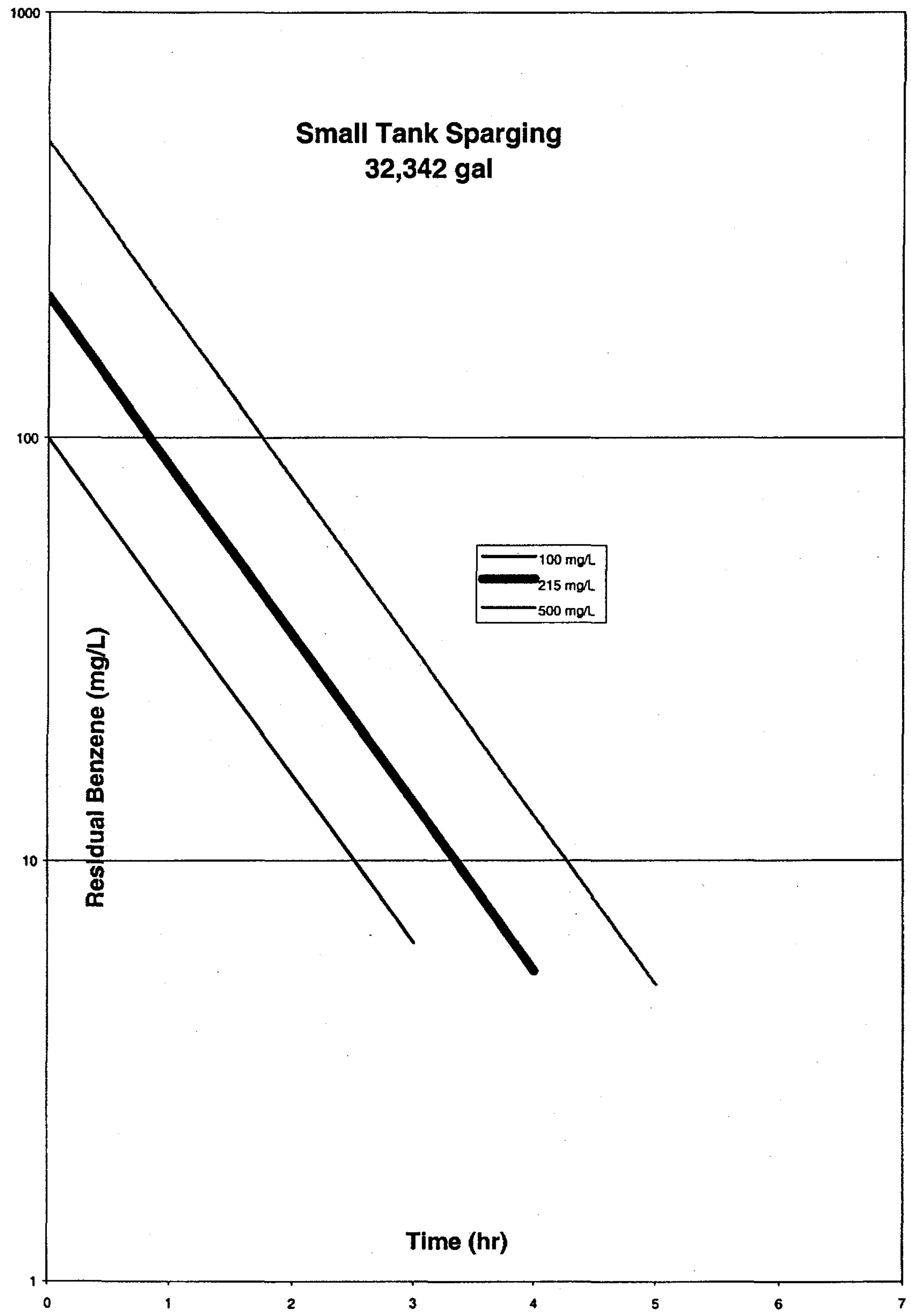
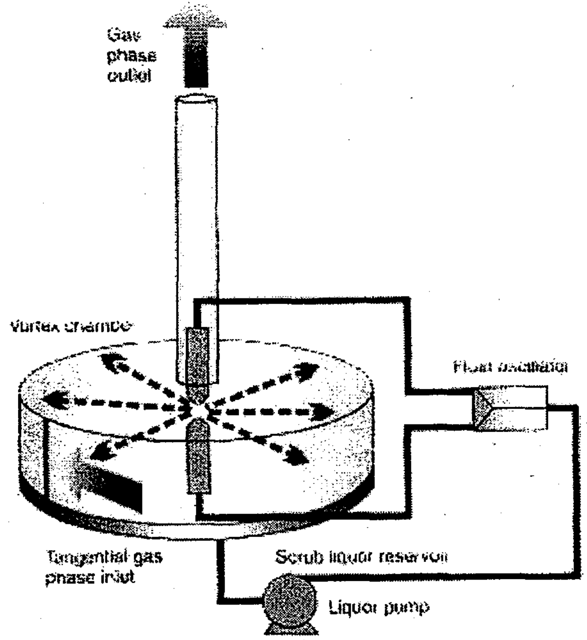
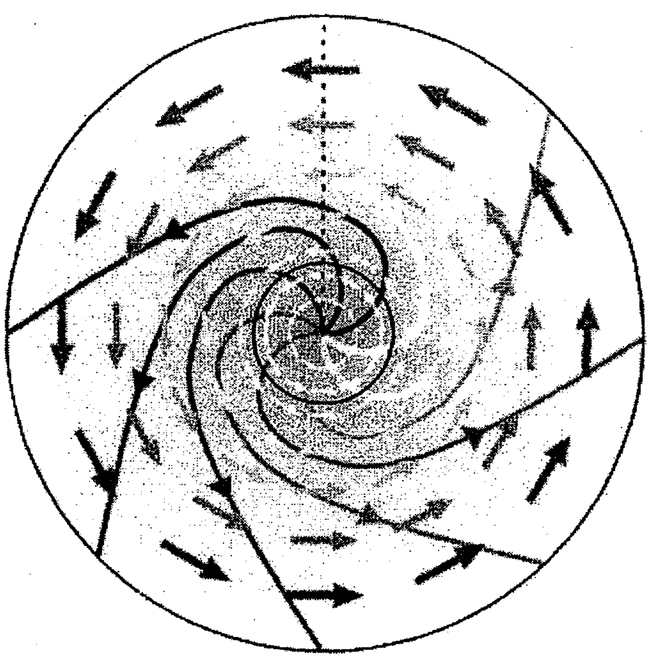


Figure 5 AEA Vortex Scrubber

AEA: *The fluidic gas-liquid contactor uses high tangential gas velocities to provide good mass transfer and disentrain liquid droplets. Vertically-opposed liquid jets supported by fluidic oscillator provide a flat symmetrical pattern of droplets.*



AEA: *Velocity vectors and droplet trajectories within the vortex chamber. As the gas spins towards the center its velocity increases, improving mass transfer and imparting tangential velocity to the liquid droplets.*



APPENDIX A
SPARGING CALCULATIONS

1. Basic Data: Small Tank TPB Precipitation Material Balance, Figure 2.

NaTPB Excess: 100%
Decon Salt Solution Flow: **22.46 gal/min**, Stream 9
Sodium Concentration: **4.70 M**

2. Physical Properties.

Solution Density @ 23°C:

$$\rho = 1.009 + 0.04454\,[Na+] \quad mg/L \quad (Ref.\ 7)$$
$$= 1.009 + 0.04454 * 4.70$$
$$\mathbf{= 1.218\ mg/L}$$

Benzene Solubility @ 23°C:

$$Solubility = 1.7689 * \exp(-0.4481\,[Na^+])\ g/L (Ref.\ 8)$$
$$= 1.7689 * 0.1217 = 0.215\ g/L$$
$$\mathbf{= 215\ mg/L}$$

Henry's Law Constant @ 23°C: (Ref. 8)

Na+	0	1	2	3	3.5	4	4.5	4.9	5	5.5	6
H	0.21	0.32	0.51	0.8	1	1.24	1.56	1.87	1.96	2.45	3.07

Henry's constant, **H = 1.80** (mg/L in Vapor)/(mg/L in Solution) for 4.70 M [Na+]

3 Vapor -Liquid Equilibrium

Partial Vapor Pressure of benzene over salt solution is given by Henry's Law:

$$P_B = H * X_B$$

Where P_B = partial pressure of benzene
X_B = mole fraction of benzene in solution
H = Henry's constant

4 Nitrogen Sparging.

Based on Late Wash solution testing (Ref. 3), the residual benzene concentration in sparged salt solution is given by:

$$C(T) = C0 \exp(-SHET/V)$$

Where C(T) = benzene concentration at time T (min), mg/L

C0 = Initial benzene concentration, mg/L
S = Nitrogen flow, SCFM (Standard ft3/min)
H = Henry's constant
V = Decontaminated solution volume, CS (ft3)
E = Stripping efficiency