Selective Absorption of H_2S from Larger Quantities of CO_2 by Absorption and Reaction in Fine Sprays

Highly selective transfer of hydrogen sulfide (H_2S) from much larger quantities of carbon dioxide (CO_2) has been accomplished by absorption and reaction into fine sprays of buffered carbonate solution. Selectivity after the intensely agitated atomization zone is dramatically improved once the droplets have formed and stagnate internally. H_2S transfer in this latter zone is virtually independent of the presence of CO_2, even at CO_2:H_2S ratios of 100; CO_2 transfer, however, is adversely affected by the presence of H_2S. The gas film resistance for mass transfer is effectively eliminated with the 50 μm mean droplet diameter. The data indicate approximately constant hydrodynamics throughout the formed droplet zone and allow good theoretical prediction of enhancement factors for this complex system. Thus the design of more efficient equipment is possible if the gas-liquid contacting method is tailored to take advantage of differences in the transfer and reaction rate properties of gases to be separated.

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SCOPE

The need for a selective separation of H_2S from CO_2 occurs as it is often too expensive and not necessary to remove large quantities of CO_2 along with the pollutant H_2S. Often no process is capable of removing the H_2S to specifications without uneconomical removal of the majority of the CO_2, thus requiring a large purification plant and large capital investment. Situations in which H_2S and larger quantities of CO_2 appear include low to medium BTU gas wells or natural CO_2 sources, and in fossil fuel plants operating under partial oxidative environments.

There have been few experimental or theoretical studies of mass transfer into a dispersed medium with subsequent intraphase reaction. Applications include topics in liquid-liquid extraction and gas purification where single components are transferred or multiple competitively or synergistically interacting components are transferred. Description of the fluid dynamics for such systems is complicated by the existence of three different regimes always present: atomization, droplet flight, and coalescence. Many atomization techniques exist and none are well-described for sprays. Atomization greatly affects the droplet flight regime as it determines the droplet size distribution, dynamics and interaction. Continuous phase backmixing and internal dispersed-phase mixing further can complicate the theoretical description of the fluid dynamics.

This article contains a summary of an experimental study on selective removal of H_2S from much larger quantities of CO_2 using a conventional liquid solution but sprayed into a chamber as a fine mist. The primary variable between runs was the amount of liquid reagent in solution. Gas and liquid samples were taken throughout the reactor.

CONCLUSIONS AND SIGNIFICANCE

There is a significant increase in selective and extensive transport of H_2S over CO_2 if the scrubbing liquid is finely divided. This advantage is particularly great outside of the atomization zone because CO_2 is physically absorbed quite well in the intense mixing of the atomization zone but does not compete well with H_2S once the droplets have been formed and become stagnant inside. This could lead to improved nozzle design for such applications. H_2S removal from CO_2 was found to be uninfluenced by the presence of CO_2:H_2S ratios of 100, the highest studied; even more severe ratios may yield similar results. With the gas film resistance effectively eliminated and with stagnant liquid droplets, it appears that a simple theoretical model may be used to adequately describe performance and for design.

Mass transfer and reaction into finely dispersed media is a relatively new area for study and may lead to innovation in unit operation. Application of the present study may be made to other systems in which focusing attention on the gas-liquid contacting to best utilize differences in reaction rates for selective transport could be important; for example, flue gas desulfurization with selective removal of SO_2 from CO_2.

INTRODUCTION

It has been known for many years that for transfer of H_2S and CO_2 into a reactive liquid in a typical contacting device, such as a packed-bed, the gas-side resistance to mass transport is dominant for H_2S and the liquid-side resistance for CO_2. Such a difference in the mass transport mechanisms is fortunate to exist as these acid gases are similar in many physical and chemical respects and thus difficult to separate. If the gas-side mass transfer resistance can be substantially reduced by fine liquid dispersion it would increase the extent of H_2S removal. If the liquid film can be made limiting

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Table 1. Survey of Processes for Selective Removal of Hydrogen Sulfide from Carbon Dioxide

Selective Process	Separation Media	Comments
Established		
Chemical Absorption		
Stretford	NA_2CO_3, plus additives (Vasan, 1978)	
Benefield	Catalyzed hot K_2CO_3 (Benson et al., 1956)	
Alkazid "DIK"	Potassium salt of diethylglycine (Wainwright et al., 1953; Pasternak, 1962)	
Giammarco-Vetrocoke	Arsenic-activated K_2CO_3 (Riesenfield and Blohm, 1962)	Arsenic hard to remove completely
Dow Chemical	Caustic process (Hohfeld, 1980)	
MDEA	Methydiethanol amine (Goar, 1980; Vidaurri and Kahre, 1977)	
Adip	MDEA or diisopropanol amine, DIPA (Ouwerkerk, 1978)	
Physical Absorption		
Selexol	Dimethyether of polyethylene	Solubility differences only a factor of 10 and directly proportional to partial pressures
Sulfinol	DIPA and Sulfulane	Expensive solution giving very low yields
Physical Adsorption		
Dry Box	Fe_2O_3	Regeneration or disposal of large sulfur tonnage is impractical
Oxorban (Lurgi)	Activated carbon	Maintaining continuous high absorption efficiency very difficult
Not Currently Commercial		
Liquid oxidation:		
UOP U.S. Patent 3,859,414 (1975)	Sodium sulfide/bisulfide	Low capacity for hydrogen sulfide
Physical adsorption:		
Molecular sieves	Molecular size and polarity differences	Regeneration problems; very high sieve initial investment; continuous high absorption efficiency
Low-temperature fractionation	Relative volatility difference	Is difficult to sustain; difficult design; high investment
Liquid membranes	Facilitated transport	Thin-film saturation

for *both* components, selectivity could possibly be increased by manipulation of the internal droplet fluid flow and reactant environment.

The term "selectivity" for removal of H_2S over that for CO_2 in a wet reactive scrubbing process has been recently defined to mean the quotient of the enhancement factor for H_2S to that for CO_2 (Aiken, 1982). However, in this paper, we shall use this term in the more usual qualitative sense, and will not calculate a selectivity factor. Selective removal processes for this application, established or new, are listed in Table 1. Wet scrubbing processes may be divided into two main classes: physical or chemical absorption. A process that makes effective use of both effects is not available. None of the processes cited can be considered superior for most selective removal applications. In particular, none of these processes could economically remove H_2S from a natural gas at 78.6 atm with 7% CO_2 and 0.01% H_2S to 4 ppm H_2S (0.0004%).

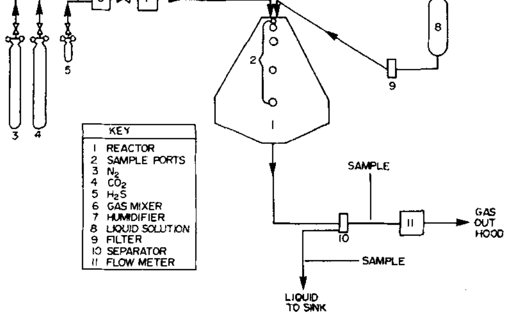

Figure 1. Spray reactor system.

Anderson and Johnstone (1955) discussed experiments on absorption and liquid phase reaction of oxygen and sulfur dioxide with catalyzed sodium sulfide solution in a venturi atomizer, but no analysis is given. Liquid combustion, fuel spray dynamics, and interaction with oxident injection has been extensively studied, although viable models are highly oversimplified (Williams, 1965); reaction in this case occurs in a spherical vapor shell outside the droplet or on the droplet surface (Bennett and Kayser, 1978). The Colstrip (Grimm et al., 1978) and Thiosorhic (Seleczi and Stewart, 1978) flue gas desulfurization process are examples of gas purification processes in which a gaseous species, in this case sulfur dioxide, is absorbed and reacted in sprays.

EXPERIMENTAL EQUIPMENT AND OPERATING PROCEDURES

Reactor System

The reactor and supporting equipment is shown in Figure 1. Scrubbing liquid and sour gas mixture enter the reactor top co-currently. Liquid is atomized in the reactor by a pressure nozzle (Spraying Systems #1/4 LN.60) at a pressure drop of 6.8 atm for a flow rate of 63.75 mL/min. This gave a spray with a most probably diameter of about 50 μm, as indicated in Figure 2, taken from Spraying Systems Corporation literature (data taken an unknown distance from nozzle, 1978). This spray is much finer than typically found in industrial spray scrubbers.

A thin liquid film extends directly outside the nozzle for about 1 cm distance, becomes unstable, and forms drops very soon thereafter. Liquid spray did not directly hit the sides of the reactor but coalesced upon meeting the conical bottom, whereafter it exited together with the gas. The gas mixture was prepared with combinations of H_2S, CO_2, or N_2 by means of a Matheson gas mixer together with high precision metering values. The gas was saturated with water before entering the reactor by passing it through a frit immersed in a column of water. A gas flow rate of 14 L/min was high enough so that the gaseous H_2S concentration in the reactor did not greatly change even for low inlet percentages. Total pressure in the reactor was just slightly in excess of atmospheric. Scrubbing solution was

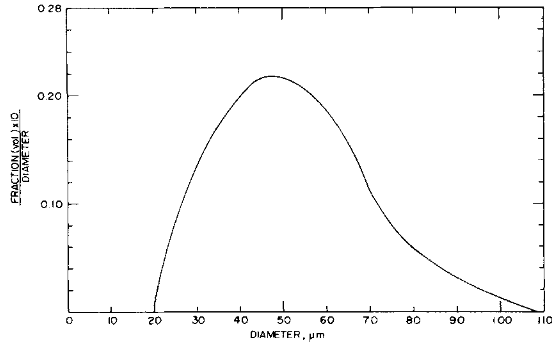

Figure 2. Droplet-size distribution (adapted from Spraying Systems Corp., data).

prepared and forced through the nozzle by keeping a constant N_2 pressure in the tank containing the liquid. Liquid solution always was 0.5M in Na^+, on the order of 80% saturation, and was filtered prior to entering the nozzle.

Sampling

Both gas and liquid were sampled at five ports within the reactor, located at distances of 0.74, 2.54, 7.62, 22.86 and 48.26 cm from the nozzle (or 0.3, 1, 3, 9, and 19 in., respectively); some samples were also taken at the reactor exit. Gas samples were taken by a thin downward pointing small diameter probe that avoided two-phase flow and coalescence at the probe tip. Axial as well as radial scans were taken to assure approximately uniform gas concentration. Collected gas was sent directly into a Tutweiler titration apparatus. Liquid samples were taken by two methods: Port 1 samples by a syringe, cut and lipped, set directly into the film near its edge. For all other ports, the liquid was collected by freezing the droplets on a liquid N_2-cooled probe, held and rotated several seconds inside the reactor. Approximately 2 mL samples could be collected in this manner. Details of sampling techniques may be found in Bendall (1981).

Analysis of Samples

Gas samples (about 50 mL each) were analyzed by the Tutweiler titration technique (Bendall, 1981). H_2S absorbed in the liquid was measured by an Orion ion-selective electrode and meter. Absorbed CO_2 in carbonate buffers is difficult to measure. Gas evolution and conductivity were found too inaccurate. CO_2 ion-selective electrodes do not operate at the ionic strength of most of our runs. A special conductivity-titration technique for comparison of CO_3^{-2} and HCO_3^- breaks with corresponding clean feed solution appears to give acceptable accuracy (Furse, 1981).

Chemical and Physical Properties

Once CO_2 and H_2S have physically absorbed into the liquid the following reaction occur within a carbonate buffer solution:

$$H_2S + OH^- = HS^- + H_2O \tag{1a}$$

$$H_2O = H^+ + OH^- \tag{1b}$$

$$CO_3^{-2} + H^+ = HCO_3^- \tag{1c}$$

$$H_2S + CO_3^{-2} = HS^- + HCO_3^- \tag{1d}$$

$$CO_2 + OH^- = HCO_3^- \tag{1e}$$

$$CO_2 + H_2O + CO_3^{-2} = 2HCO_3^- \tag{1f}$$

Equations 1b and 1c can be considered truly instantaneous (unmeasurably fast) while Eq. 1a is not by comparison, although extremely fast (the forward rate constant of Eq. 1a has been measured to be 10^{10} l/mol·sec, Karmann, 1967). Equation 1d is equivalent stoichiometrically at equilibrium to Eqs. 1a–c; equilibrium for Eqs. 1a and 1d are very much favored to the right (equilibrium constants of 9×10^6 and 2×10^3, respectively, at 25°C). Far from equilibrium, we take Eq. 1d to be the important reaction since $[CO_3^{-2}] \gg [OH^-]$ in all cases of practical interest here. Eq. 1f is stoichiometrically equivalent at equilibrium to Eqs. 1b, 1e, and 1c, but unlike Eq. 1d, the analog in the H_2S reaction sequence, is not an elementary reaction. Also Eq. 1e is much slower than Eq. 1a (Pinsent et al., 1956 give a value of 5,900/mol·s for the forward rate constant of Eq. 1e at 20°C and infinite dilution). We take advantage of these differences in reaction speeds of H_2S versus CO_2 in our experiments for selective removal.

Henry's constant for H_2S physical solubility at 25°C was taken as (Bendall, 1981):

$$H = 540\,(10^{0.079I})\text{ atm}$$

where I = ionic strength

RESULTS OF EXPERIMENTS AND DISCUSSION

Table 2 contains a summary of our experimental runs; most entries are averages of several data (Bendall, 1981). In particular,

TABLE 2. EXPERIMENTAL DATA

	Gas (atm)				Liquid (mol/L)			
	Run	H_2S^a	CO_2^b	N_2	H_2S	CO_2	CO_3^{-2c}	pH^d
Inlet	3	0.0105	1.0		0.0	0.0	0.179	9.41
Port #: 1		0.005	1.0		4.40E-3	1.06E-2		9.35
2		0.005	1.0		1.28E-2	1.5E-2	0	9.30
3		0.005	1.0		1.57E-2	1.7E-2		9.28
4		0.005	1.0		2.38E-2	2.8E-2		9.20
5		0.005	1.0		3.56E-2			9.10
Exit		—	—		—	—		—
Inlet	5	0.0104	1.0		0.0	0.0	0.553	11.30
Port #: 1		0.0015	1.0		6.68E-3	3.64E-2		10.40
2		0.0015	1.0		1.95E-2	2.5E-2		10.45
3		0.0015	1.0		2.76E-2	2.0E-2		10.45
4		0.0015	1.0		5.12E-2	4.7E-2		10.12
5		0.0015	1.0		7.42E-2	3.4E-2		10.13
Exit								
Inlet	6	0.0112	1.0		0.0	0.0	0.465	10.46
Port #: 1		0.002	1.0		4.21E-3	4.7E-2		10.11
2		0.002	1.0		1.88E-2	3.3E-2		10.14
3		0.002	1.0		2.94E-2	3.4E-2		10.10
4		0.002	1.0		5.36E-2	3.9E-2		10.0
5		0.002	1.0		7.84E-2	4.7E-2		9.9
Exit		—	—		—	—		—
Inlet	7	0.0105	1.0		0.0	0.0	0.0724	8.99
Port #: 1		0.0080	1.0		5.08E-3	2.7E-2		8.69
2		0.0080	1.0		9.92E-3	3.0E-2		8.59
3		0.0080	1.0		1.28E-2	3.1E-2		8.52
4		0.0080	1.0		1.65E-2	2.4E-2		8.58
5		0.0080	1.0		1.89E-2	2.0E-2		8.60

TABLE 2. *(Continued)*

	Run	Gas (atm)			Liquid (mol/L)			
		H_2S^a	CO_2^b	N_2	H_2S	CO_2	$CO_3^{-2\,c}$	pH^d
Exit								
Inlet	8	0.0100	1.0	1.0	0.0		0.169	9.41
Port #: 1		0.0044		1.0	4.27E-3			9.40
2		0.0044		1.0	1.42E-2			9.35
3		0.0044		1.0	1.86E-2			9.31
4		0.0044		1.0	2.67E-2			9.29
5		0.0044		1.0	3.56E-2			9.22
Exit		—		—	—			—
Inlet	10		1.0			0.0	0.167	9.40
Port #: 1			1.0			2.06E-2		9.30
2			1.0			3.00E-2		9.24
3			1.0			3.25E-2		9.24
4			1.0			3.25E-2		9.22
5			1.0			—		—
Exit			—			—		—
Inlet	11-12	0.0107		1.0	0.0		0.25	10.46
Port #: 1		0.0019		1.0	3.54E-3			
2		0.0019		1.0	1.30E-2			
3		0.0019		1.0	2.26E-2			
4		0.0019		1.0	3.16E-2			
5		0.0019		1.0	5.42E-2			
Exit		—		—	—			
Inlet	19	0.0101	1.0		0.0	0.0	0.02508	8.52
Port #: 1		0.0081	1.0		3.03E-3	5.7E-3		—
2		0.0081	1.0		3.45E-3	5.0E-3		—
3		0.0081	1.0		3.83E-3	4.9E-3		8.30
4		0.0081	1.0		4.24E-3	2.9E-3		8.31
5		0.0081	1.0		4.47E-3	4.2E-3		8.34
Exit		0.0081	1.0		7.51E-3	14.E-3		7.69
Inlet	20	0.0112		1.0	0.0			14.00
Port #: 1		0.00013		1.0	2.8E-3			(1M naOH)
2		0.0013		1.0	1.5E-2			
3		0.00013		1.0	2.8E-2			
4		0.00013		1.0	5.26E-2			
5		0.00013		1.0	7.65E-2			
Exit		0.00013		1.0	8.87E-2			
Inlet	21	0.0047		1.0	0.0			7.0
Port #: 1		0.0047		1.0	1.54E-4			(pure
2		0.0047		1.0	5.95E-4			H_2O)
3		0.0047		1.0	2.23E-4			
4		0.0047		1.0	2.54E-4			
5		0.0047		1.0	2.76E-4			
Exit		—		—	—			
Inlet	25	0.0136	1.0		0.0		1.0	(1M CO_3^{-2})
Port #: 1		0.002	1.0		4.49E-3			
2		0.002	1.0		2.32E-2			
3		0.002	1.0		3.25E-2			
4		0.002	1.0		6.99E-2			
5		0.002	1.0		0.107			
Exit								

[a] Values within the reactor represent an average.

[b] Approximate, not including H_2S^a H_2O vapor.

[c] Calculated.

[d] Measured, must be corrected for actual.

a single averaged value is given for H_2S gas concentration within the reactor. Runs 25 and 20 were conducted at very high reactant concentrations for evaluation of the gas film; Run 21 used pure water for the scrubbing medium in order to evaluate the liquid film for physical mass transfer (see ANALYSIS OF EXPERIMENTS). Runs 3, 5, 6, 7, and 19 were made to record the selectivity and extent of $H_2S - CO_2$ removal. Runs 8 and 11–12 used H_2S in N_2 to examine the effect of the competition of CO_2 or H_2S transfer; Run 10 had pure CO_2 inlet gas and illustrates the competitive effect of H_2S when compared with Run 3.

The total H_2S absorbed for each port is plotted in Figure 3. All runs seem to have the same approximate absorbed H_2S concentration at the nozzle; this is an indication that liquid composition, the main variable among the runs, is not important here and that physical absorption is the main mode of mass transfer. After the nozzle zone, the amount of H_2S absorbed increases dramatically in short distances. This rate of increase tapers off with distance but does not come to an asymptote for most runs; in this respect reactor length does seem to be important. The runs with the smaller slopes on the figure, all have the smaller $[CO_3^{-2}]$ at inlet as can be seen by reference to Table 2. This is not true for inlet pH; for example, consider Runs 6, and 11–12 both at a pH of 10.46: Run 6 with $[CO_3^{-2}]$ inlet of 0.465M is well above Run 11–12 ($[CO_3^{-2}]$ inlet of 0.25M) in Figure 4 and has a larger slope.

Figure 4 indicates competitive effects of CO_2 and H_2S transfer. The H_2S absorbed from Run 3 (CO_2 present) is compared with that from Run 8 (inert N_2 substituted for CO_2), all other parameters approximately constant. Within experimental error, the extent of absorption is identical in the two runs. Therefore, the presence of 100 times as much CO_2 as H_2S in the gas apparently does not interfere with H_2S absorption in this buffered solution. The corresponding effect on CO_2 absorption is also shown in this figure; Run

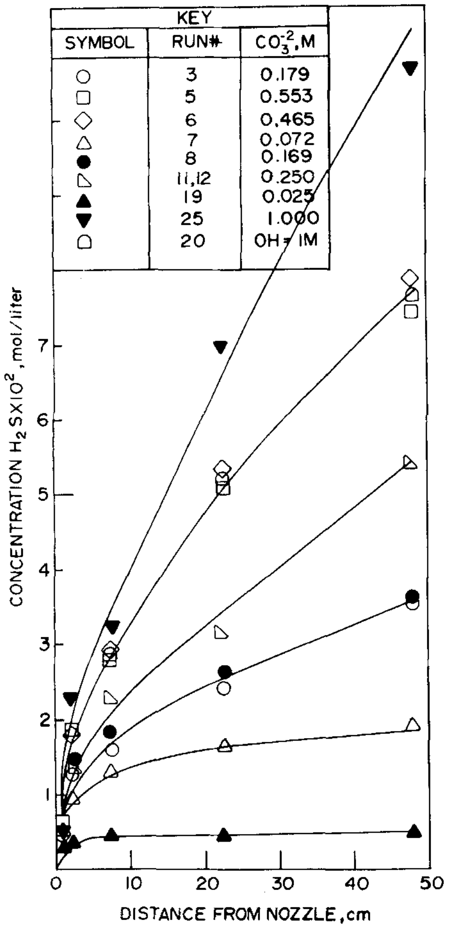

Figure 3. Absorption of hydrogen sulfide in spray reactor.

10 has no H_2S present and is equivalent in all other respects to Run 3 which has the CO_2-H_2S mixture. The CO_2 absorption is seen to be quite adversely affected by the presence of H_2S.

ANALYSIS OF EXPERIMENTS

Mass Transfer Coefficient

We define in the usual way the rate of transfer from the bulk gas at P_A (atm) to bulk liquid at A_o (mol/cm^3) as:

$$R = k_g(P_A - P_{A_i}) = Ek_L^o(A^* - A_o) \tag{2}$$

We cannot measure R from experiment. Thus define

$$a \equiv \frac{\text{interfacial area liquid}}{\text{volume of liquid}}$$

and multiply Eq. 2 by a:

$$Ra = k_ga(P_A - P_{A_i}) = Ek_L^oa(A^* - A_o) \tag{3}$$

we *can* measure Ra indirectly from experiment. We assume equilibrium between P_{A_i} and A^* at all times, given by Henry's Law:

$$P_{A_i} = HA^*/\rho \tag{4}$$

where ρ = molar density of water, 0.0555 mol/cm^3, and we assume moles $A \ll$ moles water. Danckwerts' surface resistance criterion (Danckwerts, p. 66, 1970) suggests this to be true for our spray after approximately 10^{-7} seconds (Bendall, 1981). Substitution of Eq. 4 into Eq. 3 gives

$$Ra = K_LaA \tag{5}$$

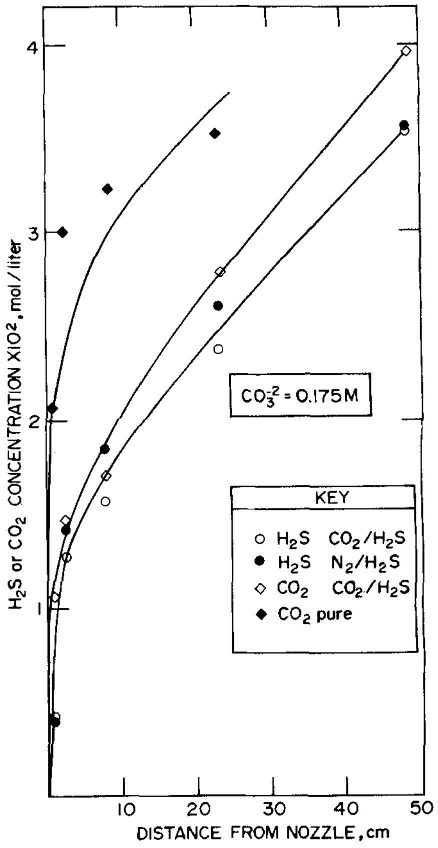

Figure 4. Competitive removal of hydrogen sulfide and carbon dioxide.

where

$$K_La = \frac{1}{\dfrac{1}{Ek_L^oa} + \dfrac{\rho}{Hk_ga}} \tag{6}$$

is the overall liquid coefficient and

$$A \equiv P_A\rho/H \tag{7}$$

is the concentration that would exist in the liquid if it were in equilibrium with the bulk gas; $A = 0$ for H_2S in the bulk. A case can be made for use of K_G rather than K_L however, because of the uncertain H of a chemically reactive system. Because we estimate this term later from experiment, this will not matter. Now define $\bar{A}$ = mixing-cup or lumped concentration of A in liquid in unreacted form so that

$$\frac{d\bar{A}}{dt} = Ra. \tag{8}$$

We now make the assumption that $\bar{A}$ is approximately the total amount of absorbed A, *viz.* the amount of A in the film is insignificant compared to the amount in the bulk. This is still not a useful form for data analysis since we cannot directly measure exposure time of the droplets at our sampling points.

In order to relate time and distance, the latter which we can measure, several different average droplet velocity experiments were conducted. Chopper experiments, double exposed still photography, and high-speed movies (Fastrax Camera, 1,000 frames/s) gave data that were averaged to produce the velocity profile in Figure 5. The magnitudes of the velocities are approximate only, a major limitation of our measurement technique being that clusters or sheets of droplets could only be monitored rather than individual droplets. The velocity at the nozzle was calculated (Bendall, 1981) from consideration of the liquid flow rate and the

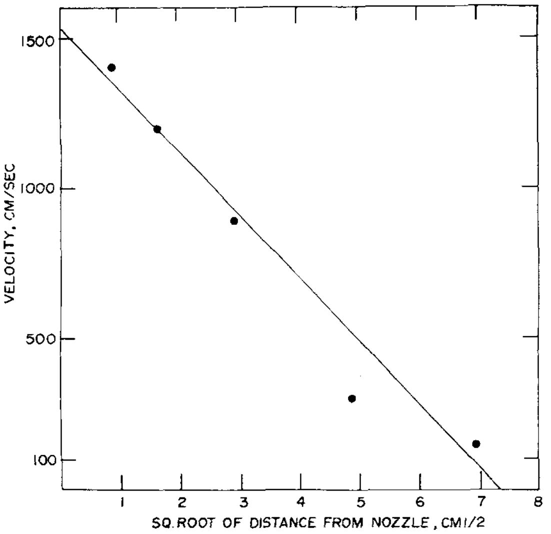

Figure 5. Average aerosol velocity in spray reactor.

orifice size including a small hollow core as measured by Arena (1973). The functional dependence is apparent from Figure 5:

$$v = v_o(1 - \alpha x^{1/2}) = v_o - v_o\alpha x^{1/2} \tag{9}$$

where v_o = velocity at orifice ($x = 0$). Since

$$v = v_o - at \tag{10}$$

where a = deceleration, we can identify time from Eqs. 9 and 10 as

$$t = \frac{v_o \alpha x^{1/2}}{a} \equiv \alpha^* x^{1/2}. \tag{11}$$

α^* has units of s/cm$^{1/2}$.

Equation 11 substituted into Eq. 8 then gives

$$\frac{dA}{dx^{1/2}} = Ra\alpha^* = K_L a\alpha^* A = \frac{(P_A/H)}{\dfrac{1}{Ek_L^o a\alpha^*} + \dfrac{\rho}{Hk_g a\alpha^*}} \tag{12}$$

The transport coefficient $K_L a\alpha^*$ is calculated from:

$$K_L a\alpha^* = \frac{1}{A}\frac{dA}{dx^{1/2}} \tag{13}$$

and are reported in Table 3. The fit of A vs. $x^{1/2}$ is quite linear for all ports after Port 1, which is in the film region and for which the velocity correlation does not apply, Figure 6 (Mandas, 1981). A was calculated from the average H_2S gas compositions of Table 2 and the Henry's constant given earlier.

The fact that A varies as $x^{1/2}$ throughout the reactor suggests that the hydrodynamics are constant and allows a single mass transfer coefficient to describe the spray reactor of formed droplets. Note the very similar $K_L a\alpha^*$ for runs 3 and 8, again demonstrating independence of H_2S on CO_2 transfer ($K_L a\alpha^*$ for run 3 is slightly greater because of a slightly higher $[CO_3^{-2}]$).

TABLE 3. OVERALL MASS TRANSFER COEFFICIENT FOR H_2S

Run	$K_L a\alpha^*$	CO_3^{-2} (Inlet) M
3	9.78	0.179
5	97.3	0.553
6	78.1	0.465
7	2.04	0.0724
8	9.24	0.169
11-12	45.6	0.25
19	0.26	0.025
20	121	OH^- = 1M
21	0.019	0
25	141	1

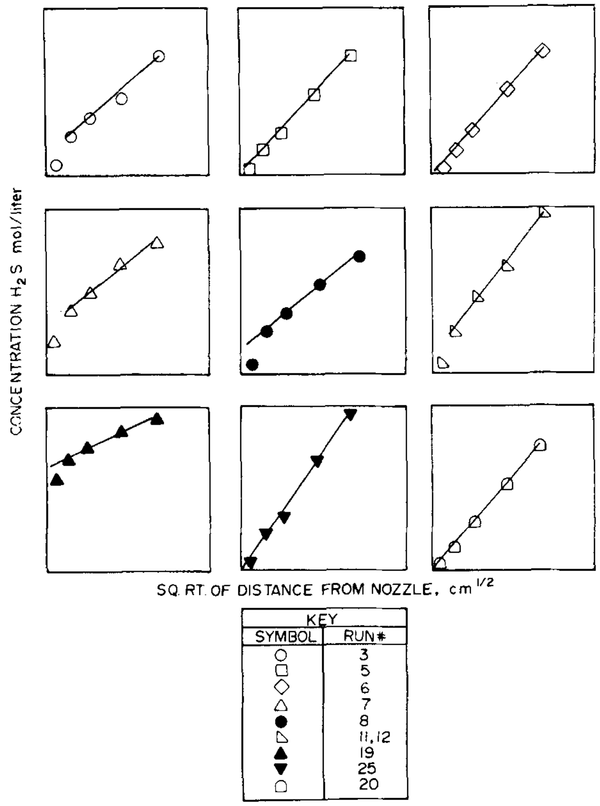

Figure 6. Concentration dependence with square root of distance for various runs.

Individual Film Resistances

With the overall coefficient $K_L a\alpha^*$, we wish now to determine the two contributing terms $1/Ek_L^o a\alpha^*$ and $1/Hk_g a\alpha^*$ corresponding to liquid and gas film resistances, respectively. We may examine a single term if the other is negligible. To obtain an estimate of the gas film term, for example, we could use conditions of minimum liquid phase resistance, like run 25, which had very concentrated reactant. This gives as an upper limit

$$\frac{1}{K_L a\alpha^*} = \frac{\rho}{Hk_g a\alpha^*} = 0.0071/\text{cm}^{1/2} \tag{14}$$

Inspection of Table 3, with runs 25 and 20 (both at high reactant concentration) representative of gas film influence, only runs 5 and 6 could have a significant gas film resistance. We shall support the assumption that Eq. 14 is true in the next section.

The liquid film resistance varies with the amount of reactant, since this affects E. We consider the gas film negligible compared to liquid film resistance for physical absorption, where also E = 1. Thus from run 21, as a lower limit

$$\frac{1}{k_L^o a\alpha^*} = 52.6/\text{cm}^{1/2}$$

Enhancement Factor

Now that we have an estimate for gas film and physical liquid film coefficients we may calculate the only other unknown parameter in Eq. 12, E the enhancement factor to physical mass transfer by virtue of reaction:

$$E = \left\{k_L^o a\alpha^* \left[\frac{1}{K_L a\alpha^*} - \frac{\rho}{Hk_g a\alpha^*}\right]\right\}^{-1} \tag{15}$$

Equation 15 represents the *experimental* enhancement factor for H_2S absorption as correlated by the two film theory. A rudimentary theory to predict H_2S absorption enhancement for a single component reacting instantaneously with a second nonvolatile component (equal diffusivities) in a stagnant semi-infinite liquid is

TABLE 4. ENHANCEMENT FOR H_2S TRANSFER

Run	*E* Experimental, Eq. 15	*E* Theoretical, Eq. 16
3	527	412
5	5,710	4,850
6	4,490	2,980
8	498	442
7	109	101
17	5,950	8,250
19	14	34
11-12	2,540	1,480
21	1	1

given for example, by Danckwerts (p. 111, 1970):

$$E = \left(1 + \frac{[CO_3^{-2}]}{A}\right) \tag{16}$$

We expect this prediction may be valid since H_2S transfer has been shown to be unaffected by the presence of CO_2. A comparison of the theoretical and experimental E is given in Table 4 and is seen to be in agreement for most runs. The agreement is rather remarkable, considering the complexity of the system and the assumptions behind Eq. 16.

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NOTATION

A = liquid concentration that exists in liquid if it were in equilibrium with the bulk gas, mol/cm^3
A^* = interfacial concentration of A, mol/cm^3
$\bar{A}$ = mixing cup or lumped concentration of A in the liquid, mol/cm^3
a = ratio of interfacial area of liquid to volume of liquid, cm^{-1}
E = enhancement factor, dimensionless
H = Henry's constant, atm
I = ionic strength
K_L = overall mass transfer coefficient based on liquid, Eq. 6, cm/s
k_L^o = physical absorption coefficient, cm/s
k_g = gas film mass transport coefficient, mol/s·cm^2 atm
P_{A_i} = interfacial (gas side) partial pressure of A, atm
R = instantaneous or steady-state mass transfer rate, mol/s·cm^2
v = Velocity of droplet cloud, cm/s
v_o = Velocity of liquid at orifice, cm/s

Greek Letters

α^* = empirical parameter relating time of average droplet in reactor with distance from nozzle, Eq. 11, s/cm$^{1/2}$
ρ = molar density of water, mol/cm^3

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