

APPENDIX B

APPENDIX B

B.1 The Single-Parameter Wilson Equation

The original Wilson equation relates the activity coefficient to the composition of the mixture. For a binary mixture of components 1 and 2 having a composition of X_1 and X_2 respectively it is written as:

$$\ln \gamma_1 = -\ln(1 - A_{21}X_2) + X_2 \left(\frac{X_2 A_{12}}{1 - A_{12}X_1} - \frac{X_1 A_{21}}{1 - A_{21}X_2} \right) \quad (\text{B-1})$$

$$\ln \gamma_2 = -\ln(1 - A_{12}X_1) - X_1 \left(\frac{X_2 A_{12}}{1 - A_{12}X_1} - \frac{X_1 A_{21}}{1 - A_{21}X_2} \right) \quad (\text{B-2})$$

where

$$A_{21} = 1 - \frac{\bar{V}_2}{\bar{V}_1} \exp(-(g_{21} - g_{11})/RT) \quad (\text{B-3})$$

$$A_{12} = 1 - \frac{\bar{V}_1}{\bar{V}_2} \exp(-(g_{12} - g_{22})/RT) \quad (\text{B-4})$$

$$g_{12} = g_{21} \quad (\text{B-5})$$

$\bar{V}_1$ and $\bar{V}_2$ are the molar volumes. The adjustable constants g_{11} , g_{22} and g_{12} are proportional to the interaction energies between either similar or dissimilar molecules. It is clear that in order to calculate activity coefficient at a composition g_{11} , g_{22} and g_{12} are the 3 basic parameters need to be known.

For g_{11} and g_{22} the energy of interaction between similar molecules, it has been proposed to be replaced by the opposite of the energy of vaporization of the pure components at the solution temperature and pressure.

$$g_{11} = -(\Delta H_{v1} - Z_1 RT) \quad (B-6)$$

$$g_{22} = -(-\Delta H_{v2} - Z_2 RT) \quad (B-7)$$

ΔH_{v1} and ΔH_{v2} , the heat of vaporization, can be related to vapor pressure by the Classius-Clapeyron equation:

$$\frac{d \ln P_1^s}{dT} = \frac{\Delta H_{v1}}{RT^2} \quad (B-8)$$

$$\frac{d \ln P_2^S}{dT} = \frac{\Delta H_{V2}}{RT^2} \quad (B-9)$$

substituting equation (B-8) and (B-9) into (B-6) and (B-7) results in:

$$g_{11} = -RT^2 \frac{d \ln P_1^S}{dT} + Z_1 RT \quad (B-10)$$

$$g_{22} = -RT^2 \frac{d \ln P_2^S}{dT} + Z_2 RT \quad (B-11)$$

The expression of vapor pressure in terms of temperature can simply be found by well-known Antoine equation:

$$\ln P^S = A + \frac{B}{T + C} \quad (B-12)$$

where A, B and C are the equation constants and can be found in several publications.

The 3 parameters are now left with only g_{12} to be

solved. This parameter can be determined if one boundary condition is known, namely, the activity coefficient at infinite dilution.

At infinite dilution, $X_1 = 0$, equation (B-1) becomes

$$\ln \gamma_1^\infty = -\ln(1 - A_{21}) + A_{12} \quad (\text{B-13})$$

substituting equation (B-3) and (B-4) gives:

$$\ln \gamma_1^\infty = 1 - \ln\left(\frac{\bar{V}_2}{\bar{V}_1}\right) + \frac{\Delta G_1}{RT} - \frac{\bar{V}_1}{\bar{V}_2} \exp\left(\frac{\Delta G_2}{RT}\right) \quad (\text{B-14})$$

where

$$\Delta G_1 = g_{21} - g_{11} \quad (\text{B-15})$$

$$\Delta G_2 = g_{12} - g_{22} \quad (\text{B-16})$$

Therefore, once the γ^∞ at one temperature is known, g_{12} can be solved implicitly from equation (B-14). The γ_1 and γ_2 at various composition can then be calculated and thus

vapor-liquid equilibrium data, such as vapor pressure of solute at the temperature of different concentration in liquid phase, can be obtained.

To solve g_{12} iterative techniques such as Newton's method can be employed. The objective function $F(g_{12})$ is defined as:

$$F(g_{12}) = 1.0 - \ln \frac{\bar{V}_2}{\bar{V}_1} + \frac{\Delta G_1}{RT} - \frac{\bar{V}_1}{\bar{V}_2} \exp\left(\frac{-\Delta G_2}{RT}\right) - \ln \gamma_1^\infty \quad (\text{B-17})$$

The new value of g_{12} from Newton's method algorithm as iteration continues is:

$$F'(g_{12}) = \frac{dF(g_{12})}{dg_{12}} = \frac{1.0}{RT} \left(1.0 + \frac{\bar{V}_1}{\bar{V}_2} \exp\left(\frac{-\Delta G_2}{RT}\right) \right) \quad (\text{B-18})$$

the derivative of the function, $F'(g_{12})$, is:

$$g_{12n+1} = g_{12n} - \frac{F(g_{12})_n}{F'(g_{12})_n} \quad (\text{B-19})$$

The criterion for the termination of iterative procedure is given by:

$$\left| \frac{g_{12n+1} - g_{12n}}{g_{12n+1}} \right| \leq \epsilon \quad (\text{B-20})$$

where ϵ is a small positive number. The computer program that solves g_{12} implicitly and further calculate the vapor pressure of solute at its various composition in liquid phase is presented in Appendix C.

APPENDIX C

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C.1 Vapor-Liquid Equilibrium Data

This section includes an interactive program which solves for the Wilson parameter, g_{12} and vapor-liquid equilibrium of absorbed gases over the solvents.

In the experiment, when a mixture of absorbed and non-absorbed gases is inject, it is assumed that the non-absorbed gas appears only in vapor phase, that is, 2 components in liquid phase, 3 components in vapor phase.

The program first asks for the necessary data such as temperature and total pressure of the system, molar volume and Antoine constants for both of absorbed gas and solvent, and activity coefficient at infinite dilution. After implicitly calculating g_{12} , then use equation (B-1) and (B-2) to calculate activity coefficient at various composition.

$$\ln \gamma_1 = -\ln(1 - A_{21}X_2) + X_2 \left(\frac{X_2 A_{12}}{1 - A_{12}X_1} - \frac{X_1 A_{21}}{1 - A_{21}X_2} \right) \quad (B-1)$$

$$\ln \gamma_2 = -\ln(1 - A_{12}X_1) - X_1 \left(\frac{X_2 A_{12}}{1 - A_{12}X_1} - \frac{X_1 A_{21}}{1 - A_{21}X_2} \right) \quad (B-2)$$

where

$$X_2 = 1 - X_1$$

With each small increment of composition of component 1 start from zero, by assuming the system is ideal gas, this program further calculate the partial pressure of component 1 and 2.

$$PP_1 = Y_1 \cdot P_t = \gamma_1 \cdot X_1 \cdot P_1^S \quad (C-1)$$

$$PP_2 = Y_2 \cdot P_t = \gamma_2 \cdot X_2 \cdot P_2^S \quad (C-2)$$

$$PP_3 = P_t - PP_1 - PP_2 \quad (C-3)$$

where P_t = total pressure of system

PP = partial pressure

X = mole fraction in liquid phase

Y = mole fraction in vapor phase

P^S = vapor pressure at system temperature.

and the subscripts:

1 is component 1, the absorbed gas (NO , NO_2 or SO_2)

2 is component 2, the solvent

3 is component 3, the nonabsorbed gas (air or N_2)

APPENDIX D

92

1.69

18.43

STOP

RT	TYPE	AREA	AREA %
1.69		463973	68.72
18.43		211156	31.28

MP 3388H
DLY OFF
RTN .30

STOP OFF
RTN 16

REJECT OFF

Solvent Loading: 17.8%

Room Temp: 25 C

Column Temp: 100 C

Inlet Pressure: 11.8 psig

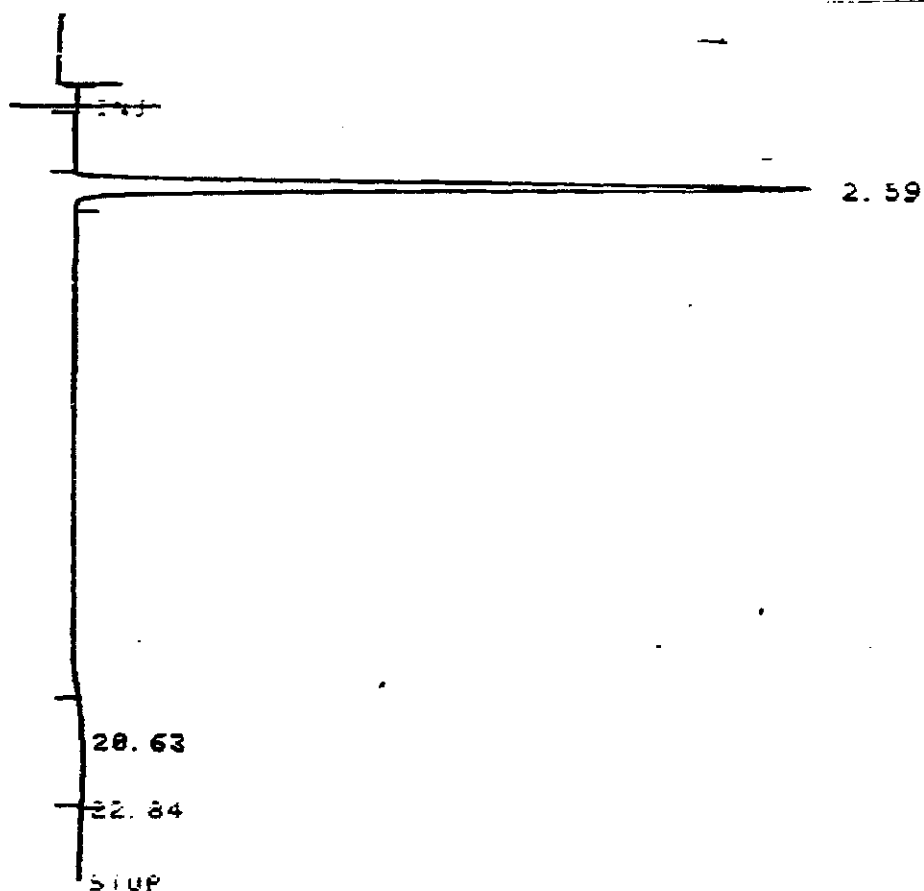
Gas Flow Rate: 27.03 cc/min

Rent. Time Diff.: 16.74 min

Henry's Constant: 2.40 atm

Act. Coefficient: 0.088

Fig. D-1 SO₂ & CHP System



RT	TYPE	AREA	PERCENT
2.59		424349	95.11
20.63		21802	4.887

HP 3380H
 DELY OFF
 MV/M .10

STOP OFF
 MIN 32

REJECT 1000

Solvent Loading: 42.68%

Room Temp: 28 C

column Temp: 60 C

Inlet Pressure: 6.5 psig

Gas Flow Rate: 11.82 cc/min

Ret. Time Diff.: 18.04 min

Henry's Constant: 17.26 atm

Act. Coefficient: 1.612

Fig. D-2 SO₂ & DETA System

RT	TYPE	NRLEN	NRLEN %
1.04		127150	75.98
7.95		40106	24.02

100-36867
 100-36867
 100-36867

SECRET 074
ATTN 32

KEJELI 2555

Solvent Loading: 42.68%

Room Temp: 28 C

Column Temp: 60 C

Inlet Pressure: 15.1 psig

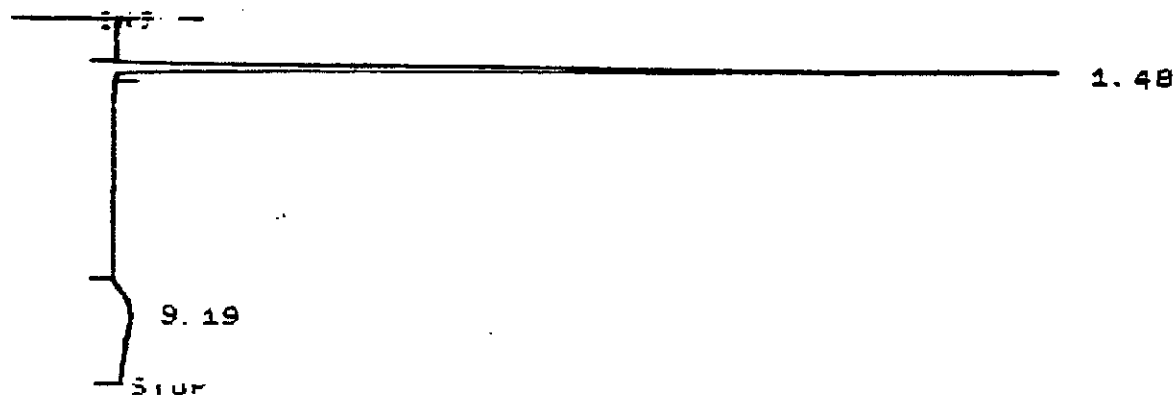
Gas Flow Rate: 39.37 cc/min

Rent. Time Diff.: 6.91 min

Henry's Constant: 17.23 atm

Act. Coefficient: 1.59

Fig. D-3 SO₂ & DETA System



RT	TYPE	AREA	AREA %
1.48		227800	76.96
9.19	1	68206	23.04

NP 3390A
 DLY OFF
 MV/M .10

STOP OFF
 ATIN ↑

REJECT 1000

Solvent Loading: 42.68%

Room Temp: 28 C

Column Temp: 70 C

Inlet Pressure: 11.0 psig

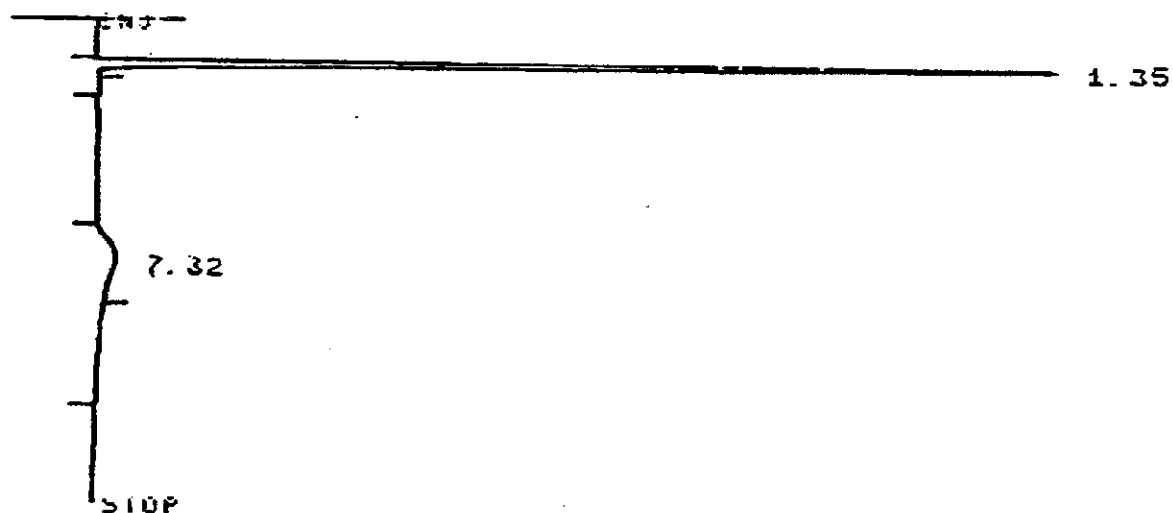
Gas Flow Rate: 24.14 cc/min

Rent. Time Diff.: 7.71 min

Henry's Constant: 22.56 atm

Act. Coefficient: 1.62

Fig. D-4 SO₂ & DETA System



RT	TYPE	AREA	AREA %
1.35		247869	86.6
7.32		36336	13.4

HF 3383A
 DLY OFF
 MV/M .30

STOP OFF
 ATTN 32

REJECT 1000

Solvent Loading: 42.68%

Room Temp: 28 C

Column Temp: 80 C

Inlet Pressure: 12.0 psig

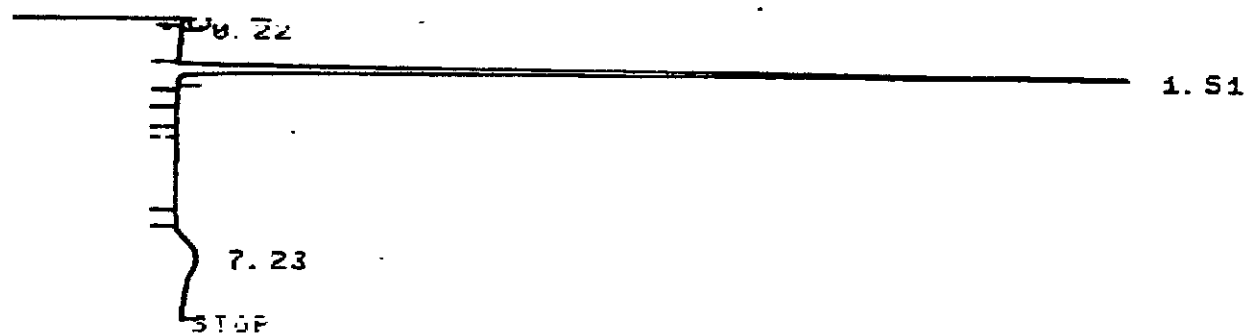
Gas Flow Rate: 26.13 cc/min

Ret. Time Diff.: 5.97 min

Henry's Constant: 27.67 atm

Act. Coefficient: 1.57

Fig. D-5 SO₂ & DETA System



RI	TYPE	AREA	AREA %
1.51		250596	84.23
7.23		46923	15.77

P 3380A
Y OFF
M T

STOP OFF DETECT 1000

Solvent Loading: 42.68%

Room Temp.: 28°C

Column Temperature: 90°C

Inlet Pressure: 10.9 psig

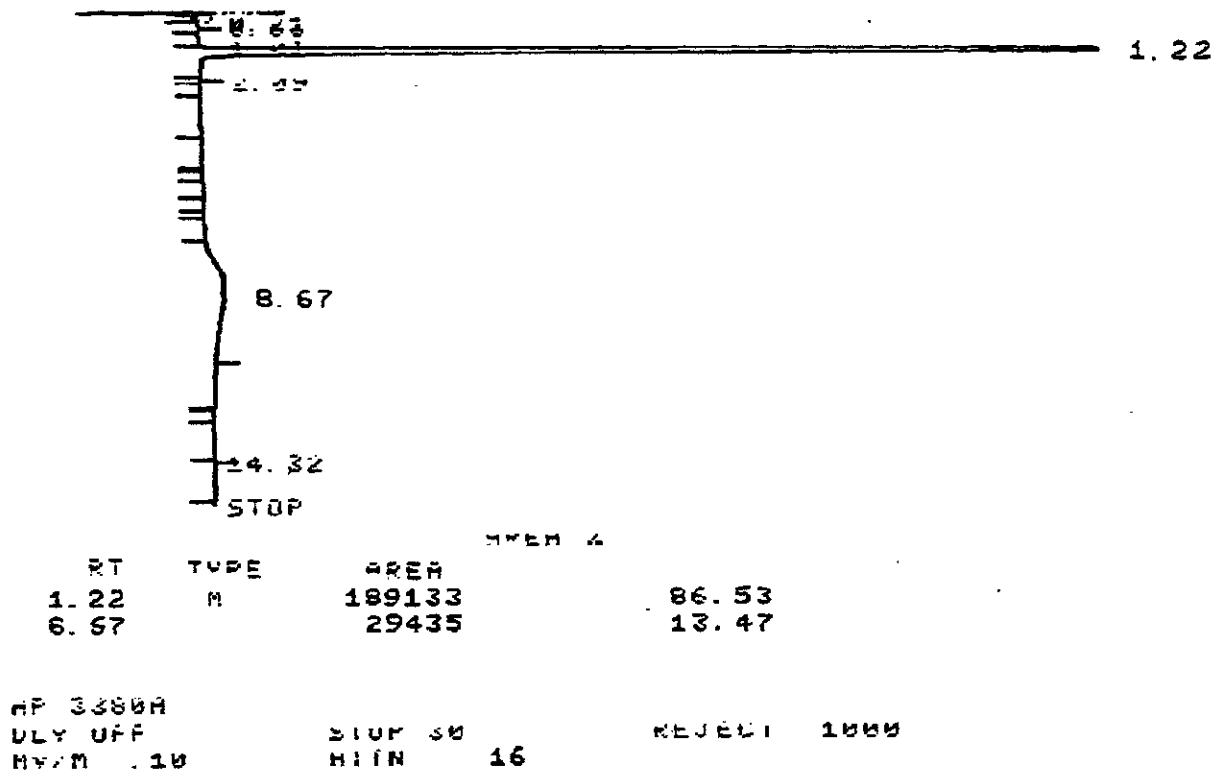
Gas Flow Rate: 22.08 cc/min

Ret. Time Diff.: 5.72 min

Henry's Constant: 33.15 atm

Act. Coefficient: 1.503

Fig. D-6. SO₂ and DETA System



Solvent Loading: 57.5%

Column Temp: 70 C

Gas Flow Rate: 26.62 cc/min

Henry's Constant: 20.26 atm

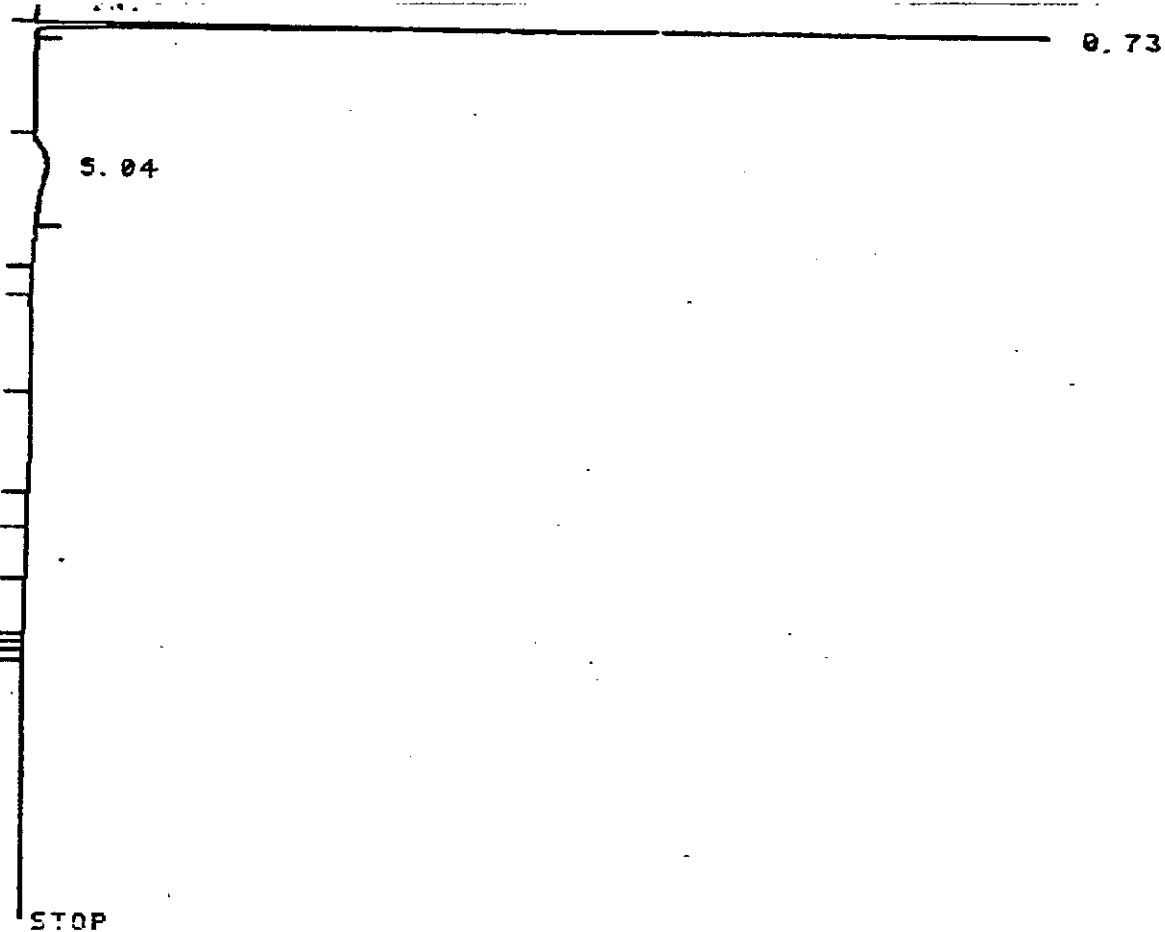
Room Temp: 23 C

Inlet Pressure: 12.2 psig

Ret. Time Diff.: 7.45 min

Act. Coefficient: 1.453

Fig. D-7 SO₂ & TETA System



RT	TYPE	AREA	AREA %
5.04		94155	85.87
		15491	14.13

P 3388M
 LY OFF
 VPM .10

STOP 30
 ATTN 16

REJECT 1000

Solvent Loading: 57.5%

Room Temp: 23 C

Column Temp: 70 C

Inlet Pressure: 20.5 psig

Gas Flow Rate: 57.47 cc/min

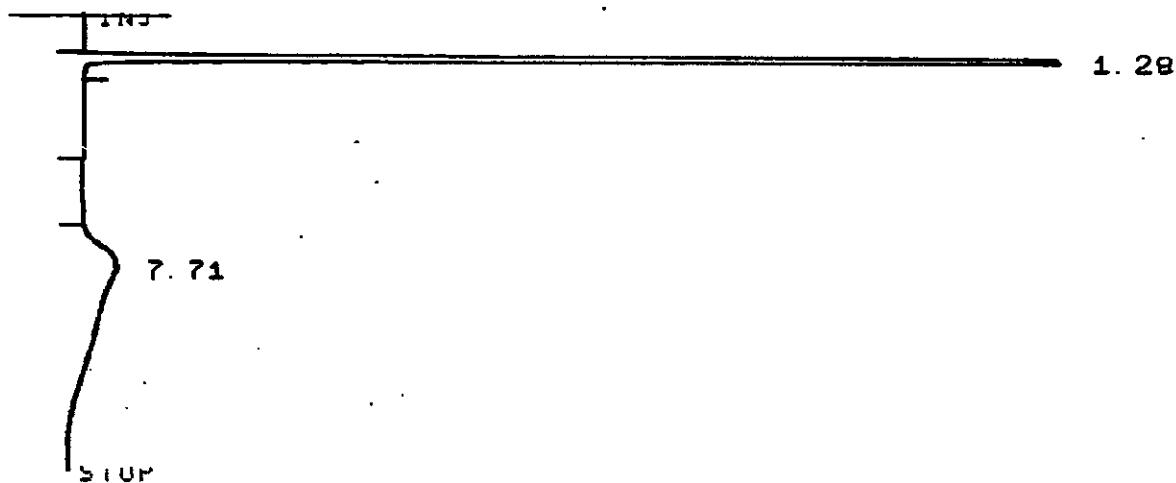
Ret. Time Diff.: 4.31 min

Henry's Constant: 19.98 atm

Act. Coefficient: 1.433

Fig. D-8 SO₂ & TETA System

100



MINEN %

RT	TYPE	MINEN	MINEN %
1.28		266764	75.22
7.71	1	87861	24.78

RT 3300H
DLY OFF
MVFM 1.28

STOP OFF
ATTN 16

REJECT 1000

Solvent Loading: 57.5%

Room Temp: 28 C

Column Temp: 80 C

Inlet Pressure: 11.5 psig

Gas Flow Rate: 24.96 cc/min

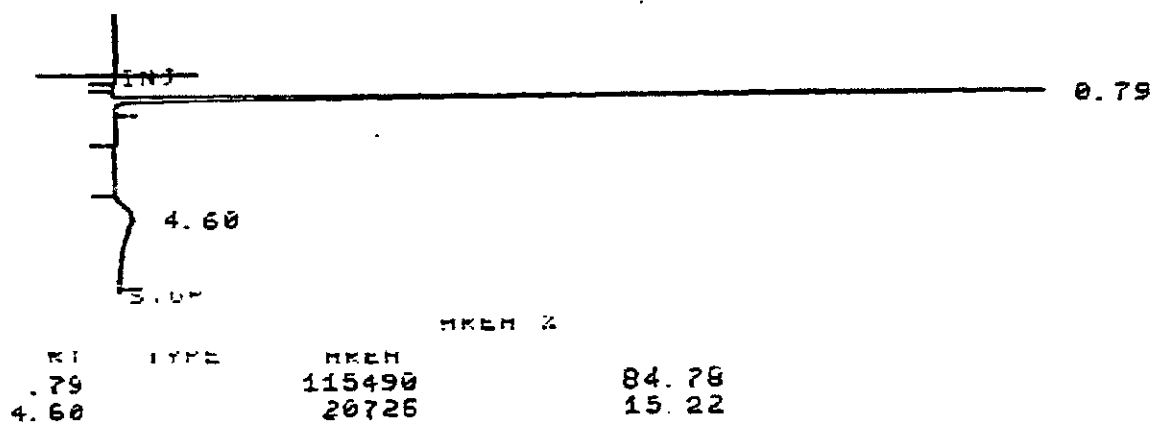
Ret. Time Diff.: 6.43 min

Henry's Constant: 25.27 atm

Act. Coefficient: 1.43

Fig. D-9 SO₂ & TETA System

101



Solvent Loading: 57.5%

Column Temp: 80 C

Gas Flow Rate: 50.93 cc/min

Henry's Constant: 25.17 atm

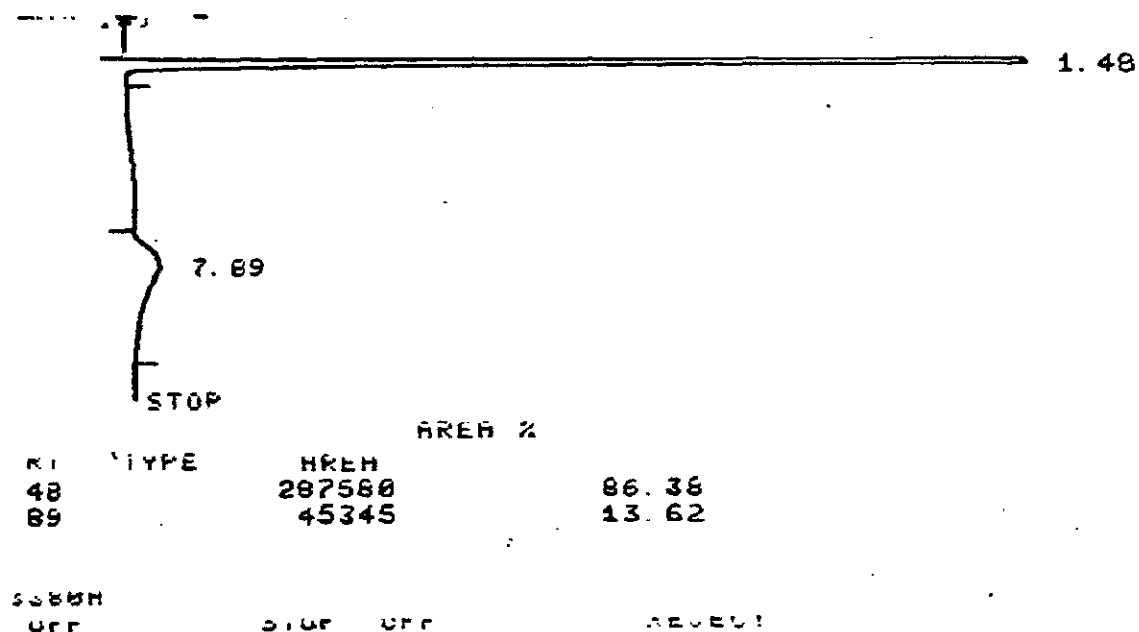
Room Temp: 28 C

Inlet Pressure: 18.8 psig

Ret. Time Diff.: 3.81 min

Act. Coefficient: 1.43

Fig. D-10 SO₂ & TETA System



Solvent Loading: 57.5%

Room Temp: 28 C

Column Temp: 90 C

Inlet Pressure: 10.4 psig

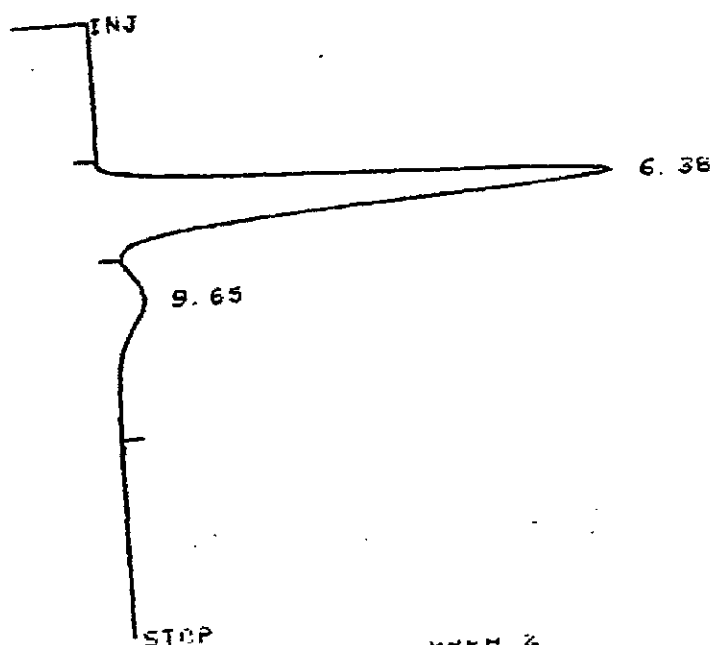
Gas Flow Rate: 20.46 cc/min

Retention Time Diff.: 6.41 min

Henry's Constant: 29.82 atm

Act. Coefficient: 1.35

Fig. D-11 SO₂ & TETA System



RT	TYPE	AREA
6.36		1299581
9.65	n	167106

HREM 2

87.41
12.59

P 3380A
LY OFF
V/M 1.18

STOP 30
MIN 32

REJECT 1000

Solvent Loading: 53.2%

Column Temp: 49 C

Gas Flow Rate: 3.37 cc/min

Henry's Constant: 245.89 atm

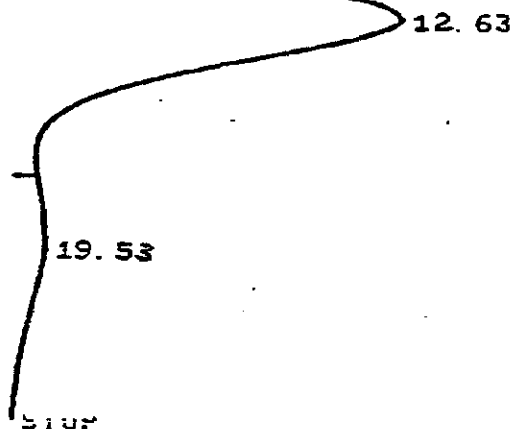
Room Temp: 25 C

Inlet Pressure: 2.2 psig

Ret. Time Diff.: 3.27 min

Fig. D-12 NO & TETA System

104

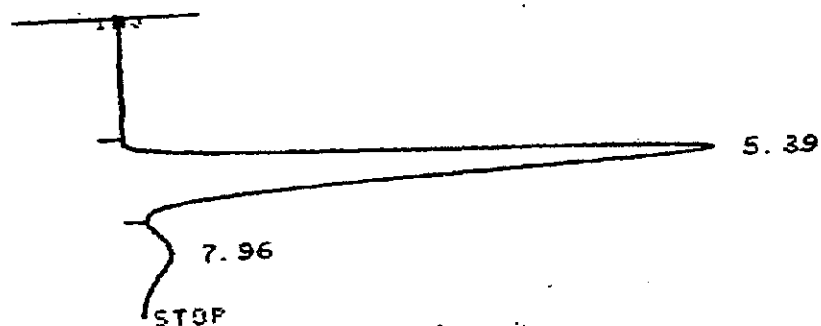


RT	TYPE	AREA	AREA %
53		2548577	84.87
53	IM	452756	15.13

3800H
OFF .10 STOP 30 MIN 32 REJECT 1000

Solvent Loading: 53.2%	Room Temp: 25 C
Column Temp: 49 C	Inlet Pressure: 0.7 psig
Gas Flow Rate: 1.62 cc/min	Retention Time Diff.: 6.90 min
Henry's Constant: 255.58 atm	

Fig. D-13 NO & TETA System



RT	TYPE	AREA	AREA %
5.39		1135140	86.06
7.96	IM	183879	13.94

1380H
T OFF
M 10

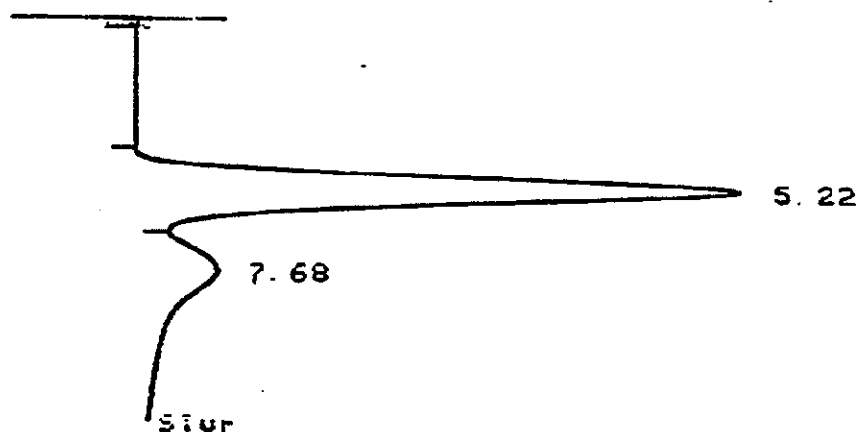
STOP 90
MIN 62

REJECT 1000

Solvent Loading: 53.2%
Column Temp: 59 C
Gas Flow Rate: 4.46 cc/min
Henry's Constant: 275.55 atm

Room Temp: 25 C
Inlet Pressure: 2.8 psig
Rent. Time Diff.: 2.57 min

Fig. D-14 NO & TETA System



RT	TYPE	AREA	AREA %
22		1027846	71.38
68	IM	412106	28.62

1000H
 OFF
 1 .10

STOP 99
 RTIN 32

REJECT 1000

solvent Loading: 53.2%

Room Temp: 25 C

column Temp: 70 C

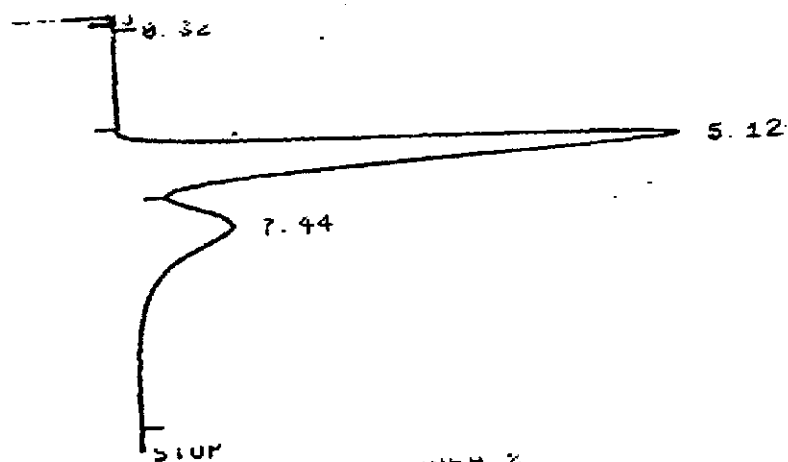
Inlet Pressure: 2.8 psig

as Flow Rate: 4.46 cc/min

Ret. Time Diff.: 2.46 min

Henry's Constant: 297.41 atm

Fig. D-15 NO & TETA System



RT	TYPE	AREA	AREA %
5.12		1059021	68.2
7.44	M	493768	31.8

3380H
Y OFF
M .10

STOP OFF
ATTN 32

REJECT 1000

Solvent Loading: 53.2%

Column Temp: 80 C

Gas Flow Rate: 4.46 cc/min

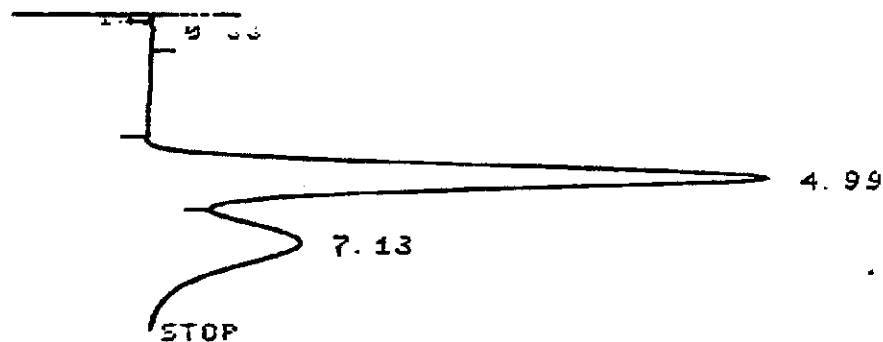
Henry's Constant: 324.55 atm

Room Temp: 25 C

Inlet Pressure: 2.8 psig

Ret. Time Diff.: 2.32 min

Fig. D-16 NO & TETA System



RT	Type	Area	Height
4.99		1036918	67.72
7.13	IM	494183	32.28

HP 3380A
 DLY OFF
 MV/M 1.10

STOP 30
 ATTN 32

REJECT 1000

Solvent Loading: 53.2%

Room Temp: 25 C

Column Temp: 90 C

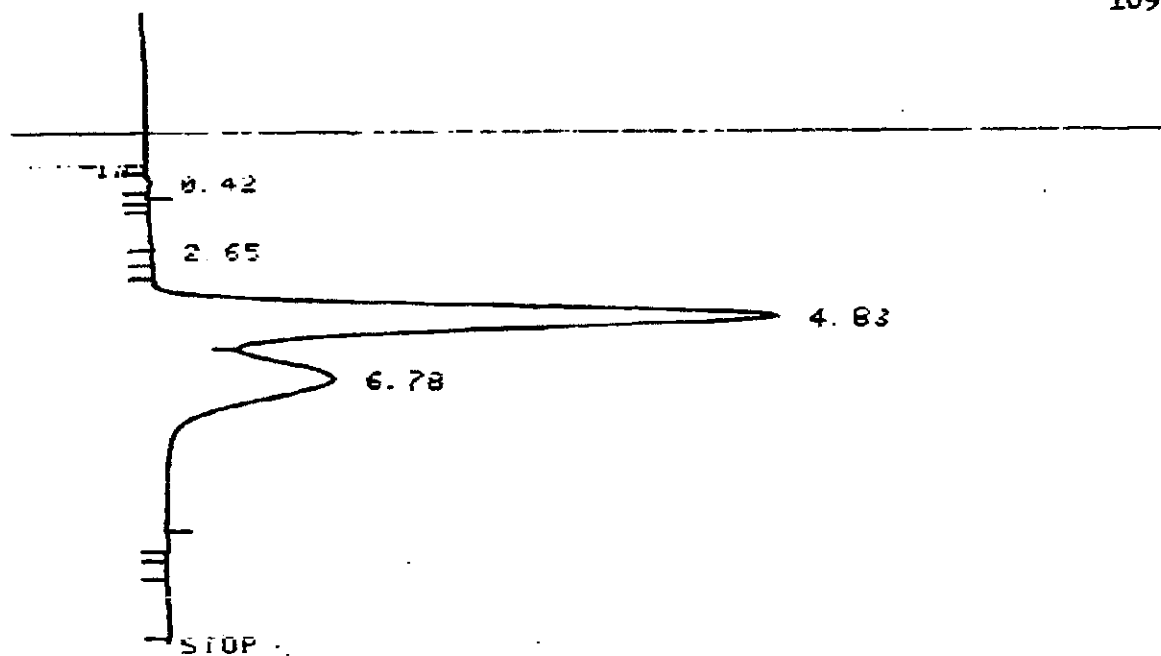
Inlet Pressure: 2.8 psig

Gas Flow Rate: 4.46 cc/min

Ret. Time Diff.: 2.14 min

Henry's constant: 361.81 atm

Fig. D-17 NO & TETA System



RT	TYPE	AREA	AREA %
0.42		1167	.075 99
4.83		1018117	66.29
6.78	M	516503	33.63

- 31800
 LY OFF STOP 30 REJECT 1000
 1/2 10 MIN 32

Solvent Loading: 53.2%

Room Temp: 25 C

Column Temp: 90 C

Inlet Pressure: 2.8 psig

Gas Flow Rate: 4.46 cc/min

Rent. Time Diff.: 1.95 min

Henry's Constant: 408.01 atm

Fig. D-18 NO & TETA System

APPENDIX E

TABLE E-1

Antoine Equation's Constants

$$\ln P^S = A + \frac{B}{T+C}$$

[P] = mmHg [K] = Kelvin

Solvent	A	B	C	Ref.
SO ₂	16.77	-2302.35	-35.97	21
CHP	21.4	-7850.9	-1.0	22
TETA	39.3	-29647.2	408.6	23
DETA	10.3	-1319.4	-182.1	23

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NOMENCLATURE

A	First Antoine equation constant
A	Wilson's equation constant
B	Second Antoine equation constant
C	Third Antoine equation constant
D	Molar conc. in gas phase
F	Gas flow rate (cc/min)
g	Binary energy interaction parameter (cal/g-mole)
H _v	Heat of vaporization
H	Henry's law constant (atm)
J	Pressure correct factor
K	Partition coefficient
M	Molecule weight (solvent)
n	Number of moles
P _i	Inlet pressure (psi or mmHg)
P _o	Outlet pressure (psi or mmHg)
P ^s	Vapor pressure (psi or mmHg)
P _t	Total pressure of a system (psi or mmHg)

P_w	Vapor pressure of water (psi or mmHg)
Q	Molar conc. in liquid phase
R	Gas constant
S	Retention ratio
T	Temperature (K)
T_c	Column temperature (K)
T_r	Room temperature (K)
t	Retention time difference (min)
t_m	Time for non-absorbed gas to pass the column
t_r	Time for absorbed gas to pass the column
V	Volume
V_s	Specific retention volume (cc/g)
V_m	Gas hold up volume
V_n	Net retention volume (cc)
V_r	Retention volume
$\bar{V}$	Molar volume (cc/g-mole)
W	Weight of solvent (g)
x	Mole fraction in liquid phase
y	Mole fraction in gas phase
z	Gas compressibility factor

Greek Letters

- ϕ Fugacity coefficient
- γ Activity coefficient
- Ω Uncertainty range

Superscript

- $^{\circ}$ Corrected parameter
- $^{\infty}$ Infinite dilution

Subscript

- 1 Component 1, or solute
- 2 Component 2, or solvent
- 3 Component 3, or inert gas
- i The solute in Henry's law constant
- j the solvent in Henry's law constant
- l Liquid phase
- g Gas phase