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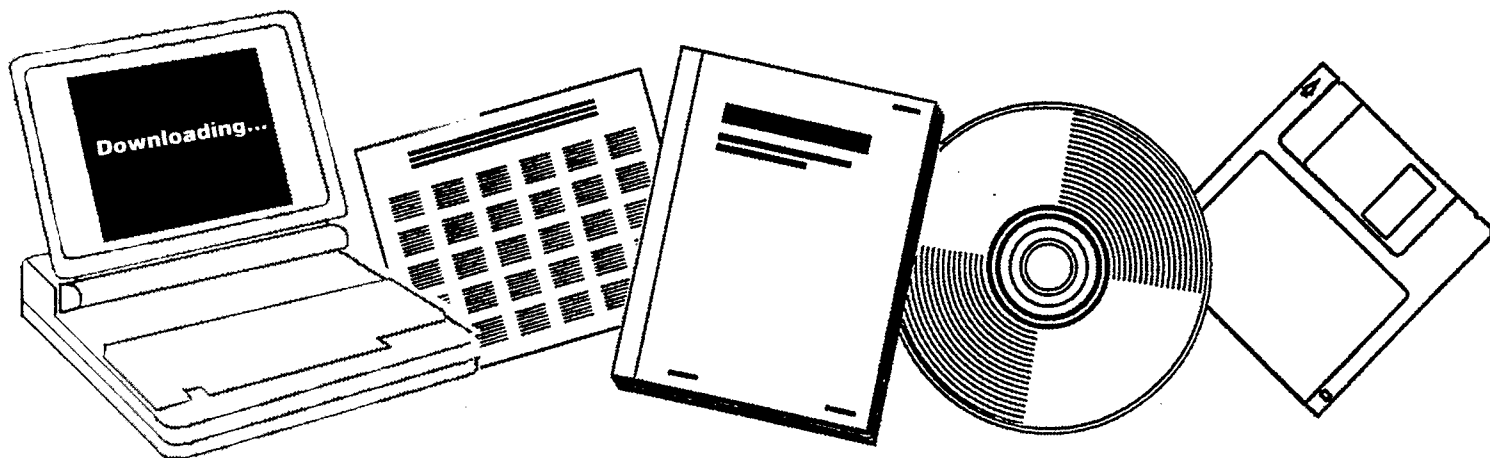
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**NOVEL EXPERIMENTAL STUDIES FOR COAL
LIQUEFACTION. QUARTERLY PROGRESS REPORT,
JANUARY 1-MARCH 31, 1986**

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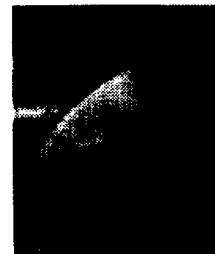
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QUARTERLY PROGRESS REPORT

NOVEL EXPERIMENTAL STUDIES
FOR COAL LIQUEFACTION

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Task 1: Thermal Behavior of Slurry Reactors Used for Indirect Coal Liquefaction

The conversion of synthesis gas to liquid products is usually carried out with the reactants in the gas phase and a solid catalyst. Because of relatively poor heat transfer from the gas to the solid, the exothermic heat of reaction is difficult to remove, and care must be taken to prevent the catalyst from overheating with loss of selectivity and activity. Slurry reactors in which the catalyst is suspended in a liquid medium and the gases are bubbled through the slurry have intrinsically better heat transfer characteristics and appear promising for indirect liquefaction processes.

Scope of Work

In Task 1 of this project the thermal behavior of slurry reactors when used for indirect coal liquefaction is being studied. Previous work with a ruthenium catalyst has shown that two distinct steady states can be found for identical operating conditions. Other catalysts and operating conditions are being studied.

Work is being done using three indirect liquefaction routes involving synthesis gas -- the Fischer-Tropsch reaction, the one-step conversion to methanol, and the two-step conversion to methanol.

Results and Highlights

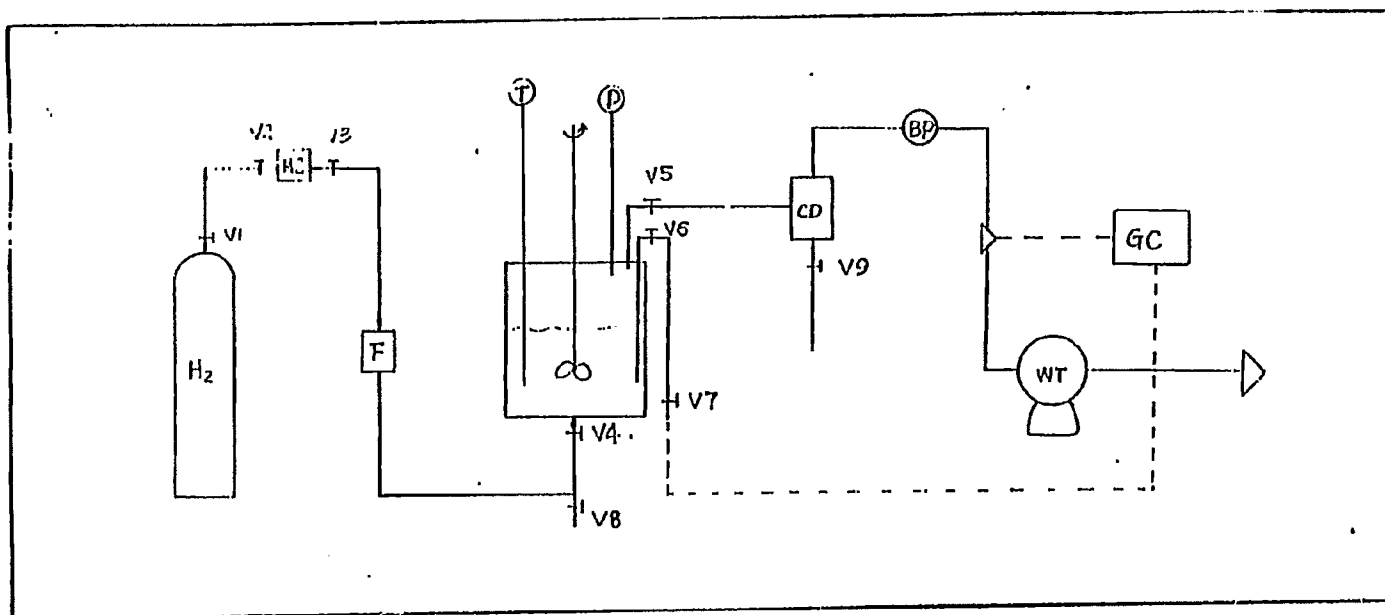
Experimental work during the last quarter was concentrated on examining the second step of the two step process -- the hydrogenolysis of the formate (in these experiments methyl formate) using a Cu-Cr (United Catalyst G-89) catalyst. During the last quarter, more than 100 data points were obtained.

The experimental procedure is described below, and a summary of the data obtained is given.

Equipment and Experimental Procedure

The reaction system is shown in Figure 1. The reactor, a standard one liter autoclave manufactured by Autoclave Engineers, is equipped with a variable speed mechanical stirrer which is driven by a magnetic coupling. An internal thermocouple allows measurement of the slurry temperature.

Initially, the reactor is filled with 600 cc of methyl formate. In order to remove air, hydrogen is introduced to the reactor until the pressure is 150 psig. After 10 minutes the pressure is lowered to 10 psig. This procedure is repeated two more times. Oxygen concentration is reduced to 0.008 volume percent (calculated assuming an ideal gas and perfect mixing). Catalyst which has been reduced in a tube outside the reactor is forced into the reactor using hydrogen. Then the reactor is sealed and the stirrer turned on. The reactor is kept at room temperature and low hydrogen pressure for about ten hours. Before reaction, the reactor is heated to the required temperature and then the pressure is increased by feeding hydrogen from a high pressure cylinder. In Figure 1, valves V1, V2, V3, V4 are kept open all the time during reaction, but valves V5, V6, and V7 are closed. A mass flow controller, "MC", is used to control hydrogen flow to maintain reactor pressure constant. Liquid samples are taken by opening valve V6 to let the high pressure inside the reactor push about 10cc of liquid into the sampling tube, and then valve V6 is closed. After cooling, the sample in the sampling tube is withdrawn by opening valve V7 and injected into a gas chromatograph for analysis. The temperature of the reactor is controlled within ± 1 K by a computer control system.



MC: mass flow controller

CD: condenser

BP: back pressure regulator

GC: gas chromatograph

WT: wet test meter

F : filter

Figure 1. Schematic diagram of apparatus used for study of hydrogenolysis of methyl formate

Experimental Conditions and Run Listing

Experiments were carried out at temperatures from 100°C to 160°C. These are lower than those used by Sorum and Onsager (1984). Ranges for experimental variables are listed in Table 1. A listing of experimental conditions by run number is given in Table 2. Some typical results are given in Table 3.

Table 1: Operating Conditions

Operating Parameter	Unit	Operating Range
Temperature	°C	100 - 160
Pressure	psig	400 - 1000
Catalyst loading	gram/l	5 - 20
Mef concentration	mole/l	0.5 - 15.0

Table 2: Listing of Runs

Run #	Temp. C	Press. psig	C _{mef} * mole/l	C _{cata} * gram/l
3-1	100.	910.	5.9	10.5
3-2	120.	910.	4.5	10.2
4-0	120.	405.	15.7	5.4
4-1	140.	515.	14.0	5.3
4-2	160.	700.	12.3	5.1
5-0	140.	900.	14.7	10.7
6-0	140.	900.	15.1	5.4
7-0	140.	450.	15.5	10.5
7-1	100.	950.	6.3	10.3
8-0	120.	650.	15.4	21.6
8-1	100.	950.	10.1	20.1
8-2	140.	950.	6.5	19.3
8-4	160.	950.	0.9	17.7
9-0	120.	650.	15.0	21.5
9-1	100.	950.	9.1	19.8
9-2	140.	950.	5.2	18.9
10-0	140.	900.	12.7	5.2
10-1	140.	900.	5.1	4.6
10-2	120.	1000.	3.2	4.5

*: Initial Concentration

Table 3: Typical Experimental Results

Run #	Temperature (°C)	Total Pressure (psig)	Concentration	
			MEF (mole/l)	Catalyst (gram/l)
8-0	393.0	650.0	15.21	21.51
	393.0	650.0	14.68	21.34
	393.0	650.0	14.21	21.18
	393.0	650.0	13.72	21.01
	393.0	650.0	13.35	20.89
	393.0	650.0	12.94	20.74
	393.0	650.0	12.52	20.59
	393.0	650.0	12.15	20.46
8-1	373.0	950.0	9.95	20.10
	373.0	950.0	9.90	20.09
	373.0	950.0	9.80	20.05
8-2	413.0	950.0	6.20	19.25
	413.0	950.0	5.54	19.02
	413.0	950.0	4.90	18.80
	413.0	950.0	4.30	18.59
	413.0	950.0	3.86	18.43
	413.0	950.0	3.44	18.28
	413.0	950.0	3.05	18.13
	413.0	950.0	2.76	18.02
	413.0	950.0	2.51	17.93
	413.0	950.0	2.24	17.83
8-4	433.0	950.0	2.07	17.76
	433.0	950.0	0.83	17.69
	433.0	950.0	0.64	17.62
	433.0	950.0	0.51	17.57
	433.0	950.0	0.41	17.54
	433.0	950.0	0.33	17.51
	433.0	950.0	0.31	17.50
433.0	950.0	0.26	17.49	

Data Analysis

The reaction was found to be very selective, with methanol being the only product detected. Equilibrium was not reached in the studied conditions.

After examining several kinetic rate expressions, the following equation was chosen for expressing the rate of hydrogenolysis of methyl formate in a three-phase reaction system. As reported in the last Quarterly Progress Report (October 1985 to December 1985), the reverse reaction is negligible at the experimental conditions, so only the forward reaction is considered.

$$R_r = -k \text{ EXP } (-E/RT) (C_{\text{mef}})^a (P_{\text{H}_2})^b C_{\text{cata}} \quad (1)$$

where

- R_r : reaction rate (mole/l min)
- k : frequency factor of reaction rate constant
- E : activation energy (J/mole K)
- R : gas constant (J/mole K)
- T : temperature (K)
- C_{mef} : concentration of methyl formate (mole/l)
- P_{H_2} : partial pressure of hydrogen (atm)
- C_{cata} : catalyst concentration (gram/l)
- a : order of reaction with respect to methyl formate
- b : order of reaction with respect to hydrogen

The reaction rate R_r is calculated by:

$$R_r = (C_{\text{mef}_i} - C_{\text{mef}_{i+1}}) / (t_{i+1} - t_i) \quad (2)$$

where C_{mef_i} and $C_{mef_{i+1}}$ are determined by gas chromatography analysis, and t_i and t_{i+1} are reaction times for samples i and $i+1$.

After rearranging and taking the logarithm of both sides, equation (1) becomes:

$$\ln(R_p/C_{cata}) = \ln(k) - (E/R)(1/T) + a \ln(C_{mef}) + b \ln(P_{H_2}) \quad (3)$$

A preliminary least squares regression using MINITAB gives the following results:

$\ln(k)$: 13.3 ± 0.90
 E/R : $-10622. \pm 373.$
 a : 1.29 ± 0.04
 b : 0.77 ± 0.09

Correlation coefficient: 92.6%

Statistical analysis and interpretation of results is continuing.

Future Work

During the next quarter work will be continued on analysis of the hydrogenolysis data and experimental measurement of the carbonylation reactions for the two step process. Measurement of the thermal effects for the Fischer-Tropsch synthesis will be resumed when a supply of Fe/Cu/K₂CO₃ catalyst is received.

References to Task 1

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Task 2: Coal Liquefaction under Supercritical Conditions

Supercritical fluid extraction is an attractive process primarily because the density and solvent power of a fluid changes dramatically with pressure at near critical conditions, and during the extraction of coal, the density of a supercritical fluid should also change the extractability of the coal. During earlier quarters a non-reacting supercritical fluid, toluene, was studied to determine the effect of density of the coal extraction/reaction process. Extractions were carried out for 2 to 60 minutes at reduced densities between 0.5 and 2.0 and at temperatures between 647 and 698 K. The data obtained can be explained by the hypothesis that coal dissolution is required preceding liquefaction reactions and that the degree of dissolution depends upon solvent density and temperature. A kinetic model shows that higher solvent densities result in faster conversion rates and in higher total conversions. Two papers have resulted from this study.

A second factor that makes supercritical extraction attractive is high mass transfer rates. At high pressures, mass transfer rates in a supercritical fluid are much higher than in a liquid, despite the fact that the supercritical fluid has liquid-like solvent powers. The objective of this work is to measure mass transfer rates for naphthalene extraction by carbon dioxide to enable us to determine how mass transfer coefficients vary with pressure, flow rate, and bed height, since these parameters will influence the design of extraction or reaction processes which utilize supercritical fluids. Ultimately, such measurement will be extended to coal/supercritical fluid systems to help define the flow rates liquid/solvent ratios that would be appropriate for a supercritical system.

Review of Mass Transfer/Equilibrium Under Supercritical Conditions

Over the past few years, significant interest has been expressed in a separation concept wherein a condensed phase (liquid or solid) is contacted with a fluid phase that is supercritical both in a temperature and pressure sense. Several industrial extraction processes have been developed such as deasphalting petroleum with supercritical propane (1) and decaffeinating coffee with supercritical carbon dioxide (2). Other examples would include deashing coal liquids (3) and regenerating activated carbon with carbon dioxide (4).

Supercritical carbon dioxide is a particularly attractive solvent for practical applications because it is nontoxic, nonflammable, environmentally acceptable, and relatively inexpensive. The critical temperature of carbon dioxide is only 304 K and thus extractions can be accomplished at moderate temperatures where thermal degradation of heat-labile extracts is minimal.

One of the cited advantages for using supercritical fluids as extractants is that the diffusion coefficients are significantly higher than for liquid systems while the viscosity more closely approximates that of a gas. Such trends would lead one to conclude that mass transfer characteristics of supercritical fluids may be better than comparable extraction carried out in the normal liquid phase.

There are few experimental data for diffusion coefficients of solutes in supercritical fluids. Those available are shown in Figure 2 where diffusion coefficients are plotted as a function of reduced pressure of the solvent gases for the systems CO₂-naphthalene, CO₂-benzene, and ethylene-naphthalene. The low-pressure data show D_{AB} to be inversely proportional to pressure, but, approximately, $D_{AB} \propto P^{1/2}$. The range of reduced temperatures shown in Figure 2 is 0.97 to 1.09. At higher temperatures, D_{AB} would increase.

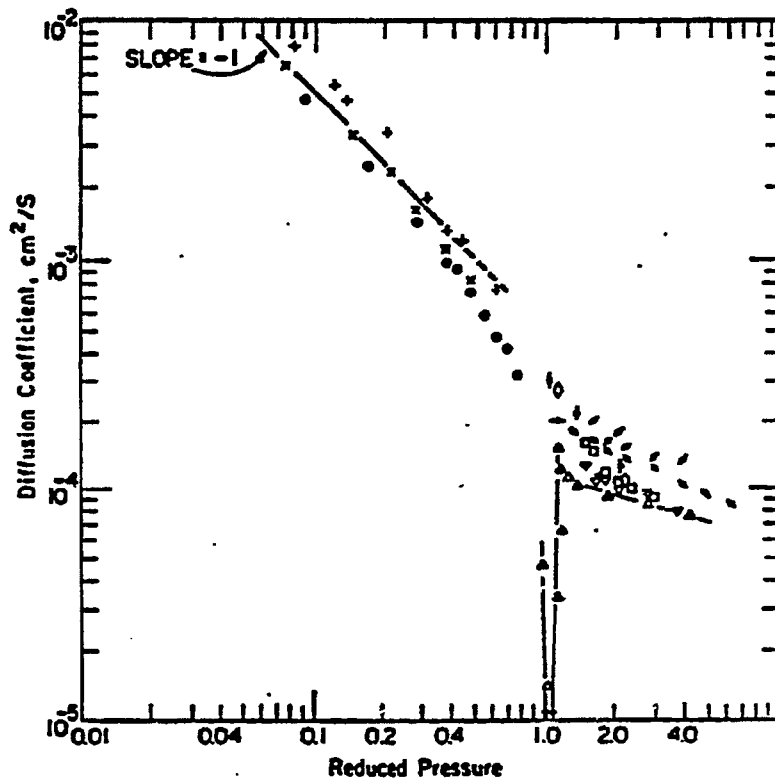


Fig.2 The Diffusion Coefficients in Supercritical Fluids

There is an interesting discontinuity in Figure 2 for the system CO₂-naphthalene at 35°C. At a pressure of about 80 ~ 81 bar ($P_r \sim 1.09$), the lower critical end point is attained and the diffusion coefficient is essentially zero (5). The change in D_{AB} , as this critical point is approached, is very abrupt. Diffusion coefficients have not been reported for systems near the upper critical end points, but judging from the general trend of liquid-liquid systems near critical points (6, 7), it would be expected that diffusion coefficients would be very small in this region. In general, even though one has few data, it would appear that the binary diffusion coefficients for solutes in supercritical gases are around 10^{-4} cm²/s for P_r values of 1 ~ 5 and T_r values of 1 ~ 1.2.

In the pressure and temperature range where most supercritical extractions would be operated, there are no generally accepted correlations for mass transfer coefficients. Storck and Coeuret (8) and Wilson and Geankoplis (9) have reviewed liquid phase relations while Gupta and Thodos (10) cover dilute gas phase system. Both Bradshaw and Bennett (11) and Pfeffer (12) point out that due to the difference in Schmidt numbers, liquid and gases would be expected to have different correlations for the mass transfer coefficient.

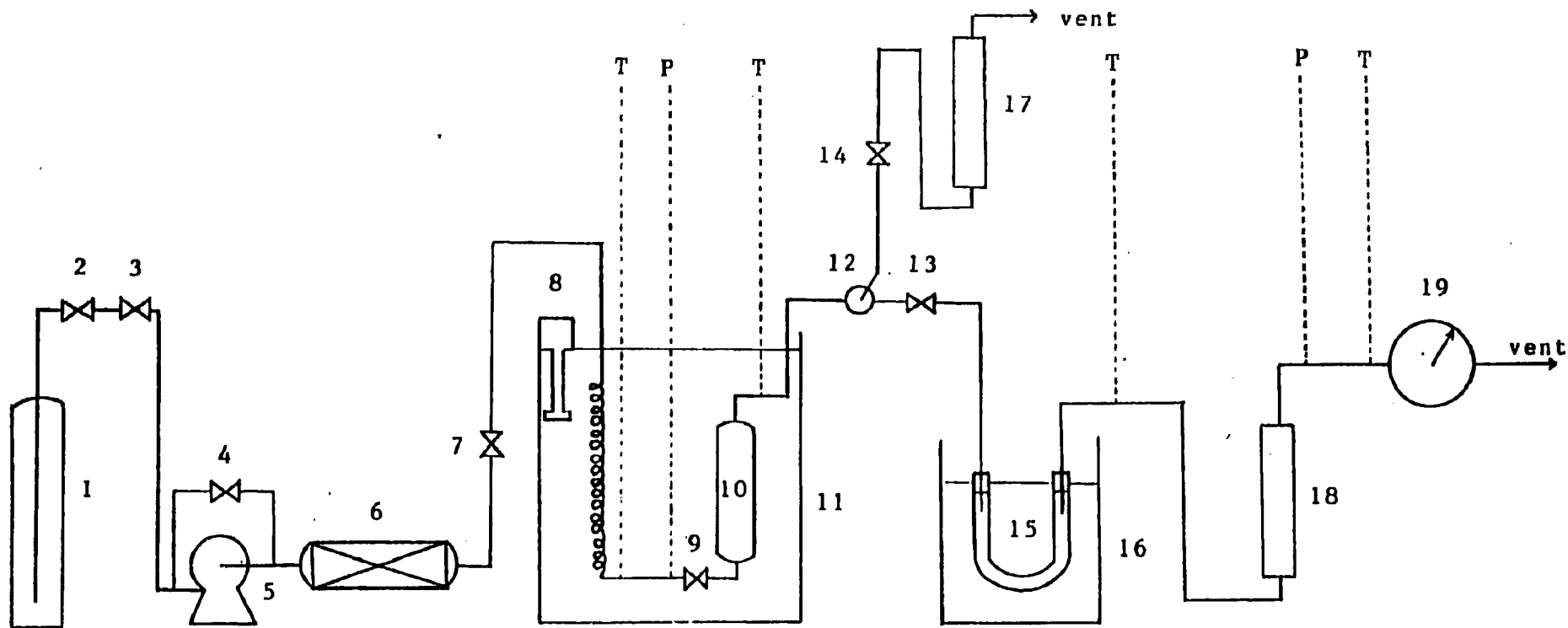
It is the purpose of this work to measure mass transfer coefficients and to compare their dependence on pressure and temperature to the corresponding dependence of diffusion coefficients. In this way, the relationship between diffusion coefficients and mass transfer coefficients can be determined.

Experimental Design

The schematic diagram of the experimental apparatus used in the past quarter is shown in Figure 3. Liquid carbon dioxide is pumped into the system via a high-pressure Milton-Roy liquid pump. Pressure is controlled to the desired pressure by using a back pressure regulator and pressure fluctuations are dampened with an on-line surge tank. The system consists of a preheater which allows the solvent to reach the desired temperature and the extraction vessel 313 cm³ in volume, 26.67 cm in length and 3.87 cm in diameter. The extraction vessel is packed with naphthalene pellets which have been made from pure naphthalene using a die. The height of the packing in the bed can be changed by using inert packing at the bottom and the top of the bed. The inert packing material being used is nylon rods of similar shape and size as the pellets. Another advantage in using the inert pellets is to get rid of end effects in the packed bed being used as the extractor. Pressure at the inlet of extractor will be measured using a pressure transducer. The temperature of the extractor will be measured at the inlet and outlet.

The fluid mixture coming out of the extractor is depressurized to atmospheric pressure by passing it through a heated metering valve. Originally, dissolved solids were to be precipitated in two U-tubes (ice temperature) in series following the valve. Both tubes were packed with glass wool at the outlet. The instantaneous flow rate of the gas leaving the extractor is measured using a rotameter and the total amount of gas flow is measured with a calibrated wet test meter. During this quarter the sample system has been modified as explained below.

The mass of precipitated solid will be found as described below. With this value and total amount of gas flow from wet test meter, the mole fraction of solids in the supercritical fluid can be readily determined. The inlet stream temperature and pressure of wet test meter are also measured.



- 1. Cylinder
- 2. Relief valve
- 3. Check valve
- 4. Back pressure regulator
- 5. Liquid pump
- 6. Surge tank
- 7. Relief valve

- 8. Temperature controller
- 9. Valve
- 10. Extractor
- 11. Water bath
- 12. Three-way valve
- 13. Metering valve
- 14. Metering valve

- 15. U-tube
- 16. Ice bath
- 17. Rotameter
- 18. Rotameter
- 19. Wet test meter

P: Pressure gauge
 T: Temperature gauge
 Data logger

Fig. 3 The Schematic Diagram of Experimental Apparatus for Supercritical Fluid Extraction

The whole apparatus is rated for a pressure of 5,000 psi. At the beginning of experiment, we use the upper vent line (valve 14 in Figure 3) in order to reach steady state for a given flow rate, temperature and pressure. After we get steady state, we can obtain data using the lower vent line (valve 13 in Figure 3). All measured temperatures and pressures are recorded on a data logger.

The parameters that are being studied are:

- a. effect of flow rate on solubility of naphthalene in carbon dioxide at different pressures and temperatures;
- b. effect of bed height on the mass transfer coefficient under supercritical conditions;
- c. effect of flow rate on the mass transfer coefficient under supercritical conditions;
- d. effect of pressure on the mass transfer coefficient under supercritical conditions.

The experimental conditions are as follows:

System: Naphthalene - Carbon dioxide

Pellet characteristics:

Material: Naphthalene

Shape: Cylindrical

Size: Length (mm) = 10

Diameter (mm) = 4.5

Height of Bed (cm): 13.3, 8.9, 6.7

Temperature of Bed ($^{\circ}$ K): 308, 328

Pressure (psi): 1470, 2205, 2940, 3675

Flow rate (Std. Lit/Min at 70⁰F and 1 atm): 4 ~ 40

Calculation of Mass Transfer Coefficients

The measurement of mass transfer coefficients is based upon the following equation:

$$dN_A = d(VY_A) = ky (Y_{iA} - Y_A) dA = ky(Y_{iA} - Y_A) a s dZ \quad (1)$$

where

- N_A : the molal flow rate of transfer of component A in mole per unit time
- V : the molal flow rate of the gas phase
- ky : individual mass transfer coefficient
- Y_A : the mole fraction of component A in bulk of gas phase
- Y_{iA} : the mole fraction of component A at gas side of interface
(assume to be equilibrium)
- A : the total area of interface for mass transfer (area of pellets in packed section)
- a : the area of interface per unit volume of packed section
- S : the total cross section area of column
- Z : the height of packed section

Here,

$$V = \frac{V'}{1 - Y_A} \quad (2)$$

where V' is molal flow rate of inert component in moles per unit time

Therefore,

$$d(VY_A) = V' d\left(\frac{Y_A}{1 - Y_A}\right) = V' \frac{dY_A}{(1 - Y_A)^2} = \frac{dY_A}{1 - Y_A} \quad (3)$$

From equations (1) and (3)

$$\int_{Y_{A.in} = 0}^{Y_{out}} \frac{dY_A}{(1-Y_A)(Y_{iA} - Y_A)} = \frac{ky a}{\bar{G}_{My}} \int_0^{Z_T} dz$$

where \bar{G}_{My} is the averaged molal mass velocity of the gas in moles per unit area per unit time.

For dilute gas (i.e., $1 - Y_A \approx 1$)

$$\int_0^{Y_{out}} \frac{dY_A}{Y_A^* - Y_A} = \left(\frac{Ky a}{\bar{G}_{My}} \right) Z_T$$

where Y_A^* is equilibrium mole fraction.

By integration and rearrangement

$$\underline{\underline{Ky = \left(\frac{\bar{G}_{My}}{a Z_T} \right) \ln \left(\frac{Y_A^*}{Y_A^* - Y_{out}} \right)}}$$

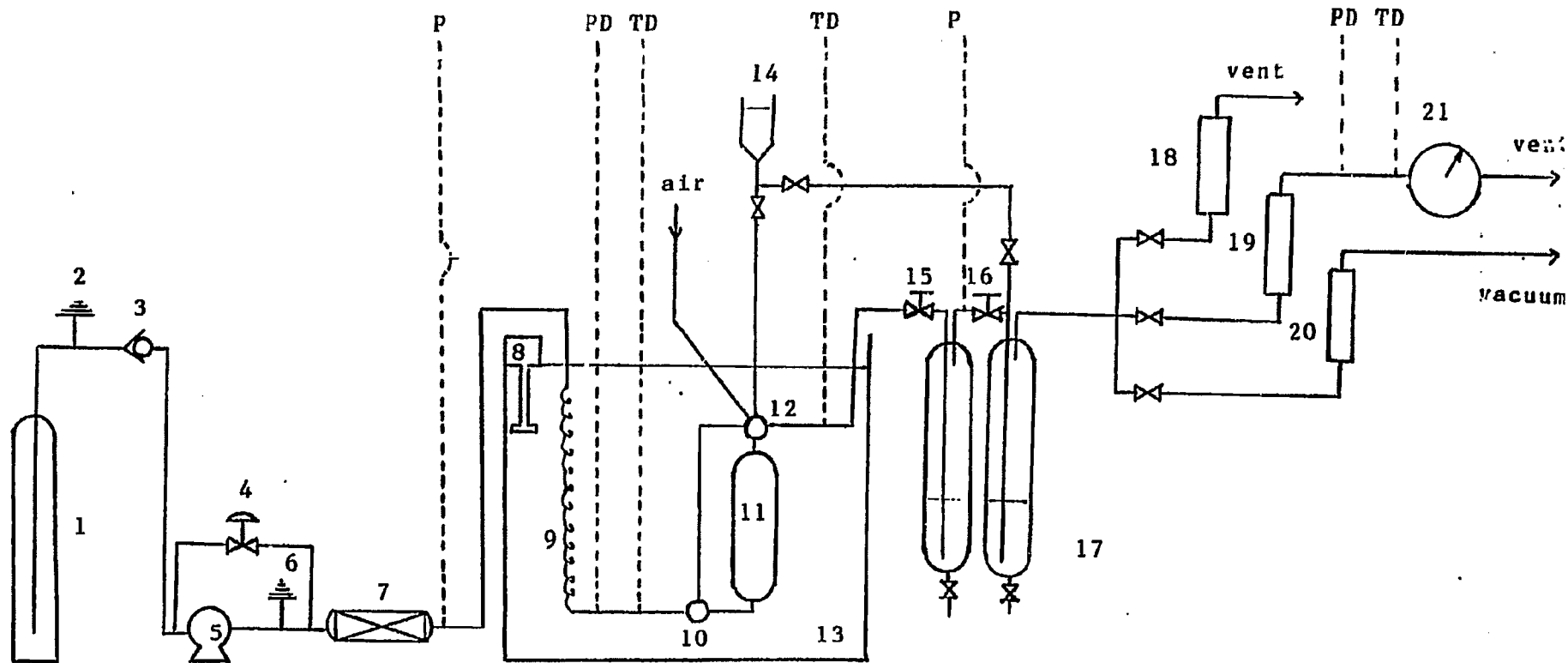
Y_{out} can be determined by the amount of naphthalene precipitated and total amount of carbon dioxide passed through the experimental apparatus during the given time.

$$Y_{out} = \frac{N_{C_{10}H_8}}{N_{C_{10}H_8} + N_{CO_2}}$$

$$N_{C_{10}H_8} = \frac{W_{C_{10}H_8}}{\text{Mol. Wt. (128)}}, \quad N_{CO_2} = \frac{PV}{ZRT}, \quad (Z = 0.995)$$

Work for Current Period (January 1 - March 30)

The schematic diagram shown in Figure 3 was built and tested. In this experiment, the solid dissolved in vessel (10), under supercritical conditions, is precipitated in a sample collector (15). The sample precipitates because its solubility decreases when the pressure is lowered. However, the expansion of carbon dioxide through valve 12, which is after the extraction vessel, resulted in three problems. First, the velocity of the gas when expanded from 4000 psia to 1 atm is too high causing entrainment of the precipitated extract and all of the extract must be collected to obtain accurate mass transfer coefficients. Second, the pressure in vessel (15) might cause the glass sample U-tube to burst. Third, the expansion causes the CO_2 to cool to the extent that it could become solid. This actually happened in the first test run. Consequently, changes were made as shown in Figure 4.



- | | | |
|----------------------------|---------------------------|--------------------|
| 1. Cylinder | 8. Temperature controller | 15. Metering valve |
| 2. Relief valve | 9. Preheater | 16. Metering valve |
| 3. Check valve | 10. Three-way valve | 17. Sample tanks |
| 4. Back pressure regulator | 11. Extractor | 18. Rotameter |
| 5. Liquid pump | 12. Five-way valve | 19. Rotameter |
| 6. Relief valve | 13. Water bath | 20. Rotameter |
| 7. Surge tank | 14. Solvent(Toluene) tank | 21. Wet test meter |

P: Pressure gauge
 PD: Pressure transmitter to Data logger
 TD: Thermocouple to Data logger

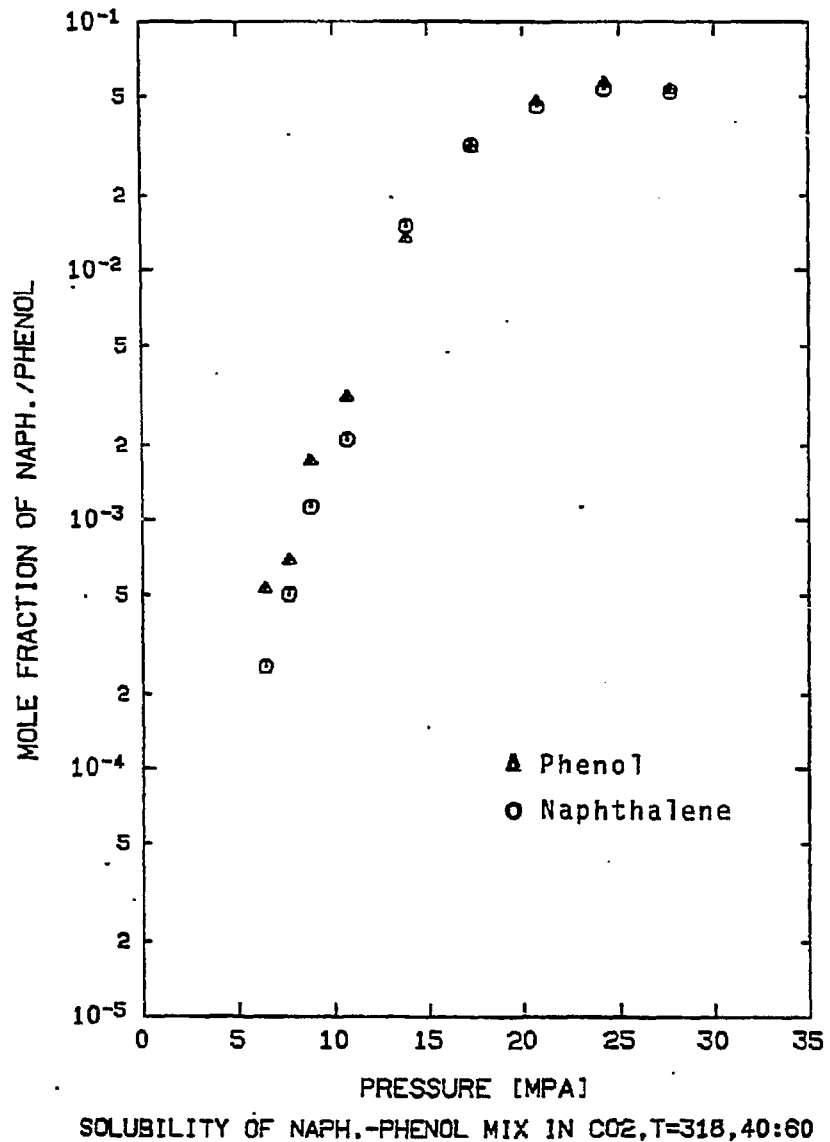
Fig. 4 Schematic Diagram of the Experimental Apparatus for Supercritical Fluid Extraction

In the revised system, the sample collectors are high pressure bombs which are heated by two 200 watt resistance heaters. Each vessel contains toluene which will help dissolve the extract. These vessels are operated at 300 to 400 psia where the solubility of the solid in the CO₂ is at a minimum (Figure 5). The second vessel is redundant and is used to guarantee that all of the extract is collected.

To determine the amount of extract collected, the amount of toluene (with dissolved extract) will be weighed. A sample of the toluene-extract solution will then be injected into a gas chromatograph to determine what portion of the solution is extract. To insure no extract remains in the line between valve 12 and the sample cylinder, valve 12 was converted to a five-way valve to allow washing of the line with toluene. Finally, the bypass, from valve 10 to 12 is designed to insure steady flow before allowing the CO₂ to pass through the extraction vessel (11).

Experimental

Figure: 5 Solubility of Solids in CO₂. Note that the mole fractions approach unity at zero pressure.



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