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Report for the Technical Data Analysis Program

Work Performed Under Contract No. AC22-82PC50046

The Pittsburg & Midway Coal Mining Co.
Denver, Colorado

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**Report for the
TECHNICAL DATA ANALYSIS PROGRAM**

**The Pittsburg & Midway Coal Mining Co.
1720 South Bellaire Street
Denver, CO 80222**

**Prepared For The
U.S. Department of Energy
Division of Energy Technology
Coal Conversion Utilization**

TABLE OF CONTENTS

	<u>Page</u>
<u>Summary</u>	1
<u>1.0 Introduction</u>	2
<u>2.0 Cold Flow Bubble Column Studies</u>	5
2.1 University of Pittsburgh	5
2.2 Gulf Science & Technology Co.	6
2.3 Conclusions from Cold Flow Tests	7
<u>3.0 SRC-II Reactor Studies</u>	10
3.1 P-99 Reactor L/D Studies	10
3.2 Ft. Lewis Dissolver Downcomer Tests	11
3.3 Ft. Lewis Radiotracer Studies	11
3.3.1 Background	11
3.3.2 February 1981 Tracer Test Program	12
3.3.3 Analytical Methodology	12
3.3.4 Inlet Injection Results	17
3.3.5 Middle Injection Results	19
<u>4.0 Discussion of Tracer Results</u>	27
<u>5.0 Conclusions</u>	29
<u>6.0 Future Considerations</u>	30
<u>References</u>	31

LIST OF TABLES

<u>Number</u>	<u>Title</u>	<u>Page</u>
Table I	Comparison of SRC-II Reactor Parameters	4
Table II	Operating Conditions During Radiotracer Injection at Reactor Inlet	15
Table III	Operating Conditions During Radiotracer Injection at Middle of Reactor	16
Table IV	Summary of Data from Radiotracer Injections at Reactor Inlet	20
Table V	Inlet Injection Gas Tracer Results	21
Table VI	Summary of Data from Radiotracer Injections into Middle of Reactor	22

LIST OF FIGURES

<u>Number</u>	<u>Title</u>	<u>Page</u>
Figure 1	Gas Holdup vs. Superficial Gas Velocity for the Air-Water-Coal System.	8
Figure 2	Gas Holdup vs. Superficial Gas Velocity for Aqueous Alcohol Mixtures.	9
Figure 3	Comparison of P-99 Dissolver Temperature Profile for the 10ft and 20ft Tall Reactors.	23
Figure 4	Ft. Lewis Radiotracer Detector Locations	24
Figure 5	Typical Detector Response for a Gas Phase Injection.	25
Figure 6	Typical Detector Response for a Liquid Phase Injection	26

ABSTRACT

Slurry phase residence time in the SRC-II dissolver is a major process variable affecting the extent of conversion to distillate products. Kinetic studies have shown that overall conversion of vacuum bottoms to lighter hydrocarbons is a zero order process with respect to vacuum bottoms. This implies that although the extent of backmixing is not a critical process variable, slurry phase mean residence time is. Experimental results from the P-99 PDU supported this.

Radioactive tracers were injected into the Ft. Lewis dissolver during normal operation to study the residence time distribution of the gas and slurry phases. Results from gas phase radiotracers showed the gas phase to approach plug flow behavior, but with much larger than expected gas holdup. Gas holdup appeared to be 20% of the reactor volume, rather than the 10-12% predicted by standard holdup correlations. Slurry phase radiotracer results indicated that the slurry phase was well-backmixed. Experiments with a downcomer designed to enhance direct slurry flow from the top to the bottom of the dissolver showed this to be a viable and controllable device.

The high level of gas holdup at Ft. Lewis suggest that standard holdup correlations must be modified to be used with confidence in the design of coal liquefaction reactors. Additional programs to define holdup relationships in high temperature, high pressure coal liquefaction reactors are recommended.

Summary

During the SRC-II Demonstration Plant design phase, concern over the effects of reactor scale-up led to additional studies which included:

- a) Bubble column tests at the University of Pittsburgh and at Gulf Science and Technology Co.
- b) Experiments with different reactor lengths at P-99 to study thermal backmixing in tall reactors.
- c) Radioactive tracer studies on the gas, liquid and solid phases in the Fort Lewis pilot plant reactor.
- d) Testing of an internal downcomer in the Fort Lewis reactor as a potential method for improving mass and thermal backmixing, if required.

The bubble column tests highlighted the inaccuracy inherent in extending existing gas holdup correlations to different gas/liquid systems. Existing correlations for air-water systems appeared to be unable to consistently predict holdup for more complex systems.

The P-99 reactor studies showed that increased dissolver height and gas velocities resulted in less efficient thermal backmixing.

Radioactive tracer studies on the Fort Lewis reactor confirmed earlier hypotheses that the slurry phase was well backmixed while the gas phase approached plug flow. The gas mean residence time and gas holdup were found to be higher than predicted by bubble column correlations. Extrapolation of the resulting loss in slurry residence time to the Demonstration Plant operating conditions suggests an increase in SRC yield of 5% mf. coal beyond the design yield might have occurred at design operating conditions.

Tests with a downcomer installed in the Fort Lewis reactor showed that it was a viable technique to recirculate slurry. Since the Fort Lewis reactor was already well backmixed, it was not possible to determine its effect on overall mixing; however, there was a slight shift toward more isothermal conditions with the downcomer in service, indicating some improvement in mixing.

1.0 Introduction

Reaction yield data for the SRC-II process were developed on four pilot plant units with reactor parameters shown in Table I. Equivalent parameters for the SRC-II Demonstration Plant dissolvers (both the single reactor per train and parallel reactors per train concepts) are also shown in Table I. The impact of the substantial scale up in the Demonstration Plant on reactor hydrodynamics and performance is somewhat uncertain due to the lack of experience with large scale, high temperature, high pressure bubble columns.

SRC-II kinetic studies on the A-1 CSTR have indicated that it may be possible to characterize the coal liquefaction reactions occurring in the dissolver as a simple zero order hydrogenation of dissolved coal ("SRC")¹. The proposed model depicts the reaction rate of SRC as a function of temperature, hydrogen partial pressure, and ash concentration. The hydrogenation of SRC to lower molecular weight hydrocarbons occurs at a low rate at typical reaction conditions and is highly exothermic. Conversion of total SRC in the dissolver feed (due to recycle slurry and SRC from initial coal dissolution) is only 30% per pass at reaction conditions of 855°F, 1700 psi hydrogen partial pressure, and 1 hour slurry residence time. Since the SRC reaction rate is temperature sensitive, conversion of SRC in the dissolver is related to the time-at-temperature history of the slurry. Successful dissolver design therefore depends on accurately predicting phase holdups in the dissolver which affect mean residence time, and backmixing which affects the temperature profile.

Operating experience with the Ft. Lewis dissolver and the 10 ft tall P-99 dissolver indicated that both systems were relatively well backmixed. This was evidenced by the near isothermal temperature profiles in these units. The backmixing in these reactors made it possible to use the heat released by exothermic hydrogenation reactions to raise the feed slurry temperature to reaction conditions by direct heat exchange, thereby reducing indirect feed preheat requirements. Conversely, the "cold" feed slurry provided a heat sink to absorb the heat of reaction, thereby limiting quench gas requirements for dissolver temperature control.

The greater vessel height and higher gas velocity in the Demonstration Plant dissolver would be expected to result in less efficient axial backmixing and higher gas holdup than experienced in the pilot plants. Less efficient backmixing would result in a wider

temperature profile, and lower average temperature. Higher gas holdup would result in lower slurry residence time. The sparsity of information on the hydrodynamics of commercial bubble columns creates an uncertainty in the magnitude of these effects. Most studies of gas holdup and backmixing in bubble columns in the past have been limited to aqueous low pressure cold flow systems, and have resulted in highly specific empirical models. Application of these models in the design of an SRC dissolver system must be viewed with caution².

To mitigate some of this uncertainty, several studies were made in support of the Demonstration Plant design to investigate gas holdup and backmixing. Cold flow bubble column experiments were conducted at the University of Pittsburgh and at Gulf Science and Technology Company (GS&TC) to elucidate the relationship between gas holdup and physical properties and operating conditions. Radiotracer studies were made on the Ft. Lewis dissolver during normal plant operation to study phase holdup, mean residence time, residence time distribution, and backmixing. Backmixing was also investigated at P-99 in a series of runs in which dissolver L/D ratio was varied.

The results from phase holdup studies are useful in interpreting the effects of operating variables on the mean residence time (MRT) of slurry in the reactor. Residence time distribution (RTD) information provides an indication of the extent of backmixing or plug flow in a reactor. In design of the SRC dissolvers, the Axial Dispersion Model was assumed to be an appropriate flow model. The MRT and RTD data obtained from the Ft. Lewis radiotracer studies provide dispersion numbers (inverse Peclet Numbers) for this model which can be compared to calculated dispersion based on literature correlations.

TABLE I
COMPARISON OF SRC-II REACTOR PARAMETERS

<u>Pilot Units</u>	<u>Flow System</u>	<u>Diameter, Inches</u>	<u>Height, Feet</u>	<u>L/D Ratio</u>	<u>Approximate Superficial Gas Velocity CM/SEC</u>
A-1 *	CSTR	-	-	-	-
Merriam	bubble column	1.1	5	55	0.5
P-99 (tall reactor)	bubble column	4.25	20	56	2
P-99 (short reactor)	bubble column	6.0	10	20	1
Ft. Lewis	bubble column	24.0	30	15	4
<u>Demo Plant Design</u>					
Single Reactor per train (3000 TPD coal/reactor)	bubble column	132	148	13	10
Two Parallel reactors per train (1500 TPD coal/reactor)	bubble column	132	66	6	5

*The A-1 unit reactor was a one liter well- agitated autoclave, representative of an ideal continuous stirred tank reactor (CSTR).

2.0 Cold Flow Bubble Column Studies

2.1 University of Pittsburgh

Comprehensive cold flow studies of factors influencing the hydrodynamic behavior of bubble columns were conducted at the University of Pittsburgh on a continuous liquid flow bubble column (6" ID x 11' high) and on a batch liquid bubble column (1' ID x 8' high)³. Gas holdup and thermal dispersion were studied at various levels of gas and liquid velocity, liquid surface tension, liquid composition, and viscosity with both Newtonian and non-Newtonian fluids. Some tests were also made with coal/water or sand/water slurries.

Gas holdup was found to increase with gas velocity, but decrease with increasing liquid velocity, solids content, and particle size, as has generally been reported in the literature. Typical results obtained with the air/water/coal system are shown in Figure 1. A major observation in this program was unexpectedly large gas holdup observed in aqueous solutions of propanol and butanol. As shown in Figure 2, at gas velocities of 10 cm/sec, the gas holdup in the aqueous propanol and butanol systems was greater than 35% of bubble column volume. The relatively low surface tension of these mixtures (45-65 dynes/cm versus approximately 70 dynes/cm for water at the test conditions) was expected to result in higher gas holdups than the air/water system, but the magnitude of the differences was expected to be small. These studies also indicated that as alcohol chain length increased, gas holdup increased at equivalent surface tension, suggesting that liquid composition played a major role in gas holdup behavior.

Qualitative observation of the gas holdup behavior of these systems and application of Zuber-Finley theory suggested that the high levels of gas holdup with aqueous propanol and butanol solutions were associated with small bubble sizes and high radial uniformity in the distribution of bubbles across the cross section of the bubble column. The lower levels of holdup with ethanol and methanol solutions exhibited increasing non-uniformity of bubble distribution across the column. The uniformity of bubble distribution and bubble size are indications of the degree of coalescence of bubbles in the column. Evidence of

small bubbles suggests minimal coalescence. Non-uniform bubble distribution suggests that a velocity gradient existed across the column cross section due to different bubble sizes and rise velocities.

Calculated thermal dispersion coefficients indicated that existing correlations, when corrected for measured holdup, provided adequate estimates of dispersion coefficients.

The general conclusion to be drawn from this study is that caution is necessary in predicting gas holdup for SRC-II dissolvers from available literature correlations. Liquid phase characteristics can dramatically affect holdup in ways that are not clearly understood. It should be noted that liquid surface tension at the SRC-II dissolver conditions is expected to be approximately 5 dynes/cm, an order of magnitude less than the water or alcohol systems tested here. This should qualitatively result in very small bubbles that are very stable, and tend not to coalesce. Gas properties in the SRC dissolver will also be significantly different than gas properties studied in cold flow tests. Application of cold flow holdup correlations should be clearly validated by testing of these models in an actual SRC- II environment.

2.2 Gulf Science & Technology Company Studies

GS&TC conducted cold flow studies on a large scale (25' high x 12" ID) plexiglass bubble column with air/water/sand systems ^{4,5}. The primary goal of these studies was to determine if the assumptions inherent in the axial dispersion model were satisfied in a tall column, and if literature correlations accurately predicted dispersion coefficients. Gas holdup was also measured.

Dispersion was qualitatively evaluated by observing backmixing after injecting dye into the column⁴, and quantitatively evaluated by using an electrolytic tracer⁵. Visual observation of dye tracers suggested that the dye appeared to concentrate vertically in distinct zones or "cells". Each cell appeared to be backmixed within itself, but mixing between cells appeared limited. Approximately two minutes was required to achieve equilibrium dye concentration in the column. Comparison of the liquid phase Peclet Number

calculated from the electrolytic tracer tests with the Peclet Number calculated from literature correlations suggested that backmixing in the bubble column was less efficient than would be predicted.

The qualitative and quantitative test results suggested that the use of standard dispersion coefficient calculations and the axial dispersion model may be inadequate in modeling backmixing in tall bubble columns.

Measured gas holdup in the GS&TC bubble column agreed with predicted values based on literature correlations developed for similar air/water systems.

2.3 Conclusions from Cold Flow Tests

Two conclusions may be drawn from the cold flow bubble column tests. First, existing gas holdup correlations based on cold flow aqueous tests appear to be highly empirical, and may produce incorrect estimates of holdup when extrapolated to different systems. Second, the application of dispersion coefficient correlations and the axial dispersion model to large bubble columns may result in misleading interpretations of the type and degree of backmixing.

FIGURE 1.
GAS HOLDUP VS. SUPERFICIAL GAS VELOCITY FOR
THE AIR-WATER-COAL SYSTEM.

FIGURE 2.
GAS HOLDUP VS. SUPERFICIAL GAS VELOCITY FOR
AQUEOUS ALCOHOL MIXTURES.

3.0 SRC-II Reactor Studies

3.1 P-99 Reactor L/D Studies

If there is a high degree of backmixing in an SRC dissolver, and the exothermic heat of reaction generated is available to heat the feed slurry to reaction temperature, it should be possible to operate at a preheater outlet temperature of approximately 720°F and to minimize dissolver quench requirements. To study the effect of dissolver length on backmixing, the P-99 PDU was operated with dissolvers of equal volume, but differing height (See Table D).

These studies were initially conducted during Runs 63-66, in which a low reactivity coal was processed⁶, and were later repeated in Runs 77-81 with a normal reactivity coal⁷. Typical dissolver temperature profiles for these runs are shown in Figure 3. Average temperature in the 20 ft reactor runs was consistently lower than in the 10 ft reactor runs as evidenced by the greater temperature range across the tall reactor. This would appear indicative of less efficient thermal backmixing in the taller reactor vessel.

Comparison of required preheater outlet temperatures indicates that for the normal reactivity coal, the reactor height did not affect the required temperature, but for the low reactivity coal, a higher preheater outlet temperature was required for the tall dissolver run.

Product yields for both reactor sizes were not significantly different, even though the average dissolver temperature for the tall reactor runs was approximately 10°F less than the average temperature in the short reactor (846°F vs. 856°F). The lack of a significant difference in product yields highlights the uncertainty that exists in the relationship between product yields and slurry temperature and residence time distribution in the dissolver. The less backmixed environment in the tall reactor appeared to make it possible to achieve comparable yields with the more backmixed short reactor at a lower temperature, suggesting some merit to having a less backmixed slurry phase.

3.2 Ft. Lewis Dissolver Downcomer Tests

The Ft. Lewis dissolver was believed to be a well backmixed system. It was possible to operate the plant with as low as a 730°F preheater outlet temperature, while maintaining the 860°F dissolver outlet temperature. The temperature profile in the dissolver was fairly flat, with the observed temperature rise only 5-15°F along the dissolver length. The extent to which such efficient backmixing would prevail on tall commercial scale reactors is uncertain. One possible technique suggested for improving top-to-bottom backmixing in tall bubble column reactors was installation of a downcomer within the reactor. A downcomer was installed at Ft. Lewis to test the feasibility of this concept in an SRC-II dissolver.

Tests with the downcomer demonstrated that it was possible to establish flow from the top to the bottom of the dissolver through such a device. The fluid temperature in the downcomer was constant along its length, and was hotter than the surrounding material in the reactor by as much as 10°F. Flow was controllable by injecting purge gas into the bottom of the downcomer. Because the Ft. Lewis dissolver was already backmixed, it was not possible to quantify the effect of the downcomer on mixing, but thermal data appeared to indicate a slight shift towards more isothermal operation with the downcomer in service.

3.3 Ft. Lewis Radiotracer Studies

3.3.1 Background

Residence time distribution and phase holdup data in an SRC-II reactor environment were obtained through radiotracer studies on the Ft. Lewis dissolver during normal operations. Gas (argon-41), liquid (bromine-42 as bromophenanthrene), and solid (flaked or colloidal gold-198) radioactive tracers were injected either into the dissolver or upstream of the dissolver and the concentration of radioactive material over time was measured with scintillation detectors at various locations along the dissolver. The unique problems associated with the injection and detection of radiotracers into a high pressure reactor, and with the interpretation of data resulted in the need to develop specific techniques for these studies. Four series of radiotracer tests were completed at Ft. Lewis from June, 1979 through February, 1981. Due to the evolutionary nature of this program, the best results were believed to be those

from the February, 1981 test series, which are discussed below. The major emphasis in the radiotracer tests was to study backmixing in the reactor. However, by analyzing the tracer concentration data, it was also possible to obtain estimates of mean residence time and phase holdup.

3.3.2 February 1981 Tracer Test Program

Ten successful injections of BR-32 and AR-41 were made during the February 1981 test program⁸. (Injections of AU-198 were not attempted because of unresolved problems with previous attempts to inject this radiotracer.) These tests were conducted to investigate flow patterns and mixing phenomena in conjunction with dissolver downcomer tests. Scintillation detectors were located at the top, middle, and bottom of the dissolver, as shown in Figure 4. Radiotracer injections were made at either the dissolver inlet or at the middle of the dissolver.

The injections at the dissolver inlet were made to obtain information on gas residence time and holdup in the reactor, and to study flow patterns. Inlet injections are particularly useful in describing flow in a plug flow system. Operating conditions during the inlet injection tests are shown in Table II.

Injections of gas and liquid radiotracers at the middle of the dissolver were made to study the extent of backmixing of each phase in the dissolver. Comparison of the responses of the top and bottom detectors to middle injections provided information on the extent of phase recirculation within the dissolver. Operating conditions during the middle injection tests are shown in Table III.

3.3.3 Analytical Methodology⁹

The dispersed plug flow, or axial dispersion model was assumed to represent flow in the dissolver. Radiotracer concentration data were analyzed using the method-of-moments technique, assuming open vessel boundary conditions.

Mean Residence Time (MRT) between detectors was determined by subtracting the peak centroid (first moment) of the inlet peak from the centroid of the outlet peak. Peak centroid (μ) was determined from the equation:

$$\mu = \frac{\sum_{i=a}^b A_i t_i}{\sum_{i=a}^b A_i}$$

Where A_i is the number of counts recorded in the i th channel of the multichannel analyzer, t_i is the time associated with the i th channel and a and b represent the boundaries of the peak.

The peak variance (σ^2), the second moment, was calculated as:

$$\sigma^2 = \frac{\sum_{i=a}^b A_i (t_i - \mu)^2}{\sum_{i=a}^b A_i}$$

The dispersion number (D/uL) was determined from the equation:

$$\frac{D}{uL} = \frac{1}{8} \sqrt{\frac{8}{(MRT)^2} (\sigma_{out}^2 - \sigma_{in}^2) + 1} - 1$$

Where D is the axial dispersion coefficient; u is the phase velocity; L is the vessel length; MRT is the mean residence time; σ_{out}^2 is the variance of the outlet peak; and σ_{in}^2 is the variance of the inlet peak.

The calculation of residence time and dispersion number are highly sensitive to

the profile of the detector measurements. The method-of-moments technique is ideally suited to systems which approach plug flow and result in tracer concentration curves that approach Gaussian distributions. As flow systems deviate from plug flow, the moment calculations are highly dependent upon tailing in the detector curves. While the gas phase response curve at Ft. Lewis exhibited a very distinct and sharp peak, as shown in Figure 5 suggesting an approach to plug flow, the slurry response curve appeared to approach the exponential decay typical of a backmixed system. The long tail of the slurry phase curve, as shown in Figure 6, can present a significant problem in a method-of-moments analysis, particularly if baseline drift occurs. The baseline drift will affect estimates of both the first and second moments, possibly leading to erroneous conclusions about residence time and the extent of backmixing. Backmixing was computed for middle injections from the signals of the "top" and "bottom" detectors at the side of the dissolver. Backmixing was defined as the ratio (in percent) of the total radioactivity passing the bottom detector compared with the total passing the top detector. That is:

$$\% \text{ Backmixing} = \frac{\sum_{i=a}^b (A_{\text{bottom}})_i}{\sum_{i=a}^b (A_{\text{top}})_i} \times (100)$$

Where $(A_{\text{bottom}})_i$ is the number of counts recorded in the i th channel of the multichannel pulse height analyzer connected to the bottom detector; and $(A_{\text{top}})_i$ is the number of counts at the top detector. The detectors were balanced each day using an 8 microcurie sealed source of cesium-137 located at the same position adjacent to each detector.

TABLE II

OPERATING CONDITIONS DURING RADIOTRACER INJECTION AT REACTOR INLET

Date	<u>2/20/81</u>	<u>2/23/81</u>	<u>2/23/81</u>	<u>2/24/81</u>	<u>2/24/81</u>
Time	1420	1451	1709	1606	2048
Calc Coal lb/hr	2163	2130	2400	2100	2100
Coal lb/hr	2099	1999	2112	1972	2000
Coal in Feed Slurry, wt %	31.3	31.3	36.4	31.9	30.9
Solvent lb/hr	909	929	935	943	941
SL to OI lb/hr	3834	3757	3257	3540*	3746
Total Slurry Rate, lb/hr	6903	6816	6592	6583	6787
H ₂ to O2 lb/hr	739	781	744	760	757
H ₂ to Preheater Inlet lb/hr	310	324	315	315	316
H ₂ to Preheater Outlet lb/hr	419	446	420	436	437
Reactor Inlet Gas lb/hr	719	770	735	751	753
H ₂ to Downcomer (low) lb/hr	2.5	2.5	2.4	0	0
H ₂ to Downcomer (high) lb/hr	3.5	3.6	3.5	19.2	19.3
H ₂ to Top Quench lb/hr	0	0	0	0	0
H ₂ to Sample PTS & Top Min. Quench	14.6	14.0	13.7	13.7	13.5
Reactor/Inlet °F	807	809	810	805	804
Reactor/Outlet °F	863	866	863	862	861
Reactor/Pressure	1868	1871	1870	1877	1882
Type Injection	Ar	Ar	Br	Ar	Br

* Manually Taken Rate

TABLE III

OPERATING CONDITIONS DURING RADIOTRACER INJECTION AT MIDDLE OF REACTOR

Date	<u>2/19/81</u>	<u>2/23/81</u>	<u>2/23/81</u>	<u>2/24/81</u>	<u>2/24/81</u>
Time	1507	1003	1625	1522	1659
Calc Coal lb/hr	2122	2100	2012	2100	2020
Coal lb/hr	2100	1909	2010	1970	1990
Coal in Feed					
Slurry, wt %	30.3	30.9	30.1	25.4	31.7
Solvent lb/hr	912	941	930	941	943
SL to O1 lb/hr	3864	3747	3740	5214*	3413
Total Slurry Rate, lb/hr	6898	6788	6682	8255	6376
H ₂ to O2 lb/hr	514	721	752	745	756
H ₂ to Preheater Inlet lb/hr	----	297	320	310	313
H ₂ to Preheater Outlet lb/hr	----	414	425	428	434
Reactor Inlet Gas lb/hr	514	711	745	738	747
H ₂ to Downcomer (low) lb/hr	2.4	2.3	2.5	0	0
H ₂ to Downcomer (high) lb/hr	3.2	3.2	3.6	19.0	19.1
H ₂ to Top Quench lb/hr	72.0	0	0	0	0
H ₂ to Sample PTS & Top Min. Quench	14.7	12.8	14.0	13.5	13.8
Reactor/Inlet °F	748	809	811	805	804
Reactor/Outlet °F	824	861	865	862	860
Reactor/Pressure	1872	1881	1866	1878	1879
Type Injection	Br	Br	Ar	Ar	Br

* Manually Taken Rate

3.3.4 Inlet Injection Results

Results from the gas and liquid phase radiotracer injections at the inlet of the dissolver are presented in Table IV⁸. Three injections of the gas phase radiotracer were made. The first and third gas injections resulted in similar estimates of mean residence time to the top detector (58.7 and 65.3 seconds) and middle detector (23.5 and 26.8 seconds). Results from the second gas tracer injection produced much different estimates of residence time. A review of the tracer concentration curves produced in the second injection revealed substantial noise in the detector signals. In light of this, and the apparent anomaly in mean residence time, the results from the second injection are not included in the discussion that follows.

The operating conditions and tracer results from the first and third gas tracer injections are summarized in Table V. Both injections resulted in significantly different estimates of residence time for the top and bottom detectors. This is an indication that gas flow through the dissolver approached plug flow, with the bulk of the radiotracer passing the bottom detector well in advance of passing the top detector. The distinct peak in the detector curves supported this conclusion. The mean residence time to the top detector is a reasonable indication of total residence time in the dissolver, as it includes approximately 95% of the dissolver volume.

To analyze the tracer curve results, it is necessary to consider the flow conditions and detection techniques at the measurement points¹⁰. The Ft. Lewis dissolver system was representative of an open vessel (undisturbed flow at measurement points) with radiotracer measurement via a "through the wall" detection technique. The first moment (Mean Residence Time) of the concentration curves produced in this system was related to actual mean residence time by the following equation:

$$\text{Actual Residence Time } (\bar{t}) = \frac{\text{Concentration Curve MRT } (\bar{t}_c)}{1 + 2 \times \text{Dispersion Number } \left(\frac{D}{uL}\right)}$$

Fractional gas holdup was calculated by the following relationship:

$$\text{Gas Holdup (E}_g\text{)} = \frac{\text{Actual Mean Residence Time } (\bar{t}) \times (\text{Volumetric Flowrate})}{\text{Vessel Volume to Detector}}$$

Actual mean residence times and fractional gas holdups for the gas phase injections are shown in Table V. Both injections produced estimates of overall gas holdup in the dissolver of approximately 20% of the dissolver volume. Both injections also produced estimates of extremely large gas holdup at the bottom of the dissolver (45%). Whether this large gas holdup was real, possibly due to the simple nozzle-type feed distributor at the dissolver inlet, or incorrect due to problems in radiotracer injection or measurement techniques is uncertain. Further studies would have been warranted to resolve this, had the SRC-II project continued.

Calculated gas holdup between the detectors, in the bulk of the dissolver volume, based on the difference in gas holdup estimated at each detector is also shown in Table V. The reliability of the estimated holdup of 14% to 18% is dependent on the accuracy of the bottom detector measurements. The overall holdup of 20% discussed above is believed to be most representative of conditions in the Ft. Lewis dissolver because it is independent of the puzzling estimates based on the bottom detector.

The estimates of gas holdup obtained from the tracer studies differed significantly from the holdup estimated by the SRC-II Demonstration Plant gas holdup correlation. Calculated holdup based on that correlation¹¹ should have been approximately 10.5% at the conditions of the gas radiotracer injections. At the 20% overall gas holdup estimated from the radiotracers, the actual slurry residence time in the Ft. Lewis dissolver was only about 90% of what it was believed to have been.

Review of the liquid injection data in Table IV indicates that tracer curve MRT values were similar for the top and bottom detectors in both liquid injections. This response would be expected for a well backmixed system. The exponential

decay of the tracer curves (Figure 6) also supports this. Further interpretation of the results from these liquid tracers is not warranted because of the highly backmixed nature of the slurry phase. The mathematical techniques for calculating dispersion number discussed above must be complemented by a qualitative review of whether the calculated model parameters were representative of the concentration curves observed. The mathematical techniques used here simply give a best estimate of curve parameters. Further work in analyzing the slurry phase measurements is needed to validate this approach. Based on the data available from these studies, the best estimate of average slurry residence time is derived from gas phase holdup estimates by assuming slurry holdup to be equal to the gas-free volume of the dissolver.

3.3.5 Middle Injection Results

The injection of radiotracers into the middle of the dissolver provided a quantitative means of observing the extent of backmixing by comparing responses of the top and bottom detectors. Results from gas and liquid injections are shown in Table VI⁸. High estimates of backmixing are indicative of comparable measurements of concentration at the top and bottom detectors. The data in Table VI for the liquid phase radiotracers show high levels of backmixing. The gas radiotracer data show much less backmixing, but the fact that radiotracer concentrations were detected at the bottom of the dissolver after injection at the middle of the dissolver suggests that some gas phase dispersion was occurring. This could have been due to either solubility of the argon radiotracer in the slurry phase or entrapment of small gas bubbles in the liquid phase as it was recirculating to the bottom of the dissolver.

TABLE IV

SUMMARY OF DATA FROM RADIOTRACER INJECTIONS AT REACTOR INLET
February 1981
SRC-II MODE

<u>Date</u>	<u>Time</u>	<u>Phase</u>	<u>Detector Location</u>	<u>Mean Residence Time</u>	<u>Dispersion Number</u>
2/20/81	14:20	Gas	Top	58.7 sec.	0.092
			Bottom	23.5 sec.	0.081
2/23/81	14:51	Gas	Top	104.0 sec	0.104
			Bottom	11.3 sec.	0.077
2/23/81	17:09	Liquid	Top	63.6 min.	0.145
			Bottom	65.6 min.	0.149
2/24/81	16.06	Gas	Top	65.3 sec.	0.076
			Bottom	26.8 sec.	0.165
2/24/81	20.48	Liquid	Top	65.4 min.	0.143
			Bottom	63.4 min.	0.154

TABLE V
Inlet Injection Gas Tracer Results

	<u>2/20/81</u> <u>Injection</u>	<u>2/24/81</u> <u>Injection</u>
<u>Plant Conditions</u>		
Feed Slurry Rate, LB/HR	6903	6583
Feed Gas Rate, LB/HR	739	760
Gas Molecular Wt.	4.35	4.50
Gas Volumetric Flow, SCFH	64471	64093
Gas Volumetric Flow, ACFH (860°F)	1278	1264
<u>Tracer Data</u>		
<u>Top Detector</u>		
Concentration Curve MRT ($\bar{t}_C$)	58.7 sec	65.3 sec
Concentration Curve Dispersion Number ($\frac{D}{uL}$)	0.092	0.076 sec
Estimated Gas Phase Mean Residence Time	49.6 sec	56.7 sec
Vessel Volume to Detector	88.4 Ft ³	88.4 Ft ³
Fractional Gas Holdup	0.199	0.225
<u>Bottom Detector</u>		
Concentration Curve MRT ($\bar{t}_C$)	23.5 sec	26.8 sec
Concentration Curve Dispersion Number ($\frac{D}{uL}$)	0.081	0.165
Estimated Gas Phase Mean Residence Time	20.2 sec	20.2 sec
Vessel Volume to Detector	16.1 Ft ³	16.1 Ft ³
Fractional Gas Holdup	0.445	0.441
Calculated Fractional Gas Holdup Between Top and Bottom Detectors	0.144	0.441

TABLE VI**SUMMARY OF DATA FROM RADIOTRACER INJECTIONS INTO MIDDLE OF REACTOR****February 1981****SRC-II MODE**

<u>Date</u>	<u>Time</u>	<u>Phase</u>	<u>Detector Location</u>	<u>Mean Residence Time</u>	<u>Dispersion Number</u>	<u>% Backmixing</u>
2/19/81	15:07	Liquid	Top	67.5 min.	0.142	73%
			Bottom	66.9 min.	0.145	
2/23/81	10:03	Liquid	Top	71.0 min.	0.130	99%
			Bottom	64.4 min.	0.152	
2/23/81	15:25	Gas	Top	41.0 sec.	0.128	24%
			Bottom	44.8 sec.	0.120	
2/24/81	15:22	Gas	Top	26.1 sec.	0.131	2%
			Bottom	33.5 sec.	0.095	
2/24/81	16:59	Liquid	Top	62.7 min.	0.154	97%
			Bottom	62.4 min.	0.154	

FIGURE 3.

COMPARISON OF P-99 DISSOLVER TEMPERATURE
PROFILE FOR THE 10 FT AND 20 FT TALL REACTORS

FIGURE 4.

FT. LEWIS RADIOTRACER DETECTOR LOCATIONS

FIGURE 5.

TYPICAL RESPONSE FOR GAS PHASE INJECTION

FIGURE 6.

TYPICAL RESPONSE FOR A LIQUID PHASE INJECTION

4.0 Discussion of Tracer Results

The gas holdup results from the Ft. Lewis tracer studies indicate that an increase in emphasis on developing holdup correlations specific to the high temperature, high pressure dissolver environment is necessary. The higher-than-expected gas holdup would have dramatic implications in a commercial SRC-II facility. Higher gas holdups lead to shorter slurry residence time in the dissolver, which results in less conversion of heavy coal liquids to hydrocarbon distillate and gas products.

At Ft. Lewis, double-the-expected gas holdup (20% vs 10% expected) would have resulted in approximately a 10% decrease in expected slurry residence time. Yield correlation models estimate that such a loss in residence time would lead to a small increase in SRC yield (loss of lighter hydrocarbon products) of 2-3% mf coal. Such a yield difference would probably not be observed as statistically significant at Ft. Lewis due to the small number of material balance runs conducted there and the wide range of process conditions and coals tested.

Extrapolation of the Ft. Lewis gas holdup results to the Demonstration Plant scale would suggest gas holdup of approximately 40% in the single vessel per train design at the high design gas velocities (10 cm/sec). Although this would be an unexpectedly large gas holdup based on most bubble column cold flow studies, the University of Pittsburgh studies discussed above indicate that such large gas holdups can occur in some systems. With a gas holdup of 40%, the actual slurry residence time in the Demonstration Plant would have been approximately 80% of the design level. The lower slurry residence time would be expected to result in an increase in SRC yield of 5% m.f. coal or more. A shift from the design yields of this magnitude would result in a significant decrease in liquid product yield, with excess vacuum bottoms production. Such a shift in yield could have become a serious long term problem in a highly integrated commercial facility, and to balance the overall plant, it might have become necessary to decrease coal throughput to lower the SRC yields. This would amount to derating the unit.

This discussion highlights the importance of developing improved gas holdup correlations in optimizing the design of a commercial facility. In the absence of an improved database in this area, it is appropriate to consider the operating and economic risks inherent in not designing for enough slurry residence time. Reconsideration of dissolver design might have

led to increasing the designed dissolver volume or selecting a multiple dissolver per train concept which would lead to lower velocity and gas holdup in each vessel.

The tracer study residence time distribution data showed that the gas phase approached plug flow, while the slurry phase approached backmixed behavior. The axial dispersion model is suited for systems which approach plug flow, and therefore can reasonably be applied to the gas phase. However, caution must be exercised in applying this model to backmixed systems. Insufficient residence time distribution data has been collected and analyzed to determine if the calculated dispersion number accurately depicts the residence time distribution of the slurry phase. Inappropriate application of this model to the slurry phase could lead to over-simplification of the environment in the dissolver and inadequate provision for slurry backmixing. While the downcomer was established as a means of mitigating this problem, its effectiveness on a large scale is unknown. An alternative in the case of poor thermal backmixing would be to raise the preheater outlet temperature above its design operating point. To keep from exceeding the preheater design tubeskin temperature and heat flux rate in a large plant, it might have been necessary to decrease flow through the preheater, thereby derating the capacity of the plant.

5.0 Conclusions

- Phase holdup and residence time distribution information for the Ft. Lewis dissolver were obtained through radiotracer injection. Locating detectors along the length of the dissolver, and injecting tracers at the inlet of the vessel and into the middle of the vessel provided residence time data which were useful in evaluating flow patterns.
- Gas holdup calculated from the Ft. Lewis tracer tests was found to be much higher than predicted from bubble column correlations. Gas and liquid composition and operating conditions are believed to have been significant factors in the high level of gas holdup observed.
- The radiotracer concentration curves developed from the tracer tests showed the slurry phase to be backmixed and the gas phase to approach plug flow behavior.
- Caution must be exercised in applying the axial dispersion model to backmixed systems. Whether the model parameters calculated from the Ft. Lewis data accurately described the degree and form of backmixing in the reactor is uncertain.
- The downcomer tested at Ft. Lewis showed promise as a way to deal with inadequate backmixing in a tall reactor, should that be a problem. Flow of slurry through the downcomer was detectable from temperature measurements, but the quantity of flow and its effect on mixing in the reactor could not be determined. Prediction of downcomer performance on a large scale unit is uncertain.
- The design philosophy of the large scale reactor vessel for an SRC-II plant should consider the effects of performance uncertainties on downstream units which are closely integrated to the reaction area. Failure to adequately consider these effects may lead to derating the overall facility to bring the process units into balance.

6.0 Future Considerations

In a commercial plant, large single reactors per train offer both capital cost and reduced complexity advantages when compared to smaller reactors operating in parallel. However, serious questions have been raised about extrapolating correlations too far past existing data. The Fort Lewis tracer studies raise serious concern about correlations for predicting phase holdup in large scale reactors. There is also concern about the reliability with which the extent of slurry backmixing can be predicted. Studies of backmixing in larger cold flow bubble columns would increase confidence in scale-up to larger size dissolvers. However, many issues cannot be fully resolved until a large scale liquefaction facility is operated, and holdup and backmixing in an actual reaction environment can be evaluated.

An engineering review of data from the other direct liquefaction programs should be made to expand current understanding and further delineate areas where additional work is needed. Future direct liquefaction pilot plants and demonstration facilities should be designed with more consideration given to obtaining information on the reactor environment.

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