Summary

This is the final report under Contract Number DE-AC22-79ET14801 titled "Gas/Slurry Flow in Coal Liquefaction Processes", covering the period 1 October 1979 to 31 March 1982. This work is a continuation of studies initiated by Air Products and Chemicals, Inc. on the fluid dynamics of 3-phase flow to support the design of a 6000 T/D commercial dissolver. Design studies on the SRC-I 6000 T/D commercial plant began in July, 1978. DOE support for the 3-phase flow studies by Air Products began 1 July 1979 under contract number DE-AC05-78-ORO-3054 and terminated 30 September 1979 at the start of this contract. A background of information developed at Air Products is included in the text.

A commercial coal liquefaction plant will employ vertical tubular reactors feeding slurry and gas concurrently upward through these vessels. In the SRC-I plant design the reactor is essentially an empty vessel with only a distributor plate located near the inlet. Because the commercial plant represents a considerable scale-up over Wilsonville or any pilot plant, this program addressed the need for additional data on behavior of three phase systems in large vessels. Parameters that were investigated in this program were studied at conditions that relate directly to projected plant operating conditions.

The design goal for commercial dissolvers is to ensure that solids passing through the reactors remain suspended in sufficient quantity and distribution to provide the necessary catalytic effect. To achieve this goal, the fluid dynamic behavior of a three-phase upflow system was studied by measuring gas and slurry holdup, liquid dispersion, solids suspension and solids accumulation. The dependent parameters are gas and liquid velocities, solid particle size, solids concentration, liquid viscosity, liquid surface tension and inlet distributor. Two reactor columns (5-inch diameter by 5 feet and 12-inch diameter by 25 feet) were used in this cold-flow simulation which included air/water/sand, nitrogen/tetralin/sand, and air/glycol-water/sand systems. In addition, a model was developed to describe the distribution and accumulation of solid particles in the reactor columns.

Gas holdup in gas/liquid and gas/liquid/solid systems were investigated both with and without liquid flow. Within the range of liquid superficial velocity from 0.0-0.5 ft/sec, gas holdup is found to be independent of liquid flow which agrees with other investigators. The results also confirm our previous finding that gas holdup is independent of column diameter when the column diameter is 5 inches or larger. This independence of column diameter has been tested with a wide range of operating conditions: 0.0-0.6 ft/sec liquid velocity, 0.0-0.4 ft/sec gas velocity, 140 minus - 20 mesh particles, 0.0-20.0 lb/ft^3 solids concentration, with and without distributor.

The gas holdup depends strongly on gas flow rate; gas holdup increases with increasing gas velocity. In general, gas holdup is found proportional to $(V_g)^{0.75}$, where V_g is gas superficial velocity. A lower exponential value (0.50-0.65) was measured when either 50% glycol-50% water and 70% glycol-30% water solutions were used.

Effect of solids particles on gas holdup depends on the gas flow rate. At low gas superficial velocities (≦0.10 ft/sec), the presence of solids did not change the gas holdup. However, at high gas flow, gas holdup was reduced slightly (3-10%) with both fine and large particles. This effect is true regardless of the type of inlet distributor.

Increasing liquid viscosity and surface tension reduce gas holdup which agrees with other investigators. Because of the complexity of the system, we could not find a single correlation to best fit all the data. However, the correlations developed by Akita and Yoshida, Pilhofer et al., and Hughmark all fit the data with a reasonable degree of accuracy. It is important to note that none of these correlations can be applied to a foaming system.

The degree of liquid backmixing markedly affects chemical changes occurring in the dissolver, such as sulfur removal, and oil and distillate formation. The liquid dispersion coefficient, which is a measure of the degree of backmixing, was found to increase with increasing gas velocity and column diameter.
Within the range of liquid physical properties employed in this work, no trend

of dependence on viscosity and surface tension was observed. In a gas/liquid two phase upflow system, a correlation developed by Baird et al., best fit our data. Several other correlations, including one developed by Air Products, underpredict our results slightly.

The liquid dispersion coefficient was found to be independent of liquid velocity and inlet distribution. However, the presence of solid particles reduced the coefficient which decreased with increasing solids concentration, regardless of the particle size. Nevertheless, the liquid dispersion results have clearly shown that the SRC-I Demonstration Plant Dissolver closely resembles a completely backmixed reactor.

The amount of hydrogen gas transferred to the bulk liquid is an important factor in the dissolver design. Chemical reactions, such as solvent rehydrogenation, will be limited by the gas/liquid mass transfer rate if the hydrogen transfer is less than the rate consumed by the reactions.

Gas/liquid mass transfer rates were investigated in both air/water and air/water/sand systems using a 5-inch and 12-inch diameter column. Our results indicate that the mass transfer rate increases with increasing gas velocity. However, its dependence on gas velocity behaves differently as the column diameter varies. In both the 5-inch and 12-inch diameter columns, the mass transfer coefficient increases with increasing gas velocity, but the rate of increase in the larger vessel decreases with increasing gas velocity whereas in the 5-inch diameter column, the rate of increase is almost constant.

The gas/liquid mass transfer coefficient is independent of column height and inlet distribution because of the highly backmixed system. However, the presence of solids reduces the transfer rate. A correlation developed by Yoshida and Akita for a gas/liquid two-phase system was found to fit our air/water data. This correlation was modified to describe the mass transfer rate in a gas/liquid/solid system.

Gas velocity is the key element for maintaining solid particles in suspension. Above the critical gas velocity, which is defined as the minimum gas velocity to achieve full solids suspension, solids accumulation is practically independent of gas velocity. However, solids accumulation was found to increase linearly with decreasing liquid velocity at a fixed gas input rate. The effect of liquid and gas velocities on solids accumulation was studied in a 5-inch and 12-inch diameter column using 60/80 and 140 minus mesh sand particles. The superficial gas and liquid velocities were varied from 0.01 to 0.43 ft/sec and from 0.0 to 0.05 ft/sec, respectively, covering the flow conditions designed for the SRC-I demonstration plant. At a half slurry turndown velocity, the accumulation of the fines (140 mesh minus) did not increase substantially but the amount of large particles (60/80 mesh) increased tremendously. Of more concern is the settling of the larger particles which have critical gas velocities higher than the half gas turndown velocity.

Increasing column diameter and liquid viscosity reduces the accumulation of solids in the dissolver. As the column diameter increases, dispersion in the system also increases, thereby reducing the solids concentration gradient and accumulation. Since particle terminal velocity decreases with increasing liquid viscosity, the solid particles can be carried through the column by the bulk liquid flow easier in a more viscous liquid. Therefore the accumulation is reduced. These effects of column diameter and liquid viscosity are more apparent on the large particles because the distribution of fine particles is nearly homogeneous whereas the large ones show a substantial concentration gradient.

A solid dispersion model was developed to predict the solids concentration gradient and accumulation in a gas/liquid/solid upflow system. This model was tested successfully with 40 runs conducted in water, tetralin, and 50% glycol-50% water solution under a wide range of operating conditions covering the SRC-I plant design. We also found that there is little particle-particle interaction to affect the solids distribution profiles of different particle sizes. Therefore the model-predicted concentration profile of each individual particle size can be superimposed together to provide a complete description of the solids distribution in the dissolver.

When the critical gas velocity of any particle exceeds the operating flow conditions, the solid particles will settle at the reactor bottom, creating an undesirable situation. Our results indicate that a solid withdrawal system located at the reactor bottom can reduce solids accumulation and significantly alter the solids distribution profile. Although all solid particles, large or small, were removed together at the bottom of the column, our data indicate that the larger particles were removed faster because they settled to the bottom more quickly. However, these results were obtained under non-steady-state operation. It is recommended to perform steady-state solids removal experiments in the future to determine the optimal operating conditions.