SECTION 1: INTRODUCTION AND BACKGROUND

The Solvent-Refined Coal (SRC) process involves the use of a liquefaction reactor in which coal is dissolved in a hydrogen donor solvent in the presence of hydrogen gas. This reactor, commonly known as the dissolver, operates at high temperatures and pressures and consists of a vertical tube containing an upward flowing three-phase mixture of coal, solvent, and hydrogen gas. The complexities associated with a three-phase flow system present a series of problems in the efficient design of the dissolver. The occurrence of unequal residence times of the phases involved, the existence of different mixing patterns of those phases, and the tendency of solids to accumulate at the bottom of the dissolver greatly complicate the reactor design equations. In order to design a technically feasible and cost-effective dissolver, the physical behavior of the three-phase system in the tubular reactor must be clearly understood.

The 6,000 tons/day (tpd) SRC-1 Demonstration Plant dissolver will be scaled up considerably from the Wilsonville and Ft. Lewis pilot plant dissolvers. The relative sizes of the dissolvers for the pilot plants and the SRC-1 Demonstration Plant are compared below at a residence time of 0.56 hr, a gas-feed rate of 20,000 scf per ton of dried coal, and a 38 wt % coal slurry:

Plant	Capacity (tpd)	Dissolver size			Superficial velocity	
		Diameter (ft)	Height (ft)	Volume (ft^3)	Liquid (fps)	Gas (fps)
Wilsonville	6-10	1.0	23	18.1	0.012	0.074
Ft. Lewis	50-61	2.0	34	106.8	0.017	0.10
SRC-1 Demo Plant	6,000	11.0	110	10,454.0	0.06	0.36

The reactor volumes and diameters differ dramatically. Thus, the scale-up to the SRC-I plant dissolver design cannot be trusted solely to be kinetic-reaction-dependent, but has to be supported by the sound practices of three-phase flow hydrodynamics. For example, the effect of differences in the superficial velocities of gas and liquid on slurry behavior should be considered. A five-fold difference in these velocities exists between the Wilsonville Pilot Plant and the SRC-I Demonstration Plant dissolvers. This difference can have a considerable impact on the process because the gas and liquid superficial velocities have a strong effect on: (a) gas void volume, (b) actual solids concentration in the dissolver, and (c) the relative degree of back-mixing. As velocity through the dissolver increases, the tendency for solids to remain behind diminishes, causing a decrease in the actual concentration of ash particles in the reactor. Those particles that do remain will tend to be larger. Considerable evidence shows that reactor solids have a definite catalytic effect. Larger particles will have less exposed surface area, and thus will probably have diminished catalytic activity. Knowing the particle sizes that can accumulate under SRC-I Demonstration Plant flow conditions will indicate the size of dissolver solids that should be examined for catalytic activity.

During the early stages of the SRC plant design, the problem of solids accumulation at the bottom of the dissolver reactor was realized. This problem has been a challenging one at the pilot plant and may be a very significant obstacle to the success of continuous operation of the SRC-I Demonstration Plant. It is a common belief that most of the coal dissolves throughout the preheater before entering the reactor. However, approximately 10% of this coal can be identified as mineral matter that does not dissolve

and presumably has a catalytic effect on the desired solvent regenerating reactions. Under the influence of gravity, these undissolved particles tend to accumulate in the lower section of the dissolver, causing tremendous complications that may eventually prevent continuous operation. A study by Exxon Corporation (1) suggested that since some particles are never going to reach the top of the reactor at the flow rates required for operation, a withdrawal system must be used to extract the particles larger than 8 mesh.

A literature survey on three-phase flow through tubular reactors led to the conclusion that bubble column slurry operations were characterized at zero liquid flow rates, and that no data exist for three-phase flow through tubular columns. Most of these publications put more emphasis on the dimensional analysis of gas-slurry systems. Although they describe most of the factors affecting flow patterns, these reports are based on information obtained from systems using significantly higher flow rates. The relevant conclusions of the reports are as follows:

1) Bubble column slurry operations are usually characterized by zero net liquid flow, and the particles are held suspended by momentum transferred from the gas phase to the solid phase via the liquid medium. (2)

2) The critical gas velocity for complete suspension of particles is mainly affected by liquid flow near the gas distribution, and therefore to obtain a small critical gas velocity, the shape of the bottom of the column and the position of the gas distributor may become important. (3)

3) Gas holdup and the concentration distribution of solid particles do not depend on gas distribution, or on the shape of the bottom of the column when solid particles are suspended completely. (3)

4) The range of gas velocity over which a bubble column slurry reactor may be operated decreases with increasing solids concentration. The lower limiting gas velocity increases because of an increasing tendency towards sedimentation, and the upper limiting gas velocity decreases because of an increasing tendency towards bubble coalescence and the corresponding increase in bubble size. (4)

5) The larger solid particles show a somewhat smaller gas holdup. This is considered to be caused by the larger rising velocity of coalesced bubbles in the presence of solid particles. In the region of high gas velocity, where large coalesced bubbles rise frequently, the effect of the concentration of solid particles on gas holdup becomes gradually smaller as gas velocity increases. Gas holdup was found to decrease by increasing the amount of solids in the reactor. (5)

6) The maximum quantity of solids that can be held in suspension in a liquid medium agitated by bubbles in a defined system is termed critical solids holdup. There are two regions of critical solids holdup with gas velocity. In the first region of low gas velocity, holdup is a function of gas distribution, and data indicate that an arrangement with about six orifices per square inch provides a reasonably uniform closed gas distribution. In the second region of higher gas velocity, critical solids holdup is independent of gas distribution. (6)

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7) Critical solids holdup decreases with increasing particle size of the
   solids and the surface tension of the liquid. (6)

8) Critical solids holdup is a function of solid surface properties, and for
   solids of different degrees of wettability in a liquid, solids holdup is
   much greater with a less wettable solid under identical operating
   conditions. (6)

9) The dispersion coefficient of suspended solid particles is the same as
   that of the liquid within the column without solid particles. (3)

10) The longitudinal dispersion coefficients of the liquid and of the slurry
    increase with an increase in superficial gas velocity and are
    proportional to the 1 to 1.5th power of the column diameter. (5)

The last two observations are related to mass and heat transfer in slurry
bubble columns:

11) The overall volumetric absorption coefficient kgA_6 was observed to
    increase with gas velocity toward an upper limiting value of about 1.3 x
    10^{-6} mole/min for the absorption of oxygen from air in aqueous sodium
    sulfite solution containing cupric ions as catalyst, as well as sand
    particles (particle size 0.015 to 0.030 cm). At low gas velocities,
    absorption was observed to decrease with increasing particle size,
    increasing amount of solids in the column, and increasing density between
    solid and liquid, whereas at higher velocities, it was nearly independent
    of these variables. The interfacial area A_6 was reported to be nearly

constant at a value of 10 cm^2/cm^3 for gas velocities corresponding to

a constant volumetric absorption coefficient. The gas velocity above

which kgA_6 is nearly constant is related by analytical and graphical

correlations to the amount of solids in the column and to the critical

gas velocity corresponding to complete suspension of the solids. (7)

12) Experiments with bubble columns containing suspended solid particles of

varying diameters have demonstrated an increase in heat transfer

coefficient with increasing particle size. (2)

One of the most important aspects of gas flowing through a tubular reactor is

its availability at the reaction site. The transfer of gas to that site, be

it in a homogeneous liquid phase transition state or a heterogeneous surface-

activated state, is ultimately related to the interfacial area between the

liquid and gaseous phases. In a two-phase gas/liquid system, the interfacial

area is related to the volume fraction actually occupied by gas. This

fraction, which is typically referred to as the gas holdup or void fraction,

is extremely important to optimum reactor design.

Numerous investigators have studied gas holdup with many different systems,

covering a wide range of liquid viscosity (0.58-152.0 cP), surface tension

(22.3-76.0 dyn/cm), and density (0.79-1.70 g/cm^3). Several correlations have

been developed to predict the gas void fraction in two-phase flow systems. In

general, the gas void fraction is expressed in terms of gas superficial

velocity, liquid surface tension, viscosity, and density. However, none of

these correlations have been proven applicable in the presence of the solid

particles, or tested to account for the gas entrance effects.

Another significant aspect of gas/liquid flow is the dispersion (back-mixing) of the fluid phase. A small amount of gas flow can trigger a high degree of dispersion in the liquid phase. In general, back-mixing in the liquid phase depends on column diameter, gas velocity, and the nature of the gas distributor plate. Most investigators agree that the axial dispersion coefficient is independent of liquid flow rate and liquid phase properties (surface tension, viscosity, and density). Many correlations have been developed to predict the axial dispersion coefficient in gas/liquid flow reactors. At low gas velocities (0.02 fps), the agreement among these published correlations is poor. However, at high gas velocities (0.33 fps), these correlations give better agreement. Because of the inconsistencies in these published correlations, guidelines are unavailable for predicting the dispersion coefficient, which is an essential value for designing a reactor.

Air Products and Chemicals, Inc. conducted a detailed study in support of the dissolver design for the SRC-I Demonstration Plant. This more than 2-year long study of the three-phase flow hydrodynamics was carried out in a systematic fashion using cold-flow modelling techniques. Almost all the tests were performed at room temperature and pressure, and the visual observation of the test section played a significant role in the data analysis and provided a valuable explanation of observed deviations.

The cold-flow modelling studies were carried out in 2-in. and 5-in. internal diameter (i.d.) Plexiglas columns. Various combinations of gas, liquids, and solids were used to simulate the process fluids in the actual dissolver as

much as was practical. In all cases, experiments were initiated by performing
two-phase (gas/liquid) flow tests to ensure that the experimental techniques
were adequate to reproduce existing data.

The cold-flow dissolver modelling was begun in a 2-in.-diam Plexiglas column
by simulating the operation of a Coal Process Development Unit (CPDU)
reactor. The CPDU is a laboratory scale SRC pilot plant at Air Products'
facilities in Trexlertown, PA. Another objective of this study was to develop
a reactor model that could be used to predict axial dispersion coefficients
and solids concentration profiles in the reactor. This model was continuously
modified based on the latest available data from a larger, 5-in.-diam
Plexiglas column. The gas holdup, solids buildup, and particle size
distribution in the column were also studied in detail.

This report summarizes the work performed and pertinent results obtained from
the cold-flow dissolver modelling studies. It also briefly describes
subsequent tasks and studies undertaken to support the SRC-I Demonstration
Plant dissolver design.