APPENDIX TO SECTION 5 - Derivation of Gas Holdup Determination Methods

There are three common methods for determining gas holdup in a two- or three-phase system. Method 1, used reliably in all of the runs, entails measuring an expanded three-phase and settled slurry height. Gas holdup is then determined from the following equation:

$$\varepsilon_G = 1 - H_s/H_e \tag{A.5.1}$$

where

ε_G = gas holdup
H_s = settled slurry height
H_e = expanded slurry height

Method 2 uses manometer tubes filled with the liquid phase. Referring to Figure 5.2.1, a force balance on each manometer tube yields:

$$\rho_L\ (h''-D) = \rho_{3\phi}\ (H_e - D) \tag{A.5.2}$$

$$\rho_L\ (h' - A - D) = \rho_{3\phi}\ (H_e - A - D) \tag{A.5.3}$$

where $\rho_{3\phi}$ = three-phase density.

Dividing Equation A.5.3 by A.5.2 gives:

$$\frac{H_e - A - D}{H_e - D} = \frac{h' - A - D}{h'' - D} \tag{A.5.4}$$

Solving for H_e,

$$H_e = \frac{h''\ (D + A) - h'D}{h'' - h' + A} \tag{A.5.5}$$

Inserting into the average gas holdup Equation A.5.1 gives the final equation

$$\varepsilon_G = 1 - \frac{H_s(h'' - h' + A)}{h''(D + A) - h'D} \tag{A.5.6}$$

The manometer tube method was investigated for the following reasons. Firstly, gas holdup profiles up the column can be obtained by using multiple manometer tubes instead of just an average gas holdup. Secondly, in Method 2 the measurement of expanded height, which is a major source of inaccuracy in Method 1, is not required.

However, Method 2 assumes that the three-phase density, $\rho_{3\phi}$, remains constant along the column height. This is only true of two-phase runs and three-phase runs using very small size (<10μm) solids. Method 3 relaxes this assumption at the cost of requiring accurate knowledge of the liquid and solid densities and of the solid weight fraction in the slurry.

In Method 3, referring to Figure 5.2.2, a manometer filled with a liquid of known density, $\rho_{3\phi}$, is connected to two parts on the column, a distance "A" apart. A force balance yields:

$$A\rho_{3\phi} = H_M\rho_{Hg} \tag{A.5.7}$$

where H_M = vertical distance between the two liquid meniscuses on the manometer. The column three-phase density can be expanded to

$$\rho_{3\phi} = \Sigma\, \varepsilon_i\rho_i = \varepsilon_s\rho_s + \varepsilon_L\rho_L + \varepsilon_G\rho_G \tag{A.5.8}$$

By definition

$$\Sigma\, \varepsilon_i = 1 = \varepsilon_S + \varepsilon_L + \varepsilon_G \tag{A.5.9}$$

Expressing ε_L and ε_S in terms of measurable quantities

$$\varepsilon_L = (1-\varepsilon_G)/(1 + w_S\rho_L/w_L\rho_S) \qquad (A.5.10)$$

$$\varepsilon_S = (1-\varepsilon_G)/(1 + w_L\rho_S/w_S\rho_L) \qquad (A.5.11)$$

Combining Equations A.5.7 through A.5.11 and noting that $\rho_G << \rho_L, \rho_S$, and $w_S + w_L = 1$ yields

$$\varepsilon_G = 1 - \frac{H_{Hg}\rho_{Hg}}{A}\left(\frac{w_S}{\rho_S} + \frac{(1-w_S)}{\rho_L}\right) \qquad (A.5.12)$$

Method 3 is applicable to three-phase runs where the column density is not constant. It does, however, require that the average solid weight fraction be known. The solid concentration profile taken at four locations was generally a straight line when plotted as its logarithm with respect to height.

A least squares fit of the natural log of the solid weight fraction vs. height yielded the following reaction:

$$\ln w_S = \ln w_{S_0} - m\,h \qquad (A.5.13)$$

where

w_{S_0} = the weight fraction measured at the column bottom assuming no settling

m = slope = change of logarithm of the weight fraction with respect to height

h = height

When solid settling was apparent, ~>60 wt%, the bottom value was not used. The average weight fraction between parts i and i + 1 was then given by

$$w_S = (w_{Si} + w_{Si+1})/2 = w_{So} (\exp -mh_{i+1} + \exp -mh_i)/2 \qquad \text{(A.5.14)}$$

For the column average gas holdup, $h_{i+1} = H_E$, the expanded bed height and $h_i = 0$ in Equation A.5.14. For intermediate gas holdup, h_{i+1} and h_i are the two column heights where the manometer tube is inserted.

APPENDIX TO SECTION 9 - Derivation of the Solid Concentration Profile

A differential application of the one-dimensional continuity equation gives

$$\frac{\partial C}{\partial t} = \frac{\partial n_S}{\partial Z} \tag{A.9.1}$$

where n_S = mass flux relative to stationary coordinates in a gravitational field. For the continuous mode,

$$N_S = \frac{C(n_S + n_L)}{\rho} - Cu_{ST} - E_{ZS} \frac{\partial C}{\partial Z} \tag{A.9.2}$$

The first term on the right-hand side accounts for the bulk slurry flow. The Cu_{ST} term accounts for the gravitational force on the particles and the final term accounts for the diffusion of particles. All velocities have a positive value.

It is assumed that

$$j_L \approx U_{SL} = \frac{n_S + n_L}{\rho} \tag{A.9.3}$$

At steady state

$$\frac{\partial C}{\partial t} = 0 \tag{A.9.4}$$

Combining Equations A.9.1 to A.9.4 gives

$$(j_L - u_{ST})\frac{dC}{dZ} = E_{ZS} \frac{d^2C}{dZ^2} \tag{A.9.5}$$

The general solution to Equation A.9.5 is

$$C = A + B \exp \; (j_L - u_{ST})\frac{Z}{E_{ZS}} \tag{A.9.6}$$

Two boundary conditions are required. At Z = 0 (bottom)

$$j_L C_H = (j_L - u_{ST})C - E_{ZS}\,\frac{dC}{dZ}_O \tag{A.9.7}$$

$$= (j_L - u_{ST})(A + B) - E_{ZS}\,\frac{dC}{dZ}_O$$

At Z = L

$$V_L C_H = (j_L - u_{ST})C_H - E_{ZS}\,\frac{dC}{dZ}_L \tag{A.9.8}$$

Differentiating Equation A.9.6 with respect to height, Z, gives:

$$E_{ZS}\,\frac{dC}{dZ} = B\,(j_L - u_{ST}) \exp \; (j_L - u_{ST})Z/E_{ZS} \tag{A.9.9}$$

Equating each boundary condition to Equation A.9.9

$$A = j_L C_H/(j_L - u_{ST}) \tag{A.9.10}$$

$$B = -C_H\,u_{ST}/(j_L - u_{ST}) \;\; \exp \; -(j_L - u_{ST})\,L/E_{ZS} \tag{A.9.11}$$

Inserting Equations A.9.10 and A.9.11 into A.9.6 gives the steady state solution Equation A.9.5

$$C = C_H/(j_L - u_{ST}) \;\; j_L - u_{ST}\exp\,(j_L - u_{ST})(Z - L)/E_{ZS} \tag{A.9.12}$$

As $u_{ST} \to 0$, the solid concentration is uniformly C_H.

In batch operation when $j_L = 0$, Equation A.9.5 becomes

$$0 = u_{ST}\,\frac{dC}{dZ} + E_{ZS}\,\frac{d^2C}{dZ^2} \tag{A.9.13}$$

giving a steady state solution

$$C = C_1 \exp \frac{-u_{ST}}{E_{ZS}} (Z - Z_1) \qquad \text{(A.9.14)}$$

APPENDIX TO SECTION 11 - OPTIMUM GAS HOLDUP DERIVATION

In a bubble column, the rate of consumption of hydrogen at the catalyst surface,

$$r = r_{H_2} = r_{CO + H_2}/(1 + U)$$

$$= K_O\, w(1 - \varepsilon_G)C/(1 + U), \qquad \text{(A.11.1)}$$

is equal to the rate at which hydrogen is transported across the gas-liquid interface.

$$r = K_L a\,(C^* - C) \qquad \text{(A.11.2)}$$

Interfacial area, a, is related to gas holdup and bubble size by

$$a = 6\,\varepsilon_G / d_{SB} \qquad \text{(A.11.3)}$$

Solving Equation A.11.2 for C and substituting it and Equation A.11.3 into Equation A.11.1 eliminates the hydrogen concentration in the liquid phase, C:

$$\text{(A.11.4)}$$

$$r = (6K_L C^*/d_B)\ \varepsilon_G/(1 + (6K_L(1 + U)/K_O w d_B)\varepsilon_G/(1 - \varepsilon_G)$$

To find the gas holdup, ε, that allows for the maximum consumption rate of hydrogen, the derivative of Equation A.11.4 is taken with respect to ε_G and the resulting expression is set equal to zero.

$$dr/d\varepsilon_G = 0 = (1 - 6K_L(1 + U)/K_O W d_B)\varepsilon_G{}^2\text{opt}$$

$$- 2\,\varepsilon_G\,\text{opt} + 1 \qquad \text{(A.11.5)}$$

Solving Equation A.11.5 quadratically yields one physically realistic solution:

$$\varepsilon_G \text{ opt} = 1/(1 + (6K_L(1 + U)/K_0 w d_B)^{1/2}) \qquad \text{(A.11.6)}$$