

3. Nonagglomerating catalyst particles $\leq 60 \mu\text{m}$ should be suitable for bubble column batch operation, as no unsuspended solids were observed for either solid system studied.
4. Silicon oxide concentration profiles were unaffected by the presence of plain tube heat transfer internals.

10.0 Heat Transfer

Heat transfer coefficients were measured because this affects the amount of cooling surface required. At Rheinpreussen, the heat transfer surface accounted for about 6% of the reactor volume (Ruckenstein & Smigelschi; 1965; Burkel, 1972). Thus, the effect of more precise data on overall production rates should be small.

The heat of reaction may be removed from the slurry system by (a) cooling tubes immersed in the slurry (Kolbel & Ackerman, 1951; Kolbel & Ralek, 1977; Fair, 1967b), (b) circulating the slurry through an external cooling system (Fair, 1967b; Hall, 1952), or (c) partially condensing overhead vapor and returning cold condensate (Kolbel & Ackerman, 1951; Fair, 1967b; Hall, 1952; Schlesinger et al., 1951). It is believed that heat transfer internal to the reactor will be preferred in commercial-size units. Internal heat transfer precludes the need for (a) an external heat exchanger, (b) a large and expensive slurry pump, and (c) large piping for the slurry circuit. Also, the operational problems of erosion and plugging caused by slurry circulation are minimized. Lastly, the agitation produced by the gas phase within the reactor produces heat transfer coefficients that are higher than those produced by gas or slurry alone. Thus, the total heat transfer surface required using internal heat transfer is equal to or less than that required with an external exchanger.

Although immersed cooling tubes should be the most thermally efficient means for recovering the heat of reaction, their use is the least understood (Fair, 1967b). This study was undertaken, in part, to

better quantify internal heat transfer in slurry bubble columns over a broad range of FT operating conditions.

10.1 Theory

Previous investigators have examined heat transfer in gas-liquid dispersions (Shaykhutdinov, 1971), two-phase bubble columns (Fair, 1967b; Kolbel, 1958a,b; Ruckenstein, 1965; Burkel, 1972; Mansmann, 1977; Kolbel, 1968) and three-phase bubble columns (Kast, 1962; Kolbel, 1960; Deckwer, 1980). Kolbel (1958a), who developed the slurry phase FT process at Rheinpreussen, originally correlated his data to obtain:

$$Nu = 43.7 Re_H^{0.22} \quad (10.1.1)$$

This empirical relation, which uses the heater length as the characteristic dimension, cannot be extrapolated to other systems. Ruckenstein and Smigelschi (1965) correlated their results for a two-phase bubble column system, using liquid (L) physical properties to obtain:

$$h = 1.59 k_L (g c_p^2 C_p / \mu k)_L^{1/3} \quad (10.1.2)$$

However, the relation is limited by using gas holdup, ϵ , instead of gas velocity. It is the agitation caused by the gas velocity, not the gas holdup, that determines the degree of heat transfer.

Kast (1962) realized that the bubble motion, breaking up the boundary layer at the wall surface, provided the heat transfer mechanism and proposed the following functional relationship:

$$St = F(Re Fr^2 Pr^2)^{-0.22} \quad (10.1.3)$$

Deckwer (1980) applied Higbie's surface renewal (1935) and Kolmogoroff's isotropic turbulence mechanism (Tennekes & Lumley, 1972)

to three-phase bubble column heat transfer. From this different approach, Deckwer obtained a relationship similar to Kast's:

$$St = f(Re \ Fr^2 \ Pr^2)^{-0.25} \quad (10.1.4)$$

The present work was undertaken to extend the understanding of slurry phase FT operation. Because internal cooling tubes are expected to be used commercially, simulated cooling tubes were used. A wider range of solid loadings were studied in anticipation of future FT operations, which would seek to maximize space-time yields. Also, the solid oxides which were used were more representative of possible catalyst supports. Larger scale equipment was used to evaluate its effect on heat transfer. Lastly, to maximize column production, higher gas velocities and viscosities were studied to assess their effects on heat transfer.

10.2 Experimental

Seven 5.72 cm OD simulated cooling tubes were inserted parallel to the 30.5 cm ID column and extending the column length (see Figure 10.2.1). In three of the tubes, electrical resistance heaters (see Figure 10.2.2) were placed.

Heater A was located inside the center heat transfer internal about 3 m above the distributor plate, or about 2/3 of the distance between the distributor and the top of the bubble column. Two temperatures were measured at the heater midpoint, 150 degrees from each other. Heaters B and C, 3 and 1.5 m above the distributor respectively, were located in two of the six outer tubes with the surface thermocouple oriented toward the column center.

Procedure

The heat transfer coefficient, h , was determined using the rate equation

$$h = Q / (sL(T_{SM} - T_{SL})) \quad (10.2.1)$$

The wattage applied across the electric heater was a quantitative measure of Q . The length, L , was the axial length of the coil windings inside the heater, about 1 cm shorter than the heater length. The temperature difference was taken from thermocouples at the heater midpoint, just below the heater surface, and in the bulk liquid. About a half hour was needed to obtain a constant temperature difference. The three heaters were measured sequentially, in random order.

If a cylindrical heater is long enough, the heater midpoint temperature at the tube surface, T_{SM} , should be unaffected by heat leak from the heater top and bottom. However, if the heater is too short a significant fraction of the total heat leaves at the heater ends, leading to a lower value of T_{SM} and, by Equation 10.2.1, a higher apparent heat transfer coefficient.

The temperature profile of a cylindrical heater with both ends of the cylinder at the bulk slurry temperature has been derived by Kalb (1982):

$$T_{SM} - T_{SL} = \frac{Q}{hA} \left[1 - \frac{\exp(-(1-N)b^{1/2}) + \exp((-N)b^{1/2})}{1 + \exp(-b^{1/2})} \right] \quad (10.2.2)$$

where

$$b = \frac{hL^2}{kD}$$

N = dimensionless heater length, x/L

The factor $Q/(hA)$ in Equation 10.2.2 has units of temperature and is equal to the temperature difference, $T_{SM} - T_{SL}$, which would exist if no end heat losses occurred. Therefore, the bracketed expression in Equation 10.2.2 must be close to unity for the heat error to be ignored. At the heater midpoint N equals $1/2$ for the 5.72 cm OD carbon steel cooling tube casing. If the heat transfer coefficient were $30 \text{ W/m}^2\text{K}$, the heater length would need to be 29 cm for a 0.01% error due to heat leak. As heat transfer coefficients increased, the required length decreased. Since the heaters used were 48 cm long, the end heat losses were ignored.

10.3 Results and Discussion

Some trends can be determined from a plot (see Figure 10.3.1) of every fifth data point of the heat transfer coefficient vs. gas velocity for each liquid/solid/particle size combination. Larger heat transfer coefficients, h , were measured in water-based slurries. This can be attributed to larger values of density (ρ), thermal conductivity (k), and heat capacity (C_p) for water. Also, the value of h is seen to increase beyond a gas velocity of 10 cm/s. This is contrary to the behavior observed by Deckwer (1980), who observed no further increase in h above 10 cm/s. A difference between the silicon oxide and iron oxide slurries can also be reported. Again, the higher k of silicon oxide may be responsible.

The data was statistically analyzed for each solid oxide to yield the following correlations:

$$St_{Fe} = (0.063 \pm 0.009)(ReFr^2 Pr^{1.60 \pm 0.12})^{-0.25 \pm 0.01} R^2 = 0.96 \quad (10.3.1)$$

$$St_{Si} = (0.045 \pm 0.007)(ReFr^2 Pr^{0.93 \pm 0.11})^{-0.28 \pm 0.02} R^2 = 0.89 \quad (10.3.2)$$

The accuracy in Equations 10.3.1 and 10.3.2 is expressed as \pm one standard deviation. A comparison of these heat transfer results to the Deckwer (1980) relation, Equation 10.1.4, is shown in Figure 10.3.2 for a typical solid loading of 20 wt% silicon oxide or iron oxide in water or isoparaffin. Good agreement is obtained for the iron oxide systems, being about 10% lower for water and agreeing identically for isoparaffin. The silicon oxide heat transfer predictions were 32 to 56% higher in water and 1 to 19% higher in isoparaffin than Deckwer's.

To better examine the reasons for this disagreement, it is useful to recast Equations 10.3.1 and 10.3.2 in dimensional form using slurry, not liquid properties:

$$h_{Fe} = 0.063 V_G^{0.25} \rho^{0.75} C_p^{0.59} \mu^{-0.15} g^{0.25} k^{0.41} \quad (10.3.3)$$

$$h_{Si} = 0.045 V_G^{0.16} \rho^{0.72} C_p^{0.74} \mu^{-0.02} g^{0.28} k^{0.26} \quad (10.3.4)$$

Comparing these exponents to those values that would be predicted using surface renewal and isotropic turbulence,

$$h \propto V_G^{0.25} \rho^{0.75} C_p^{0.50} \mu^{-0.25} g^{0.25} k^{0.50} \quad (10.3.5)$$

agreement is generally satisfactory. Disagreement is greatest between the calculated and theoretical viscosity exponential terms. It is possible that the high shear rates assumed to be taking place at the heater surface predicted by Nishikawa (1977) may be lower than predicted. For example, at 15 cm/s the shear ($\dot{\gamma}$) is 387 s^{-1} from Equation 4.0.7 and the viscosity (μ) is 8 cp from Equation 4.0.4. Had the shear rate been 92 s^{-1} , then the viscosity would be 3.48 cp and Equation 10.3.3 would have been $h \propto \mu^{-0.25}$ instead of $h \propto \mu^{-0.15}$.

Another possible reason for disagreement is that the silicon oxide thermal conductivity, k , was not accounted for correctly. Silicon oxide, or quartz, has an anisotropic thermal conductivity depending on direction in its lattice structure. Also, its thermal conductivity is over ten times that of water and over fifty times greater than that of isoparaffin. Equation 4.0.2 does not take large differences in k between liquid and solid properly into account. For example, by Equation 4.0.2 a 50 vol% mixture of isoparaffin ($k=0.13 \text{ W/m}^\circ\text{K}$) and silicon oxide ($k = 6.95 \text{ W/m}^\circ\text{K}$) would have $k_{SL}=0.48 \text{ W/m}^\circ\text{K}$. A secondary reason may be that the literature value of k may not agree with the actual value of k for the silicon oxide used in this study.

Finally, some of the previous investigators may not have properly accounted for end heat loss, as discussed in the experimental section, or surface area. It is incorrect to average the temperature across the heater length to obtain the surface temperature. Such an approach will lead to higher calculated h values. It is also incorrect to use the heater housing length or heater ends to calculate the surface area. Rather, the actual heat source length should be used.

11.0 Engineering Evaluation

The gas holdup, bubble diameter, mass transfer, solid concentration, liquid dispersion and heat transfer relationships developed in this work have been incorporated into a bubble column computer model proposed by Deckwer (1982) in order to predict the effect of these hydrodynamic parameters on reactor space-time yield.

The substitution of the cold flow correlations for those in the original Deckwer model caused some changes in predicted column performance. While the effect at low gas velocities was negligible, at 9 cm/sec. the space-time yield doubled (see Table 11.0.1 vs. Table 11.0.2). The lower predicted gas holdup allowed for more catalyst per unit volume. The Rheinpreussen base case was not

simulated with the new gas holdup correlation because it's conversion and space time yield were measured quantities (Kolbel & Ralek, 1980), while the reaction kinetics were calculated assuming the Deckwer (1980) gas holdup correlation. Thus, any change to the gas holdup correlation would necessitate a change of the inferred reaction kinetics in order to yield the same hydrogen conversion and space-time yield.

The lower gas holdup predictions had a desirable effect on reactor space-time yield because the mass transfer resistance was not predicted to be limiting.

Compared to Rheinpreussen, overall space-time yields, STY, or production rates per unit volume using these catalysts was lower, although catalyst improvements are anticipated in future work. Comparing fuel fraction yields (see Figure 11.0.1), it is seen that catalysts A and B produce several times more diesel fuel than was produced at Rheinpreussen. If shift activity can be added to catalyst B as anticipated, the diesel fraction will be almost five times that obtained at Rheinpreussen, reducing the size of downstream refining equipment.

The Rheinpreussen operating conditions are not necessarily optimized for running the new APCI catalysts.

In bubble column operation there exists an optimum gas holdup which will maximize column space-time yield. Gas holdups higher than this optimum will be reaction rate-limited while those lower than this will be mass transfer-limited. The optimum gas holdup will be affected by the bubble size (d_{SB}), intrinsic kinetic rate (K_O), catalyst weight loading (w), the rate of mass transfer across the gas-liquid interface (K_L), and the usage ratio (U).

The optimum gas holdup is given by the expression

$$\alpha_{opt} = 1 / (1 + (6K_L(1 + U) / K_O w d_{SB})^{1/2}) \quad (11.0.1)$$

In Equation 11.0.1 it is seen that increasing the intrinsic kinetic rate constant, the catalyst loading or the Sauter mean bubble diameter all result in increasing the optimum gas holdup value. Conversely, as the rate of mass transfer (K_L) increases, the optimum gas holdup decreases.

It is interesting to note that while C^* , the hydrogen solubility, directly increases the space-time yield, it does not affect the optimum gas holdup value.

Using Equation 11.0.1 the optimum gas holdup, resulting gas velocity, and relative space-time yields for a variety of values of K_0 , W , and d_{SB} are given in Table 11.0.3. The relative effect on space-time yield can be determined from the following equation (see Appendix A.11 for derivation):

$$r = (6K_L C^* / d_{SB}) \propto 1 / (1 + (6K_L (1 + U) / K_0 W d_{SB}) \propto 1 / (1 - \alpha) \quad (11.0.2)$$

Going from a bubble size of 0.07 cm (case 1) as measured by Deckwer (1980) to 0.3 cm (case 4) results in roughly doubling of the optimum gas holdup value.

Fortunately, running both conditions at their optimum, the space-time yield is only 13% less at the larger bubble size. Increasing the weight loading by a factor of 3 (case 1 vs. 2) or (case 4 vs. 5) results in roughly a 60% increase in optimum gas holdup value, a 6-fold increase in the optimum gas velocity to achieve that gas holdup, and a 2.5-fold increase in space-time yield. Higher gas velocities lead to a more economical operation by requiring fewer tall bubble columns to do the same job.

Thus, when operating at optimum hydrodynamic conditions, differences in parameters that adversely affect the mass transfer resistances can be compensated to minimize their impact. Also, almost full advantage can be taken of parameters that positively affect the chemical reaction rate.

11.1 Staged Reaction Systems

Bubble column space-time yield can be improved by staging the gas phase as shown in Figure 11.1.1. This staging can be modeled on the computer in either of two ways. In the first method, a number of idealized continuous stirred tank reactors (CSTRs) are simulated with almost complete recycle of the liquid and catalyst phase and no recycle of gas phase. This would be comparable to the recirculating cell model used in the literature. In the second method, the extent of backmixing of the gas phase is modeled by applying an axial dispersion model (Levenspiel, 1972). In this model, a low dispersion coefficient refers to many CSTRs in series, while a high dispersion coefficient refers to very few CSTRs in series.

These two methods for simulating staged reactors have been related by Carberry (1976). He showed that

$$N = V_G L / 2D \quad (11.1.1)$$

Thus, either way of expressing staged reaction systems can be used. Fortunately, the Deckwer computer model already incorporates the axial dispersion model for the gas, liquid, and catalyst phases. To simulate five stages for the gas phase, it is necessary only to solve for D in Equation 11.1.1 and input that value for the gas phase dispersion coefficient. For the liquid and catalyst phases, it is assumed desirable to keep them as well backmixed as before.

The results of simulating the Rheinpreussen-size column as several stages having the same total volume as the original single-stage column are shown in Table 11.1.1 and Figure 11.1.2.

Three systems were simulated consisting of 1, 5, and 20 CSTRs in series. It is seen that five stages result in roughly a 5% increase

in space-time yield. No further space-time yield improvement was obtained with 20 stages.

While Table 11.1.1 does indicate the improvement that could be expected by staging the gas phase, there are several factors that the simulation did not take into account. At each stage, the gas phase would be redistributed into that stage. Distributor hole size would thus play a major factor in highly staged systems. It would be possible to reduce bubble size considerably, greatly reducing the mass transfer resistance. This is simulated in column 4 of Table 2, where it is assumed that small-size distributors result in a Sauter diameter of 0.07 cm. Another opportunity with staged systems is to introduce additional syngas at each stage to overcome the effect of gas contraction as the syngas is consumed in the reaction. This further could increase space-time yield by roughly 25%, although at the expense of excess piping and slightly lower conversion.

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13.0 Nomenclature

A	= constant, g cm^{-3}
A	= distance between manometer ports, cm
A	= heater surface area, m^2
a	= constant
a	= interfacial area, cm^2 surface area cm^{-3} expanded slurry
B	= constant, g cm^{-3}
Bo	= $\text{gD}^2\rho_L/\sigma$ = Bond number
C	= concentration, g cm^{-3}
C	= concentration of hydrogen gas, $\text{mol H}_2 \text{ cm}^{-3}$ slurry

C^*	= Equilibrium slurry concentration of hydrogen gas, mol H_2 cm^{-3} slurry
C_N	= carbon number
C_p	= specific heat, $J\ g^{-1}\cdot K^{-1}$
CZ	= Probability of observing a bubble chord length less than λ = cumulative gamma distribution function
D	= column diameter, cm
D	= dispersion coefficient, $cm^2\ s^{-1}$
D_L	= Diffusivity of diffusion component in liquid, $cm^2\ s^{-1}$
DX	= distance between bubble tops, cm
d	= diameter, cm
d_{SB}	= Sauter mean bubble diameter
E_z	= axial dispersion coefficient, $cm^2\ s^{-1}$
Fl	= $\rho_L^2 \sigma^3 / (\mu_L^4 (\Delta \rho) g)$ = Fluid number
Fr	= $V_G / (g d_S)^{1/2}$ = Froude number
Fr_p	= $V_G / g d_p$ = particle Froude number
f	= Fanning friction factor
Ga	= $g D^3 / \nu_L^2$ = Galileo number
g	= gravitational acceleration constant, $cm\ s^{-2}$
H	= liquid height in manometer tube, cm
HS	= hole size, cm
h	= heat transfer coefficient, $W\ m^{-2}\cdot K^{-1}$
j_{21}	= drift flux of component 2 relative to component 1, $cm\ s^{-1}$
j_T or j	= superficial velocity of liquid and gas, $cm\ s^{-1}$
j_i	= superficial velocity or flux of component i , $cm\ s^{-1}$
K_L	= mass transfer coefficient, $cm\ s^{-1}$
K_O	= Intrinsic reaction rate constant, $sec^{-1}\ wt\%^{-1}$
Ku	= $\rho_L / (\sigma (\rho_L - \rho_G) g)$ = Kutateladze number
k	= thermal conductivity, $W\ m^{-1}\cdot K^{-1}$
k_{SL}	= $k_L (2k_L + k_S - 2V_S (k_L - k_S)) / (2k_L + k_S + V_S (k_L - k_S))$ = slurry thermal conductivity, $Wm^{-1}\cdot K^{-1}$
L	= heater length, m
L	= bubble column effective height, cm
LT	= lag time, s
m	= constant

N	= molar consumption rate, mol s^{-1}
N	= x/L , dimensionless heater length
N	= number of stirred tanks in series
Nu	= $h(d_H/k)$ = Nusselt number
n	= mass flux relative to stationary coordinates in a gravitational field, $\text{gcm}^{-2}\text{s}^{-1}$
n,s	= cumulative gamma distribution parameters
Pe	= $V_G d_C/E_Z$ = Peclet number
Pr	= $(\mu c_p/k)_{SL}$ = Prandtl number
Q	= volumetric flowrate, $\text{cm}^3 \text{s}^{-1}$
Q	= power output of heater, W
R^2	= correlation coefficient
Re	= $d_s j_G \rho_{SL} / \mu_{SL}$ = Reynolds number
Re_{SL}	= $u_{ST} d_s \rho_{SL} / \mu_{SL}$ = particle Reynolds number based on slurry properties
Rep	= $u_{ST} d_s \rho_{SL} / \mu_L$ = particle Reynolds number based on liquid properties
$r_{CO + H_2}$	= rate of disappearance of $CO + H_2$, mol cm^{-3} slurry
r, r_{H_2}	= rate of disappearance of H_2 , mol cm^{-3} slurry s^{-1}
SF	= Strobe frequency, s^{-1}
Sc	= $\mu_L / (\rho_L D_L)$ = Schmidt number
St	= $h / \rho_{SL} c_{pSL} V_G$ = Stanton number
s	= heater perimeter, cm
T	= temperature, $^{\circ}\text{K}$
t	= time, s
U	= N_{CO}/N_{H_2} , usage ratio
U_{GL}	= relative gas to liquid velocity, cm s^{-1}
u	= actual rise velocity
u_{SG}	= relative solid to gas velocity, cm s^{-1}
u_{ST}	= single particle terminal settling velocity, cm s^{-1}
V	= velocity, cm s^{-1}
V	= volume, cm^3
V_G	= gas superficial velocity, cm s^{-1}
V_R	= volume of expanded slurry, cm^3 expanded slurry

v	= volume fraction
W	= weight fraction
W	= catalyst weight loading, wt%
We	= $d_B j_G^2 \rho_L / \sigma$ = Weber number based on gas bubble
We _s	= $d_S j_G^2 \rho_S / \sigma$ = Weber number based on solid particle
w	= weight fraction
X, Y	= variables of integration
x	= length along heater, m
y	= gas phase concentration of O ₂ , mol cm ⁻³
Z	= distance above distributor, cm

Greek Letters

α, ϵ	= gas holdup, volume fraction gas, vol fraction
λ	= bubble chord length, cm
μ	= viscosity, g cm ⁻¹ s ⁻¹
ρ	= density, g cm ⁻³
γ	= shear rate, s ⁻¹
σ	= surface tension, dyne cm ⁻¹ = g s ⁻²
Δ	= finite difference
ν	= μ/ρ = kinematic viscosity, cm ² s ⁻¹

Subscripts

A	= actual
B	= bubble
E	= at equilibrium
Fe	= iron oxide
G	= gas
H	= heater
H _g	= mercury
L	= liquid
M	= measured or manometer

P = solid particle
S = solid
SB = Sauter mean bubble
SL = slurry
SM = heater surface midpoint
Si = silicon oxide

14.0 References

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