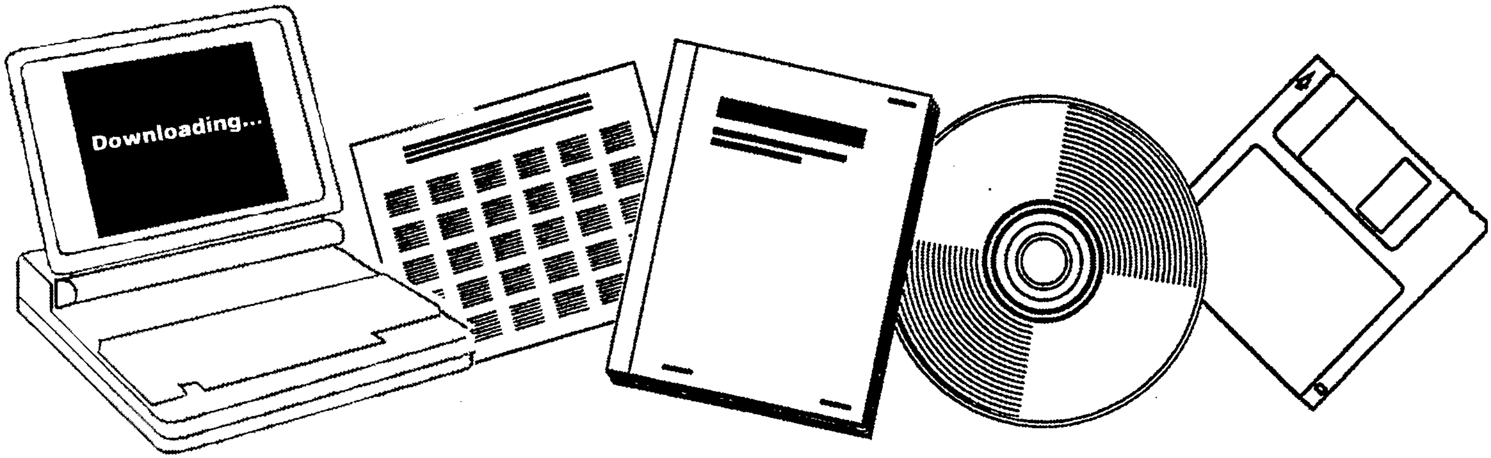
DE92008959

DESIGN OF SLURRY REACTOR FOR INDIRECT LIQUEFACTION APPLICATIONS. QUARTERLY TECHNICAL PROGRESS REPORT, JULY 1990--SEPTEMBER 1990

VIKING SYSTEMS INTERNATIONAL, INC.
PITTSBURGH, PA

1990



One Source. One Search. One Solution.

NTIS

Providing Permanent, Easy Access to U.S. Government Information

National Technical Information Service is the nation's largest repository and disseminator of government-initiated scientific, technical, engineering, and related business information. The NTIS collection includes almost 3,000,000 information products in a variety of formats: electronic download, online access, CD-ROM, magnetic tape, diskette, multimedia, microfiche and paper.

Search the NTIS Database from 1990 forward

NTIS has upgraded its bibliographic database system and has made all entries since 1990 searchable on **www.ntis.gov.** You now have access to information on more than 600,000 government research information products from this web site.

Link to Full Text Documents at Government Web Sites

Because many Government agencies have their most recent reports available on their own web site, we have added links directly to these reports. When available, you will see a link on the right side of the bibliographic screen.

Download Publications (1997 - Present)

NTIS can now provides the full text of reports as downloadable PDF files. This means that when an agency stops maintaining a report on the web, NTIS will offer a downloadable version. There is a nominal fee for each download for most publications.

For more information visit our website:

www.ntis.gov

U.S. DEPARTMENT OF COMMERCE
Technology Administration
National Technical Information Service
Springfield, VA 22161

DOE/PC/89870--T9

DE92 008959

DE92008959

QUARTERLY TECHNICAL PROGRESS REPORT

July 1990 - September 1990

DESIGN OF SLURRY REACTOR FOR INDIRECT LIQUEFACTION APPLICATIONS

by

Anand Prakash
Prakash G. Bendale

Viking Systems International
2070 William Pitt Way
Pittsburgh, PA 15238

Prepared for the
U.S. Department of Energy

Work Performed under Contract No. DE-AC22-89PC89870

Introduction

The objective of this project is to design a conceptual slurry reactor for two indirect liquefaction applications;production of methanol and production of hydrocarbon fuels via Fischer-Tropsch route. The work will be accomplished by the formulation of reactor models for both the processes and use computer simulation. Process data, kinetic and thermodynamic data, heat and mass transfer data and hydrodynamic data will be used in the mathematical models to describe the slurry reactor for each of the two processes. The cost of current vapor phase reactor systems will be compared with cost estimated for the slurry reactor systems. For the vapor phase systems, upstream and downstream processing equipments may have to be included during cost analysis for a meaningful cost comparison.

Finally, we will point out any inadequacies in the technical database currently available for a commercial design of the slurry reactor and identify research needs to improve upon the slurry reactor design. Assumptions used in the design will be documented.

Project Status

During this quarter, work was performed in the following areas:

- Mathematical formulation of reactor model with no external recirculation of slurry.
- Selection of gas holdup correlation, using experimental data from Air Products and Chemical's PDU operation.
- Selection of correlation for volumetric mass transfer coefficient.
- Development, testing and debugging of computer codes.

Reactor Model

The latest reactor configuration at Air Products and Chemicals does not include external recirculation of slurry. The model equations change slightly in absence of slurry recirculation.

The model assumptions are the same as detailed in previous reports.

Model Equations:

The following equations describe the reactor model with no external recirculation of slurry.

Gas Phase Mass Balance:

The gas phase mass balance equations do not change.

Liquid Phase Mass Balance:

The liquid phase mass balance for component i can be written as:

$$\frac{d^2x_i}{dz^2} + \frac{k_{L,i}a\,L^2}{D_L\,\epsilon_l}(y_i - x_i) + \Sigma\alpha_{i,k}\epsilon_l C_{cat} r_k (H_i L^2/D_L P) = 0$$

Boundary conditions

$$\frac{dx_i}{dz} = 0 \qquad \text{at } z = 0$$

$$\frac{dx_i}{dz} = 0 \qquad \text{at } z = 1$$

Solid Phase Mass Balance:

$$\frac{d^2w}{dz^2} + Pe_s' \frac{dw}{dz} = 0$$

with boundary conditions:

$$w = 0 \qquad z = o$$

$$wdz = 1$$

An analytical solution to the above equation is possible and is given below:

$$w = \frac{Pe_s' \exp(-Pe_s' z)}{[1 - \exp(-Pe_s')]}$$

Heat Balance

Due to near isothermal conditions of operation in the reactor, the heat balance equations need not be incorporated.

Selection of Gas Holdup Correlations:

The experimental gas holdup values for the PDU operation of Air Products and Chemicals were obtained using the available information from different reports and the data provided by APCL engineers. Figure 1 shows a plot of the experimental gas holdups as a function of gas velocity. The figure also shows a mean value line with deviations from mean. Most of the deviation from the mean can be attributed to changes in slurry concentration. The high experimental gas holdups (> 0.25) obtained in the heterogeneous regime (U_G > .1m/s) indicate that the system is noncoalescing. In a coalescing system, the gas holdup in the heterogeneous regime usually does not exceed 25%. In a noncoalescing system, the average bubble size is reduced due to reduction in bubble coalescence rate. A smaller average bubble size in the dispersion results in higher gas holdup.

Various literature correlations for the estimation of gas holdup (see Appendix-A) were tested against the experimental gas holdup data. Table 1 gives average relative error, average absolute relative error and minimum and maximum errors for different correlations. it can be seen that three correlations (Hughmark (1967);Hikita et al. (1980);Sauer and Hempel (1987)) give a reasonable prediction of gas holdup for the given system with average absolute error of less than 15%. In the reactor model the gas holdups are estimated in a subroutine called 'HOLDUP'. All the three correlations have been included in the subroutine with proper comments. For a given run, any of these correlations can be selected for testing.

Correlation of Sauer and Hempel

Correlation based on data obtained in a slurry bubble column. It includes a slurry viscosity term to

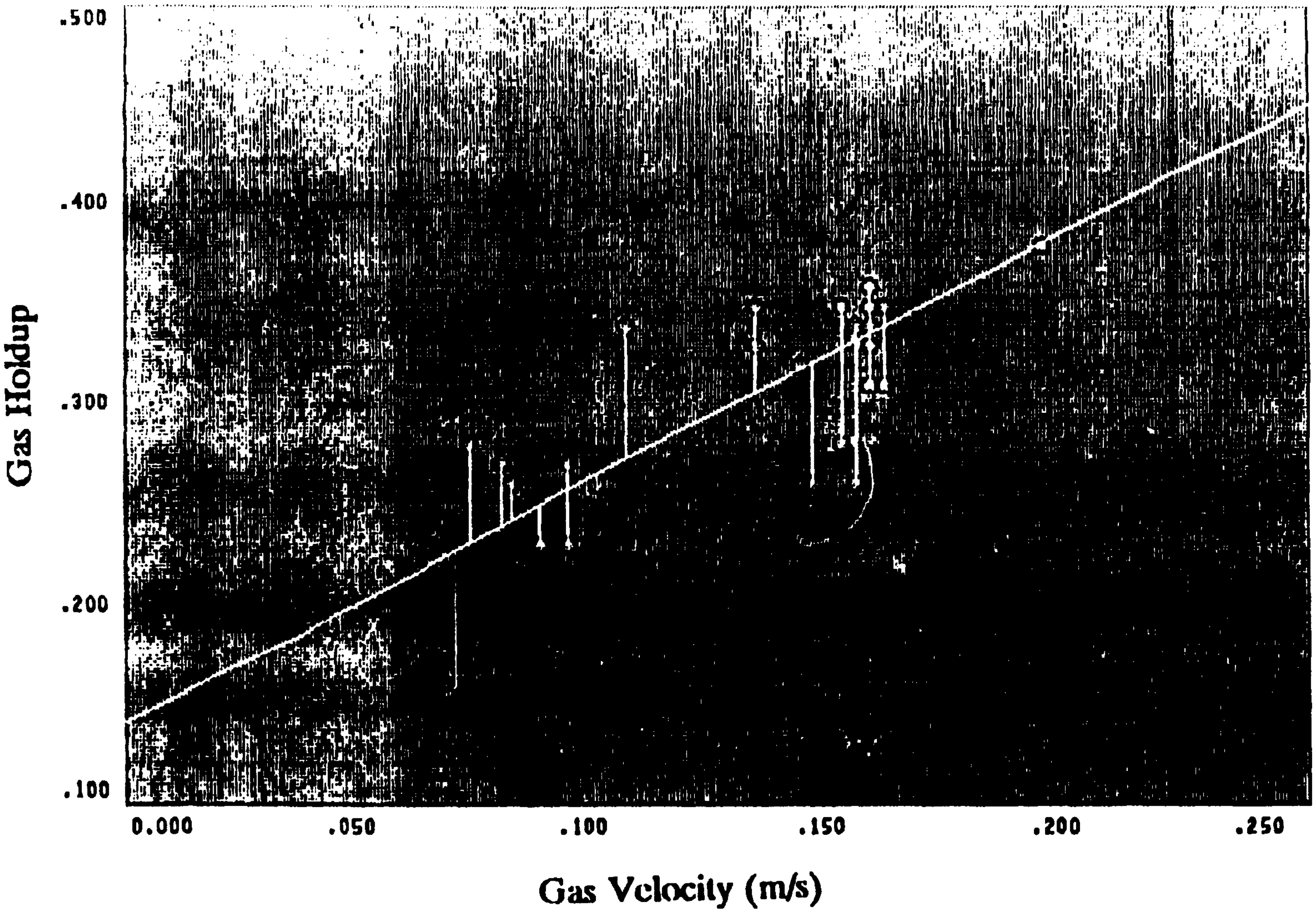


Figure 1. Gas Holdup (Exptl.) vs Gas velocity

account for the effects of solid particles on gas holdups.

Correlation of Hughmark and Hikita et al.

These correlations are based on the data obtained in solid-free bubble columns. These correlations may, therefore, have only limited application in slurry bubble columns. Application of these correlations over a wide range of operating conditions needs to be tested further. Smith and Ruether (1985) observed that the correlation of Hughmark (1967) could predict the gas holdups in their slurry bubble column with an average error of about 7%-when the liquid density in the original correlation was replaced by slurry density. The modified correlation of Hughmark should therefore provide reasonable estimates of gas holdups over a wide range of operating conditions in slurry bubble columns.

The modified correlation of Hughmark was therefore selected for the estimation of gas holdups in the slurry reactor model. The correlations of Sauer and Hempel and Hikita et al. will be used for a few cases for comparison purposes only.

Table 1. Comparison of various literature correlations

Correlation by	Avg. Abs. RelativerError (%)	Avg. Relative Erros (Min & Max) (%)
Koide et al. (1984)	34.4	34.4 (-47.0, -9.4)
Sada et al. (1986)	21.0	14.0 (-35.0,55.7)
Sauer and Hempel (1987)	12.0	-3.6 (-22.5,34.0)
Hughmark (1967)	13.4	-9.0 (-27.0,21.4)
Akita and Yoshida (1973)	31.6	-31.6 (-45.6,-7.2)
Deckwer et al. (1980)	190.0	190.0 (39.0,317.0)
Hikita et al. (1980)	10.4	0.5 (-22.3,32.6)

Selection of Correlations for Volumetric Mass Transfer Coefficient:

The correlations of Akita and Yoshida (1973) and Nguyen-tien et al. (1985) were selected for the estimation of volumetric mass transfer coefficient. These correlations were subsequently modified slightly in view of available information.

Akita and Yoshida (1973)

$$(k_L a D_c^2/D_i) = 0.6\, e_G^{1.1} (\mu_L/\rho_L D_i)^{0.5} (g D_c^2 \rho_L/\sigma)^{0.62} (g D_c^3 \rho_L^2/\mu_L^2)^{0.31}$$

This correlation includes a gas holdup term which can account for changes in volumetric mass transfer coefficient due to changes in gas holdup. The gas holdup can change with gas velocity, solid particles density,size and concentration and with changes in coalescing behavior of the liquid. This correlation,however, has a reactor diameter term which can result in overprediction of volumetric mass transfer coefficient in large diameter reactors. For a given gas and liquid system, it gives

$$k_L a \propto D_c^{0.17} e_G^{1.1}$$

From the available literature data, it can,however, be observed that the effect of reactor diameter on volumetric mass transfer coefficient becomes insignificant for reactor diameter larger than 0.15 m. The correlation of Akita and Yoshida will thus overpredict the volumetric mass transfer coefficient for reactors much larger than 0.15m. Therefore, the value of reactor diameter in this correlation was fixed at 0.15m for larger reactors.

Nguyen-tien et al. (1985)

$$k_L a = 0.39\,(1 - \epsilon_{s,sus}/0.58)U_G^{0.67}$$

This correlation is based on data obtained for oxygen mass transfer in aqueous solutions. For other systems $k_L a$ can be corrected as below:

$$(k_L a)_i = (k_L a)_o (D_i/D_{o,w})^{2/3}\,(\mu_w/\mu_L)^{0.3}$$

This correlation has been found to be applicable over a wide range of particle size and concentration in three-phase contactors. It is, however, based on data obtained in coalescing liquids. It is, therefore, expected to provide only conservative estimates of volumetric mass transfer coefficient in noncoalescing systems. In order to improve its applicability for noncoalescing systems, this correlation was modified slightly - the solids fraction term in the original equation was replaced to represent solids fraction in three-phase system.

The modified correlation of Akita and Yoshida and Nguyen-tien et al. predicted volumetric mass transfer coefficient within 12% of each other for the reactor used in PDU operation.

Development of Computer Codes:

Computer codes for the reactor model have been developed for the following cases;

With external recirculation of slurry

1) Gas plug flow; Liquid axial dispersion
2) Gas axial dispersion; Liquid axial dispersion

No external recirculation of slurry

3) Gas plug flow; Liquid axial dispersion
4) Gas axial dispersion; Liquid axial dispersion

Most of the runs will be carried out for case 4. A comparison of the results for cases 3 and 4 will indicate the importance of gas phase backmixing on reactor performance when there is no external recirculation of slurry.

The effects of following variables will be studied:

* Reactor diameter and length
* Feed gas composition
* Reactor temperature and pressure

- Gas flow rate
- Catalyst particle size
- Slurry concentration

In addition parametric sensitivity analysis will be carried out to study the influence of the following on reactor performance.

- Liquid dispersion coefficient
- Gas dispersion coefficient
- Solid dispersion coefficient
- Gas holdup variations along reactor height

Notations:

a : gas-liquid interfacial area, m^{-1}

C_{cat} : catalyst concentration, gm/cc slurry

$C_{cat,avg}$: average catalyst concentration, kg/m^3 slurry

$C_{cat,b}$: catalyst concentration at column bottom, kg/m^3 slurry

$C_{G,i}$: gas-phase concentration of component i, $kmol/m^3$

$C_{L,i}$: liquid-phase concentration of component i, $kmol/m^3$

C_G : total gas-phase concentration, $kmol/m^3$

D_G : gas-phase dispersion coefficient, m^2/s

D_i : diffusivity of component i in the reactor liquid (m^2/s)

D_L : liquid-phase dispersion coefficient, m^2/s

$D_{o,w}$: diffusivity of oxygen in water (m^2/s)

D_S : solid-phase dispersion coefficient, m^2/s

H_i : Henery's constant for component i ($kgmoles/m^3.atm$)

H_o : static slurry height (m)

H_R : heat of reaction, kJ/kmol

k_{eff} : effective heat dispersion coefficient, kJ/m.s.K

$k_{L,i}$: liquid-side mass transfer coefficient for component i, m/s

$k_L a$: volumetric mass transfer coefficient (s^{-1})

$(k_L a)_i$: volumetric mass transfer coefficient for component i (s^{-1})

$(k_L a)_o$: volumetric mass transfer coefficient for oxygen in aqueous phase (s_{-1})

L : reactor length, m

m_i : solubility coefficient for component i, $(mol/m^3)_G/(mol/m^3)_L$

P : total pressure, atm

R : universal gas constant, 0.082 m^3-atm/kmole/K

r_k : reaction rate for kth reaction, kmol/kg.s

T : temperature, K

T_o : inlet temperature, K

T_w : reactor wall temperature, K

U_G : gas superficial velocity, m/s

U_{Go} : inlet gas superficial velocity, m/s

U_L : liquid superficial velocity, m/s

U_S : settling velocity of catalyst particles in swarm, m/s

v : dimensionless gas-phase superficial velocity (U_G/U_{Go})

w : dimensionless catalyst concentration

x_i : dimensionless liquid-phase concentration of component i ($C_{L,i}H_i/P$)

y_i : mole fraction of component i in the gas phase

z : dimensionless axial distance (x/L)

Dimensionless numbers

Bo : Bodenstein number $(U_S H_o / D_g)$

Fr_G : Froude number for gas $(U_G / gD_c$

Pe_G : gas-phase Peclet number $(U_{Go} L / D_G e_G)$

Pe_H : Peclet number for heat transfer $(U_L L / E_L e_L)$

Pe_L : liquid-phase Peclet number $(U_L L / D_L e_L)$

Pe_S : solid-phase Peclet number $[(U_S - U_L / e_L)(L / D_S)$

Pe_S' : solid-phase Peclet number with no liquid flow $(e_L U_S L / D_S)$

Pe_p : particle Peclet number $(U_G D_c / D_s)$

Re_G : gas Reynolds number $(U_G D_c o_l / \mu_l)$

Re_p : particle Reynolds number $(U_t d_p o_l / \mu_l)$

St_G : gas-phase Stanton number $(K_{L,i} aL / U_{Go})(RT / H_i)$

St_H : Stanton number for heat transfer $(ha_H L / pC_p U_L)$

$St_{L,i}$: liquid-phase Stanton number for component i $(K_{L,i} aL / U_L)$

Greek

α_{ik} : stoichiometric coefficient of component i in reaction k

ϵ_G : gas holdup

e_L : liquid holdup

e_s : solids holdup

$e_{s,sus}$: solids holdup in the slurry

μ_G : viscosity of gas phase (kg/m.s)

μ_L : viscosity of liquid phase (kg/m.s)

ν_L : kinematic liquid viscosity (m^2/s)

ν_{sus} : kinematic viscosity of slurry (m^2/s)

$\nu_{eff,rad}$: radial momentum transfer coefficient (m^2/s)

ρ_G : density of gas phase (kg/m^3)

ρ_L : density of the liquid phase, kg/m^3

ρ_{sus} : density of pseudo-homogeneous slurry (kg/m^3)

σ : surface tension of liquid (kg/s^2)

Subscripts

L : liquid phase

G : gas phase

i : component i

k : kth reaction

w : wall

o : inlet condition

S : solid phase

REFERENCES

Akita, K., and Yoshida, F., "Gas Holdup and Volumetric Mass Transfer Coefficient in Bubble Columns", Ind. Eng. Chem. Proc. Des. Dev., Vol. 12, p. 76 (1973).

Deckwer, W.D., Louisi, Y., Zaidi, A., Ralek, M., Ind. Eng. Chem. Process Des. Dev., Vol. 19, p. 699 (1980).

Hikita.H., Asai, S.,Tanigawa,K., Segawa, K., Kitao, M., "The Volumetric Liquid Phase Mass Transfer Coefficient in Bubble Columns", Chem. Eng. J., p.61 (1980).

Hughmark, G.A., "Holdup and Mass Transfer in Bubble Columns", Ind. Engng. Chem. Proc. Des. Dev., Vol. 6, p. 218 (1967).

Koide, K., Takazawa, A., Komura, M., Matsunaga, H., "Gas Holdup and Volumetric Liquid Phase Mass Transfer Coefficient in Solid-Suspended Bubble Columns", J. Chem. Eng. Jpn., Vol. 1, p. 459 (1984).

Nguyen-Tien, K., Patwari, R.N., Schumpe, A., Deckwer, W.D.,"Gas-Liquid Mass Transfer in Fluidized Particle Beds", AIChE J., Vol. 31, p. 194 (1985).

Sada, E., Kumazawa, H., Lee, C., Iguchi, T., "Gas Holdup and Mass Transfer Characteristics in Three-Phase Bubble Columns", Ind. Eng. Chem. Proc. Des. Dev., Vol. 25, p. 472 (1986).

Sauer, T., Hempel, D.C., "Fluid Dynamics and Mass Transfer in a Bubble Column with Suspended Particles", Chem Eng. Technol., Vol. 10, p. 180 (1987).

Gas Holdup Correlations:

Koide et al. (1984)

$$\frac{\epsilon_G}{(1-\epsilon_G)^4} = \frac{0.277\left(\frac{U_G\mu_L}{\sigma_L}\right)^{0.918}\left(\frac{g\mu_L^4}{\rho_L\sigma_L^3}\right)^{-0.252}}{1+4.35\left(\frac{\epsilon_S}{\rho_S}\right)^{0.748}\left(\frac{\rho_S-\rho_L}{\rho_L}\right)^{0.881}\left(\frac{D_T U_G\rho_L}{\mu_L}\right)^{-0.168}}$$

This correlation is based on a large number of data points.

Sada et al. (1986)

$$[\epsilon_G/(1-\epsilon_G)^3]_{III} = 0.41\ U_t^{\ 1/16}\ \epsilon_{s,sus}^{\ -(0.125\ U_t^{0.19})} \qquad [\epsilon_G/(1-\epsilon_G)^4]_{II}$$

here

$U_t = g(\rho_p - \rho_l)d_p^2/(18\mu_l)$

units of U_t and U_G are in cm/s

Sauer and Hempel (1987)

$$[\epsilon_G/(1-\epsilon_G)] = 0.0277\ [U_G/(\nu_{sus}\ gU_G)^{1/4}]^{0.844}\ [\nu_{sus}/\nu_{eff,rad}]^{-0.136}\ [C_{cat,avg}/C_{cat,b}]^{0.0392}$$

$\nu_{sus} = \nu_l\ (1 + 2.5\epsilon_s + 10.05\epsilon_s^2 + 2.73\text{x}10^{-3}\ \exp(16.6\epsilon_s))$

$\nu_{eff,rad} = 0.011\ D_c(gD_c)^{0.5}\ (U_G^3/\nu_{sus}g)^{1/8}$

$C_{cat,avg}/C_{cat,b} = [1 - \exp(-Bo)]/Bo$

$Bo = U_sH_o/D_s$

This correlation accounts for the effect of suspension viscosity on gas holdup in slurry bubble column.

Deckwer et al. (1980)

$$\epsilon_G = 0.053\ U_G^{1.1}$$

This correlation is based on the data obtained with molten wax slurry system applied in the FT synthesis.

Hughmark (1967)

$$\epsilon_G = [2 + (0.35/U_G)(\rho_L\ \sigma/72)^{1/3}]^{-1}$$

Smith and Ruether (1985) observed that incorporation of the slurry density rather than the liquid density in the above equation reduced the residual error for slurry bubble columns.

Akita and Yoshida (1973)

$$[\epsilon_G/(1-\epsilon_G)^4] = 0.2\left(\frac{gD_c^2\rho_L}{\sigma}\right)^{1/8} (gD_c^3/\nu_L^2)^{1/12}\ U_G/(gD_c)^{0.5}$$

This correlation is based on the data obtained with a solid-free bubble column. For a slurry bubble column this correlation may be applicable only for suspension of very fine particles (< 10 μm) at high gas velocities (> 0.1 m/s).

Hikita et al. (1980)

$$\epsilon_G = 0.672(U_G\mu_L/\sigma)^{0.578}\ (\mu_L^4 g/\rho_L\sigma_L^3)^{-0.131}(\rho_G/\rho_L)^{0.062}\ (\mu_G/\mu_L)^{0.107}$$

This correlation is also based on the data obtained from solid-free bubble column, its applicability may, therefore, be limited to fine particles suspension.

SATISFACTION GUARANTEED

NTIS strives to provide quality products, reliable service, and fast delivery. Please contact us for a replacement within 30 days if the item you receive is defective or if we have made an error in filling your order.

▶E-mail: info@ntis.gov
▶Phone: 1-888-584-8332 or (703)605-6050

Reproduced by NTIS

National Technical Information Service
Springfield, VA 22161

This report was printed specifically for your order from nearly 3 million titles available in our collection.

For economy and efficiency, NTIS does not maintain stock of its vast collection of technical reports. Rather, most documents are custom reproduced for each order. Documents that are not in electronic format are reproduced from master archival copies and are the best possible reproductions available.

Occasionally, older master materials may reproduce portions of documents that are not fully legible. If you have questions concerning this document or any order you have placed with NTIS, please call our Customer Service Department at (703) 605-6050.

About NTIS

NTIS collects scientific, technical, engineering, and related business information – then organizes, maintains, and disseminates that information in a variety of formats – including electronic download, online access, CD-ROM, magnetic tape, diskette, multimedia, microfiche and paper.

The NTIS collection of nearly 3 million titles includes reports describing research conducted or sponsored by federal agencies and their contractors; statistical and business information; U.S. military publications; multimedia training products; computer software and electronic databases developed by federal agencies; and technical reports prepared by research organizations worldwide.

For more information about NTIS, visit our Web site at http://www.ntis.gov.

NTIS

Ensuring Permanent, Easy Access to U.S. Government Information Assets

U.S. DEPARTMENT OF COMMERCE
Technology Administration
National Technical Information Service
Springfield, VA 22161 (703) 605-6000