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Sujatha Degaleesan

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Nomenclature

a	dilation parameter
AR	amplitude ratio
A_s	surface area defined by Equation 3.9
b	translation parameter
Bo	Bodenstien Number
c	constant in Equation 2.7
C	liquid tracer concentration, mol/cm^3
C_0, C_1	constants in Equation 2.2
C_μ	constant in Equation 4.8
d	distance, cm
d_b	bubble diameter, cm
d_o	orifice diameter, cm
d_R	diameter of heat exchange tubes, cm
D_1, D_2	Average axial eddy diffusivities in region 1 and 2 (Figure 6.22), cm^2/s
$\mathbf{D}$	turbulent eddy diffusivity tensor
D_{ax}	axial dispersion coefficient, cm^2/s
D_c	column diameter, cm
D_{eff}	effective axial dispersion coefficient defined by Equation 6.58, cm^2/s
D_m	molecular diffusivity, cm^2/s
D_{rr}, D_{zz}, $D_{\theta\theta}$	diagonal elements of eddy diffusivity tensor, cm^2/s
D_{rz}, $D_{r\theta}$, $D_{z\theta}$	off-diagonal elements of eddy diffusivity tensor, cm^2/s
$\overline{D}_{rr}$, $\overline{D}_{zz}$	average radial and axial eddy diffusivities, cm^2/s
D_{ri}	radial eddy diffusivity at inversion point, cm^2/s
e	particle position error, cm,
	entropy
$E(\omega)$	energy spectrum
f	time series

f	frequency index
f_i	ratio factor for distribution of kinetic energy along i^{th} direction
f_p	polynomial function
$f_t(t)$	input function to simulate tracer injection, mol/cm^3
F_i	interfacial coupling term
F	volumetric flow rate, cm^3/s
Fr	Froude Number
g	acceleration due to gravity, cm^2/s
G	band pass filter
H	low pass filter
$H(t)$	Heaviside step function
I	Intensity of radiation
j_{GL}	drift flux, cm/s
J	concentration flux, mol/cm^2.s
k	turbulent kinetic energy per unit volume, dyne/cm^2
k_z	constant in Equation 6.48, cm^{-1}
K_r	constant in Equation 2.46
K_g	constant in Equation 2.51
K_T	constant in Equation 6.59
l	length scale, cm
l_r, l_z	turbulent length scales along r and z, cm
L	dispersion height, cm
L_{min}, L_{max}	axial levels (range) for axial averaging of CARPT parameters, cm
m	void fraction exponent (Equation 2.7),
	discretized dilation parameter
n	discretized translation parameter
n_c	intensity counts per sampling time
$n(t)$	noise (Chapter 3.7)
$\vec{n}$	unit normal vector
N	length of data set (signal)
N_c	dimensionless capacitance number
N_{exp}	number of statistically independent experiments in a compartment
N_R	number of tubes in reactor
N_s	Stokes Number
N_T	number of time steps

N_{vel}	Number of occurrences of tracer in a compartment
P	pressure, atm
P_o	pressure at orifice, atm
p	ratio of particle response time to Lagrangian integral time scale, position index (Chapter 3.7)
PA	phase angle
Pe	Peclet number
P_m	specific energy dissipation per unit mass, cm^2/s^3
P_2	second order polynomial (Equation 5.12)
P_4	fourth order polynomial (Equation 5.11)
r_p	radius of curvature for controlled motion experiment, cm
R	column radius, cm, Lagrangian correlation coefficient, cm^2/s^2
Re	Reynolds number
R'_L	Lagrangian autocorrelation coefficient (Equation 4.12)
R_s	radius of curvature in Equation 3.9
s	scale index (Chapter 3.7)
st	energy threshold for wavelet filtering
t	time, s
T_{dim}	number of azimuthal subdivisions which define sampling compartments
T_{lag}	number of time steps to ensure statistically independent experiments
T_s	viscous shear stress, $dyne/cm^2$
u	Eulerian velocity, cm/s
$\vec{u}$	Eulerian velocity vector
$\bar{u}_1$, $\bar{u}_2$	average liquid velocity in upflow and downflow regions (Figure 6.22), cm/s
$u_{b\infty}$	bubble rise velocity, cm/s
$\vec{u}_i$	interfacial velocity vector
u_{Lc}	liquid circulation velocity, cm/s
$\bar{u}_{rec}$	mean recirculation velocity (Equation 5.3), cm/s
u_s	slip velocity, cm/s
U_g	superficial gas velocity, cm/s
U_{ge}	equivalent gas superficial velocity, cm/s
U_l	superficial liquid velocity, cm/s
U_{trans}	transition gas velocity, cm/s
v	Lagrangian velocity, cm/s

V	volume, cm^3
V_{f-p}	v_f - v_p, cm/s
V_{lb}	large bubble rise velocity, cm/s
V_{small}	small bubbles rise velocity, cm/s
$\vec{x}$	position vector
X_k	phase (k) function
y	displacement of fluid due to fluctuating Lagrangian velocity, cm
y^2	mean square displacement due to fluctuating Lagrangian velocity, cm

Greek symbols

α_k	local holdup (ensemble averaging) of phase k
β	defined by Equation 3.3
β_k	local holdup (volume averaging) of phase k
δ	dirac impulse input
δ_{ij}	Kronecker delta
ϵ	energy dissipation
$\bar{\epsilon}_1$, $\bar{\epsilon}_2$	average liquid holdups in upflow and downflow region (Figure 6.22)
ϵ_g, ϵ_l	local holdup of gas, liquid
$\bar{\epsilon}_g$	cross sectional (column) average gas holdup
$\hat{\epsilon}_g$	line average gas holdup
ϵ_{lb}	holdup of large bubbles
ϵ_{trans}	gas holdup at transition (small bubbles)
ϕ_s	relative free area in chapter 6
λ	wavelet packet coefficient
μ	viscosity, cP,
	linear attenuation coefficient, cm^{-1}
ν	kinematic viscosity, cm^2/s
ν_t	turbulent viscosity, cm^2/s
ω	angular frequency, radians/s,
	solid angle
ρ	density, g/cm^3
ψ	mother wavelet
$\psi_{a,b}$	wavelet defined by Equation 3.19
σ	surface tension, dyne/cm

σ_n^2	variance of noise
τ	Reynolds stress, dyne/cm^2
τ_L	Lagrangian integral time scale, s
τ_p	particle response time, s
τ_{rr}, $\tau_{\theta\theta}$, τ_{zz}	turbulent normal stresses. dyne/cm^2
τ_{rz}, $\tau_{r\theta}$, $\tau_{z\theta}$	turbulent shear stresses, dyne/cm^2
ξ	nondimensional radius

subscript

a, b	perfectly mixed regions A, B (Figure 6.22)
ch	sparger chamber
eff	effective
f	fluid
g, G	gas
l, L	liquid
k	phase
n	noise
p	particle
r,θ,z	cylindrical coordinates
x,y,z	Cartesian coordinates
sl	slurry

superscript

x	phase weighted
$x\rho$	mass weighted
$'$	fluctuating quantity (time averaged - instantaneous)

Chapter 1

Introduction

Bubble columns are cylindrical vessels wherein gas is sparged, via a distributor, into a medium of liquid or liquid-solid suspension, in the form of bubbles. Bubble columns serve as multiphase contactors and reactors in the chemical, petrochemical, biochemical and metallurgical industries. As reactors, bubble columns are used for chemical processes involving oxidation, chlorination, alkylation, polymerization and hydrogenation reactions (Shah et al. 1982; Fan 1989). Other processes that employ bubble columns include hydro treating and conversion of petroleum residues (Lunin et al. 1985), coal gasification and coal liquefaction processes such as the Exxon Donor Solvent (EDS) process (Tarmy et al. 1984) in the direct production of liquid fuels from coal. Bubble columns have been identified as a suitable type of reactor for a variety of gas conversion processes involving the production of liquid fuels from synthesis gas (that is produced by coal gasification). Typical examples are the Fischer-Tropsch process (Kolbel and Ralek 1980), synthesis of methanol and other synthetic fuels that offer environmental advantages over petroleum derived fuels. Technology for the liquid phase methanol synthesis, LPMEOH^{TM}, in slurry bubble columns, is being developed by Air Products and Chemicals. Bubble columns have also found wide application as fermenters, in waste water treatment and in a variety of metallurgical operations such as leaching of metal ores. Good heat and mass transfer characteristics, lack of moving parts and therefore reduced wear and tear, higher durability of catalyst, ease of operation, compactness and low operating and maintenance costs are the advantages that render bubble columns as an attractive reactor choice for various multiphase processes, especially for processes involving highly exothermic reactions.

Bubble columns usually operate with a length to diameter ratio, or aspect ratio, of at least five. They are operated in either semi-batch (zero liquid throughput) or continuous mode (co-current or counter-current), with liquid superficial velocities lower than the gas superficial velocity by at least an order of magnitude. As a result, it is the flow of gas that controls the hydrodynamics of the individual phases in these systems. In the presence of a solid (catalyst) phase, these reactors are commonly referred to as slurry bubble columns. The size of particles used in slurry bubble columns are typically in the range of 5 to 100 μm, with a loading of up to 50 % by weight (Krishna et al. 1997a). Heat exchanger tubes may be inserted into the reactor for cooling the system and maintaining isothermal conditions, especially for highly exothermic reactions. In addition, in some cases the column may be sectionalized using baffles to inhibit liquid backmixing (Shah et al. 1982; Deckwer and Schumpe 1993).

In general, the design and scale-up of bubble column reactors depends on the quantification of three main phenomena: 1) heat and mass transfer, 2) mixing characteristics and 3) chemical kinetics of the reacting system. The first two factors are system dependent and intimately linked with the fluid dynamics of bubble columns. Although simple in construction, due to the complex hydrodynamics and its influence on transport characteristics (Figure 1), the design and scale-up of bubble columns has involved considerable empiricism, and has been the subject of extensive ongoing research (Shah et al. 1982; Deckwer et al. 1987; 1993; Fan 1989; Saxena 1995). Reported studies indicate that the accurate and successful design and scale-up of bubble column reactors requires an improved understanding of the multiphase fluid dynamics and its influence on phase holdup distribution, mixing and transport characteristics.

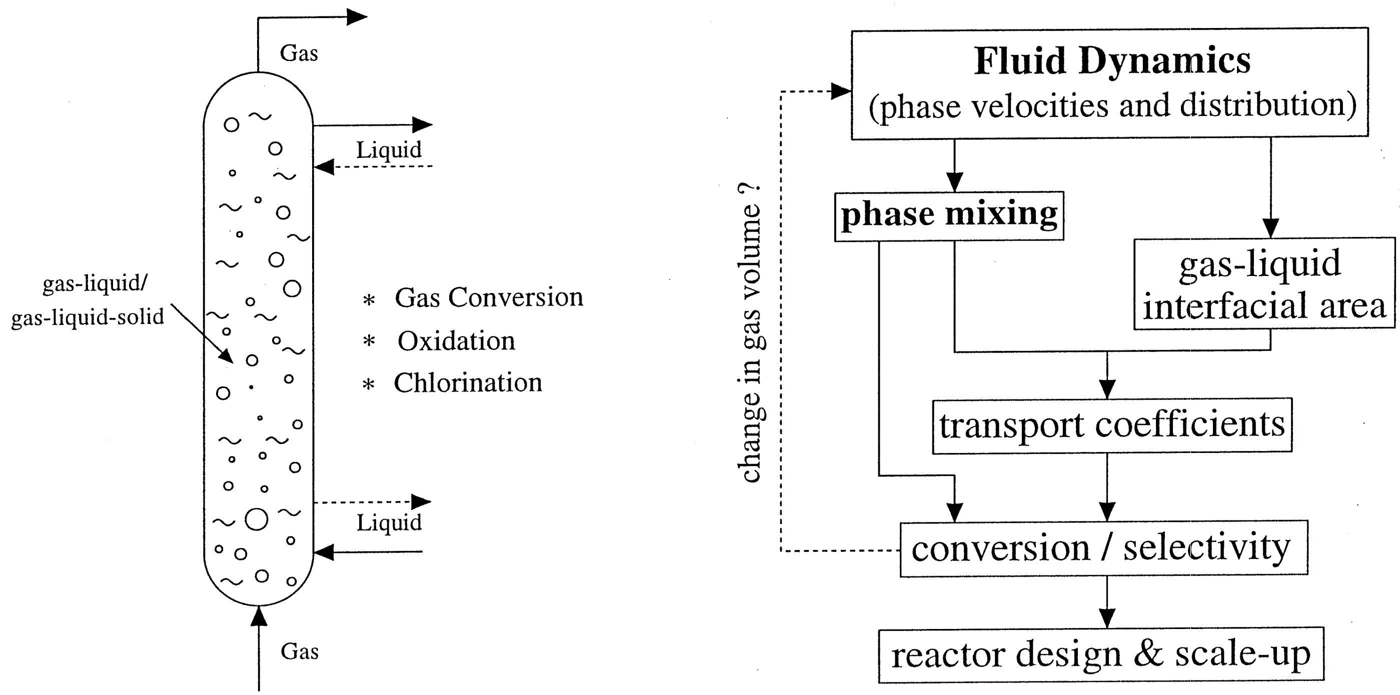

Figure 1.1: Bubble Columns and Slurry Bubble Columns

In recent years considerable effort has been directed towards fundamental fluid dynamic modeling of two phase flows in bubble columns with the aim of using these models as tools in the design of bubble column reactors. This has resulted in a number of computational fluid dynamic (CFD) codes (Svendsen et al. 1992; Ranade 1992; Grienberger and Hofmann 1992; Sokolichin and Eigenberger 1994; Lapin and Lubbert 1994; Delnoij et al. 1997). However, the uncertainty regarding the various phase interaction terms and turbulence closure schemes, in addition to the computational difficulties associated with computation of large flow fields have delayed the full implementation of these codes in practice. Simulation of the churn-turbulent flow regime, which is preferred for industrial bubble columns, poses further difficulties due to the complications arising from the multiple bubble sizes and bubble interactions. It is widely recognized that the present fluid dynamic models require experimental data as input and for model verification. Hence, the efforts in modeling have to be supplemented with reliable experimental data for the fluid dynamic parameters describing phase interaction and turbulence. Providing such a reliable experimental database for the liquid phase fluid dynamic parameters under various conditions, using the Computer Automated Radioactive Particle Tracking (CARPT) technique is one of the goals of this investigation. The CARPT technique was adapted by Devanathan (1991) for tracking the liquid phase in bubble columns. This is a non-invasive technique used to measure the liquid velocities and turbulence parameters, such as the intensity of turbulence, Reynolds stresses and the turbulent eddy diffusivities.

While one awaits full verification of fundamentally based CFD codes, there is a need to develop phenomenological models that mechanistically relate phase mixing and other transport characteristics to the fluid dynamics in the system. Liquid phase mixing is one of the important factors in the design of bubble columns reactors, which not only governs the residence time distribution of the liquid, but also affects that of the gas phase, and in addition, determines the mean driving force for mass transfer. Since with the CARPT technique it is possible to measure all the fluid dynamic parameters of the liquid phase, including the Lagrangian based turbulent eddy diffusivities, such experimental information can be utilized to study and model the effect of fluid dynamics on liquid phase mixing in bubble columns.

1.1 Objectives

The objectives of this research work are two fold: (i) improvement of the CARPT technique and measurement of pertinent fluid dynamic parameters in bubble columns and (ii) quantification of liquid mixing in bubble columns. The specific research goals are outlined below:

- **CARPT Technique and Experiments:**

  Improvement of the CARPT technique to obtain more reliable data for liquid phase velocities and turbulence parameters in bubble columns is one of the objectives of this study. This goal is pursued by (i) the development of a reconstruction algorithm for CARPT calibration which takes into account the effects of solid angle and (ii) implementation of a wavelet-based filtering algorithm to reduce the white noise in the instantaneous position data that arises due to statistical fluctuations in the gamma radiation. The details of these developments along with a description of the experimental facility are presented in Chapter 3.

  Another goal is to extend the CARPT database by conducting CARPT experiments in bubble columns of three sizes, 14 cm, 19 cm and 44 cm in diameter, to study the effects of scale and operating conditions on the fluid dynamic parameters in air-water systems. The parameters measured using CARPT (in the entire flow field) include the time averaged liquid velocities, turbulence parameters such as the turbulent Reynolds stresses and turbulent kinetic energy, and the turbulent eddy diffusivities. The experimental conditions along with results are discussed in Chapter 4.

In order to meet the second objective of quantifying liquid mixing in bubble columns reactors, scale-up relations are developed for the mean liquid recirculation velocity and the average eddy diffusivities for air-water systems in the churn-turbulent flow regime, based on the database generated in this work along with measurements of Kumar (1994) for the void fraction and literature data (Nottenkamper et al. 1983; Menzel et al. 1990). Further, a procedure is sought based on a unified characterization of churn-turbulent bubble columns (of large diameters) for estimation of the mean liquid recirculation and average eddy diffusivities in industrial bubble column reactors under different process conditions. These scale-up issues are addressed in Chapter 5.

- **Liquid Mixing in Bubble Columns:**

Another objective of this research is to utilize the experimental information for the hydrodynamics to model liquid mixing in bubble columns. For this purpose, a fundamental two dimensional convection-diffusion model for liquid mixing in bubble columns is developed, in the attempt to tie the experimental fluid dynamic data from CARPT and CT (computed tomography) to liquid mixing. The details of the model are provided in Chapter 6. One of the goals in this regard, is to test the two dimensional model along with the proposed scale-up methodology to phenomenologically describe liquid (slurry) mixing in an industrial bubble column. This is accomplished for the case of the methanol synthesis slurry bubble column reactor.

To promote its use in industrial practice the full blown two dimensional model is further simplified to a one dimensional representation, referred to as the recycle with cross flow and dispersion model or the two-compartment convective diffusion model.

Due to the widespread use of the axial dispersion model, it is deemed appropriate to relate the axial dispersion coefficient to the fundamental fluid dynamic parameters. For this purpose, a Taylor type analysis of the two dimensional convection-diffusion equation is performed to arrive at an expression for an effective axial dispersion coefficient (in the one dimensional axial dispersion model) in bubble columns. In combination with the scale-up rules developed, this analysis provides a basis for evaluation of the axial dispersion coefficient in industrial bubble column reactors which usually operate under high pressure with internal heat transfer tubes.

1.2 Structure of Thesis

The thesis is organized as follows: A brief background on the experimental and modeling aspects of bubble columns is given in Chapter 2. An introduction to the CARPT technique and the improvements resulting from this work are described in Chapter 3. Details of the experiments conducted and the results obtained are discussed in Chapter 4. Scale-up issues are addressed in Chapter 5. The modeling of liquid mixing in bubble columns is discussed in Chapter 6. A summary of the research findings along with recommendations for future work is provided in Chapter 7.