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Scale-up of Bubble Column Reactors: A Review of Current State-of-the-Art

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ABSTRACT: In multiphase flow reaction systems, in general, an extrapolation of small diameter behavior to larger ones is always an important and challenging task. The critical issue in such an extrapolation remains to be mixing and hydrodynamic characteristics. It needs reliable similarity criteria that would result in similar mixing and hydrodynamics and hence transport and performance in two different scales. Numerous experimental and computational studies have been performed to investigate the flow behavior of bubble column reactors for a proper design and scale-up. Experimental techniques vary from simple visual observation to more advanced noninvasive diagnostic techniques. On computational front the progress has been made from simple reactor models to fundamentally based Computational Fluid Dynamics (CFD). Such studies ultimately provide a knowledge that help in understanding the hydrodynamic and mixing characteristics of these reactors and would aid in its scale-up. Based on these studies, various methodologies have been proposed in literature for scale-up and/or to maintain their hydrodynamic and mixing similarity. This article attempts to review the current state of reported dynamic similarity and scale-up methods of bubble column reactors. It mostly covers the methods reported in open literature. The scale-up practices in industry appear to be proprietary for obvious reasons.

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1. INTRODUCTION

In a bubble column reactor, a gas phase is bubbled through a column of liquid to promote a chemical or biochemical reaction in the presence or absence of a catalyst suspended in the liquid phase (Figure 1). A bubble column offers numerous advantages

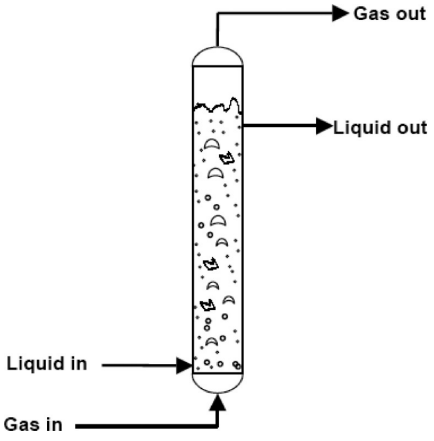


Figure 1. Schematic diagram of a bubble column.

such as good heat and mass transfer, no moving parts, ease of operation, and low operating and maintenance costs. The main disadvantage of bubble column reactors is backmixing, which adversely affects product conversion. In these reactors, momentum is transferred from the faster, upward moving gas phase to the slower liquid/slurry phase. The operating liquid superficial velocity (in the range of 0 to 2 cm/s) is an order of magnitude smaller than the superficial gas velocity (1 to 50 cm/s). Hence, the hydrodynamics of such reactors are controlled mainly by the gas flow. There exists mainly two flow regimes in relatively large diameter bubble columns, i.e., homogeneous and heterogeneous (Shaikh and Al-Dahhan[1]).

These reactors are of considerable interest in chemical, petrochemical, and petroleum industries for various processes. Examples of such processes are partial oxidation of ethylene to acetaldehyde, wet-air oxidation (Deckwer[2]), liquid phase methanol synthesis (LPMeOH), Fischer−Tropsch (FT) synthesis (Wender[3]), and hydrogenation of maleic acid (MAC). In biochemical industries, bubble columns are used for cultivation of bacteria, cultivation of mold fungi, production of single cell protein, animal cell culture (Lehmann et al.[4]), and treatment of sewage (Diesterweg[5]). In metallurgical industries, it can be used for leaching of ores. The most popular present day application of bubble columns is for energy conversion process where 'stranded gas' is being converted to liquids called Gas-to-Liquid (GTL) Fischer−Tropsch (FT) synthesis, when syngas is produced from natural gas. Syngas can also be produced from coal or biomass in which case this conversion is called Coal-to-Liquid (CTL) and Biomass-to-Liquid (BTL), respectively. The popularity of such a conversion is a response to the *postulated* future energy crisis.

Different reactor configurations were utilized for FT processes in earlier days while the design of bubble columns has been considered for low temperature FT processes since Kolbel's[6] pioneering work in the 1950s. Krishna and Sie[7] showed that a bubble column reactor may achieve a productivity of 2500 times higher than that of the classical FT reactors used in the 1940s such as fixed bed reactors,

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multibed reactors. However, there are considerable reactor design and scale-up problems associated with such energy conversion processes involving bubble columns. In general, for any multiphase reaction system, an extrapolation of small scale behavior to larger ones is always an important and challenging task. The critical issue in such an extrapolation remains to be mixing and hydrodynamic characteristics. It needs reliable similarity criteria that would result in similar mixing and hydrodynamic which would provide similar transport and performance (conversion and selectivity). Therefore, numerous experimental and computational studies have been performed to investigate the flow behavior of bubble column reactors for proper design and scale-up.

In order to achieve economically high space-time yields in the GTL-FT reactors, high slurry concentration (typically 30−50% vol.) needs to be employed. To suspend such a high amount of catalyst, high energy input is needed that can be provided by high superficial gas velocities which make the reactor operate in a churn-turbulent flow regime. The process operates under high-pressure conditions (typically 10−80 bar). To remove high exothermic heat of reaction, an efficient means of heat removal is needed, and also it needs to be operated in a churn-turbulent flow regime that can be generally achieved at high superficial gas velocities. Finally, the large gas throughputs necessitate the use of large diameter reactors (typically 5−8 m), and to obtain high conversion levels, large reactor heights, typically 20−30 m tall, are required. Successful commercialization of the bubble column reactors is crucially dependent on the proper understanding of its hydrodynamics and the scale-up principles.

Thus, various methodologies and approaches have been used and reported in the open literature for scale-up of bubble columns. This overview examines current state-of-the-art of scale-up of bubble column reactors reported in the literature. Most of these studies have focused on developing either scaling rules or an approach that would provide a platform for design and scale-up of bubble columns.

2. OVERVIEW OF REPORTED SCALE-UP METHODOLOGIES AND PROCEDURES

The following is a summary and review of the current state of the scale-up of bubble column reactors reported in the literature. We do not discuss development of phenomenological/CFD models, nor do we provide details of experimental techniques used in those studies.

2.1. Wilkinson et al.[8] Wilkinson et al.[8] proposed three criteria discussed below based on matching overall gas holdup for scale-up of high pressure bubble column reactors. A correlation for overall gas holdup was proposed based on their own as well as literature data that accounts for the effect of gas density and incorporates the flow regime transition.

Wilkinson et al.[8] performed experiments for scale-up purposes in two different column diameters (15 and 23 cm) at operating pressures varying between 0.1 to 2 MPa. Three different liquids were used: n-heptane (density = 0.684 g·cc^{-1}, viscosity = 0.00041 Pa·s, surface tension = 20 dyn·cm^{-1}), monoethylene glycol (density = 1.113 g·cc^{-1}, viscosity = 0.021 Pa·s, surface tension = 4 8 dyn·cm^{-1}), and water (density = 0.998 g·cc^{-1}, viscosity = 0.001 Pa·s, surface tension = 72 dyn·cm^{-1}). The hydrodynamics were quantified based on overall gas holdup. In addition, the authors have performed literature survey regarding the effect of liquid height and sparger design on gas holdup. Based on their own results and literature data, it was argued that the gas holdup is virtually independent of the column dimensions and the sparger layout (for low as well as high pressures) provided that the following three criteria are fulfilled:

1) The column diameter has to be larger than 15 cm.

2) The column height to diameter ratio has to be in excess of 5.

3) The hole diameter of the sparger has to be larger than 1−2 mm.

Wilkinson et al.[8] concluded that a scale-up procedure in which the gas holdup, volumetric mass transfer coefficient, and interfacial area for a large scale industrial bubble columns are estimated on the basis of experimental data obtained in a pilot plant bubble column with small dimensions (aspect ratio < 5, D < 15 cm) or with porous plate type of spargers will generally lead to a considerable overestimation of these parameters.

Wilkinson et al.[8] compared the predictions of selected reported correlations (Hikita et al.;[9] Hammer et al.;[10] Idogawa et al.;[11] Reilly et al.;[12] Idogawa et al.[13]) with their own data and literature data. The reported correlations were developed either at high operating pressure or for gases with different densities. Wilkinson et al.[8] found that none of these correlations could predict the gas holdup within statistical confidence. Reilly et al.[12] correlation showed the maximum error in this comparison. Wilkinson et al.[8] argued that the behavior of gas holdup varies according to the operating flow regime; hence, they developed a gas holdup correlation that incorporates flow regime transition. In addition, the effect of gas density and liquid phase properties on regime transition and gas holdup was studied.

The experimental data were incorporated with a gas holdup model developed by them, which is similar but slightly different than the one proposed by Krishna et al.[14] For superficial gas velocities less than transition velocity, the gas holdup increases proportionally to the superficial gas velocity, hence the following relationship was written for homogeneous flow

for $U_G < U_{trans}$:

$$\varepsilon_G = AU_G \tag{1}$$

which can be then written in the following form

$$\varepsilon_G = U_G/U_{s,b} \tag{2}$$

where $U_{s,b}$ is rise velocity of bubbles.

On the basis of dimensional analysis, Wilkinson et al.[8] proposed the following equation for bubble rise velocity

$$\frac{U_{s,b}\mu_L}{\sigma} = C\left[\frac{\sigma^3\rho_L}{g\mu_L^4}\right]^{n1}\left[\frac{\rho_L}{\rho_G}\right]^{n2} \tag{3}$$

for $U_G > U_{trans}$:

$$\varepsilon_G = U_{trans}/U_{s,b} + \frac{U_G - U_{trans}}{U_{l,b}} \tag{4}$$

In eq 4, $U_{l,b}$ is the rise velocity of large bubbles, which should be greater than $U_{s,b}$.

To hold this condition, the following dimensionless equation was chosen:

$$\frac{\mu_L U_{l,b}}{\sigma} = \frac{\mu_L U_{s,b}}{\sigma} + C\left[\frac{\mu_L(U_G - U_{trans})}{\sigma}\right]^{n3}\left[\frac{\sigma^3\rho_L}{g\mu_L^4}\right]^{n4}\left[\frac{\rho_L}{\rho_G}\right]^{n5} \tag{5}$$

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On the basis of rational explanation of behavior of transition velocity with the change in liquid properties and gas density, an empirical equation for transition velocity has been proposed

$$U_{trans} = F(\sigma, \rho_G, \rho_L, \mu_L) \tag{6}$$

Based on their own experimental data as well as published data from the literature, optimal values of parameters in eq 3, 5, and 6 were determined with the aid of nonlinear regression analysis. The regression analysis leads to the following correlations to predict transition velocity and holdup.

$$\frac{U_{s,b}\mu_L}{\sigma} = 2.23\left[\frac{\sigma^3\rho_L}{g\mu_L^4}\right]^{-0.273}\left[\frac{\rho_L}{\rho_G}\right]^{0.03} \tag{7}$$

$$\frac{\mu_L U_{l,b}}{\sigma} = \frac{\mu_L U_{s,b}}{\sigma} + 2.4\left[\frac{\mu_L(U_G - U_{trans})}{\sigma}\right]^{0.757}\left[\frac{\sigma^3\rho_L}{g\mu_L^4}\right]^{-0.077}\left[\frac{\rho_L}{\rho_G}\right]^{0.077} \tag{8}$$

$$\frac{U_{Gtrans}}{U_{s,b}} = \varepsilon_{Gtrans} = 0.5\ \exp(-193\rho_G^{-0.61}\mu_L^{0.5}\sigma_L^{0.11}) \tag{9}$$

Equations 7, 8, and 9 give the values of $U_{s,b}$, $U_{l,b}$, and U_{trans}. Using these values in eqs 2 and 4, one can predict overall gas holdup in homogeneous and heterogeneous regimes, respectively. The average error of prediction using the newly developed correlation was found to be 10% with a maximum error of 40%. The range within which these physical constants are valid is σ = 20−72 dyn·cm^{-1}; μ_L = 0.0004−0.055 Pa·s; ρ_L = 683−2960 kg·m^{-3}; ρ_G = 0.09−38 kg·m^{-3}. Hence, it was recommended to use the developed correlation of overall gas holdup in bubble columns for scale-up purposes.

The method proposed by Wilkinson et al.[8] relies completely on similarity of global hydrodynamics such as overall gas holdup. As described in the later part of this paper, Shaikh[15] and Shaikh and Al-Dahhan[16] have shown that such similarity may not hold in a heterogeneous flow regime.

2.2. Degaleesan.[17] Degaleesan[17] proposed a scale-up method that was based on the assumption that any gas−liquid/slurry would exhibit the similar hydrodynamic behavior as an air−water system if both the systems have the same overall gas holdup. The procedure involves measuring (or evaluating based on the suitable correlation) the overall gas holdup in a scaled up unit at its operating conditions and then calculating the equivalent superficial gas velocity, u_{Ge} that provides the same overall gas holdup in an atmospheric air−water system as the estimated one in the scaled up unit. Hence, it was suggested that hydrodynamics and mixing at the equivalent superficial gas velocity, u_{Ge}, in an atmospheric air−water system would represent the hydrodynamics and mixing in a scaled up unit.

To facilitate this, Degaleesan[17] developed a two-dimensional convection-diffusion model for liquid mixing to interpret the tracer response in an 18-in. diameter slurry bubble column reactor for liquid phase methanol synthesis at La Porte, Texas. The convection-diffusion model needs knowledge of liquid axial velocity profile, eddy diffusivities profile, and gas holdup profile to predict the tracer responses. The available fluid dynamic measurements in an industrial unit were gas holdup radial profile measured using Nuclear Gauge Densitometry (NGD). The liquid axial velocity profile and eddy diffusivities profile were not available in an industrial scale unit at reaction conditions. The experimental measurements of these fluid dynamic parameters were available only in a laboratory scale bubble column (diameter = 14, 19, 44 cm) at ambient conditions in an air−water system. Degaleesan[17] developed scaling rules to extrapolate available laboratory data to an industrial unit, to predict the needed fluid dynamic parameters,

The proposed approach was facilitated by the following correlations:

Overall Gas Holdup. To predict overall gas holdup in an atmospheric air−water system, Degaleesan[17] developed a correlation using literature as well as her own data in different column diameters. The proposed correlation is as follows:

$$\bar{\varepsilon}_G = 0.07U_G^{0.474-0.000626D}\ (\text{cgs units}) \tag{10}$$

The developed correlation showed a good comparison with Reilly et al.[12] and Hammer et al.[10] correlations at low superficial gas velocities. While at high superficial gas velocities, it captured the effect of diameter similar to the data of Nottenkamper et al.[18] However, based on eq 10, the effect of scale on overall gas holdup appears to be weak.

Liquid Recirculation. Experimental evidence shows that there exists a single circulation cell in a time-averaged sense with liquid upflow in the center and downflow near the wall. In a fully developed flow (L/D > 2), the liquid axial velocity appears to be axially invariant (Degaleesan[17]). The general picture of a liquid axial velocity profile in a fully developed flow is shown in Figure 2. The velocity is maximum in the center of

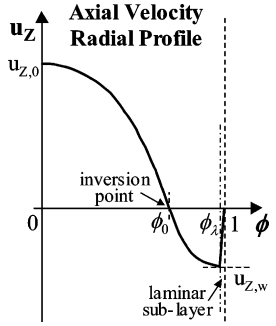


Figure 2. Radial profile of liquid axial velocity. Reprinted with permission from ref 19. Copyright 1979 John Wiley & Sons, Inc.

the column and monotically decreases with negative values at the wall. The dimensionless radial point where velocity reaches zero is called an 'inversion point' and is generally around 0.6−0.7 (Ueyama and Miyauchi[19]).

The strength of liquid recirculation is described in terms of mean liquid recirculation velocity which is defined as

$$\bar{u}_{rec} = \frac{\int_0^{\phi^*} u_z(\phi)[1 - \varepsilon_G(\phi)]\phi d\phi}{\int_0^{r^*}[1 - \varepsilon_G(\phi)]\phi d\phi} \tag{11}$$

where ϕ^* is the radial position of flow inversion.

In eq 11, the value of gas holdup radial profile is obtained using Computed Tomography (CT) while liquid axial velocity profile is obtained using Computer Automated Radioactive Particle Tracking (CARPT). The details of CT and CARPT are available elsewhere (Kumar;[20] Devanathan[21]) and will not be repeated here. Based on her own CARPT and Kumar[20] CT

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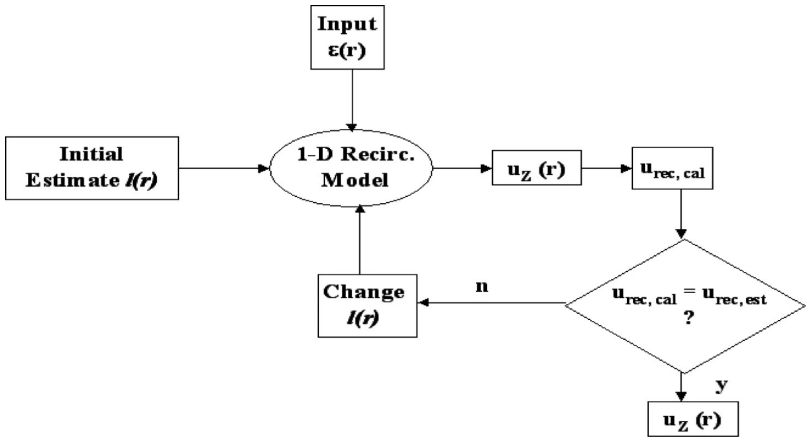


Figure 3. Determination of liquid velocity profile using the 1-D model and the known gas holdup radial profile (reproduced from ref 17).

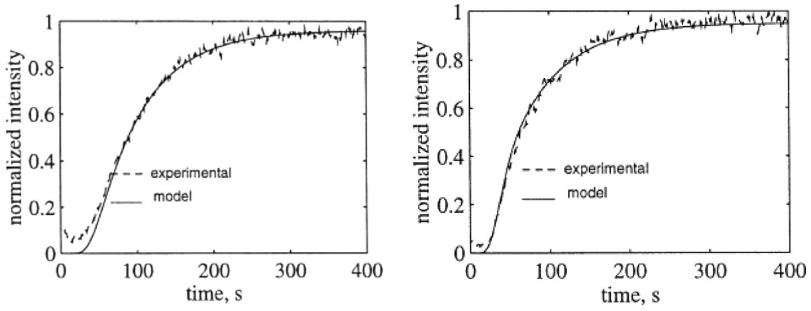


Figure 4. Comparison of model predictions and experimental data in 18-in. diameter column at liquid phase methanol synthesis conditions and Ug = 36 cm/s. Reprinted with permission from ref 17. Copyright 1997 Washington University.

data and literature data from Nottenkamper et al.,[18] the following correlation was developed for mean liquid recirculation velocity in an air−water system

$$\bar{u}_{rec} = 2.2D^{0.4}U_G^{0.4} \tag{12}$$

Eddy Diffusivities. Similar to the liquid recirculation velocity, correlations were developed for cross-sectionally averaged axial and radial eddy diffusivities using CARPT data in an air−water system as follows:

$$\bar{D}_{zz} = -\frac{2325}{D^{0.8}} + 106.6D^{0.3}U_G^{0.3} \tag{13}$$

$$\bar{D}_{rr} = -\frac{350}{D^{0.8}} + 13.0D^{0.3}U_G^{0.3} \tag{14}$$

The correlations suggest the weak effect of scale on gas holdup, while the effect of scale on mean liquid recirculation velocity and eddy diffusivities appears to be strong. Additionally, the correlations to predict radial profiles of eddy diffusivities using fourth and second order polynomials were developed as follows:

$$D_{zz}(\phi) = \bar{D}_{zz}\{-3.4979\phi^4 + 3.2704\phi^3 + 0.4693\phi^2 + 0.00503\phi + 0.5847\} \tag{15}$$

$$D_{rr}(\phi) = \bar{D}_{rr}\{-5.0929\phi^2 + 5.0717\phi + 0.1653\} \tag{16}$$

Degaleesan[17] developed the following generalized method to estimate liquid axial velocity profile and eddy diffusivity profiles in a scaled up unit using the available laboratory scale data and the above developed correlations:

i. For a given process condition, with prior knowledge of overall gas holdup (either by measurement or correlation prediction), calculate an equivalent superficial gas velocity, U_{Ge}, in an air−water system using eq 10.

ii. At the equivalent superficial gas velocity, calculate mean liquid recirculation velocity using eq 12.

iii. Calculate the radial profile of liquid velocity at equivalent superficial gas velocity using the 1-D recirculation model (Kumar[20]) as explained in Figure 3.

iv. At the equivalent superficial gas velocity, calculate eddy diffusivities profiles using eqs 13 to 16.

The liquid recirculation in its simplest form was modeled using the 1-D recirculation model to predict the liquid axial velocity profile. This model requires two inputs: a gas holdup radial profile and a closure for the turbulent shear stress (using either eddy viscosity or mixing length). In the current case, gas holdup radial profiles were obtained from experimental measurements using CT. The other unknown is mixing length, to which the model was found to be very sensitive (Kumar[20]). Also, Kumar[20] showed that there is no universal expression for eddy viscosity or mixing length that can be used over a wide range of operating and design conditions.

Hence, Degaleesan[17] used a guessed mixing length and iterated until mean liquid recirculation velocity calculated from the profile generated by 1-D recirculation model compares well with (i.e., converged to the) mean liquid recirculation velocity predicted using the developed correlation [eq 12].

Degaleesan[17] suggested that the liquid axial velocity profile and eddy diffusivities profile estimated using this procedure at equivalent superficial gas velocity indicates the mixing characteristics in an industrial scale unit as both the systems

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have similar overall gas holdup. She compared the prediction of a two-dimensional convection-diffusion model with the experimental tracer data obtained in an 18-in. diameter slurry bubble column for liquid phase methanol synthesis at LaPorte, TX. As shown in Figure 4, the model predictions are close to the experimental data.

The proposed method provides a systematic approach to characterize recirculation and mixing in an industrial scale bubble column using an atmospheric air−water data. However, similarity based on only overall gas holdup is not sufficient in a churn-turbulent flow regime as shown by Shaikh.[15] In addition, this method needs *a priori* knowledge of gas holdup and its distribution. The method relies on the use of a 1-D model to estimate circulation parameter. With advances in modeling techniques, one needs to utilize a more realistic description of the system.

2.3. Inga and Morsi.[22] Inga and Morsi[22] developed a scale-up/scale-down methodology for bubble and slurry bubble columns based on similarity of the relative importance of mass transfer resistance in the overall reaction resistances. They have demonstrated their procedure for FT synthesis where it was shown that the experimental results obtained in a laboratory scale stirred tank reactor could be extrapolated to design an industrial scale slurry bubble column.

The relative importance of mass transfer resistance was defined in terms of a dimensionless parameter, β_i, which represents the balance between k_La (mass transfer coefficient) and k_0 (rate of consumption, pseudokinetic constant for first order) as follows:

$$\beta_i = \frac{(1/k_La)}{(1/k_La) + (1/k_0)} \tag{17}$$

Accordingly, maintaining the same β in two reactors will result in the same reactant concentration and catalyst activity and thereby the similar conversion and selectivity in two reactors.

The procedure suggested by Inga and Morsi[22] can be summarized as follows:

1. Measure k_La and k_0 in a stirred tank reactor at the given operating conditions.
2. Calculate the value of dimensionless parameter, β_i, using eq 17.
3. For a given industrial scale slurry bubble column reactor, calculate the value of k_La using a correlation proposed by Inga and Morsi.[22] Adjust the operating conditions such that the value of β_i in the industrial scale unit is the same as the laboratory scale stirred tank.
4. Calculate the values of gas and liquid axial dispersion coefficients for an industrial scale slurry bubble column reactor using available correlations. In their case, the correlation proposed by Field and Davidson[23] was used.
5. Predict the performance of industrial scale slurry bubble column reactor using an axial dispersion model (ADM) at the adjusted conditions.

Inga and Morsi[22] demonstrated their proposed method for FT synthesis using a laboratory scale 4-L stirred tank reactor operating at 20 Hz and 5% wt which was simulated to a conceptual industrial scale slurry bubble column reactor. The conceptual slurry bubble column reactor with 7 m diameter and 30 m height operating at 30 bar, 523 K, and 20 cm/s was modeled using ADM. The simulations were performed to maintain similar β_i as in a stirred tank reactor. The authors found the same productivity in both the reactors when the values of β_i (= 0.2) were the same.

The method proposed by these authors shows that maintaining a relative contribution of transport parameter the same in two systems results in the same performance even if one system is impeller-agitated (CSTR) and the other one is gas-agitated (slurry bubble column). Although the Inga and Morsi[23] procedure combines transport and reaction kinetics, the use of overall mass transfer coefficient is not enough to describe the mixing as it represents global transport parameter. The authors described phase mixing in an industrial scale bubble column using ADM. The flow patterns in the bubble column are far more complex than plug flow superimposed with axial nonidealities. In addition, ADM needs reliable prediction of an axial dispersion coefficient in both gas and slurry phase.

2.4. Fan et al.[24] Fan et al.[24] proposed a criterion for hydrodynamic similarity in bubble and slurry bubble columns based on the overall gas holdup. A correlation was developed to predict overall gas holdup in bubble and slurry bubble columns in terms of three dimensionless numbers. It was argued that maintaining these numbers the same in two systems would lead to similar overall gas holdup and hence similar mixing and hydrodynamics.

Fan et al.[24] studied the effect of various operating parameters that have a profound effect on overall gas holdup. Based on the vast range of data collected from the literature and their own data, an empirical correlation was proposed to estimate the gas holdup in bubble column reactors

$$\frac{\varepsilon_G}{(1-\varepsilon_G)} = \frac{2.9\left(\frac{u_G^4\rho_G}{\sigma_L g}\right)^{\alpha}\left(\frac{\rho_G}{\rho_L}\right)^{\beta}}{[\cosh(Mo_{SL}^{0.054})]^{0.41}} \tag{18}$$

where

$$\alpha = 0.21Mo_{SL}^{0.0079};\ \beta = 0.096Mo_{SL}^{-0.011}$$

$$Mo_{SL} = \frac{g(\rho_{SL}-\rho_G)(\xi\mu_L)^4}{\rho_{SL}^2\sigma_L^3}$$

$$\ln(\xi) = 4.6C_V\{5.7C_V^{0.58}\sinh[-0.71\exp(-5.8C_V)\ln Mo^{0.22}] + 1\}$$

The average error of prediction of overall gas holdup over the collected databank was 13% with a maximum error of 53%.

The physical meaning of the dimensionless group of $u_G^4\rho_G/\sigma g$ in the above equation was based on the maximum stable bubble size. Using the Davis-Taylor relation, the rise velocity of the maximum stable bubble size is

$$u_{max} = C\left[\frac{\sigma g}{\rho_G}\right]^{1/4} \tag{19}$$

If one substitutes this equation into the dimensionless group it becomes

$$\frac{u_G^4\rho_G}{\sigma g}\alpha\left(\frac{u_G}{u_{max}}\right)^4 \tag{20}$$

This dimensionless number, therefore, signifies the contribution of large bubbles to overall gas holdup. As the correlation covers the wide range of operating conditions

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within acceptable statistical error, Fan et al.[24] suggested that hydrodynamic similarity requires three dimensionless numbers to be the same, i.e., u_G/u_{max}, Mo_{SL}, and ρ_G/ρ_L.

This proposed similarity rule has not been evaluated experimentally. The suggested dimensionless numbers do not account for the effect of column diameter on gas holdup. The reason for not accounting for the column diameter lies in the reported studies which found that, above 15 cm, column diameter has a negligible effect on the overall gas holdup. However, such studies were performed mostly but not exclusively in an air−water system. van Baten and Krishna[25] reported that in concentrated slurries such an observation does not quite hold as shown in Figure 5. Additionally, similarity criteria based only on overall gas holdup is not sufficient in a churn-turbulent flow regime as shown by Shaikh.[15]

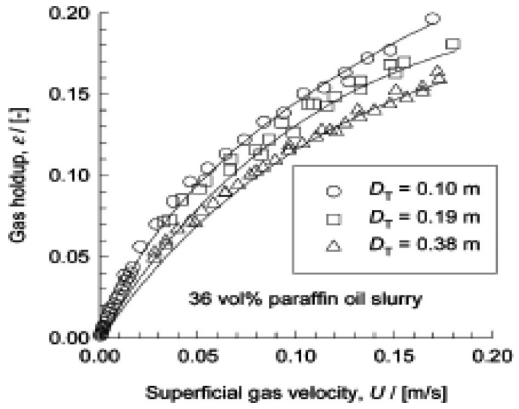


Figure 5. Overall gas holdup curve in three different column diameters using air−paraffin oil slurry. Reprinted with permission from 25. Copyright 2004 American Chemical Society.

2.5. Safonuik et al.[26] and Macchi et al.[27] Matching the dimensionless hydrodynamics groups or numbers derived from mass and momentum balances such as Reynolds (*Re*) and Froude (*Fr*) numbers has been used as a conventional scaling rule. For example, van den Bleek and Schouten[28] reported that in order to maintain dynamics and geometric similarity, *Re*, *Fr*, and dimensionless geometric numbers such as L/D should be maintained constant.

Accordingly, based on such an approach, Safonuik et al.[26] presented a scale-up method for three phase fluidized beds for dynamic similarity. In the proposed method, Safonuik et al.[26] identified the dimensionless numbers with the aid of the Buckingham-Pi theorem that have a profound effect on hydrodynamics of these reactors. The authors suggested that the dimensionless hydrodynamic parameters such as overall gas holdup in two independent systems would be the same if the identified dimensionless numbers were matched in these systems. Although the particle sizes used in their study, i.e., 0.8 and 1 mm, were outside the range generally employed for slurry bubble column reactors, this method is presented here as the concept that can be extended for bubble and slurry bubble column reactors.

Safonuik et al.[26] identified eight variables that have a significant effect on the hydrodynamics of three-phase fluidized beds as superficial gas velocity, superficial liquid velocity, liquid viscosity, surface tension, particle diameter, liquid density, particle density, and buoyancy ($\Delta\rho g$). Using these significant variables that involve three fundamental dimensions (mass, length, time), five independent dimensionless groups (Morton number, *Mo*; Etovos number, *Eo*; Reynolds number, *Re*; density ratio; superficial gas and liquid velocity ratio) were formed according to the Buckingham-Pi theorem as follows:

$$Mo = \frac{g\Delta\rho\mu_L^4}{\rho_L^2\sigma_L^3}, \quad Eo = \frac{g\Delta\rho d_p^2}{\sigma_L}, \quad Re_L = \frac{\rho_L d_p u_L}{\mu_L},$$

$$\beta_d = \frac{\rho_p}{\rho_L}, \quad \beta_u = \frac{u_G}{u_L}$$

In addition, the sixth group (ratio of gas to liquid density) was suggested for high pressure conditions.

The experiments were performed in two different column diameters. One column was an industrially operated cold flow unit of diameter 0.91 m. The gas, liquid, and solid phases used in this column were hydrogen, kerosene, and ceramic particles, respectively. The laboratory scale column had a diameter of 0.0826 m. The gas, liquid, and solid phases used in this column are air, aqueous magnesium sulfate solution, and cylindrical alumina particles. The geometric similarity between the particles was maintained by using the particles of the same length to diameter ratio, i.e., 2.6. The gas holdup was measured based on the bed expansion and pressure measurements at various axial locations. In the case of pressure measurements, it was assumed that solids holdup is independent of height throughout the column in the expanded bed.

The effect of gas and liquid Reynolds number appears to be consistent with the previous literature findings (Figure 6).

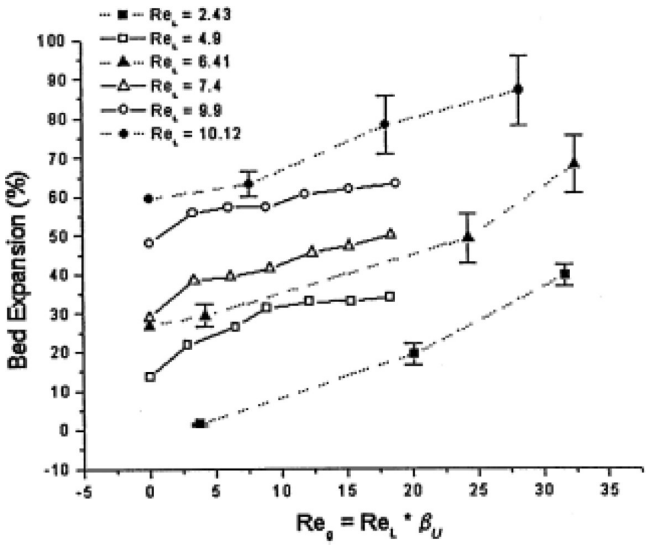


Figure 6. Bed expansion versus Reynolds number in two columns. Filled-in point: 0.0826 m diameter column, open points: 0.91 m diameter column. Reprinted with permission from ref 26. Copyright 1999 Elsevier Ltd.

Although the exact operating conditions could not be matched for this set of experiments, the trends obtained in both columns are in line with the expectations. Whenever the dimensionless numbers were close, the overall gas holdup was found to be close to each other. In addition, the gas holdup in the freeboard region was measured in both columns. The gas holdups in these two systems were found to agree well.

Later, Macchi et al.[27] tested the scaling approach proposed by Safounik et al.[26] for three phase fluidized beds based on

dynamic and geometric similitude with five dimensionless groups. The similarity of overall gas holdup in two systems was investigated by maintaining these numbers the same by using a monocomponent liquid in one system and a multicomponent liquid in the other one. Essentially, they tested the ability of Safonuik et al.[26] approach to capture coalescing behavior.

Macchi et al.[27] used air, 55% wt glycerol solution, and spherical borosilicate particles as gas, liquid, and solid phases, respectively, in one system (System I). While in another system, air, silicone oil, and spherical porous silica−alumina particles were gas, liquid, and solid phases, respectively (System II). The experiments with both systems were performed in an acrylic column of 0.127 m diameter and 2.58 m long. The overall gas holdup was measured using a differential pressure drop. They found negligible axial variation of phase holdups in the bed and freeboard region at all conditions except the highest gas and liquid velocities. The set of five dimensionless numbers proposed by Safonuik et al.[26] was maintained the same as in the 0.127 m diameter column.

Figure 7 shows the comparison between the gas holdup in two systems at various gas and liquid Reynolds number. Both

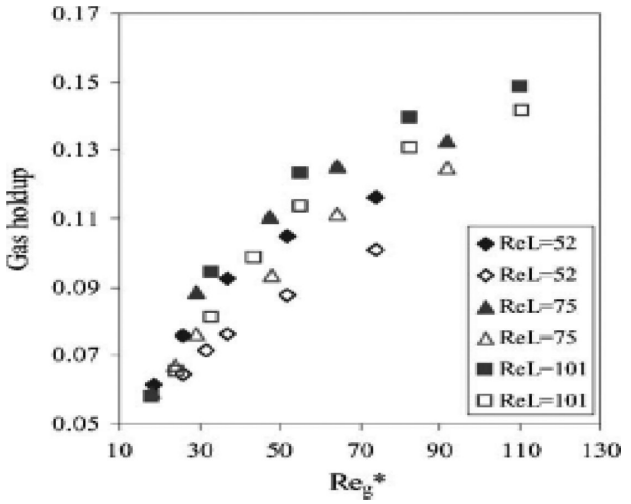


Figure 7. Gas holdup in two systems: filled-in points − System I, open points − System II. Reprinted with permission from ref 27. Copyright 2001 Elsevier Ltd.

the systems show the same trends. However, the gas holdup in System II was slightly lower than in System I. The statistical analysis concludes that the gas holdups in the two systems are within 11% root mean standard deviations. Hence, it does provide a reasonable basis for hydrodynamic similarity in the two systems. However, the pressure fluctuations studies revealed that the power spectra in the two systems are different (Figure 8).

Macchi et al.[27] concluded that the mismatch is likely due to the difference between coalescence in monocomponent and multicomponent liquid that results in different bubble distribution. In the pure liquids, gas holdup decreases with an increase in liquid phase viscosity, while in liquid mixtures it goes through maxima. An initial increase in gas holdup with an increase in viscosity in liquid mixtures is due to less coalescence rate than that of pure liquids (Wilkinson[29]). Hence, they suggested that more than five dimensionless groups are needed to fully characterize the system.

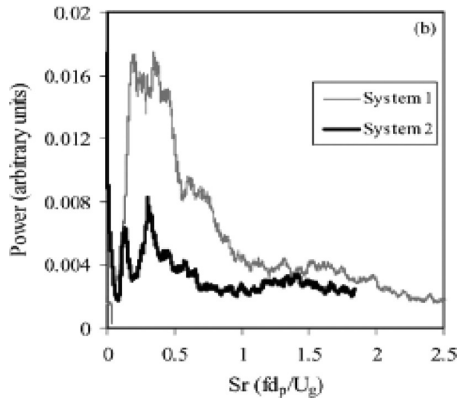


Figure 8. Power spectra in the two systems at operating conditions with the same dimensionless numbers. Reprinted with permission from ref 27. Copyright 2001 Elsevier Ltd.

2.6. Krishna et al.[30] Krishna et al.[30] developed a strategy for scaling laboratory reactors to commercial ones based on developing empirical correlations of various hydrodynamic parameters to be incorporated into Computational Fluid Dynamics (CFD) to design a commercial size bubble column reactor. The developed procedure can be described as shown in Figure 9.

They have performed dynamic gas disengagement (DGD) experiments in columns with different diameters i.e. 10, 19, and 38 cm. using an air−paraffin oil−silica system at ambient conditions. Based on DGD results, they have analyzed the effect of different operating and design parameters on 'small' and 'large' bubble holdups. The bubble column was divided into 'dense' and 'dilute' phases similar to gas−solid fluidized beds. The 'dilute' phase consists of the large bubbles, while the 'dense' phase consists of liquid phase along with solid particles and small bubbles. The 'dense' phase holdup was found to be independent of column diameter but varied with solids loading as shown in Figure 10. 'Large' bubble rise velocity was found to be the function of column diameter. Using these experimental results and their earlier findings, Krishna et al.[31] proposed various phenomenological models/correlations to predict bubble diameter, rise velocity, and holdups.

The dense phase holdup and small bubble rise velocity were correlated as follows

$$\varepsilon_{df} = \varepsilon_{df_0}\left[1 - \frac{0.7}{\varepsilon_{df_0}}\upsilon_S\right] \tag{21}$$

$$V_{small} = V_{small_0}\left[1 + \frac{0.8}{V_{small_0}}\upsilon_S\right] \tag{22}$$

where $\varepsilon_{df_0} = 0.27$, and $V_{small_0} = 0.095$ for paraffin oil slurries (corresponding to $\upsilon_S = 0$).

The superficial gas velocity through dense phase can be calculated as

$$u_{df} = V_{small}\varepsilon_{df} \tag{23}$$

To calculate the rise of single large bubble in an infinite volume of liquid, Krishna et al.[31] modified the classical Davis-Taylor relation and proposed the following correlation

$$V_{b,single} = 0.71(\mathrm{SF})(\mathrm{AF})\sqrt{gd_b} \tag{24}$$

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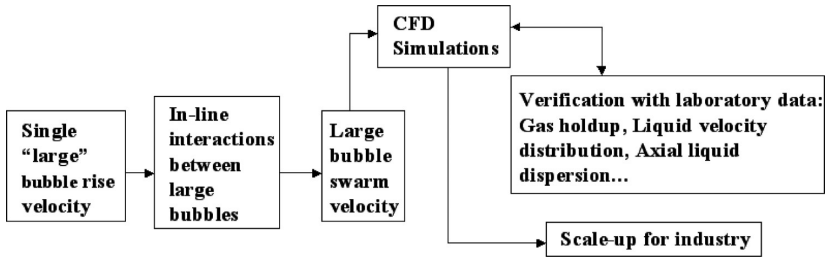


Figure 9. Scale-up strategy for bubble column reactors. Reproduced from ref 30.

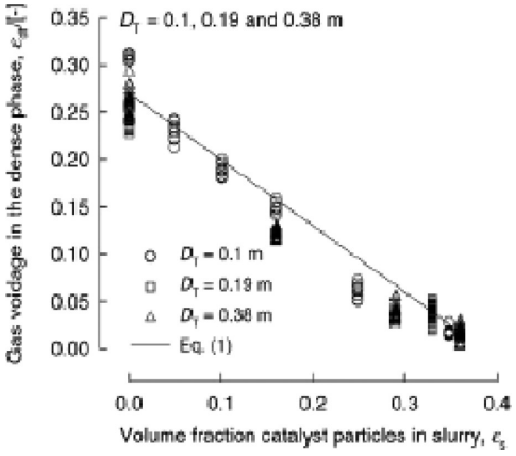


Figure 10. Effect of column diameter and slurry concentration on dense phase gas holdup. Reprinted with permission from ref 30. Copyright 2001 Elsevier Ltd.

The scale correction factor, SF, which accounts for influence of column diameter was given as

$$\mathrm{SF} = \begin{cases} 1 & \text{for } d_b/D < 0.125 \\ 1.1\exp(-d_b/D) & \text{for } 0.125 < d_b/D < 0.6 \\ 0.496\sqrt{D/d_b} & \text{for } d_b/D > 0.6 \end{cases} \quad (25)$$

The acceleration factor, AF, that accounts for an increase in large bubble rise velocity over that of a single bubble rise velocity was given as

$$\mathrm{AF} = 2.25 + 4.09(u_G - u_{df}) \text{ for air–Tellus oil} \quad (26)$$

The following empirical correlation was proposed for the average bubble size in the swarm

$$d_b = 0.069(u_G - u_{df})^{0.376} \quad (27)$$

These developed correlations were incorporated into CFD using Eulerian description of fluid phases. The authors adopted a two-phase model in this case assuming pseudohomogeneity. In the experimental studies, they found that air–paraffin oil–silica particles with 35% vol. solids loading shows gas holdup curve close to that of air–Tellus oil (Figure 11). Interestingly, the pseudoviscosity of the slurry is close to the viscosity of Tellus oil.

Hence, slurry phase was modeled in their simulation as pseudoliquid phase with Tellus oil properties. No turbulence model has been used to calculate velocity field inside large bubbles. The overall gas holdup values predicted from CFD were compared to experimental values for three different column diameters and found to be in a good agreement (Figure 11). In addition, they performed experiments to measure liquid velocity profile in an air–water and air–Tellus oil system using Pavlov tube. The reasonable agreement has been found in experiments and simulations. Based on this confirmation, Krishna et al.[32] simulated the behavior of column diameters up to 6 m and found that overall gas holdup decreases and centerline velocity increases with an increase in diameter. The strong reduction in large bubble holdup was also observed with an increase in column diameter. However, no comparison of CFD predictions with experimental data was shown at these conditions. The simulated centerline velocities showed a good agreement with Riquart[33] correlation. Additionally, to predict liquid phase backmixing, a correlation proposed by the authors was recommended. Based on these observations, they suggested the Eulerian simulation model developed in their study to be a valuable tool to predict the hydrodynamics of bubble column reactors at commercial scale.

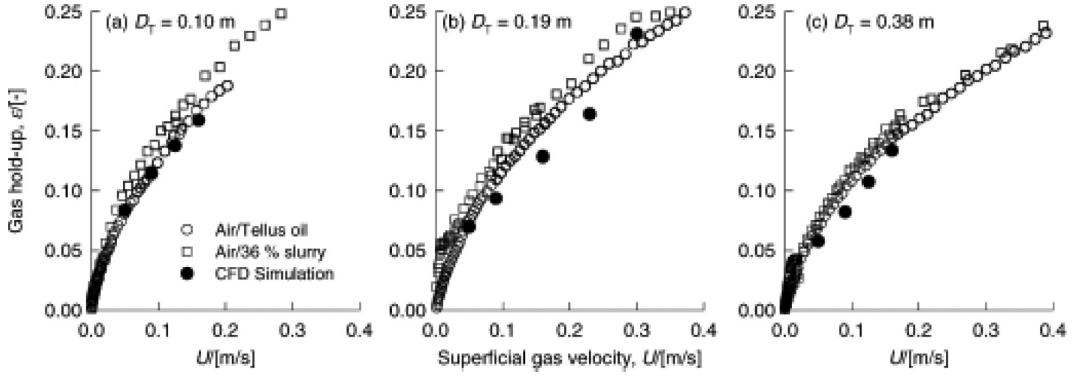


Figure 11. Gas holdup in air–paraffin oil slurry and air–Tellus oil in three different diameter columns. Reprinted with permission from ref 30. Copyright 2001 Elsevier Ltd.

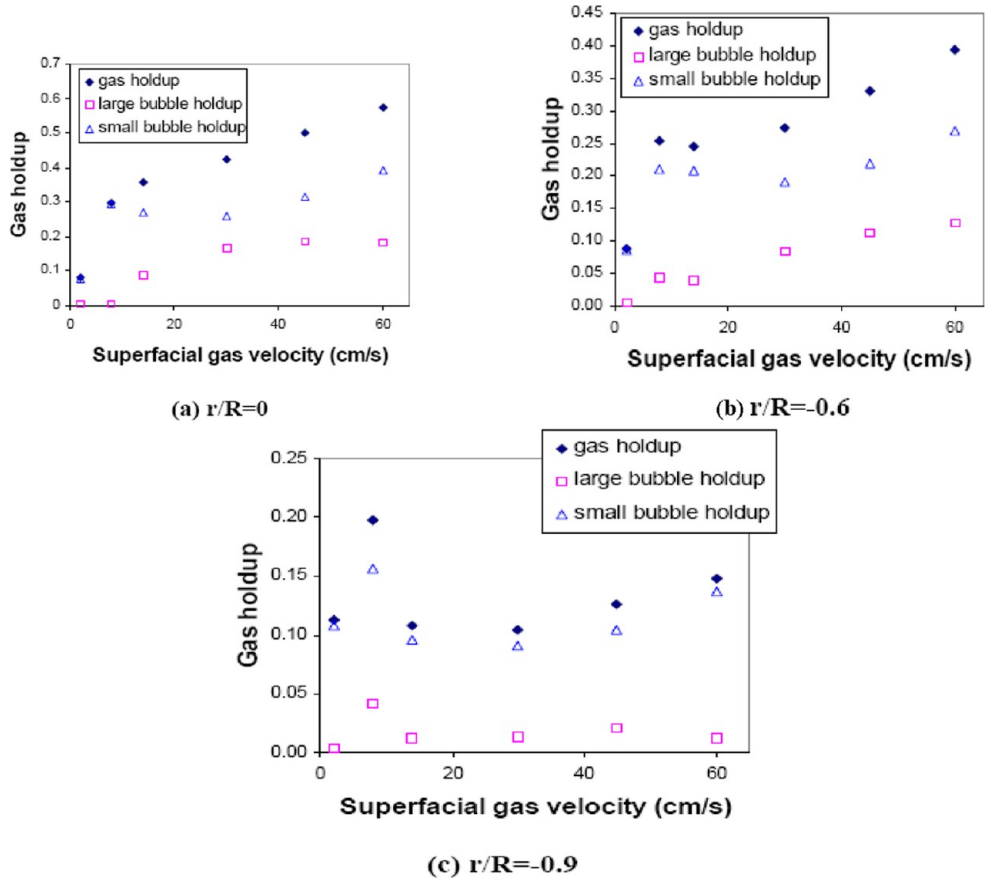


Figure 12. Variation of overall, large, and small bubble gas holdup with superficial gas velocities at ambient pressure. Reprinted with permission from ref 35. Copyright 2004 Washington University.

The correlations for prediction of bubble diameter, rise velocity, and holdups (eqs 21-27) were developed using Dynamic Gas Disengagement (DGD) studies with an assumption that the small bubble holdup remains constant in a heterogeneous flow regime. However, Jordan et al.[34] performed DGD experiments at atmospheric as well as high pressure and analyzed the obtained results using independent disengagement, sequential disengagement, and constant slip velocity assumptions. In contradiction to previous findings, they observed that the small bubbles holdup increases with an increase in superficial gas velocity. The experiments performed in 10 cm diameter column using butanol showed 100% and 67% increase in small bubble holdup from 5 to 20 cm/s using an independent (and constant slip) and sequential disengagement technique, respectively. In addition, Xue[35] studied bubble dynamics in an air−water system at atmospheric and high pressure using a four point optical probe. As shown in Figure 12, at dimensionless radius locations of 0, 0.6, and 0.9, an increase in large and small bubble population with an increase in superficial gas velocity was observed. Lee et al.[36] studied the assumptions of the DGD technique in a two-dimensional bubble column and a slurry bubble column with the help of Particle Image Velocimetry (PIV). They found that the assumptions in DGD that there is no bubble−bubble coalescence and breakup during disengagement and disengagement of bubble classes are not affected by each other and are not valid particularly in a heterogeneous regime. However, such limitations shown by Lee et al.[36] have to be checked in a three-dimensional bubble column. Hence the correlations developed using DGD need to be revisited.

In addition, CFD data shown in this work were validated in laboratory scale reactors and were extrapolated to design a 6 m reactor.

2.7. van Baten et al.[37] van Baten et al.[37] proposed a new method for scale-up of bubble columns where they have demonstrated how the hydrodynamic behavior in a 1 m diameter column can be estimated using the experimental data in a 5.1 cm diameter column. They developed a procedure to study the flow behavior in commercial scale bubble columns based on CFD in Eulerian framework by considering only a momentum exchange term. A method was devised to replace the need for a closure equation that may mar the predictions of CFD. They calculated a drag coefficient and a bubble diameter utilizing only overall gas holdup data in a small diameter column (5.1 cm).

Van Baten et al.[37] performed experiments in a 5.1 cm diameter column using an air−ethanol−cobalt (Co) system at various solids loading i.e. 4.4, 8.5, 12.5, and 16.4% wt. Insignificant change in the overall gas holdup was found between 12.5 and 16.4% wt solids loading system as shown in Figure 13. The CFD model was developed in an Eulerian framework for operating conditions with 16.4% wt solids loading assuming pseudohomogeneity. Based on the work of Sanyal et al.[38] and Sokolichin and Eigenberger,[39] the added mass and lift force terms were neglected. The momentum exchange term was written following the work of Pan et al.[40] as

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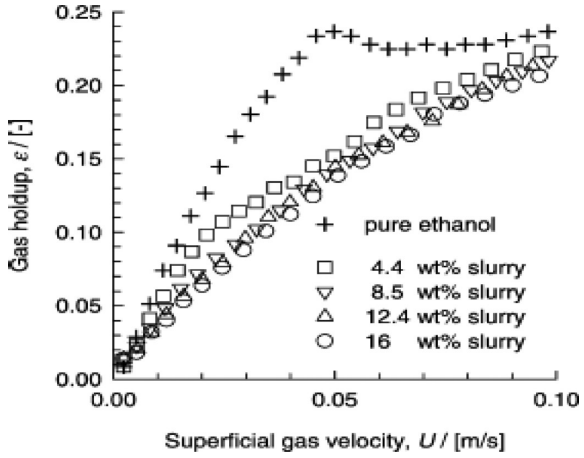


Figure 13. Gas holdup curves in air−ethanol−Co system at various solids loading in 5 cm diameter column. Reprinted with permission from ref 37. Copyright 2003 Elsevier B.V.

$$M_{L,G} = \left[\frac{3}{4}\frac{C_D}{d_b}\rho_L\right]\varepsilon_G\varepsilon_L(u_G - u_L)|u_G - u_L| \tag{28}$$

For a bubble swarm rising in a gravitational field, drag force balances the difference between weight and buoyancy so that the bracketed term in eq 28 can be substituted as

$$\frac{3}{4}\frac{C_D}{d_b}\rho_L = (\rho_L - \rho_G)g\frac{1}{V_{b,0}^2} \tag{29}$$

where $V_{b,0}$ is the rise velocity of bubble swarms at low superficial gas velocities.

Equation 29 clearly shows that the knowledge of the bubble rise velocity in an infinite volume of liquid is the only unknown parameter in the model. The authors proposed the following method to calculate the bubble rise velocity:

i. Perform overall gas holdup experiments in small diameter column (5.1 cm, in the current case).

ii. Calculate average bubble swarm velocity, V_b as

$$V_b = u_G/\varepsilon_G \tag{30}$$

iii. Plot average bubble swarm velocity, V_b versus superficial gas velocity (Figure 14).

iv. Extrapolate V_b data to low superficial gas velocity as shown in Figure 14.

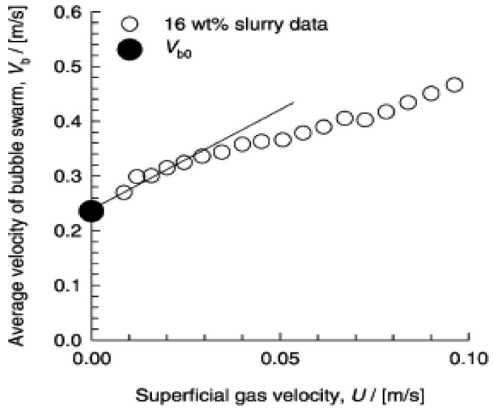


Figure 14. Average bubble-swarm velocity in air−ethanol−Co. Reprinted with permission from ref 37. Copyright 2003 Elsevier B.V.

v. The value of bubble swarm velocity at vanishingly small superficial gas velocity is $V_{b,0}$. Substitute this value in eq 29.

This way, one does not need to know the bubble size in the momentum exchange term to predict the drag coefficient. The overall gas holdups predicted from CFD along with the developed approach was compared to the one obtained from experiments and were found to be in a good agreement. Based on this comparison, van Baten et al.[37] estimated the behavior of a 1 m diameter bubble column.

Later, van Baten and Krishna[25] demonstrated their approach for different column diameters. As shown in Figure 15, for vanishingly small velocity, practically the same bubble velocity was observed for all three different diameter columns. Utilizing the developed approach, van Baten and Krishna[25] found good agreement between the overall gas holdup obtained from experiments and CFD simulations for three different diameter columns (10, 19, and 38 cm). They studied the hydrodynamic behavior of 1 to 10 m diameter bubble column by extending the same approach.

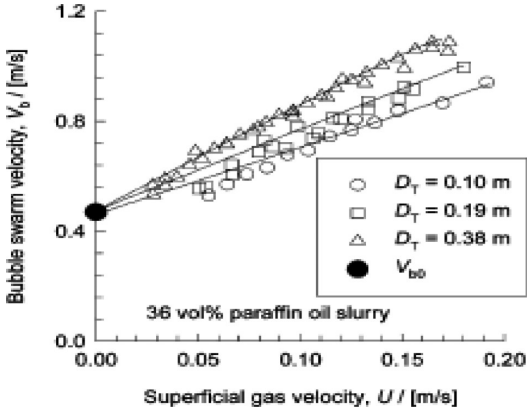


Figure 15. Average bubble-swarm velocity in air−paraffin oil slurry in three different diameter columns. Reprinted with permission from 25. Copyright 2004 American Chemical Society.

The authors proposed a simple approach to study and design the flow behavior in large diameter bubble columns, as it does not require *a priori* knowledge of bubble diameter and also it does not need drag force coefficient closure. The only knowledge needed is simple overall gas holdup experimental data in a laboratory scale column. However, the validation of CFD simulation results with experiments in large diameter columns need to be established. In addition, this method can be applicable only in cases where the bubble size does not increase significantly with superficial gas velocity. Vermeer and Krishna[41] showed that the ratio of k_La/ε_G is constant (~0.5) in a churn-turbulent flow regime in the air−Turpentine 5 system. Godbole et al.[42] reached the similar conclusions in air−water and air−Soltrol 130 systems. Vandu and Krishna[43] also found the similar results in three different diameter columns (10, 15, and 38 cm) using an air−water system (Figure 16) and concluded that the constant value of ratio of k_La/ε_G is due to the fact that the effective bubble diameter is independent of superficial gas velocity. Based on these findings, van Baten and Krishna[25] rationalized the application of the developed approach for scale-up of bubble columns. Chaumat et al.[44] developed a gas tracer technique to study Residence Time Distribution (RTD) and mass transfer in 0.2 m diameter and 1.6 m long bubble column. Various gases (nitrogen, carbon

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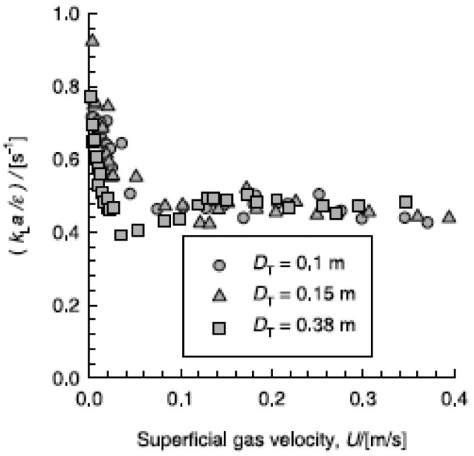


Figure 16. Variation of k_La/ε_G with superficial gas velocity in three different diameter columns using an air−water system. Reprinted with permission from ref 43. Copyright 2004 Elsevier B.V.

dioxide) and liquids (water, cyclohexane) were used at ambient pressure. However, they concluded that the relation between mass transfer and hydrodynamics appears to be more complex than simplified linear relation (Figure 17). In light of such conflicting results, such an approach may need to be revisited.

2.8. Zhang and Zhao.[45] Zhang and Zhao[45] proposed a scale-up strategy for low temperature methanol synthesis in a continuous slurry bubble column reactor. It involved studying hydrodynamics in cold flow units, catalyst performance evaluation in an autoclave, and process investigation in pilot-scale continuous slurry bubble column reactor.

The study was performed over the following three steps:

1) Hydrodynamics in a cold flow unit with different column structure using the three-phase systems

2) Catalyst preparation and performance evaluation in an autoclave

3) Process exploration in continuous slurry bubble column reactor with a capacity of 2 tons per year.

Hydrodynamics experiments were performed in three different (diameter = 0.042, 0.05, and 0.1 m) continuous slurry bubble column reactors and tapered bubble column reactor. The methanol synthesis was characterized by gas volumetric contraction; hence the tapered bubble column was explored. It has a conical and cylindrical section such that the diameter of the conical section increases from 0.1 m at the bottom to 0.2 at the top. Above the conical section, a cylindrical section of 0.2 m diameter was placed. The different gas−liquid (air−water, nitrogen-actual liquid medium) and gas−liquid−solids (air−water−quartz sand, nitrogen-actual liquid medium-Cu based catalyst powder) systems were studied. The actual liquid medium was sodium methoxide and xylene. Hydrodynamic parameters studied were overall gas holdup, gas holdup axial profile, solids holdup axial profile [using Sedimentation-Dispersion Model (SDM)], and flow regime transition.

Catalyst performance evaluation was accomplished by performing experiments in a 1-L autoclave where the catalyst preparation method was studied. In addition, the effect of liquid phase medium, temperature, and pressure was evaluated. Based on these experiments, xylene was chosen as liquid phase medium and temperature was chosen to be lower than 120 °C.

In the process development part, preliminary experiments were performed in a slurry-batch bubble column reactor of 50 mm × 5 mm diameter and 4500 mm height. They concluded that reaction and separation of liquid products is essential for a commercial methanol synthesis process. Hence, a continuous slurry bubble column system that consists of a slurry bubble column reactor, a separator, and a slurry recycle unit was recommended for synergy of coupling of two-step methanol synthesis. A continuous circulating slurry bubble column reactor with capacity of 2 tons per year was manufactured. The syngas composition was maintained at CO/H_2 = 2. The operation was carried out for 100 h. The catalyst started deactivating after 50 h. The effect of temperature and pressure was studied. They the observed uniform catalyst concentration profile at the methanol synthesis conditions. The experiments were also performed in a 1-L autoclave at the same conditions as that of theh circulating slurry bubble column reactor. The comparison of the results in these two systems showed that the scale-up efficiency of the circulating bubble column reactor was about 80% of that in the autoclave.

The authors provided a comprehensive roadmap for development of the low-temperature methanol synthesis in the circulating slurry bubble column reactor. The strategy tied flow behavior and catalysis studies with that of process engineering. However, no guidelines were provided regarding hydrodynamic similarity in cold and hot units.

2.9. Shaikh[15] and Shaikh and Al-Dahhan.[16] Shaikh[15] and Shaikh and Al-Dahhan[16] proposed a new hypothesis for hydrodynamic similarity in bubble column reactors. They evaluated the proposed hypothesis utilizing advanced diagnostic techniques such as gamma-ray Computed Tomography (CT) and Computer Automated Radioactive Particle Tracking (CARPT). The details of these techniques are available elsewhere (Degaleesan;[17] Kumar[20]) and hence will not be repeated here. Essentially, CT provides time-averaged cross-sectional distribution of phase holdups and CARPT provides

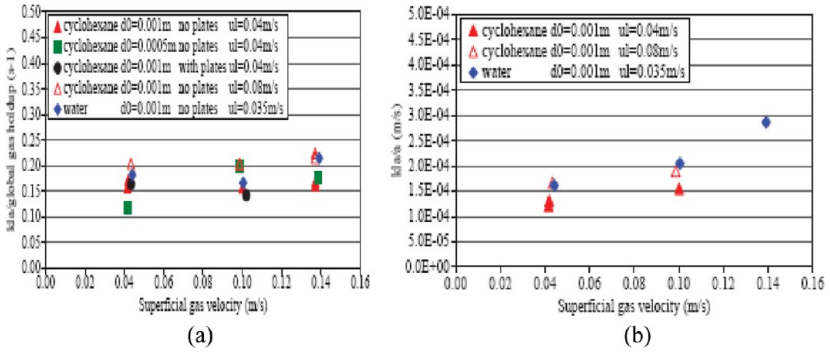


Figure 17. Relation between mass transfer coefficient and hydrodynamics. Reprinted with permission from 44. Copyright 2005 Elsevier Ltd.

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instantaneous liquid velocity and its radial profile as well as a turbulent parameters profile.

The combination of two studies was the basis of a new hypothesis for their studies. These studies were detailed analysis of reported scale-up procedures of the bubble column and the results from Kemoun et al.[46] and Ong.[47] Based on the detailed analysis of reported hypotheses and scale-up procedures, they found that most of these procedures inherently used similarity of global parameters such as overall gas holdup and/or mass transfer coefficient. In addition, they combined the results from the work of Kemoun et al.[46] and Ong[47] as shown in Figure 18. It shows the gas holdup radial profile obtained using CT in an air−water system (Ong;[47] Kemoun et al.[46]) at different operating conditions in a churn-turbulent flow regime with the overall gas holdup of 0.41. Although these systems have similar overall gas holdups, they have different gas holdup radial profiles that will clearly lead to different flow patterns and mixing intensities.

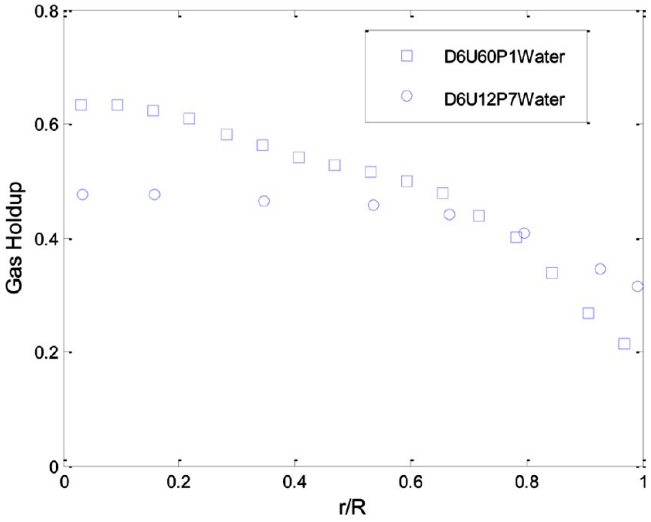


Figure 18. Comparison of gas holdup radial profile in a 6" column using an air−water system at two different operating conditions [D6U12P7Water: 7 bar, 12 cm/s, an air−water (Kemoun et al.[46]); D6U60P1Water: 1 bar, 60 cm/s, and an air−water (Ong[47])] with similar overall gas holdups (~0.41).

The conclusions of Macchi et al.[28] and Figure 18 suggested that the two systems can have similar overall gas holdups but different flow patterns and mixing intensities. This indicates that two systems can be globally similar in nature but have different local hydrodynamics. Hence, similarity based only on overall gas holdup does not appear sufficient.

Based on this, Shaikh[15] and Shaikh and Al-Dahhan[16] proposed a new hypothesis as follows:

> *"Overall gas holdup and its time-averaged radial profile or cross-sectional distribution should be the same for two reactors to be dynamically similar."*

The proposed hypothesis was evaluated using similarity and mismatch conditions. Similarity conditions are the ones that have similar overall gas holdup as well as gas holdup radial profile. While mismatch conditions are the ones that have similar overall gas holdup and mismatch gas holdup radial profiles. These conditions were identified using overall gas holdup measurement and gamma-ray CT. Later, CARPT experiments were performed at identified similarity/mismatch conditions to measure the detailed hydrodynamics (radial profile of liquid axial velocity and turbulent parameters) in order to assess the proposed hypothesis.

They demonstrated that to be hydrodynamically similar it is necessary to have similarity of both overall gas holdup and gas holdup radial profile. Figure 19 shows one such result at a similarity condition. Figure 19a shows similar gas holdup profiles at the same overall gas holdup (0.35) which results in a similar liquid axial velocity profile (Figure 19b) and turbulent kinetics energy (TKE) profile (Figure 19c). Thus, the similarity conditions showed that similar overall gas holdup and gas holdup radial profiles resulted in close liquid recirculation and mixing intensity in both systems. Figure 20 shows experimental results at one of the mismatch conditions. Figure 20a shows mismatch gas holdup profiles at the same overall gas holdup (0.35) which results in mismatched liquid axial velocity profile (Figure 20b) and turbulent kinetics energy (TKE) profile (Figure 20c). Thus, the mismatch experiments had similar overall gas holdups but mismatched profiles and resulted in varied liquid recirculation and mixing intensity. This clearly shows that maintaining similar overall gas holdup alone can lead to different recirculation and mixing, if gas holdup radial profiles are not matched.

They also showed that the condition that two systems must operate in the same flow regime to be hydrodynamically similar is ***necessary*** but not ***sufficient***. It showed the importance of matching gas holdup radial profiles or cross-sectional distributions in two systems, even if both the systems operate in the same flow regime. Using the available data of Ong,[47] Shaikh and Al-Dahhan,[49] and Shaikh,[15] they showed that traditionally used criterion for hydrodynamic similarity, based only on global parameter, can be specifically applicable if both the systems operate in bubbly flow.

In addition, dimensionless groups reported for hydrodynamic similarity were evaluated for available sets of similarity conditions. They concluded that within the range of studied experimental conditions, no consistent set of dimensionless group exhibit similarity and hence the application and limitations of dimensionless approach for bubble column reactors need to be investigated in detail.

Although it was out of scope of their work, Shaikh and Al-Dahhan[16] outlined a tentative procedure for extrapolating hydrodynamic behavior. It involved a combination of the proposed hypothesis and state-of-the-art correlations for needed hydrodynamic parameters developed using a wide range of operating and design parameters. These hydrodynamic parameters are overall gas holdup, radial profile of gas holdup and liquid axial velocity, and center-line axial velocity. The correlation developed for overall gas holdup based on Artificial Neural Network (ANN) by Shaikh and Al-Dahhan[50] is one such example.

The focus of their work was to propose and evaluate the hypothesis for hydrodynamic similarity using CT and CARPT. The development of reliable hydrodynamic similarity criteria is a goal of any scale-up procedure. Although, the proposed hypothesis was successfully evaluated at different operating conditions and physical properties, it needs to be evaluated using different column diameters and have the potential to apply in various multiphase reactors.

2.10. Youssef.[51] Youssef[51] proposed a scale-up method based on the horizontal scale-up (i.e., scale-in-parallel or scale out) approach where the large scale unit consists of a multiplication of the small units and hence, geometry, flow pattern, and flow regime are kept the same. His method was

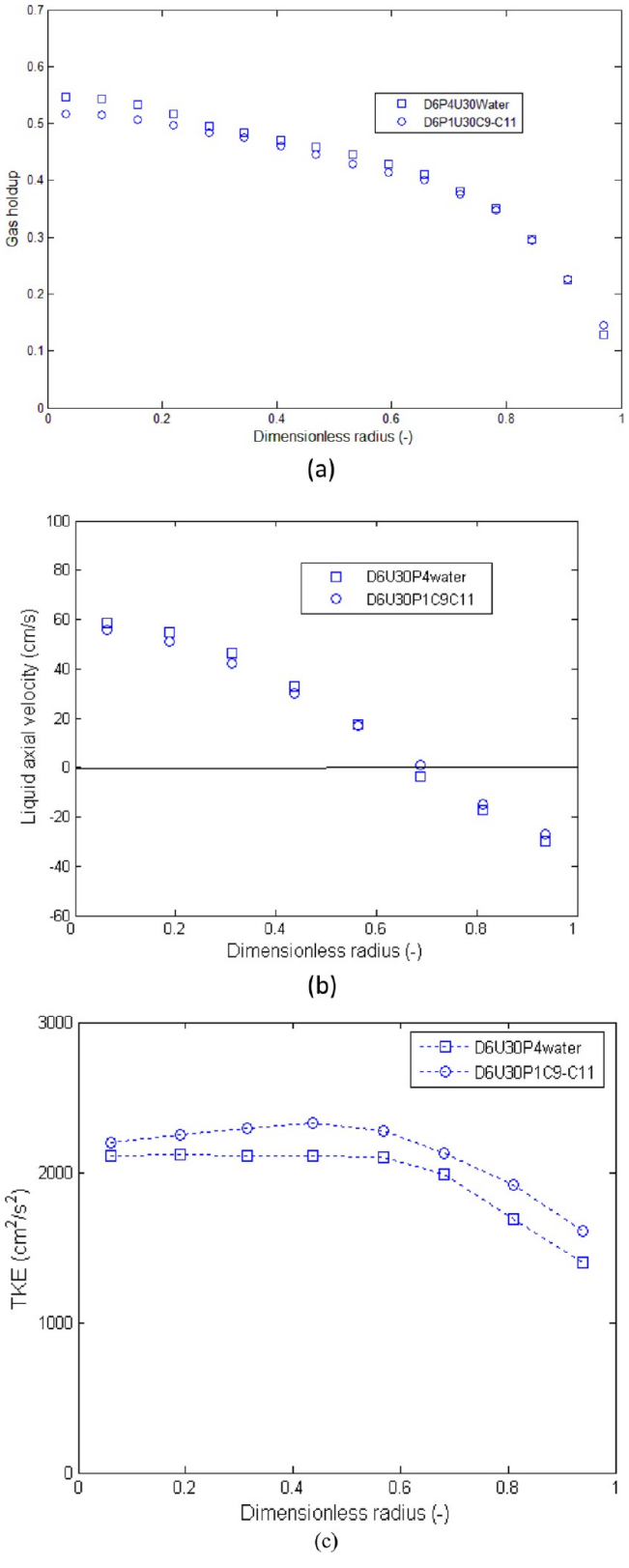


Figure 19. a) Gas holdup and b) axial liquid velocity c) TKE radial profile in a 0.162 m diameter stainless steel column [D6P1U30 C_9–C_{11} oil: 6 in. diameter column, 0.1 MPa, 30 cm/s (Han[48]), and an air–C_9–C_{11} fluid system, D6P4U30water: 6 in. diameter column, 0.4 MPa, 30 cm/s, and an air–water system] [overall gas holdup ~0.35] (Shaikh[15]).

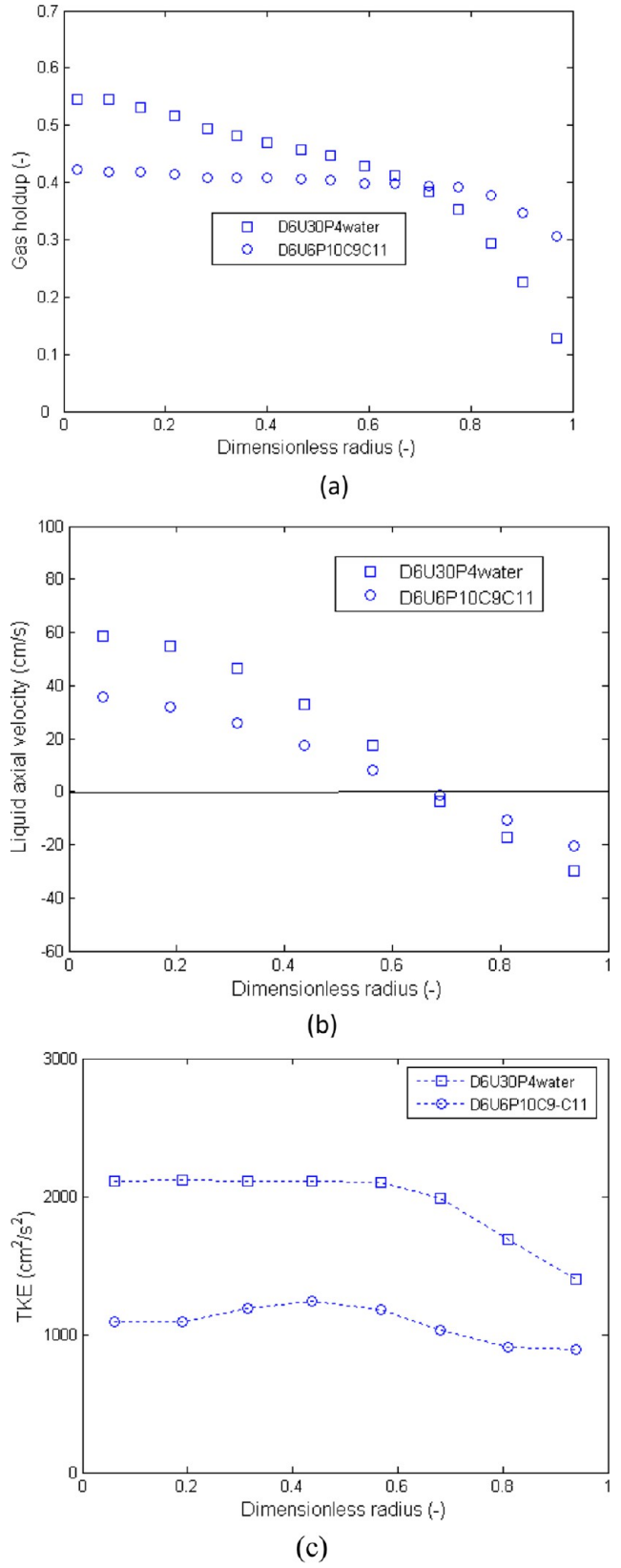


Figure 20. a) Gas holdup and b) axial liquid velocity c) TKE radial profile in a 0.162 m diameter stainless steel column (D6P4U30water: 6 in. diameter column, 0.4 MPa and 30 cm/s, an air–water; D6P4U16C_9–C_{11}: 6 in. diameter column, 0.4 MPa and 16 cm/s, air–C_9–C_{11}) [overall gas holdup ~0.35] (Shaikh[15]).

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based on the approach of Kolbel and Ackermann[6] in which the reactor space was subdivided using similar vertical shaft, and the flow of gas was uniformly distributed among the equally spaced compartments. The heat exchanging tubes were placed either inside the shaft or entirely or partly inside the spaces between the shafts. Such configuration was made to reduce the intensity of liquid recirculation encountered in bubble and slurry bubble columns. Along the same concept of compartmentalization, the recent Sasol patent (Steynberg et al.[52]) proposed a solution for the scale-up risk of the Fischer−Tropsch slurry bubble column reactor by creating zones to mimic the behavior of small reactor diameter. They used a group of discrete channels inside the reactor shell separated by heat transfer medium flow spaces. They claimed the feasibility of scaling process, minimizing microscale mixing patterns and improving heat transfer.

For relatively high exothermic reactions where bubble and slurry bubble column reactors are favored including Fischer−Tropsch synthesis, a large number of heat exchanging tubes that would cover a noticeable percent of the reactor cross sectional area is needed. For example, Fischer−Tropsch synthesis requires heat exchanging internals that would cover ~22−25% of the reactor cross sectional area. Accordingly, for these conditions Youssef[51] proposed an arrangement of the heat transfer tubes in a form of small bubble/slurry bubble columns inside the reactor shell. In this case, the reactor is compartmentalized in a form of small columns using heat transfer tubes. The question that has escalated is whether such a small column with a wall consists of tubes mimic the behavior of the same column size with a solid wall. Therefore, Youssef[51] studied the hydrodynamics of an air−water system in a 6-in. diameter bubble column with a wall formed by 1 in. diameter tubes placed inside an 18-in. diameter column using a 4-point optical probe that measured the radial profile of gas holdup, specific interfacial area, bubble cord length, and bubble velocity (upward and downward). The tube bundles were mounted 5 in. above the gas distributor. The results were compared with independent results obtained by Xue[35] and Wu[53] in a separate 6 in. diameter bubble column setup with a solid wall. A higher gas holdup was obtained at each corresponding radial location in the tubes bundles compared to those obtained in a solid wall column. The average relative difference of about 15% was reported between them, particularly at the radial locations within the central region of the columns. However, the shape of the radial profile of the gas holdup inside the tubes bundle column is about the same as that inside the solid wall column. The bubble cord length and bubble velocity distributions at the center of the tube bundle are close to that of the solid wall column at 45 cm/s superficial gas velocity with the tubes bundle mounted at about 5-in. above the gas distributor.

However, at the region close to the tube bundle there is a clear variation from that close to the solid wall. These variations could have been caused by the interaction between the inside and outside the tube bundle that represent the column of 6-in. diameter inside the 18 in. diameter column. Although Youssef[51] claimed that such methodology for scale-up seems feasible, he suggested that further work is required.

It is obvious that this method has many uncertainties and technical difficulties associated with the vertical space between the bottom edge of the tubes and the gas sparger and when the configuration is duplicated inside a larger diameter column; there will be gaps between tube bundles that promote mixing and interactions between inside and outside the tubes. To avoid the latter issue, hexagonal type arrangement of the tubes has been recommended (Kolbel and Ackermann;[6] Youssef[51]).

It is noteworthy that whether this method is feasible or not including the approach of Sasol (Steynberg et al.[52]), the methodology of Shaikh[15] discussed above (Shaikh and Al-Dahhan[16]) can be used to ensure hydrodynamics similarity whether in a column of tube bundle or solid wall and for both scale-in-parallel (horizontal scale-up) or vertical scale-up by increasing the size. This is because the gas dynamics and its radial profile dictate the hydrodynamics of the columns as demonstrated above (Shaikh and Al-Dahhan[16]).

3. REPORTED STATUS OF SCALE-UP IN INDUSTRY

The status of scale-up of bubble column reactors in industry is not widely reported in the literature for obvious proprietary reasons. Tarmy and Coulaloglou[54] discussed fundamental development and scale-up issues and related them through an example of the coal liquefaction process. The traditional approach for scale-up is shown in Figure 21 where kinetic

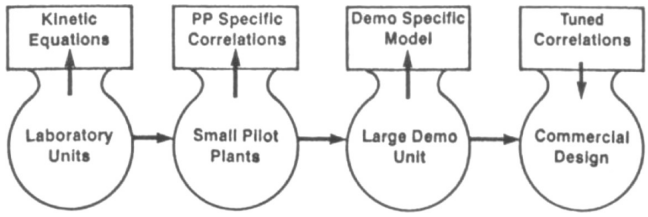


Figure 21. Traditional linear scale-up development. Reprinted with permission from ref 54. Copyright 1992 Elsevier Ltd.

constants obtained from the laboratory data assists in correlating the small pilot plant results. The pilot plant studies are then followed by demonstration in a relatively larger unit. They presented an evolving interactive scale-up strategy (Figure 22) that makes more efficient use of resources, time, and technology.

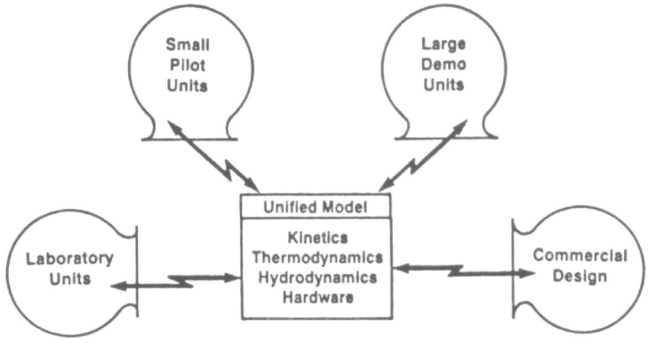


Figure 22. Evolving scale-up strategy. Reprinted with permission from ref 54. Copyright 1992 Elsevier Ltd.

Tarmy and Coulaloglou[54] mentioned that understanding of hydrodynamics and related issues to be the most critical element in development and scale-up of these reactors. The industrial example of scale-up demonstrated by them involves hydrodynamic studies based on overall phase holdups and liquid backmixing in terms of the Peclet number measured using pressure transducers and radioactive tracers. The highest superficial gas velocity in the pilot reactor at process conditions

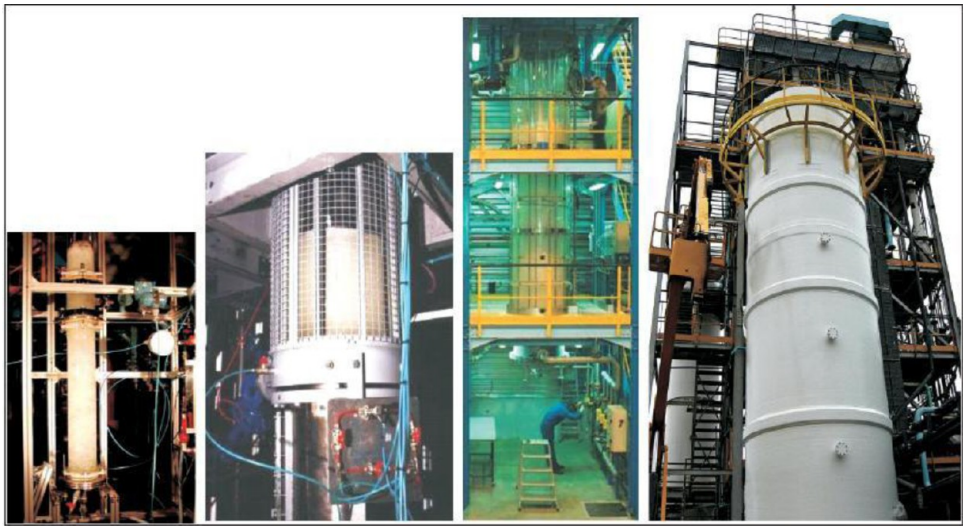


Figure 23. Eni/IFP hydrodynamic facilities. Reproduced from ref 59.

was 8 cm/s. Based on Tarmy et al.[55] studies, this condition appears to be in bubbly flow.

Espinoza et al.[56] discussed gas holdup prediction and scale-up for Sasol slurry phase reactor. Espinoza et al.[56] modified the gas distribution theory proposed by Toomey and Johnstone[57] for slurry bubble column reactors where it was divided into dense and dilute phases. The minimum fluidization velocity was replaced by transition velocity from homogeneous to heterogeneous flow regime. It was observed that dense phase holdup was not affected by the column geometry, while for small diameter columns the dilute phase holdup is determined by the column geometry. For large diameter columns (>1 m), the dilute phase holdup was constant for all the systems, while dense phase holdup was different depending upon gas and slurry properties. Using the modified two-phase theory, they developed an approach similar to the one proposed by Krishna and Ellenberger.[58] Espinoza et al.[56] mentioned that during scale-up of a 1 m diameter FT reactor to a 5 m diameter one, such an approach was not used. However, an importance of gas and slurry mixing characteristics in a slurry bubble column was emphasized.

Zennaro[59] has briefly discussed the development of Eni/IFP GTL FT and upgrading technology. Their strategy is based on three targets:

- development of an innovative FT/upgrading technology based on tailored catalysts, reactor design, and optimized product upgrading
- engineering studies of a fully integrated GTL complex
- minimization of scale-up risk by developing appropriate tools

As shown in Figure 23, experimental facilities of varied scale for hydrodynamic studies were developed to facilitate easy and low-risk scale-up of reactor technology. In addition, hydrodynamic as well as mass and heat transfer, thermodynamics, and kinetics have been described in detailed reactor model, validated up to 2500 bpd equivalent size.

4. REMARKS

An overview shows that there have been numerous attempts toward developing procedures for hydrodynamic similarity and subsequently for scale-up of bubble column reactors. Every proposed method has its own advantages and disadvantages; however, these studies with convergent objectives help in improving our understanding of scaling rules and approaches to be applied to bubble column reactors. Due to the complexity of flow in bubble columns, the scale-up of these reactors is still unanswered. The purely empirical approach for such a purpose should be strictly avoided. An ideal choice for designing commercial bubble column reactors would be fundamentally based CFD models. However, Rafique et al.[60] showed that interfacial closures are still an unresolved issue, and tuning the coefficients to a known field is still a state-of-the-art practice. Additionally, available closures do not account for the effect of turbulence. Chen[61] implemented the Population Balance approach where the bubble population balance equation was solved simultaneously with a solution of flow field. In a churn-turbulent flow regime, the breakup rate was enhanced 10 times the predicted one to match experimental flow field results. However, by increasing the break up rate 10 times for all his simulations, he found a good agreement between model predictions and experimental data in different air−liquid systems. This shows that, even after incorporating detailed *science*, there are still unresolved issues. Hence, the scale-up of bubble columns, like other multiphase reactors, is still more *art* than the *science*. However, an application of *art* needs a thorough understanding of prevailing hydrodynamic phenomena. Therefore, in the absence of fully evaluated CFD, a reliable similarity rule needs to be developed based on a phenomenological approach.

Most of the reported scale-up procedures so far utilize the similarity of a global parameter such as overall gas holdup in two columns for hydrodynamic similarity. Such similarity based on global parameters is not surprising because over the years bubble column hydrodynamics have been quantified mostly based on the global parameters such as overall gas holdup and mass transfer coefficient. However, as shown by Shaikh[15] and Shaikh and Al-Dahhan,[16] the generalization of hydrodynamic similarity based on an overall gas holdup can be fatal and needs to be exercised with prudence.

The ultimate hope for design and scale-up of bubble columns remains on fully validated and verified CFD. This hope turning into reality depends on a number of issues related to CFD. In this regard, Grace and Taghipour[62] provided a comprehensive outline and comments. On an experimental front, the

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researchers have improved measurement techniques from simple visual observation to advanced diagnostic techniques such as PIV, CARPT, CT, etc. These improved techniques provide us with a better understanding of prevailing phenomena than before. However, the combination of an experimentally evaluated scaling rule based on a phenomenological approach and CFD would provide a promising avenue for scale-up of bubble column reactors and needs to be explored.

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Notes

The authors declare no competing financial interest.

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NOMENCLATURE

C_D	drag coefficient, dimensionless
C_V	volumetric solids loading, dimensionless
D	column diameter, m
d_b	bubble diameter, m
d_p	particle diameter, m
D_G	gas dispersion coefficient, $m^2 \cdot s^{-1}$
D_L	liquid dispersion coefficient, $m^2 \cdot s^{-1}$
D_{rr}	radial eddy diffusivity, $m^2 \cdot s^{-1}$
D_{zz}	axial eddy diffusivity, $m^2 \cdot s^{-1}$
Eo	Etovos number, dimensionless
g	gravity constant, $m \cdot s^{-2}$
k_0	pseudo-first-order rate constant, s^{-1}
k_La	volumetric mass transfer coefficient, s^{-1}
L	column length, m
$M_{L,G}$	interphase momentum exchange term, $N \cdot m^{-3}$
Mo	Morton number, dimensionless
r	radial location in the column, m
Re	Reynolds number, dimensionless
U	superficial gas velocity, $m \cdot s^{-1}$ [in Figures 13, 14, 15, and 16]
U_G	superficial gas velocity, $m \cdot s^{-1}$
U_{Ge}	equivalent superficial gas velocity (eq 10), $m \cdot s^{-1}$
U_{Gtrans}	superficial gas velocity at flow regime transition, $m \cdot s^{-1}$
U_L	superficial liquid velocity, $m \cdot s^{-1}$
$U_{l,b}$	large bubble rise velocity, $m \cdot s^{-1}$
u_{df}	bubble velocity in dense phase, $m \cdot s^{-1}$
u_{max}	rise velocity of maximum stable bubble size, $m \cdot s^{-1}$
u_{rec}	liquid mean recirculation velocity, $m \cdot s^{-1}$
$U_{s,b}$	small bubble rise velocity, $m \cdot s^{-1}$
u_z	liquid axial velocity, $m \cdot s^{-1}$
$V_{b,single}$	single large bubble rise velocity, $m \cdot s^{-1}$
V_b	bubble swarm velocity, $m \cdot s^{-1}$
$V_{B\infty}$	terminal bubble rise velocity, $m \cdot s^{-1}$
$V_{b,0}$	bubble rise velocity at vanishingly small velocity, $m \cdot s^{-1}$
V_L	axial liquid velocity, $m \cdot s^{-1}$
V_{small}	small bubble velocity, $m \cdot s^{-1}$

Greek letters

ε_{df}	gas holdup in the dense phase, dimensionless
ε_{df_0}	gas holdup in the dense phase corresponding to no solids holdup, dimensionless
$\varepsilon_{G_{trans}}$	overall gas hold up at transition point, dimensionless
ε_G	overall gas hold up, dimensionless
ε_L	overall liquid hold up, dimensionless
ε_s	overall solids hold up, dimensionless
ξ	constant in eq 18, dimensionless
α	parameter in eq 18, dimensionless
ϕ	ratio of radial location to column radius, dimensionless
ϕ^*	radial location at inversion point, dimensionless
β_d	ratio of particle to liquid density, dimensionless
ρ_G	gas phase density, kg m^{-3}
β_i	dimensionless parameter in eq 17
ρ_L	liquid phase density, kg m^{-3}
ρ_P	particle density, kg m^{-3}
σ_L	liquid surface tension, N m^{-1}
μ_L	liquid viscosity, kg m^{-1} s^{-1}
ν_S	solids loading, dimensionless
ρ_{SL}	slurry phase density, kg m^{-3}
β_u	ratio of superficial gas to liquid velocity, dimensionless
β	parameter in eq 18, dimensionless

Abbrevations

AF	acceleration factor
ANN	artifical neural network
CARPT	computer automated radioactive particle tracking
CFD	computational fluid dynamics
CT	computed tomography
DGD	dynamic gas disengagement
FT	Fischer−Tropsch
GTL	gas-to-liquids
LDA	laser doppler anemometry
LPMeOH	liquid phase methanol synthesis
PBM	population balance model
PIV	particle image velocimetry
SF	scale factor

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