HYDRODYNAMICS CHARACTERISTICS AND GAS-LIQUID MASS TRANSFER IN A THREE PHASE FLUIDIZED BED REACTOR

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Abstract. This paper presents the experimental characterization of hydrodynamics and gas-liquid mass transfer in a three-phase fluidized bed containing polystyrene and nylon particles. The influence of gas and liquid velocities on phase holdups and volumetric gas-liquid mass transfer coefficient was investigated for flow conditions similar to those applied in biotechnological process. The phase holdups were obtained by the pressure profile technique. The volumetric gas-liquid mass transfer coefficient was obtained adjusting the experimental concentration profiles of dissolved oxygen in the liquid phase with the predictions of the axial dispersion model. According to experimental results the liquid holdup increases with the gas velocity, whereas the solid holdup decreases. The gas holdup increases significantly with the increase in gas velocity, and it shows for the three-phase fluidized bed comparable values or larger than those of bubble column. The volumetric gas-liquid mass transfer coefficient increases significantly with an increase in the air velocity for both bubble column and fluidized beds. In addition, in the operational condition of high liquid velocity, the presence of low-density particles in the bed increased the gas-liquid mass transfer, and thus the volumetric mass transfer coefficient values obtained in the fluidized bed were comparable or larger than those of bubble column.

Key words: Hydrodynamics, Mass transfer, Bubble column, Three-phase fluidized bed

1. INTRODUCTION

Three-phase fluidization is an operation used to bring into contact gas, liquid, and solid particles. The solid particles are fluidized by upflow liquid, which is the continuous phase, and cocurrent dispersed gas bubbles. The increased application of three-phase fluidized bed reactors in the chemical and biochemical processing fields has led to an increase in studies concerned with fully defining the characteristics of such reactors.

An interesting phenomenon is the bed expansion or contraction upon injecting gas into a liquid fluidized bed while the liquid flow rate is kept constant. With large particles, the bed height increases monotonically as gas velocity increases. However, an initial decrease of bed height exists if small particles are used. This phenomenon is believed to be caused by the wake trailing behind the bubble (Jean & Fan, 1986; Lee & De Lasa, 1987).

For the successful design and operation of three-phase fluidized beds, it is important to know the hydrodynamics and mass transfer characteristics of the fluidization process. For example, the design of a reactor depends on the expansion/contraction of the fluidized bed, which is in turn affected by the bed porosity. The bubble size, gas residence time, and consequently the gas-liquid mass transfer are influenced by the phase holdups (phase volume fractions). Therefore the bed porosity and the phase holdups are among the important informations needed for the design of a three-phase fluidized bed reactor.

Quite recently three-phase fluidized beds gained increasing importance in the area of biotechnology particularly in fermentations and wastewater treatment because three-phase fluidized beds provide favourable mixing and mass transfer properties combined with low shear stressing of the biological material. In these processes the mass transfer rates in three-phase fluidized beds from the gaseous to the liquid phase are sufficiently high.

The characteristics of three-phase fluidized bed reactors have been reviewed by Wild *et al*. (1984), Muroyama & Fan (1985) e Fan (1989). Gas, liquid and solid phase holdups, bed expansion, pressure drop, minimum fluidization velocity, and volumetric gas-liquid mass transfer coefficient are just some of the many aspects of fluidization process which have attracted the attention of many researchers. Most of these characteristics, however, have not been fully clarified (especially to low-density particles) which has motivated continued studies aimed at completely defining these systems (Tang & Fan, 1989,1990; Han *et al*., 1990; Hirata *et al*., 1995).

In this study, the experimental characterization of hydrodynamics and gas-liquid mass transfer in a three-phase fluidized bed containing polystyrene and nylon particles are investigated. The influence of gas and liquid velocities on phase holdups and volumetric gas-liquid mass transfer coefficient was investigated for flow conditions similar to those applied in biotechnological process.

1.1 Phase holdups in three-phase fluidized beds

According to Wild *et al*. (1984) and Silva (1995) the bed porosity and the individual phase holdups have been determined by means of pressure gradient (Efremov & Vakhrushev, 1970; Han *et al*., 1990), electro-resistivity probe (Tang & Fan, 1989), optical fiber probe methods (Lee & De Lasa, 1987), and by simultaneous closure of the gas and liquid feeds (Saberian-Broudjenni *et al*, 1987).

From the probe methods, it has been observed that the solid and the liquid holdups remained quite constant in the lower part (approximately two-thirds) of the fluidized bed. In the upper part of the bed, the solid holdup decreased rapidly, while the liquid holdup increased correspondingly as the top of the bed was approached (Lee & De Lasa, 1987).

However, in the main fluidized bed region, the phase holdups are very uniformly distributed across the bed. Therefore, we shall consider the mean values of the holdups and bed porosity in the bed. In addition, the gas holdup values obtained from the probe methods are somewhat lower than those from the pressure gradient method (Yu & Kim apud Han *et al*., 1990).

The evaluation of the overall phase holdups based on the pressure gradient method can be obtained through the following equations:

$$-\frac{dP}{dz} = (\varepsilon_g \rho_g + \varepsilon_l \rho_l + \varepsilon_s \rho_s) g \qquad (1)$$

$$\varepsilon_g + \varepsilon_l + \varepsilon_s = 1 \qquad (2)$$

$$\varepsilon = \varepsilon_g + \varepsilon_l \qquad (3)$$

$$\varepsilon_s = 1 - \varepsilon = \frac{M_s}{\rho_s AH} \qquad (4)$$

where: A – cross-sectional area of fluidized bed; g – gravitational acceleration; H – fluidized bed height; M_s – total weight of solid particles; P – static pressure at height, z; z – axial coordinate; ε - bed porosity; ε_g – global gas holdup; ε_l – global liquid holdup; ε_s – global solid holdup; ρ_g – density of gas phase; ρ_l – density of liquid phase; ρ_s – density of solid phase.

In this method, ε_s can be directly obtained from Eq. (4) with the height of bed expansion measured either by visual observation or by the pressure gradient method (Wild *et al.*, 1984; Muroyama & Fan, 1985) while ε_l and ε_g can be calculated from Eqs. (1) and (2) simultaneously with the experimentally measured static pressure gradient. This phase holdup measurement is based on the assumption of a homogeneous fluidized bed.

An alternative method of calculation was proposed by Efremov & Vakhrushev (1970). In this method the gas holdup can be measured by means of two manometer tubes. The pressure tap of the first tube was located directly above the distributor and the second pressure tap was located at the level of the upper boundary of the liquid-solid fluidized bed. When gas is passed into the bed, the liquid levels in the tubes fall by amounts Δh_1 e Δh_2. The gas holdup of the three-phase bed is determined from the ratio of the difference between the tube readings and the bed height:

$$\varepsilon_g = \frac{\Delta h_1 - \Delta h_2}{H} \qquad (5)$$

1.2 Gas-liquid mass transfer in a three-phase fluidized bed

In practice, the mass transfer rate across the gas-liquid interface can be described by the product of three terms: the liquid-side mass transfer coefficient (K_L), the interfacial area (a), and the concentration difference (ΔC). In a three-phase fluidized bed both the liquid-side mass transfer coefficient and the gas-liquid interfacial area inherently depend on the bed hydrodynamics. The liquid-side mass transfer coefficient incorporates the effects of the liquid flow field surrounding the rising gas bubbles. The interfacial area reflects the system bubble behavior. Consequently, the dependency of the bubble behavior on system properties such as gas and liquid velocities and particle size and density must carry over to the interfacial area. The most common approach in treating gas-liquid mass transfer is to combine the mass transfer coefficient and interfacial area terms into a single volumetric mass transfer coefficient ($K_L a$) averaged over the entire column height (Fan ,1989).

Much work has been done on gas-liquid mass transfer coefficient for three-phase fluidized bed of heavy particles (Ostergaard & Suchozebrski, 1971; Nguyen-Tien *et al.*, 1985; Chang *et al.*, 1986). It was found that increases in gas velocity, particle size, and solid concentration might result in an increase in $K_L a$. However, information on gas-liquid mass transfer behavior in three-phase fluidized beds of low-density particles is scarce (Tang & Fan, 1990; Miyahara *et al.*, 1993; Riedel & Gimenes, 1996).

The determination of the $K_L a$ is based on the evaluation of the oxygen concentration profiles measured along the column under steady-state conditions. The hydrodynamic behavior of three-phase fluidized beds was described with the axial dispersion model.

The differential equation of the axial dispersion model for the liquid phase can be given as (Deckwer *et al.*, 1982):

$$\frac{1}{Pe}\frac{d^2 C_{DO}}{dx^2} - \frac{dC_{DO}}{dx} + St(C^* - C_{DO}) = 0 \tag{6}$$

The boundary conditions for Eq. (6) are

$$C_{DO_0} = C_{DO}\big|_{x=0} - \frac{1}{Pe}\frac{dC_{DO}}{dx}\big|_{x=0} \qquad \text{at } x=0 \tag{7}$$

$$\frac{dC_{ODO}}{dx} = 0 \qquad \text{at } x=1 \tag{8}$$

Solving Eq. (6) using the above boundary conditions (Deckwer *et al.*, 1983):

$$C_{OD} = A_1 \exp(r_1 x) + A_2 \exp(r_2 x) + a' + bx - \frac{b}{St} \tag{9}$$

where:

$$x = \frac{z}{H} \tag{10}$$

$$St = \frac{K_L a H}{v_l} \tag{11}$$

$$P_{TS} = P_T(1 - \alpha_2) \tag{12}$$

$$\alpha_2 = \frac{\varepsilon_l^b \rho_l g(H_C - H)}{P_T} \tag{13}$$

$$\alpha_3 = \frac{\varepsilon_g \rho_g + \varepsilon_l \rho_l + \varepsilon_s \rho_s}{P_{TS}} \tag{14}$$

$$Pe = \frac{v_l H}{\varepsilon_l E_{ZL}} \tag{15}$$

$$C^* = \frac{y P_{TS}}{He}[1 + \alpha_3(1 - x)] \tag{16}$$

$$a' = \frac{-y P_{TS}}{H}(1 + \alpha_3) \tag{17}$$

$$b = \frac{-y P_{TS}}{H}\alpha_3 \tag{18}$$

$$A_1 = \frac{[\,Br_2\,exp(\,r_2\,)-br_1\,]}{N} \qquad\qquad (19)$$

$$A_2 = \frac{[\,Br_1\,exp(\,r_1\,)-br_2\,]}{N} \qquad\qquad (20)$$

$$B = \left[a'-C_{OD_o} - \frac{b}{St}\right]Pl - b \qquad\qquad (21)$$

$$N = r_1^2\,exp(r_1)- r_2^2\,exp(r_2) \qquad\qquad (22)$$

$$r_1 = \frac{Pe}{2}\left[1+\sqrt{1+\frac{4St}{Pe}}\right] \qquad\qquad (23)$$

$$r_2 = \frac{Pe}{2}\left[1-\sqrt{1+\frac{4St}{Pe}}\right] \qquad\qquad (24)$$

C_{DO} - axial dissolved oxygen concentration; C_{DO_o} - C_{DO} at the inlet; C* - equilibrium C_{DO} at the gas-liquid interface; E_{ZL}- axial liquid dispersion coefficient; H_C – total height of the column; He – Henry's constant for oxygen; P_T – pressure at the top of bubble column; P_{TS} – pressure at the top of three-phase zone; Pe – liquid phase Peclet number; St – Stanton number; v_l – superficial liquid velocity; x – dimensionless axial coordinate; y – gas-phase mole fraction of oxygen; z – axial coordinate; ε_l^b - liquid holdup at two-phase zone; α_2, α_3 - constants; a', b, A_1, A_2, B, N, r_1, r_2 - parameters defined by Eqs. (19) to (26).

2. EXPERIMENTAL

The details of the apparatus and experimental procedures are described elsewhere (Silva, 1995). Briefly, the three-phase system consists of air as the gas phase; 2.2 mm polystyrene (ρ_s = 1.05 g/cm^3) and 2.7 mm nylon (ρ_s = 1.15 g/cm^3) as the solid phase; and tap water as the liquid phase. The column consisted of a 6.3-cm inside-diameter and 200-cm high transparent acrylic-resin tube. Ten pressure taps were mounted into the side of the column wall. The static pressure at each of these points was measured with a water manometer. Gas holdup was calculated from the knowledge of bed pressure drop and expanded bed height by using the method proposed by Efremov & Vakhrushev (1970). Liquid samples were withdrawn at 10 axial positions. The dissolved oxygen concentrations in water at various axial positions in the column were measured by using the Winkler's method (APHA, 1985). The oxygen in the feed solution was stripped by pure nitrogen in the reservoir.

3. RESULTS AND DISCUSSION

3.1 Phase holdups

Figure 1 shows the gas holdup in a bubble column (gas-liquid two-phase system) as a function of superficial gas velocity. Gas holdup increased almost linearly with increasing superficial gas velocity. It can be seen that with increasing gas velocity from 0.6 to 2.4 cm/s,

the change of gas holdup is 3.6 times. Figure 1 also includes a comparison of gas holdup data obtained from this study with the predicted gas holdup using Akita & Yoshida's (1973) and Kumar *et al.*'s. (1976) correlations. The Akita & Yoshida's correlation underestimate and Kumar *et al.*'s correlation overestimate the gas holdup by about 23% and 10%, respectively, under the experimental conditions.

Akita & Yoshida (1973):

$$\frac{\varepsilon_g}{(1-\varepsilon_g)^4} = 0.2 \left(\frac{D_c^2 \rho_l g}{\sigma} \right)^{1/8} \left(\frac{D_c^3 \rho_l^2 g}{\mu_l^2} \right)^{1/12} \left(\frac{v_g}{\sqrt{gD_c}} \right) \tag{25}$$

Kumar *et al.* (1976):

$$\varepsilon_g = 0.728U - 0.485U^2 + 0.0975U^3 \tag{26}$$

where:

$$U = v_g \left(\frac{\rho_l^2}{\sigma(\rho_l - \rho_g)g} \right)^{1/4} \tag{27}$$

D_C – column diameter; v_g – superficial gas velocity; σ - surface tension of liquid phase; μ_l – viscosity of liquid phase.
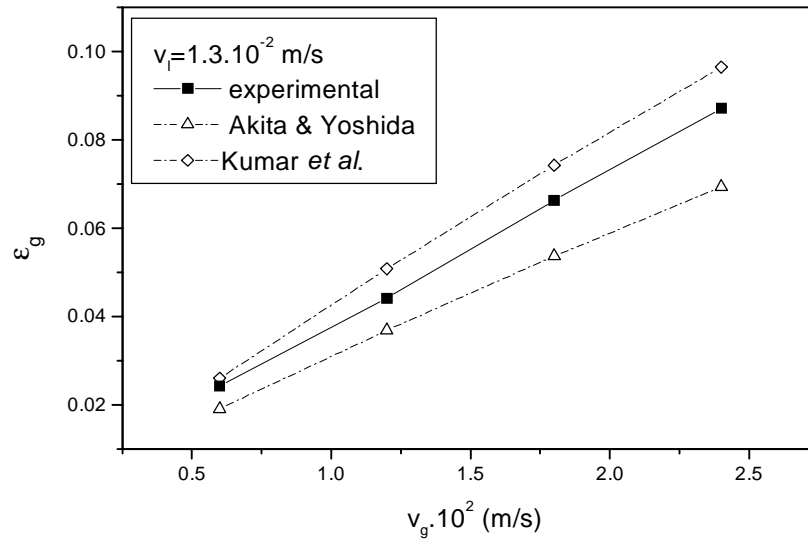

Figure 1 – Comparison of gas holdup in a bubble column with the correlations proposed by Akita & Yoshida (1973) and Kumar *et al.* (1976).

Figure 2 shows the effect of gas velocity on liquid and solid holdup for the three-phase fluidized bed of polystyrene particles when liquid velocity is 1.3 cm/s. As can be seen, liquid holdup slightly increases (16%); however, solid holdup decreases 36% with increasing gas velocity from 0.6 to 2.4 cm/s. Gas holdup in three-phase fluidized beds showed trends similar to that found in a bubble column.

As shown in Figure 3, gas holdup increases significantly with an increase in gas velocity in three-phase fluidized bed. As can be seen increasing gas velocity from 0.6 to 2.4 cm/s, the change of gas holdup is 3.7 times. Note that no hysteresis effects were observed in the gas holdup with respect to gas velocity under the conditions of this study: essentially, at a given gas velocity, the same gas holdup was obtained independent of whether the proceeding gas velocity used was lower or higher. Also, at liquid velocity of 1.3 cm/s three-phase fluidized bed generally has a lower gas holdup than bubble column. This effect was more pronounced in three-phase fluidized bed of nylon (higher density). However, at liquid velocity of 2.5 cm/s gas holdup values in three-phase fluidized bed were comparable or larger than those of bubble column and, it can be seen that with increasing liquid velocity from 1.3 to 2.5 cm/s, the gas holdup values increased 50% and 30% for nylon and polystyrene particles, respectively.
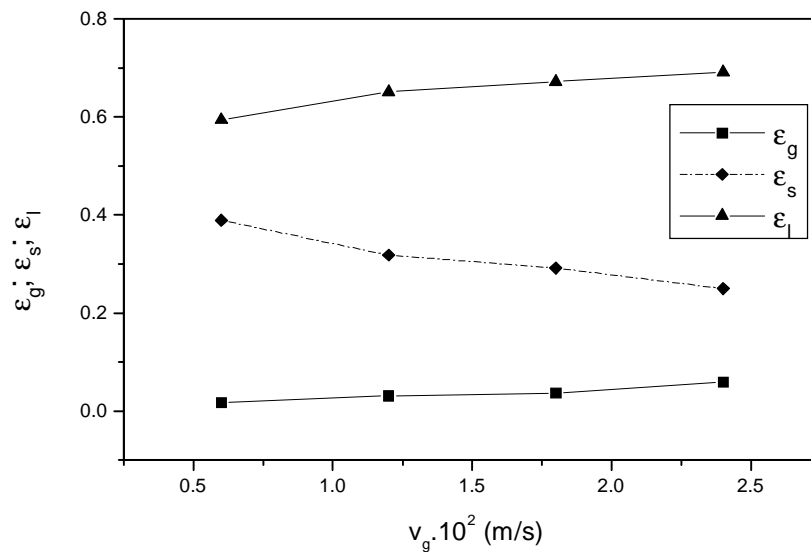

Figure 2 – Effect of gas velocity on the solid, liquid and gas holdup for the three-phase fluidized bed of polystyrene particles at liquid velocity of 1.3 cm/s.
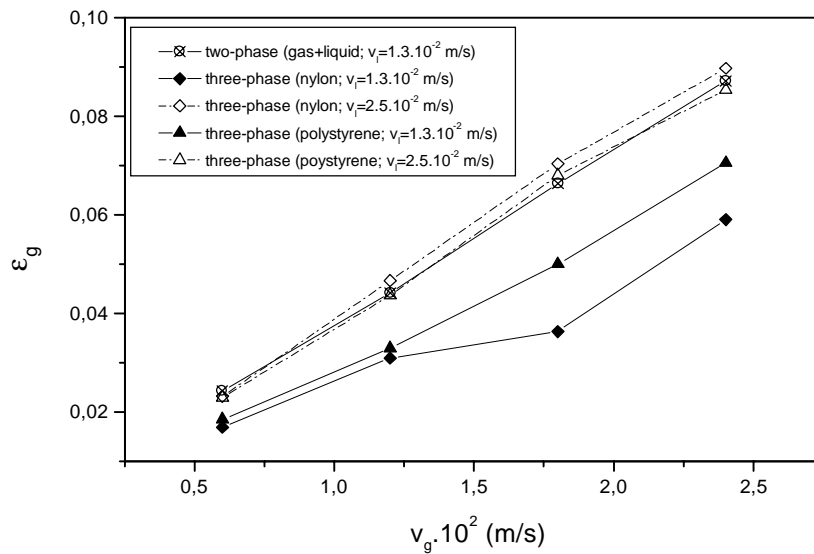

Figure 3 – Effect of gas velocity on gas holdup for three-phase fluidized bed containing low-density particles.

3.2 Gas-liquid mass transfer

In the present study, in order to obtain the K_La values in a bubble column and in a three-phase fluidized bed, Eq. (9) – axial dispersion model, was fitted to the measured dissolved oxygen profile in the liquid phase by using Marquadt's optimizing technique (Giudici apud Silva, 1995).

Table 1 shows the K_La and E_{ZL} values obtained here by parametric fitting for a bubble column. As can be seen K_La, increases significantly with an increase in gas velocity in the bubble column, but E_{ZL} is almost independent of the gas velocity used. According to Deckwer *et al*. (1982) the values of K_La obtained from fitting Eq. (9) to the dissolved oxygen concentration profile are not very sensitive to variation in E_{ZL}.

Table 1 - K_La e E_{ZL} values obtained by parametric fitting for a bubble column

$v_l.10^2$ (m/s)	$v_g.10^2$ (m/s)	$K_La.10^2$ (s^{-1})	$E_{ZL}.10^4$ (m^2/s)
1.3	0.6	0.58	14.45
	1.2	1.43	20.03
	1.8	2.55	16.31
	2.4	3.91	18.83

Figure 4 shows that K_La increases significantly with an increase in the gas velocity for both bubble column and three-phase fluidized bed of polystyrene and nylon particles. According to Lamont & Scott apud Miyahara *et al*. (1993) mass transfer at a gas-liquid interface is due to tiny eddies which are formed in the gas-liquid flow. Presumably, as the gas velocity is increased, the gas holdup increases, resulting in intense wake shedding from the bubble forming large numbers of small eddies as the gas flows in, enhancing, thereby, mass transfer at the gas-liquid interface and, as a result, increasing K_La. It has been observed in the present study that K_La values in three-phase fluidized beds are smaller than those in bubble column in the operational condition of small liquid velocity. This effect is undoubtedly due to the bubble coalescence phenomenon, to a greater extent, in low liquid velocity.

The effect of liquid velocity on K_La in bubble columns and three phase fluidized beds can be seen in Figure 5. As can be seen, K_La slightly decreases with an increase in liquid velocity in bubble column; however, a reverse trend is observed in three-phase fluidized beds. In the operational condition of high liquid velocity, the presence of low-density particles in the bed increased the gas-liquid mass transfer through not only an increase in holdup (or the interfacial area) but an increase in K_L, probably due to the increase turbulence intensity, and thus the K_La values obtained in the fluidized bed were comparable or larger than those of the bubble column.

The K_La data for three-phase fluidized bed obtained from this study are in qualitative agreement with those reported by Ostergaard & Suchozebrski (1971) for 1 and 6 mm glass beads, Tang & Fan (1990) for polystyrene, acrylic, acetate and nylon particles with sizes from 1 to 2.5 mm, and Riedel & Gimenes (1996) for PVC particles.

4. CONCLUSIONS

From the results for three-phase fluidized bed containing low-density particles, the following conclusions can be drawn.

(1) The liquid holdup increases with the gas velocity, whereas the solid holdup decreases.

(2) The gas holdup increases significantly with the increase in gas velocity, and it shows for the three-phase fluidized bed comparable values those of bubble column.

(3) The volumetric gas-liquid mass transfer coefficient increases significantly with an increase in the air velocity for both bubble column and fluidized beds.

(4) In the operational condition of high liquid velocity, the presence of low-density particles in the bed increased the gas-liquid mass transfer, and thus the volumetric mass transfer coefficient values obtained in the fluidized bed were comparable or larger than those of bubble column.
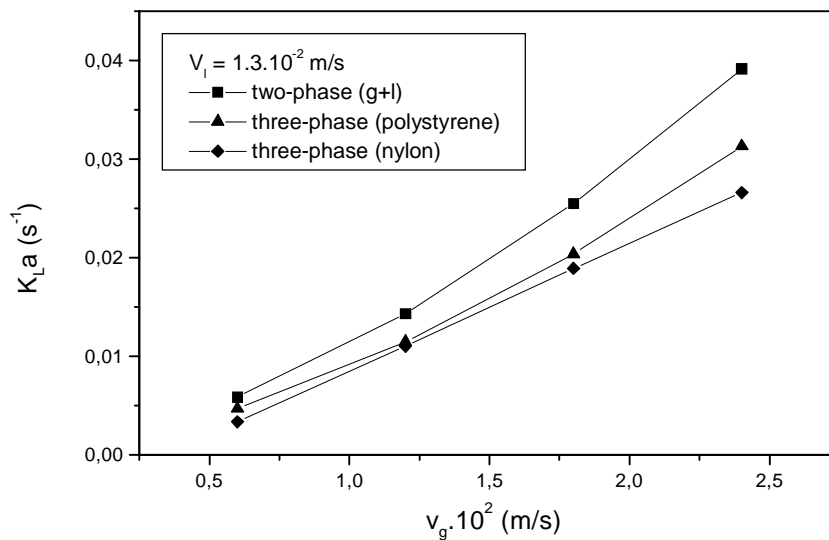

Figure 4 – Effect of gas velocity on K_La for three-phase fluidized bed of polystyrene and nylon particles
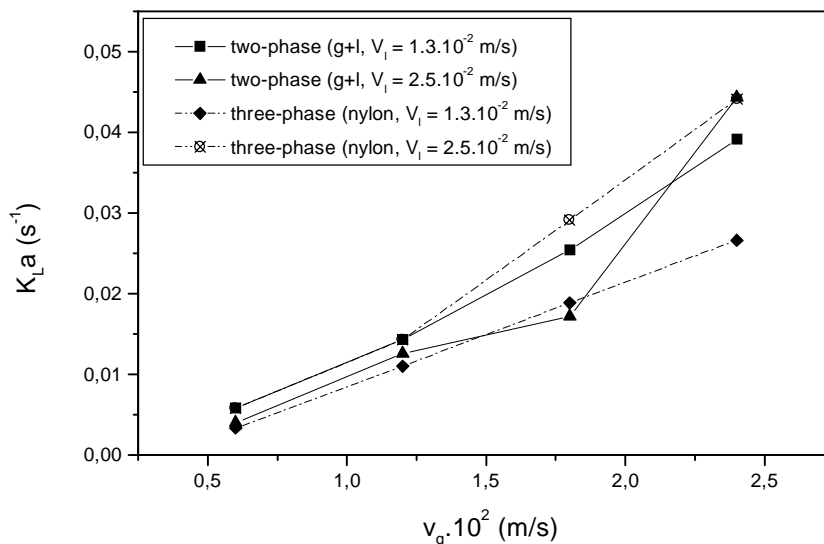

Figure 5 – Effects of gas and liquid velocities on K_La for bubble column and three-phase fluidized bed of nylon particles

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