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# *sLiquid-Phase Back mixing in Bubble Columns*

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## Abstract

Liquid-phase axial dispersion coefficients have been measured for air-water system in bubble columns of 10, 15 and 30 cm diameter. The experiments are carried out using a transient method (the tracer response method). Dispersion coefficient is obtained by adjusting the experimental profiles of tracer concentration with the predictions of the model. The experimental results show that one-dimensional axial dispersion coefficient, $D_{ax,L}$, reveal strong scale dependence. Backmixing of liquid phase increases with the increase of reactor diameter and superficial gas velocity. Axial dispersion coefficient for large column reactors can be easily predicted from the developed relation $D_{ax,L} = 24.48\ Ug^{0.29}\ D_T^{0.69}$. Comparison of calculated with the experimental data and with the published data of other authors shows good agreement which ensure the reliability and confusability of the adopted correlations to be used in further design and scale-up purposes.

**Keywords: Bubble columns, Column diameter influence, Backmixing, Axial dispersion Coefficient**

# الخلط ألرجوعي للطور السائل في الأعمدة الفقاعية

## الخلاصة

**تم قياس معامل التشتت المحوري (Axial dispersion coefficient) للطور السائل لثلاث أعمدة فقاعية مختلفة الأقطار ( 10 ، 15 ، 30 سم ) باستخدام نظام ماء – هواء . تم إجراء التجارب بإتباع أسلوب الاستجابة النبضية للدليل (tracer response experiments ) .**

**بينت النتائج بان معامل التشتت المحوري هو مقياس قوي ويمكن الاعتماد عليه في حسابات الخلط ألرجوعي (backmixing) للطور السائل في الأعمدة الفقاعية. ومن النتائج المختبرة وجد بان الخلط ألرجوعي للطور السائل يزداد بزيادة قطر الأنبوب (column diameter) وسرعة الغاز ( superficial gas velocity).**

**بالاعتماد على النتائج تم وضع علاقة رياضية لحساب معامل التشتت المحوري والتي يمكن من خلالها التنبؤ بمعامل التشتت المحوري للأعمدة الفقاعية. أظهرت النتائج لهذا الموديل تطابقا جيدا مع البيانات المنشورة في الأدبيات العلمية مما يعطي الاعتمادية والموثوقية لاستخدام الموديل في الحسابات التصميمية للاعمدة الفقاعية.**

**الكلمات الدالة: الأعمدة الفقاعية، تأثير قطر الأنبوب، الخلط ألرجوعي، معامل التشتت المحوري**

## Notation

| | |
|---|---|
| $C_0$ | Final concentration achieved when $t = \infty$ , kg/m |
| $C_L$ | Liquid concentration, kg/m$^3$ |
| $D_{xx}$ | Average axial turbulent eddy diffusivity according to Degaleesan et al. (1997), m$^2$/s |
| $D_{rr}$ | Average radial turbulent eddy diffusivity according to Degaleesan et al. (1997), m$^2$/s |
| $D_{ax,L}$ | Liquid phase axial dispersion coefficient, m$^2$/s |
| $D_T$ | Column diameter, m |
| g | Acceleration due to gravity, m/s$^2$ |
| H | Total height of the column, m |
| $H_0$ | Total liquid height in the column, m |
| Hd | Dispersion height, m |
| L | Total liquid height in the column, m |
| Pe | Peclet number |
| t | Time, s |
| $U_g$ | Superficial gas velocity ( m/s ) |
| z | Axial coordinate, m |
| $\varepsilon_G$ | Fractional gas hold-up |

## Introduction

Bubble-column reactors are widely used in chemical, petrochemical, biochemical and metallurgical industries. Their lack of moving parts and excellent heat and mass transfer characteristics are some of the predominant advantages that render them particularly attractive for various multiphase exothermic reactions. Bubble columns are often designed with a length-to-diameter ratio, or aspect ratio, of at least 5. They are operated in either semi batch mode (zero liquid throughput), such as in liquid-phase methanol synthesis, or continuous mode (co-current or countercurrent) such as in Fischer - Tropsch synthesis, with liquid superficial velocities lower than the gas superficial velocity by at least an order of magnitude. As a result, it is the gas flow that controls the fluid dynamics of the individual phases in these systems. This in turn controls liquid mixing and interphase mass transfer, which subsequently influence conversion and selectivity.

Mixing of liquid phase has to be attributed to various phenomena such as turbulent vortices, liquid entrainment in the wakes of rising bubbles, large-scale liquid circulation, radial exchange flows. All these are obviously interrelated and are primarily dependent on bubble size and rise velocity distribution, gas hold-up profiles, bubble-bubble interactions and liquid circulation flow [1].

For gas-liquid systems type, dispersion can be defined as a stochastic exchange process by which gradients of intensive quantities such as concentration and temperature are broken down, as a direct effect, the concentration in the reactor is reduced [2].

In bubble column reactor, the gas generates significant backmixing in the liquid phase. After a thorough analysis it can be noticed that the backmixing of the dense phase is caused by the eddies which derive their energy from large, fast-rising bubbles. The maximum size of the eddies, for vessels with $H/D_T > 1$, is limited by the column diameter, $D_T$. The induced circulation patterns have a profound effect on mass transfer and productivity of these systems and they are especially important in eliminating concentration gradients within the vessel. The design of the bubble column reactor always requires consideration of the backmixing in the liquid phase [2].

Since the axial dispersion model characterizes the back mixing by only a single parameter, its simplicity made it the most widely used representation of the non-ideal mixing behavior for each phase in bubble column reactors. The time variation of the liquid phase concentration of a tracer is given by Fick's law:

$$\frac{\partial C_L}{\partial t} = D_{ax,L}\,\frac{\partial^2 C_L}{\partial z^2} \qquad \text{……... (1)}$$

Where the axial dispersion coefficient ($D_{ax,L}$) is characterized as a unique parameter for the degree of back mixing during process. The term 'axial' is used in order to mark clearly the difference between the mixing in the direction of flow and the mixing in the lateral or radial direction**[1]**. In bubble columns these two quantities are quite different in magnitude, so that the axial dispersion coefficient is significantly exceeding the radial dispersion coefficient **[3].**

Though there is a large variety of experimental data on liquid phase back mixing available in the literature for different column diameters and superficial gas velocities, it is difficult to compare the results of one author with those of others for the scale up task. The reasons are: (1) differences in the physical properties of the liquids used in various studies, (2) presence of impurities in the liquid phase, and (3) the fact that each published study is often restricted to one column with a diameter often smaller than 0.3 m.

Axial dispersion coefficients of the liquid phase in vertical gas-liquid contactors have been reviewed by Shah et al., (1978)**[4].** The dispersion coefficient is expressed in dimensionless form as Peclet number, its value denoting the degree of back mixing in the column. If Pe = 0 back mixing is complete and if Pe = ∞ plug flow prevails. Most of reported empirical correlations indicated the dispersion coefficient to be dependent on the gas velocity and column diameter. A significant influence of the flow direction (i.e. co-current or counter-current) has not been pointed out. If the range of liquid velocities used in industrial operation is considered, the superficial liquid velocity $U_l$ appears to have no influence on liquid-phase dispersion. Steady state measurements by Deckwer et al., (1973)**[5],** Badura et al., (1974)**[6]** and Towell et al., (1972)**[7]** confirmed this fact. It is usually assumed that the dispersion coefficient does not depend on the column height. However, studies of Deckwer et al., (1973)**[5]** and Schugerl et al., (1977)**[8]** show that dispersion coefficients may differ along the column height, decreasing from top to bottom.

The degree of axial dispersion is also affected by vessel internals and surface active agents that delay the coalescence**[1]**. Konig (1978)**[9]** demonstrated the effects of surfactants and sparger type by experimenting with weak alcohol solutions using three different porous spargers. They clearly indicated that the interaction of surfactants and sparger can be very complex. Surfactants can produce either much more or much less back mixing than surfactant-free systems, depending on the bubble size, which, in turn, depends on the sparger used.

The influence of the physical properties of the liquid on the dispersion coefficient has been investigated by several authors **[10,11,12,13 and 14]** but little effect had been observed.

Ichikawa (1967)**[15]** and Chen (1989)**[16]** found insignificant effect of superficial liquid velocity on $D_{ax,L}$, whereas other authors such as Schugerl et al., (1977)[8] and Palaskar et al., (2000)[17] described much more significant effect of superficial liquid velocity.

Baird et al., (1975)[18] modeled the most useful relation for reactor scale-up purposes and vertical cylindrical vessels with pure liquids without coalescence inhibitors. They proposed the following equation:

$$D_{ax,L} = 0.35\, g^{1/3} D_T^{4/3} U_g^{1/3} \quad \text{- - - - (2)}$$

Furthermore, the authors extended their correlation to fluidized beds and liquid-liquid spray columns, which proved to be partially successful.

The dependency of $D_{ax,L}$ on $D_T$ and $U_g$ as expressed by Eq. (2) has been confirmed by many studies and can be also transcript in dimensionless terms of Pe and Fr numbers as represented in the following Eq.

$$\frac{D_{ax,L}}{D_T U_g} = k\left(\frac{g D_T}{U_g^2}\right)^{1/3} \quad \text{- - - - - - - (3)}$$

Where the constant $k$ have different values for different authors.

In 1996 Degaleesan[19] developed a phenomenological model for liquid mixing, mainly based on the monitored flow circulation and turbulent eddy diffusion. The model was called "recirculation and cross flow with dispersion" (RCFD) and accounts for two mixing mechanisms. The first one is the convective recirculation, due to the large scale liquid motion. In this context, the column can be simply considered as a big recirculation cell, with liquid moving upwards in the central core zone and descending along the annular region, near the walls of the column. Besides this, fluctuated motion of the liquid elements superimposed, due to the bubble turbulence. This second mixing contribution takes into account the complex motion of the gas bubbles, and therefore the random motion of fluid elements in axial, radial and azimuthal direction caused by the wakes of fast-rising large bubbles. The up flow and down flow, characterized by averaged interstitial velocities are connected to each end by two regions which are assumed well-mixed. The turbulent axial mixing is accounted for by an axial dispersion coefficient in each section; the radial mixing is incorporated into an exchange coefficient between the two sections (i.e. the up flow region and the down flow region). The input parameters of the model (such as local liquid velocities and void fraction) were obtained using Computer Automated Radioactive Particle Tracking (CARPT) and Computed Tomography (CT) as experimental techniques.

The model was further developed and extended also for bubble columns slurry reactors by Degaleesan et al., (1997)[20]**.** The two-compartment convective-diffusion model has been formulated by considering a finite volume discretization of the two-dimensional axis-symmetric convective-diffusion model developed for bubble columns flows. The same experimental techniques were used for the database, together with data from literature. The existing databases were limited to air-water systems, certain column sizes and superficial gas velocities. The methodology aimed to extrapolate the data giving the following cross sectional averaged eddy diffusivities $\overline{D}_{xx}$ and $\overline{D}_{rr}$ :

$$\overline{D}_{xx} = -\frac{0.00584}{D_T^{0.8}} + 0.1689\left(D_T U_g\right)^{0.3} \quad \text{- - (4)}$$

$$\overline{D}_{rr} = -\frac{0.000879}{D_T^{0.8}} + 0.0206\left(D_T U_g\right)^{0.3} \quad \text{-- (5)}$$

for $U_g \geq 0.05$ m/s.

The authors limited the applicability of the above correlations to air-water systems in churn turbulent regime only at atmospheric pressure. When the axial term was compared with experimental data from this work, they showed a good agreement for low gas velocity only. This could not be attributed only to the fact that the radial contribution was also taken into account, as long as this term is much smaller, but to the currently limited database, as mentioned by the authors. They advised that for higher gas velocities, the equations presented above have to be verified in compliance with experimental data.

Our objectives are to study the influence of column diameter on liquid phase back mixing, operating in the bubbly and churn-turbulent regime at superficial gas velocities in the range 0.26-10.8 cm/s. The results of our study can be expected to be useful for scale-up purposes.

## Experimental Works

Experiments are carried out in three batch type bubble columns with internal diameters of (10, 15, 30) cm and (174, 160, 150) cm in height respectively. The 10 cm column is made of PVC incorporated with glass window for the purpose of visual inspection, the 15 cm column is made of glass type (QVF), and the third column is made of Perspex. The columns are opened at the top; hence the pressure corresponded with is ambient conditions. Perforated plate spargers of identical design are used in all three columns to distribute the gas phase. The distributor plates are made of plastic plate with holes of 2 mm diameter. Air is used as the gas phase and demineralized water as liquid phase. The gas is introduced from the bottom of the columns. The experiments are carried out at various gas velocities, carefully adjusted and controlled using a calibrated rotameters. Before starting acquisition of data for a given gas flow rate, the system is given time to achieve steady state. A typical experimental set-up is shown in Fig. 1 for the 10 cm column.

Residence time distribution (RTD) of the liquid phase is measured using different amounts of saturated solution of NaCl as a tracer. Different volumes of tracer are used to obtain the optimal amount of tracer that corresponds to optimal signal within the operating range of conductivity cell. This optimal amount of a saturated solution of NaCl is found equal to 3.38 wt %.

The conductivity probes used in this work is manufactured by Philips Company of 1cm in diameter and 15 cm long. They simply consist of two electrodes, approximately 3 mm apart, and encapsulated in plastic tubing. The probes are properly calibrated by measuring their responses to solutions of different known tracer concentrations.

The signals from the electrodes are transmitted to conductance meter (of Philips type), of range 100 μs to 1000 ms which provide a reading in units of conductance. The meters are connected with an interface to a personal computer.

Tracer is injected as a pulse input. Local changes in tracer concentration are displayed and saved continuously on PC. Three electric conductivity probes are placed 2 cm away from the inside wall, located at different heights as shown in Fig. (2), each of them is connected to PC via interface circuit. The distance from the injection to the measuring points, $L_1$, $L_2$, $L_3$ and Hd are given in Table (1).

Time for each experiment is large enough in order to reach final concentration in the column.

The constructive details of the backmixing experiments, in the three bubble columns, are specified in Table (1). The operating conditions used for the performed experiments are given in Table (2).

Fig. (3) shows typical transient tracer concentrations from the 10 cm column, operated at 4.68 cm/s superficial gas velocity. These signals are fitted using the analytic solution to the diffusion equation presented in reference [21]. In this way, for a given experiment, only one variable was adjusted, i.e. the axial dispersion coefficient, $D_{ax,L}$.

From literatures investigation, it is found that there are two different measuring techniques widely used along the time. Since the stationary method is rather time consuming and expensive it is convenient to use a non- steady-state technique, such as the pulse-response method (the mixing time determinations) **[10,6,22]**. The mixing time is defined as time necessary to achieve the homogeneity in the column, after all amount of tracer was completely mixed with the liquid. Siemes and Weiss (1957)**[23]** are the first using the pulse method for measuring the dispersion in bubble columns. They are followed by Ohki et al., (1970)**[24]**, Hikita et al., (1974)**[11]**. The partial differential equation based on one-dimensional model has been solved analytically by Siemes and Weiss (1957)**[23]**. The boundary condition for equation (1) are:

$$\frac{\partial C_L}{\partial z} = 0 \quad \text{at} \quad z = 0 \quad \text{and} \quad z = L$$

and the initial conditions are:

$C_L(z,0) = C_0$ for $0 \le z \le \lambda$
$C_L(z,0) = 0$ for $z \ge \lambda$

Where $\lambda$ is the height at which tracer is injected. The solution of Eq.(1) under consideration of the corresponding boundary conditions gives**[21]**:

$$\frac{C_L}{C_0} = 1 + 2\sum_{n=1}^{\infty}\left[\left(\cos\frac{n\pi}{L}L_z\right)\exp\left(-\frac{n^2\pi^2}{L^2}D_{ax,L}\,t\right)\right] \qquad \text{----- - - (6)}$$

Where $C_0$ is the final concentration when $t = \infty$ and $L_z$ is the distance to the measuring point ($L_1$, $L_2$ and $L_3$).

The axial dispersion coefficient $D_{ax,L}$ is obtained by adjusting the experimental profiles with the solution of Eq. (6) model. A number of n = 20 terms are found to be sufficient. As it can be seen, the evaluation of $D_{ax,L}$ from the curves produced by the pulse method is fast and reasonably good.

## Results and Discussion

The experimental measurements for axial dispersion coefficients of the liquid phase show a strong function of the column diameter; Fig. (4). Liquid phase turbulence, induced mainly by the movement of bubbles and the existence of large-scale liquid internal circulation, are the main causes of liquid mixing in bubble columns. Joshi, (1980)**[32]**, and Degaleesan et. al., (1997)**[20]** indicated in their studies the presence of a large-scale liquid circulation cell in bubble columns, with liquid ascending at the central region and descending at the wall region. This liquid internal circulation is mainly driven by non-uniform radial gas distribution in the column. In homogeneous bubbly flow regime, there is no pronounced large-scale liquid circulation in the column and the liquid phase turbulence induced by rising bubbles is the main reason for liquid mixing. The scale of turbulence in homogeneous bubbly flow regime depends on the bubble size, as the gas velocity increases, the bubble size increases thus the bubble-induced turbulence increases which result in a rapid increase in the axial dispersion coefficient, as shown in Fig. (4). In churn-turbulent flow regime, both the convective liquid circulation and the liquid turbulent fluctuations play important roles in determining the mixing behavior of the liquid phase which causes liquid phase dispersion and backmixing.

Once again, the limited laboratory conditions confine in a relatively small area far from the real industrial dimensions.

The measured axial dispersion coefficients of air-water system are compared in Fig. (5) with the available literatures data at same operating conditions for the three columns. Fig. (6) shows the measured axial dispersion coefficient for the three columns at a different superficial gas velocity. The strong influence of the column diameter on the axial dispersion is evident.

The axial dispersion coefficients can be predicted in terms of superficial gas velocity, as a characteristic velocity and column diameter, as a characteristic length. Thus our suggestion is to use the following correlation for the estimation of Dax,L:

$$D_{ax,L} = 24.48\ Ug^{0.29}\ D_T^{0.69} \qquad \text{- - - - (7)}$$

The estimated model works very well not only for present experimental data but also for data culled from the literature, (see Figs. (7& 8). Equation (7) presents a reliable and accurate method of predicting the axial dispersion coefficients in case of large diameter columns.

## Conclusions

The main results presented in this work are:

1-The column diameter significantly influences the back mixing of the liquid phase. The axial dispersion coefficient of the liquid phase increases with the diameter of the reactor and with superficial gas velocity.

2-The measurements of the axial dispersion coefficient in liquid phase show that this parameter can simply be predicted as being proportional to the product of the superficial gas velocity, Ug, and the column diameter.

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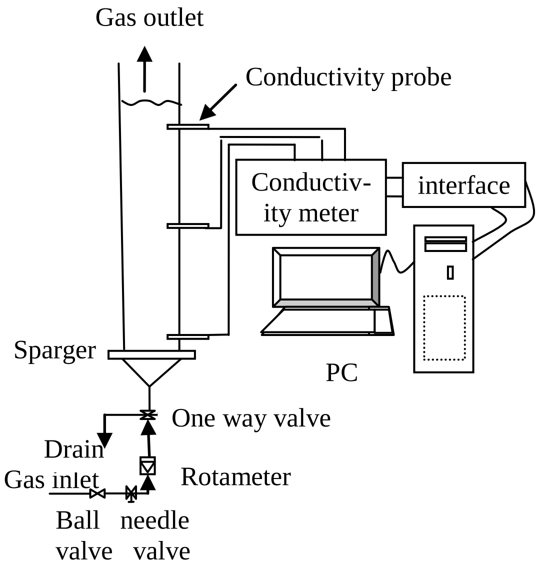


**Figure (1) Typical experimental set-up for the 10 cm diameter column**.

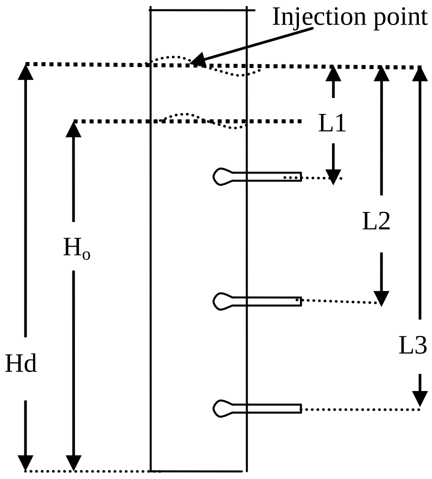


**Figure (2) Distances to the measuring points in the column.**

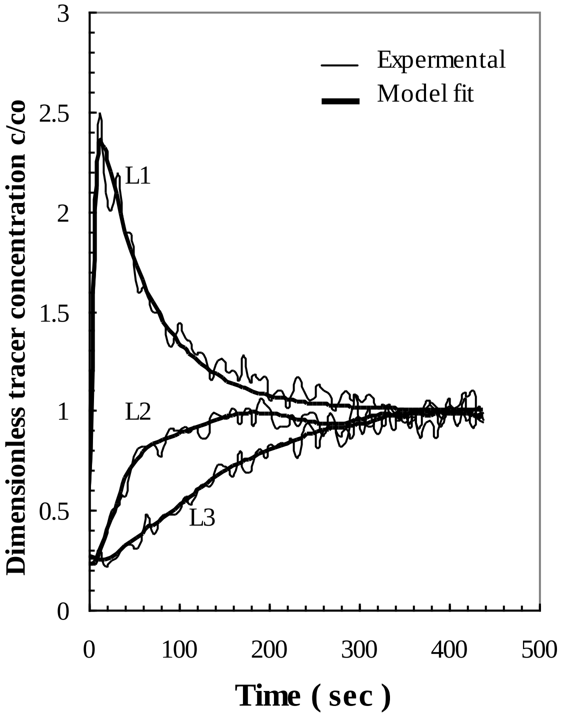


**Figure (3) Normalised liquid- phase tracer concentration measured at three different locations along the height of the column in response to pulse tracer injection. The smooth curves represent the fits to the curves from fitting a diffusion model presented in [21].**

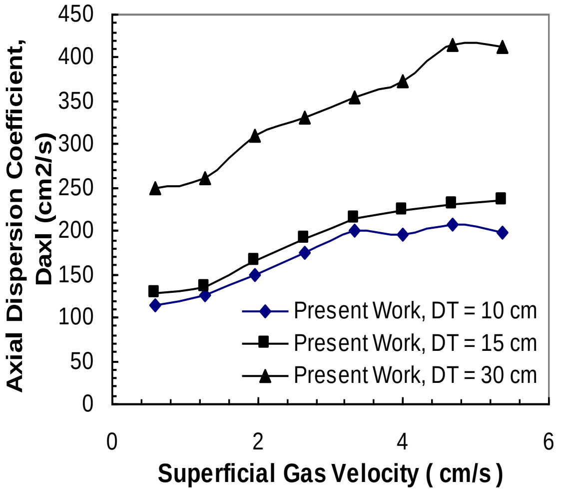
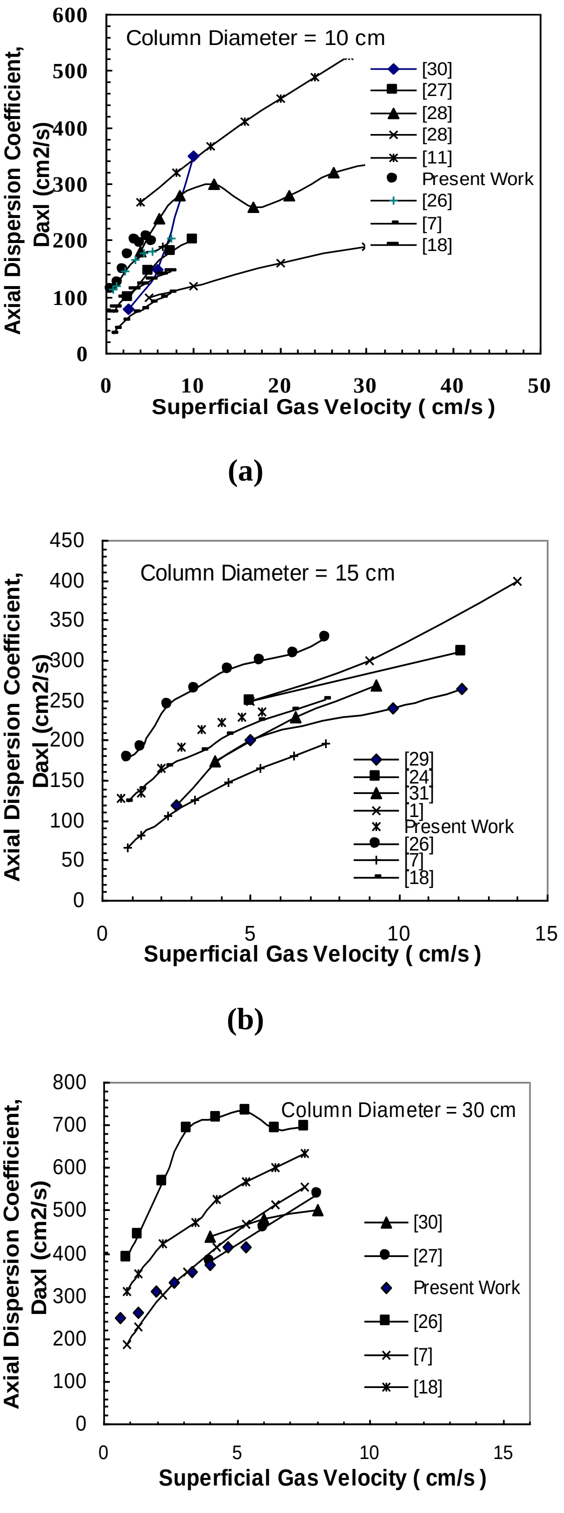


**Figure (4) Axial dispersion coefficient of the liquid phase measured in three columns, as a function of superficial gas velocity.**



(a)

(b)

(c)

**Figure (5) Axial Dispersion Coefficients of the Liquid Phase Measurement in Column with Diameter 10,15 and 30 cm , Comparison with published literature**

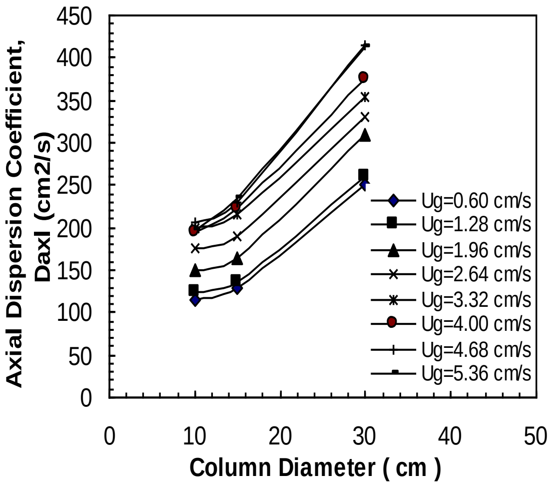


**Figure (6) Axial Dispersion Coefficient of the Liquid Phase as a Function of Columns Diameter for Different Superficial Gas Velocity**

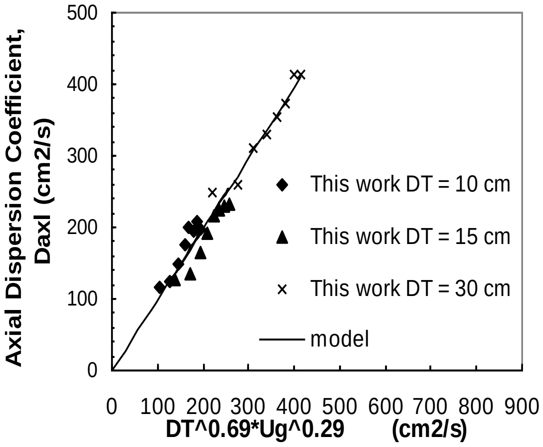


**Figure (7) Comparison between the measured axial dispersion coefficient Dax,L and the predictions**

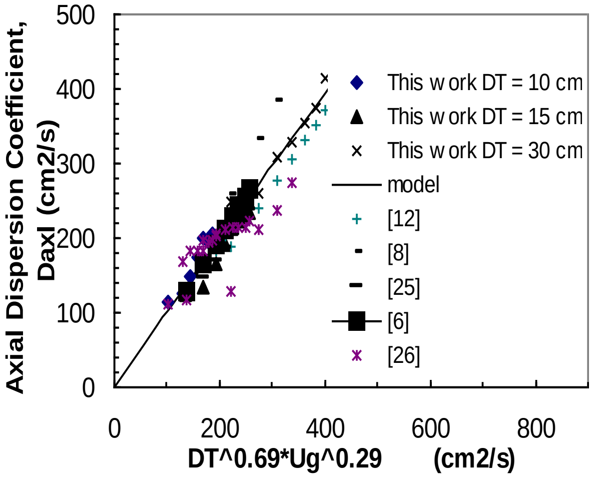


**Figure (8) Comparison between the measured axial dispersion coefficient $D_{ax,L}$ (our data and from the literature) and predictions of the estimated model by Eq. (7)**

**Table (1) Constructional detail about the backmixing experiments**

| **Operating conditions** | **Column diameter** | | |
|---|---|---|---|
| | **$D_T$ = 10 cm** | **$D_T$ = 15 cm** | **$D_T$ = 30 cm** |
| Liquid height $H_0$ /cm | 131 | 135 | 100 |
| Distance to the measuring point / cm | $L_1$ = 3.8<br>$L_2$ = 59<br>$L_3$=112.8 | $L_1$ = 3.5<br>$L_2$ = 55<br>$L_3$= 100 | $L_1$ = 3<br>$L_2$ = 54<br>$L_3$ = 79 |

**Table (2) Operating conditions used for measuring the axial dispersion coefficient**

| **Column diameter $D_T$ (cm)** | **Superficial gas velocity range, $U_g$ (cm/s)** | **Gas holdup range $\varepsilon_G$** |
|---|---|---|
| 10 | 0.26-10.8 | 0.037-0.20 |
| 15 | 0.87-7.54 | 0.058-0.27 |
| 30 | 0.87-7.54 | 0.053-0.26 |