# HYDRODYNAMICS OF THREE PHASE FLUID DISPERSE COLUMNS

Károly MOLNÁR

Department of Chemical and Food Engineering Technical University of Budapest H-1521 Budapest, Hungary

Received: April 11, 1994

## Abstract

In industry, the separation of various gases from mixtures is often necessary. In such procedures absorbent liquids are used to drive out undesirable components, either in continuous or gradual phase contact installations. The same operational units are adequate for the separation of two or multiple component mixtures, using rectification procedures. The contact of liquid and gas (vapour) may be made very efficient using the so-called three phase fluid disperse systems the features of which will be discussed in the present paper.

Keywords: absorption, desorption, distillation, fluidisation, diffusion, hydrodynamics.

# Introduction

In the chemical and food industries, in agriculture and in the ever more important environmental protection, various procedures and installations that may perform efficient separations (absorption of gases, desorption, separation of powders, cleaning of industrial gases, distillation, etc.) are often used.

A main characteristic of these operations is that the continuous contact of the gas (vapour) phase and the liquid phase is mandatory in order to obtain the degree of separation desired. In the case of these operations, components are transferred from one phase to another by mass transfer. The dimensions of the installation and thus the cost of the investment are determined by the rate of mass transport. Thus, the type of installation used for the continuous (or gradual) contact of the phases is of great importance. The above mentioned operations are traditionally performed in equipment called packed columns. In these towers, the disperse liquid phase prepared in spraying columns appears as a laminar flowing film or drop which flows in counter current with the continuous gas (vapour) phase.

The mass transfer occurs at the boundary layer of the two phases, and the degree of the process is determined by the diffusion rate within the phase. The rate of mass transfer is limited in many cases by the diffusion resistance on the liquid side. In such cases the concentration of the disperse, elementary unit phase boundary layer formed in the traditional installations reaches equilibrium very rapidly. However, practically, a molecular material transport takes also place, either from the main mass of the liquid phase towards the phase boundary layer or the other way round. As a result of this, the rate of the process is considerably slowed down. The rate of mass transfer may be increased only if in the installations used a large phase contact surface and/or a high diffusion rate within the phases can be ensured. This latter means an increase of the mass transfer factor within the phases.

The packed columns with resting layers (Raschig, Berl saddle, ordered aggregates, etc.) which are used traditionally for the formation of the disperse liquid phase determine the establishment of a large phase boundary surface. The use of 'Spray' columns results in even larger phase boundary surfaces. However, this does not improve the mass transfer factors on the liquid side. Therefore, when the diffusion rate is determined by the resistance of the liquid film or drop, in order to promote the mass transfer, besides increasing the phase boundary surfaces, another step forward could be made by making use of the advantages offered by turbulent diffusion. The diffusion is said to be turbulent, if, instead of single molecules, groups of molecules are transferred from the surface of the phase boundary layer into the mass of the liquid or the other way round. This may be achieved by a continuous splitting of the phase boundary layer and the mixing of the split parts in the main mass of the liquid. Thus, the surface is continuously renewed and, as a result, turbulent diffusion is achieved.

This may be furthered by various packing elements with special shapes (i.e. Telerette, Nutter Ring, NORPAC, etc.). Usually these are shattered bodies with many sharp edges for reforming the phase boundary layer of the disperse liquid phase and mixing this with the main mass.

The use of the above mentioned resting columns with fillings is rather limited in the cases when the liquid or the gas phase contains some solid material (e.g. powders in smoke gases). This is explained by the fact that since the filling bed acts as a filter, it becomes soiled or clogged and as a result the pressure drop of the fluid flow through the bed increases.

The rate of the turbulent diffusion cannot be further improved by newer packing elements with special shapes. Their costs are already high.

The installations and procedures based on floating packing elements that have a considerably lower density than that of the liquid phase, aim at reducing the resistance to mass transfer on the gas side but the most they can achieve is improvement of the separation efficiency for the solid pollutants present in the gas phase. The role of the above mentioned installations and procedures is to fragment the disperse gas phase, to renew the surface of the gas bubbles and to increase the turbulence on the gas side. This is achieved by the use of a packing material that floats on the continuous liquid phase, which hinders the raising movement of the gas phase being bubbled through and renewes the bubbles. In the last 10-20 years, numerous such procedures and installations have been described.

In procedures described previously, the absorption of gases, the wet dust separation and the efficiency of the simultaneous gas and dust separation are not satisfactory. As a result, application of the above procedures and installations involves a considerable increase of the operating costs.

# The Fluid Disperse Procedure and Equipment

The use of the three phase disperse column suggested by K. MOLNÁR [1] eliminates the above shortcomings. The fluid disperse procedure and installation are based on the fact that with the enhancement of the mass transfer, the phase boundary surface and the mixing of the disperse liquid phase are also increased. Fluidised solid packing elements are used with the continuous gas phase, and the liquid phase is introduced as a disperse phase. In such cases, the mass formed by the packing material fluidised with the gas phase continuously fragments, renews and mixes the liquid phase that forms on its surface. As a result, the phase boundary surface increases, the renewal of the phase boundary layer is continuous and the turbulent diffusion increases in the liquid phase.

The counter current flow phase contact is achieved in traditional columns and the installation is called 'Fluid disperse columns' (FDC). The operating principle of the FDC is illustrated in Fig. 1. In the case of installations for the mass transport, the axial remixing of the liquid phase is an undesirable side effect. In order to eliminate this shortcoming, we have designed the multiple stage fluid disperse column shown in Fig. 1b.

### Conclusions of the Scientific Literature on FDC

In the last 20 years, numerous publications dealt with three phase fluid disperse systems. Comprehensive critical appraisals have been published by MUROYAMA and FAN [2] and K. MOLNÁR [3].

Studying the literature, it may be established that the use of three phase counterflow fluidization provides an efficient means for the achievement of the continuous contact of gas (vapour) and liquid phases and for the mass transport between the phases. This efficient procedure and installation may be adequate for industrial separations while at the same time fulfilling environmental control tasks. An in-depth study of the literature



Fig. 1. Fluid disperse column

will also reveal that the more widespread use of these procedures is hindered by the fact that we do not know the true relationships that would propel us towards finding the optimal solution. In many cases we miss even the empirical relationships. Researches referring to the effect of scaling are also lacking almost completely. It would be desirable to study the influence of the shape and density of the packing material, the influence of the surface active materials. But first of all, we need relationships that would permit the design of economical equipment to be used in various industrial absorption and desorption processes and in environmental control.

## Hydrodynamic Properties of the FDC

When resorting to the use of the three phase fluid disperse method, one has to know the evolution of the pressure drop in the bed, in function of the physical variables and geometrical characteristics. For this purpose, the knowledge of the retained liquid hold up is necessary. In order to design the optimum equipment, one also has to know the gas and liquid flow rate region in which the solid packing particles are in a fluidised state. The minimum fluidization rate and the flooding rate should be also known. The knowledge of the above will allow the determination of the necessary operational velocity, which in the case of a given gas and liquid flow rate will determine the required cross-section of the equipment. Many specialists discuss the subject of axial remixing of the liquid phase within the framework of hydrodynamic properties. Since the overwhelming majority of the models describing the axial remixing is based on the turbulent diffusion theory, we think that this can be better discussed within the framework of the material transfer properties of the FDC.

# **Description of the Measuring Station**

In order to examine the hydrodynamic properties outlined in the title more thoroughly, we set up a pilotplan, experimental FDC measuring station. The detailed description of the measuring station can be found in the work of K. MOLNÁR [3]. FDCs proved to be suitable for building up a multilevel system, thus making possible the study of the influence of the cascade number. This latter is rather poorly treated in the literature. The main characteristics of the packing (spheres) used in the FDC were the following:

 characteristic size (diameter):	$d_p$	=	8 –	25	$\mathbf{m}\mathbf{m}$	
 density of the sphere:	$\sigma_s$	=	880		1050	$kg/m^3$

The density of the packing used by us is generally greater than that mentioned in the literature. According to our experience, the density of the packing material plays an important role from the point of view of the stable operation. This is explained by the fact that the lower the density of the solid phase as compared to that of the liquid phase, the more often the undesirable side effects occur which are manifest in the tendency of packing elements to float. This leads to a premature flooding and offsets the other advantages gained from the operation. At the same time it is obvious that if we desire to obtain a small-size column, we have to allow a relatively greater gas velocity in the system. Consequently, we need packings with greater size and density in order to ensure the fluidised state.

The experiments were carried out in an FDC with  $D_c = 130$  mm diameter and the results obtained were checked on an FDC with  $D_c = 500$  mm diameter, industrial size FDC. Based on the above considerations, the region of validity of the experiments is:

$$\frac{D_c}{d_p} = 5.2 - 40.$$

The hydrodynamic characteristics were determined with an air-water system and for the study of the influence of the dynamic viscosity of the liquid phase, aqueous glycerine solutions were used.



Fig. 2. Pressure drop of the gas phase in function of gas flow rate

Fig. 2 shows the evolution of the pressure drop in function of the gas and liquid loading. It can be seen from the figures that, as compared to the dry characteristic curve, the curves obtained are shifted. The pressure drop at the same gas loading is increased, while with the increase of the liquid loading, the minimum fluidization rate decreases. It can be also seen that in the region of fluidization at any liquid loading, following the increase of the gas loading, the pressure drop decreases until flooding is triggered, after which the pressure drop increases rapidly. We believe that this phenomenon may originate from the decrease of the liquid hold up quantity. From this we conclude that when examining the hydrodynamics of the FDC, in contrast to what is mentioned in the literature, we have to take the flow rate and material characteristics of the gas phase into account.

In Fig. 3, we show the influence of the cascade number upon the pressure drop in the fluidization region. It can be seen from the figure that due to an increase of the cascade number which is accompanied by the decrease of the bed height within a given step and while considering



Fig. 3. Influence of cascade number on pressure drop

 $H_{st} = nh_{st} = \text{constant}$ , we find that the pressure drop has decreased. This may be explained again by the decrease of the quantity of liquid hold up.

## Study of the Liquid Hold Up

Based on the characteristic curves shown above and considering the research reports of K. MOLNÁR [4] containing the results of a large number of hydrodynamic studies, it can be established that besides the geometrical design and the liquid loading, the quantity of liquid hold up also depends on the flow rate and material characteristics of the gas phase and the cascade number.

The quantity of liquid hold up was measured by a cubage method. Alternatively we have measured this quantity by the analysis of the response function obtained for the interference of a unit impulse. Almost 250 experimental measurement series were made. As an example, in *Figs.* 4 and 5 we show the evolution of the liquid hold up in function of the gas and liquid loading of the system.

The relations used for the calculation of the liquid hold up quantity were reviewed by K. MUROYAMA and L. S. FAN [2]. We have summarized these in *Table 1* giving also the regions of validity of each relation. We have examined the applicability of the suggested relationships, keeping in mind that in many cases their validity region considerably differs from their own measurement region and not forgetting that they refer only to a singlestage bed. In *Figs. 6, 7, 8, 9*, and 10 we show the measured liquid hold up



Fig. 4. Liquid hold up in function of gas flow rate

quantities in function of the values calculated using the relationships from *Table 1*. From the examination of the figures, we conclude that none of the suggested relationships is applicable to FDC.

In order to devise a relationship that could be used for the calculation of the liquid hold up quantity, we used the method of dimension analysis. Our choice was dictated by the fact that even though we do not know the mathematical model of the phenomenon under investigation, we do know the characteristic physical variables. We used the conclusion of the similarity method which says that the solution of the mathematical model describing the phenomenon may be given in the form of a functional relationship consisting of dimensionless numbers. Thus, the primary task was the selection (i.e. choice) of the set of dimensionless variables from the physical variables characteristic to the phenomenon.

The set of the physical variables is:

$$\varepsilon_{L,st} = \varepsilon_{L,st}(d_p, D_c, n, h_{st}, \rho_L, \mu_L, \nu_L, \rho_G, \mu_G, \nu_G, \rho_s, g).$$
(1)



Fig. 5. Liquid hold up in function of liquid loading



Fig. 6. Comparison of calculated and experimental data (GELPERIN)

Following the dimensional analysis, the set of the dimensionless variables is:

$$\varepsilon_{L,st} = \varepsilon_{L,st}(Re_L, Re_G, Fr_L, \frac{D_c}{d_p}, \frac{h_s t}{d_p}, \frac{\rho_G}{\rho_L}, \frac{\rho_s}{\rho_L}, \frac{\mu_G}{\mu_L}, n).$$
(2)

In light of the above, the constants of the exponential function below should be determined experimentally:

$$\varepsilon_{L,st} = CRe^a_GRe^b_LFr^c_L(\frac{D_c}{d_p})^d(\frac{h_{st}}{d_p})^{f^*}(\frac{\rho_s}{\rho_L})^{g^*}n^{h^*}$$
(3)



Fig. 7. Comparison of calculated and experimental data (KITO I)

where:

$$Re_G = \frac{v_G d_p \rho_G}{\mu_G} = \frac{G_G d_p}{\mu_G} \tag{4}$$

is the Reynolds number referring to the gas phase charge,

$$Re_L = \frac{v_L d_p \rho_L}{\mu_L} = \frac{G_L d_p}{\mu_L} \tag{5}$$

is the Reynolds number referring to the liquid phase charge, and:

$$Fr_L = \frac{v_L^2}{d_p g} = \frac{G_L^2}{d_p g \rho_L^2} \tag{6}$$



Fig. 8. Comparison of calculated and experimental data (KITO II)

is the Froude number referring to the liquid phase charge. The quantity of liquid hold up referring to the static bed height, as featured in Eq. (3):

$$\varepsilon_{L,st} = \frac{\sum_{k=1}^{n} V_{L,k}}{H_{s,t} \cdot T},\tag{7}$$

where

$$T = \frac{D_c^2 \cdot \Pi}{4}$$

is the empty cross-section of the FDC.



Fig. 9. Comparison of calculated and experimental data (UYSAL)

From the numerous experimental results mentioned earlier, we have determined the constants of Eq. (3). The details of the determination may be found in the work of K. MOLNÁR [3].

When using fluid disperse systems with stages, for the calculation of the quantity of liquid hold ups we suggest the following relation:

$$\varepsilon_{L,st} = 186.23 Re_G^{-0.33} Re_L^{-0.075} Fr_L^{0.1625} (\frac{h_{st}}{d_p})^{-0.35}$$
$$\cdot (\frac{\rho_s}{\rho_L})^{0.18} (\frac{D_c}{d_p})^{-0.485} n^{-0.53}$$
(8)



Fig. 10. Comparison of calculated and experimental data (VUNJAK-NOVAKOVIC)

The region of validity of the above equation is:

$$f \le 0.7,$$
  
 $0.88 < \frac{\rho_s}{\rho_L} < 1.15$   
 $6.5 < \frac{D_c}{d_p} < 20,$   
 $1 \le n \le 10, \text{ and}$ 



Fig. 11. Comparison of the measured and calculated data

the velocity region of the gas and liquid phases which ensure the fluidised state.

In Fig. 11 the adequacy of Eq. (8) for the description of the measurement points is demonstrated. For the calculation of all the measurement points, Eq. (8) may be used with an accuracy of  $\pm 10\%$ .

### Calculation of the Pressure Drop in the Fluidization Region

In order to calculate the pressure drop similarly to BARILE and MAYER [7], KITO et al. [8] as well as VUNJAK-NOVAKOVIC et al. [10], we have assumed



Fig. 12. Comparison of the measured and calculated pressure drop values

that in the equilibrium stage, the pressure drop may be calculated using the following equation:

$$\Delta p_c \cong (\rho_s \varepsilon_s + \rho_L \varepsilon_L) g H = (\rho_s \varepsilon_{s,st} + \rho_L \varepsilon_{L,st}) g H_{st}. \tag{9}$$

The relation (9) may be used for the calculation of the pressure drop when we know  $\varepsilon_{L,st}$  – which represents the quantity of the liquid hold up. For the calculation of the quantity of liquid hold up in the FDC, we devised Eq. (8). Thus, if the mass and the characteristics of the solid aggregated charge are known, then the pressure drop may be calculated:

$$\frac{\Delta p_c}{H_{st}} \cong (\rho_s \varepsilon_{s,st} + \rho_L \varepsilon_{L,st}) \cdot g, \tag{10}$$



Fig. 13. Evolution of the pressure drop in the case of a suspension

where:

$$\varepsilon_{s,st} = \frac{V_s}{H_{st}T} = \frac{m_s}{\rho_s H_{st}T} \tag{11}$$

and  $\varepsilon_{L,st}$  may be calculated from Eq. (8).

During our experimental studies, we have measured the pressure drop in more than 500 cases and we compared the results with the calculations obtained from Eq. (10). The comparison of the experimental and calculated values is shown in Fig. 12. From this illustration can be seen that the pressure drop values calculated based on Eq. (10) are within  $\pm 10\%$  of the measured values. Thus, we conclude that the calculation of the pressure drop is adequate enough for practical applications. Since the pressure drop values may be expressed with the above equation, we conclude that the relation (8) devised for the calculation of the quantity of liquid hold up may be also used successfully.

One of the main advantages of the FDC systems as compared with the towers with resting bed charges is that they allow the contact of the



Fig. 14. Comparison of the calculated and measured values in the case of a suspension

gas (vapour) and the suspension phases. This is possible because the presence of the solid particles in the liquid phase, in the present case, does not block the charge. This phase contact method becomes ever more important in solving environmental problems. Because of the above, we have extended our studies to comprise such phase contact cases and we have determined the influence of the solid particles present in the liquid phase upon the evolution of the pressure drop within the system. These results are summarized in *Fig. 13*.

We have found that because of their continuous movement and the resulting collisions, the fluidised charge particles have a self-cleaning effect, thus preventing the blockage. We have adjusted the relationship (10) given for the calculation of the pressure drop so that it may be used even in the cases when the liquid phase contains solid particles:

$$\frac{\Delta p_c}{H_{st}} \cong (\rho_s \varepsilon_{s,st} + \rho_{L+sz} \varepsilon_{L+sz,st}), \tag{12}$$

where the average density of the suspension is:

$$\overline{\rho}_{L+sz} = \frac{1}{\frac{1}{\rho_{sz}}\omega + \frac{1}{\rho_L}(1-\omega)}$$
(13)

and the concentration of the suspension is:

$$\omega = \frac{G_{sz}}{G_L + G_{sz}}.$$
 (14)

In relation (12) we noted with  $\varepsilon_{L+sz,st}$  the amount of the suspension retained, which may be calculated from (8) in such a way that we replace the liquid density with the suspension density as calculated from (13). In *Fig. 14* we show the comparison of the results of the above calculations with experimental data. The figure proves the applicability of relation (12).

## Conclusions

For designing and dimensioning a fluid disperse column, we have to know the hydrodynamic and diffusional (mass transport) characteristics. With the knowledge of the hydrodynamic characteristics, one is able to design the necessary flow cross-section of the installation, calculate the operating range and the liquid hold up quantity as well as the pressure drop of the gas phase. We have worked out calculating relationships between the hydrodynamic characteristics in order to estimate the liquid hold up quantity and the pressure drop. We have proven that the FDC may be used very efficiently when the liquid phase contains solid particles. For the latter case we have also devised a relationship for the calculation of the pressure drop. Further hydrodynamic and material transfer characteristics will be discussed in a subsequent publication.

#### Notations

- a constant
- b constant
- c constant
- C constant
- $d_p$  diameter of packing element
- $D_c$  diameter of column
- e constant
- f free cross-section factor of sustaining net
- $f^*$  constant
- Fr Froude number
- g constant
- $g^*$  gravity field
- G mass flow density
- h within bed cascade height
- $h^*$  constant
- H bed height
- n cascade number
- Re Reynolds number

- v velocity
- V hold-up volume

## Indexes

- c referring to the column
- G gas (vapour) phase
- L liquid phase
- k referring to cascade k.
- *p* referring to particle
- s solid phase
- st static
- sz suspension

#### References

- 1. MOLNÁR, K.: Fluidised Bed Columns for Gas/Liquid Countercurrent Contacting, World Cong. III. of Chem. Eng., Tokyo, Japan, Vol. II. p. 684 (1986).
- 2. MUROYAMA, K. FAN, L. S.: AIChE Journal Vol. 31, No. 1. pp. 1-35 (1985).
- 3. MOLNÁR, K.: Three-Phase Fluid Disperse Systems, PhD Thesis, Budapest, (1990).
- MOLNÁR, K. ÖRVÖS, M. SKRIBA, Z.: Transport Phenomena in Fluid Disperse Systems I. Part. OTKA 5-104. Research Report, BME Vegyipari Gépek és Mg. Tanszék, (Department of Chemical and Food Engineering)(In Hungarian) Bp. (1988).
- 5. CHEN, B. H. DOUGLAS, W. J. M.: Can. J. Chem. Eng., Vol. 46, p. 245 (1968).
- GEL'PERIN, N. J. SAVCHENKO, V. J. GRISHKO, V. Z.: Theor. Found. Chem. Eng., Vol. 2, p. 65 (1968).
- 7. BARILE, R. G. MEYER, D. W.: Chem. eng. Prog. Symp. Ser., No 119, p. 67, p. 134 (1971).
- 8. KITO, M. TABEI, K. MURATA, K.: Ind. Eng. Chem. Process Des. Dev. Vol. 17, p. 568 (1978).
- 9. KITO, M. et al.: Kagaku Kogaku Ronbunshu, Vol. 2, p. 476 (1976).
- UYSAL, B. Z.: Hydrodynamic and Particulate Recovery Studies in Mobile-Bed Contacting, Ph. D. Thesis, McGill Univ. (1978).
- 11. VUNJAK-NOVAKOVIC, G. V. VUKOVIC, D. V. : Fluidization, G. R. Grace and G. M. Matsen, Eds. 253, Plenum Press, New York (1980).