

# A COMPARATIVE STUDY OF INTERFACIAL AREA OBTAINED BY PHYSICAL AND CHEMICAL METHODS IN A BUBBLE COLUMN

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## ABSTRACT

Determination of interfacial areas in absorption processes have been experimentally investigated in a bubble column using a physical method (photographic method) and a chemical method (Danckwerts' method). Experiments were conducted in a vertical column 11.3 cm in diameter and 108.6 cm in height. The range of variables used is common for both methods. All experiments were carried out in batch regime. All values of area were correlated with variables grouped in dimensionless modules that reproduce experimental values with deviations below 9%, and these values obtained by the photographic method are of the same magnitude as those obtained by the chemical method, maximum deviation of these values being 10 %.

## INTRODUCTION

For the experimental determination of the mass transfer coefficient ( $k_L$ ) it is inevitable to first know the gas-liquid interfacial area of the contact system. One could think that knowing the value of the product  $k_L a$  would be sufficient for the calculation and design of gas-liquid absorbers and reactors. However, the knowledge of the area can be so important as the knowledge of the mass transfer coefficient to analyse thoroughly the mass transfer process and the influence of the different variables. Interfacial area can be determined of different ways depending on the gas-liquid contact system employed. In many cases it is possible to determine it by geometric considerations, while in other cases, it is necessary to determine it experimentally. The determination of interfacial areas is possible by one of the existing methods [1-5].

If the absorption process is accompanied by a fast chemical reaction, the interfacial area becomes the principal design criteria. The area can vary in a wide range, depending on the type of contact equipment, physical and chemical properties of the gas-liquid system, and on the operation conditions. Thus, specific techniques are

necessary for its determination, numerous methods being proposed, each one with its advantages and disadvantages. The different methods used in the determination of interfacial areas can be classified as physical [6-11] or chemical methods [12-14]. The first methods are based on modifying some physical property, while the second methods are based on carrying out a chemical reaction of known kinetics, in such a way that the absorption rates be proportional to the area. The results obtained using different methods, physical and chemical, have sometimes given contradictory results [12, 15].

In the present study, the interfacial area is determined by two different methods, the Danckwerts' method and the photographic method, analysing the influence of the different variables on the area and obtaining the corresponding correlations.

## MATERIAL AND METHODS

### Mass transfer measurements

The absorption experiments were carried out in the set-up that, with the exception of the contact device, has been described in detail in previous studies [16]. The bubble column, (Fig. 1), made of methacrylate, 108.6 cm high and 4 mm thick. The internal and external diameters of the column are 11.3 cm and 14.8 cm, respectively. The gas distributor was a porous plate, 4 cm in diameter. The size of the bubbles is modified by using porous plates of different equivalent pore diameter, allowing an important variation in the interfacial area. Table 1 shows the relation between porosity and the equivalent pore diameter for each of the porous plates employed. The liquid phase enters the column through the top via a vertical glass tube that is slightly bent at the lower end to avoid gas leakage.

*Table 1. Relation between porosity and the equivalent pore diameter for each of the porous plates employed*

Plate	Equivalent pore diameter · 10 <sup>6</sup> (m)
0	150-200
1	90-150
2	40-90

The gas, pure CO<sub>2</sub>, is led through a humidifier, where, by bubbling, is saturated water vapor at 25 °C. Once saturated, it passes through a bubble flow-meter, where the flow into the column is measured. The gas not absorbed, after making contact with the liquid phase, exits the column, passes to a second bubble flow-meter determining the gas flow at the exit. The amount of gas absorbed is determined by the difference of flow entering and exiting the column. In our experiments, we used inflow between 3 and 8.5 · 10<sup>-4</sup> mol/s. The liquid, previously thermostated to 25 °C, is introduced into the column in loadings of 10.3 l.

### Photographic method

This method consists in photographing different zones of the column during the absorption process and determining the diameter and number of bubbles in this zone. Once this is known, we can calculate the total number of bubbles and area occupied by these that would correspond to the gas-liquid contact area. This method was employed by several authors to determine interfacial areas in a column of spheres and cylinders [17], or to study the reaction of carbonic gas with carbonate-bicarbonate solution in the presence of hypochlorite as catalyst [18].

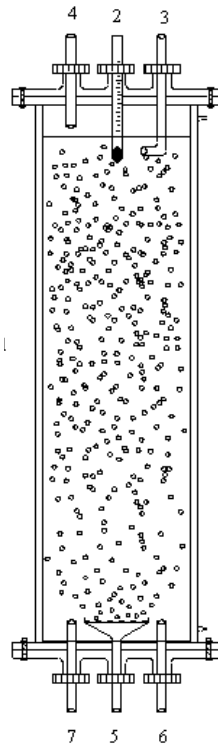


Figure 1. Bubble column scheme: 1. Bubble column; 2. Thermometer; 3. Inflow of liquid ; 4. Outflow of gas; 5. Porous plate; 6 and 7. Outflow of liquid

### Danckwerts' method

The method proposed by Danckwerts requires that the gas absorbed undergo a moderately fast pseudo-first order reaction with some of the solutes of the liquid phase. The value of interfacial area ( $A$ ) is obtained from the slope of the straight line that results from the graphic representation of absorption flux per volume ( $N^2$ ) versus pseudo-first order kinetics constant ( $k_1$ ), while the value of  $k_L$  is calculated from the ordinates at the origin. This method has the advantage that appreciable differences of  $k_1$  are obtained using very small catalyst concentrations thereby not affecting the physical properties of the absorbent liquid [19].

## RESULTS AND DISCUSSION

### Photographic method

Upon application of the photographic method experiments were designed in advance to analyse the influence of the different variables on contact area and to determine the minimum number of experiments needed.

In the last few years, several investigators have used factorial designs to cover problems of this type in other research fields [20, 21]. We have employed a two-level factorial design,  $2^3$ , for the determination of area by the photographic method. We have focused the study on three independent variables of greater influence over interfacial area, superficial gas velocity ( $X_1$ ), pore size ( $X_2$ ) and surfactant concentration ( $X_3$ ), whose maximum and minimum values are: Superficial gas velocity:  $0.8 \cdot 10^{-3} - 1.8 \cdot 10^{-3}$  m/s; Porous plate: 0 - 2; Concentration of sodium lauryl sulphate (SLS):  $5 \cdot 10^{-5} - 5 \cdot 10^{-4}$  % mass. For mathematical studies it is necessary to use normalised independent variables. The correlation of the variables will be given, depending on the mathematical model, in the following form:

$$y = b_0 + b_1 X_1 + b_2 X_2 + b_3 X_3 + b_{12} X_1 X_2 + b_{13} X_1 X_3 + b_{23} X_2 X_3 \quad (1)$$

where:  $y$  is the dependent variable;  $X_i$  are the normalised independent variables;  $b_i$  and  $b_{ii}$  are the model coefficients.

Photographs were taken of the column during the absorption process, for different operating conditions, gas flow, liquid phase, and pore size. Several photographs were taken for each system and in different zones of the column. For each photograph, a count of the bubbles was taken in a prefixed interval, and their diameters were determined (see fig 2). Contact area was calculated with these data. Each experiment was performed five times.

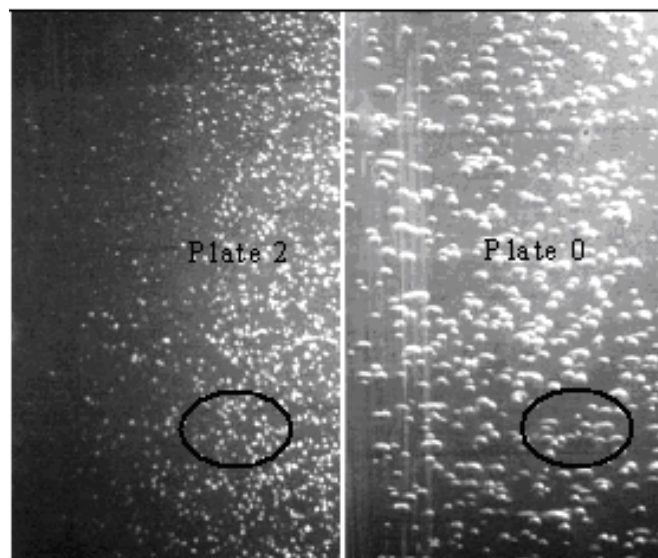


Figure 2. Photograph of bubbles of porous plate 2 and 0.

The data obtained from the photographs is collected, along with those of calculation of the gas-liquid interfacial area. The proposed equation is the following:

$$y = 11.520 + 4.690 X_1 + 1.095 X_2 - 0.470 X_3 + 0.513 X_1 X_2 - 0.310 X_1 X_3 + 0.100 X_2 X_3 \quad (2)$$

Pareto Chart is shown in figure 3, it is a histogram showing the influence of each independent variable over the dependent. The principal effects are presented standardised (effects divided by standard error). All those effects that possess a value superior to level 0.05 (value indicated on the graph) are significant at that level.

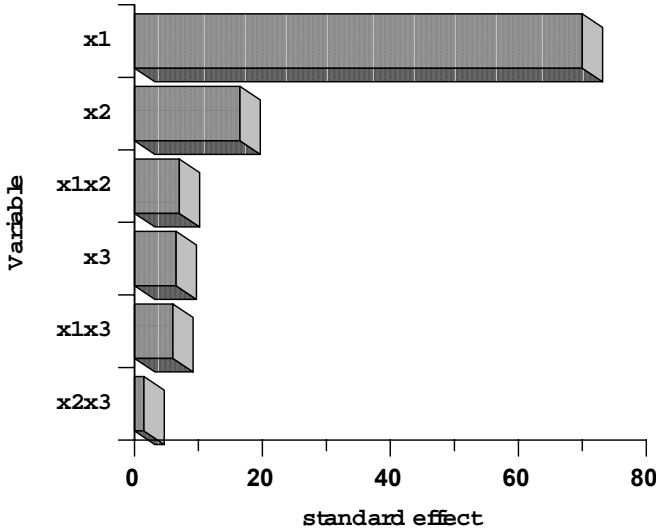


Figure 3. Pareto chart.

Another way of analysing the influence of the three variables is by response surfaces, in which the influence of two variables over the parameter of study is analysed simultaneously. Next, the response surfaces are presented as a function of superficial gas velocity and of plate, of superficial gas velocity and SLS concentration, and of plate and SLS concentration (Fig. 4). In these graphs, we observe that as we increase the levels of the variables, yield becomes greater, becoming pronounced when one of the variables is superficial gas velocity.

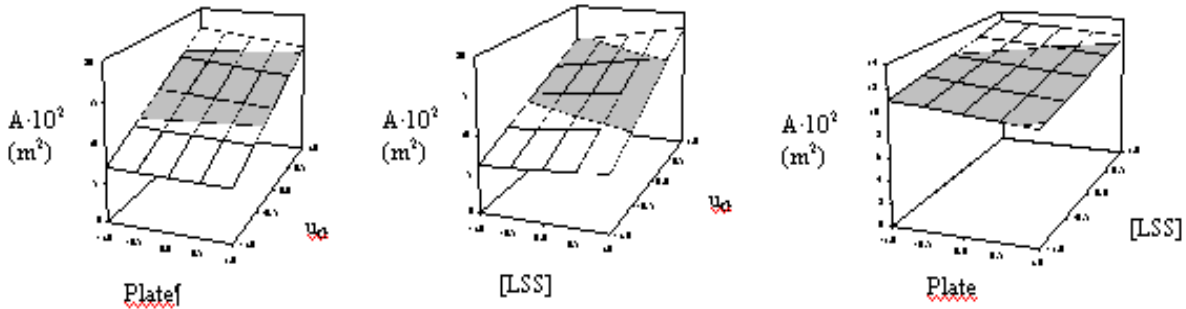


Figure 4. Response plots

### Danckwerts' method

For its application, experiments in sodium carbonate-bicarbonate buffer solutions (0.5 M-0.5 M) with different surfactant concentrations were made. We observe that interfacial area decreases with increasing concentration of sodium lauryl sulphate and with pore diameter, and increases with gas flow. This behaviour being the same as that observed in the photographic method. In figure 5, as an example, interfacial area versus surface tension is shown. The variation indicates a decrease with SLS addition; this behaviour is due to that when surfactant is present in the medium, turbulence decreases and therefore area is lower. Interfacial area varied proportionally with gas flow; this behaviour is similar for all solutions and porous plates employed. In figure 6 this variation is manifested:

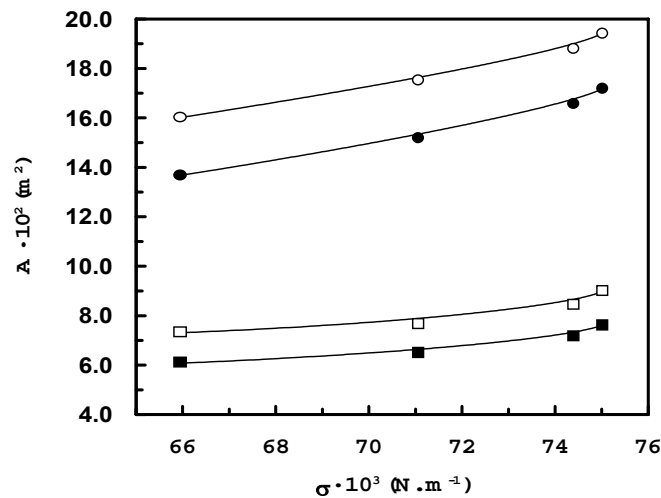


Figure 5. Dependence of interfacial area,  $A$ , on surface tension, pore diameter and gas flow rate: (●)  $1.81 \cdot 10^{-3} \text{ m/s}$ , plate 0; (■)  $0.87 \cdot 10^{-3} \text{ m/s}$ , plate 0; (○)  $1.81 \cdot 10^{-3} \text{ m/s}$ , plate 2; (□)  $0.87 \cdot 10^{-3} \text{ m/s}$ , plate 2.

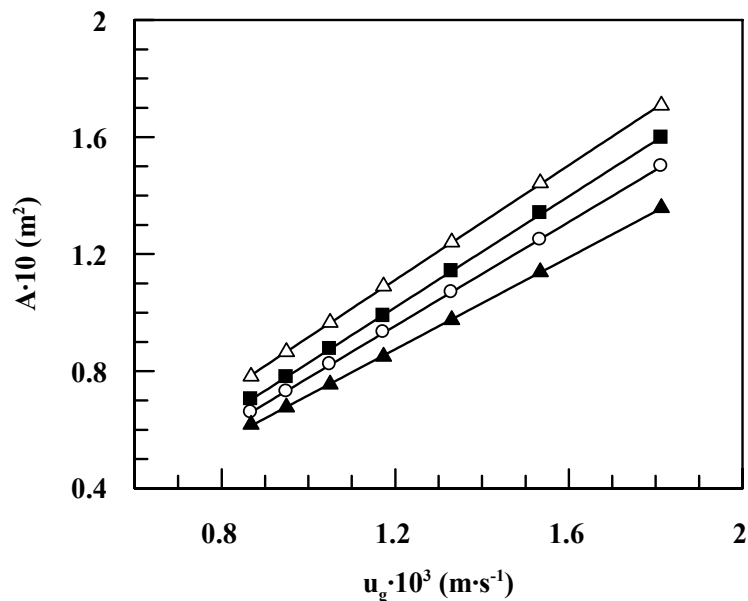


Figure 6. Dependence of  $A$  on superficial velocity of gas and surfactant concentration for plate 0, (△) 0 % mass SLS; (■)  $0.5 \cdot 10^{-4} \text{ % mass SLS}$ ; (○)  $1 \cdot 10^{-4} \text{ % mass SLS}$ ;

(▲)  $1.5 \cdot 10^{-4}$  % mass SLS.

To complement this analysis, we have undergone a fit for interfacial area as a function of dimensionless numbers. The equation obtained is the following:

$$A = K_1 \cdot Re^{0.98} Sc^{0.57} Fr^{0.09} Bo^{-0.70} \left( \frac{d_p}{d_c} \right)^{-0.19} \quad (3)$$

where  $K_1$  has a fixed value equal to  $6.12 \cdot 10^{-2} \text{ m}^2$  for all solutions and plates. The values calculated using Eq. (3) for the three plates, reproduce experimental values with deviations below 8.6 %.

The results obtained by both methods were compared, finding better results in the reproduction of experimental values of area from the chemical method equation than from the photographic method, as shown in figure 7. In this figure we observe that small area values have lower concordance between the values obtained by the physical method and those obtained by the chemical method, these differences being more pronounced for smaller pore size.

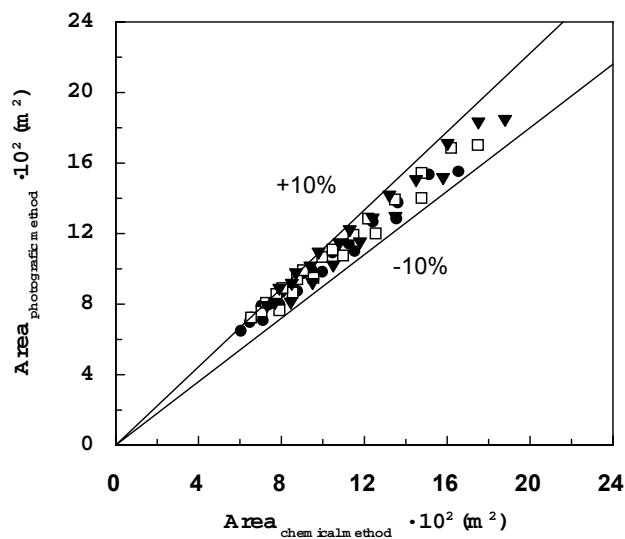


Figure 7. Interfacial area calculated with photographic method against interfacial area calculated with chemical method. (○) plate 0, (□) plate 1, (▽) plate 2.

## CONCLUSIONS

Interfacial areas were determined by the photographic method and a chemical method. A non-linear regression statistical analysis was performed to analyse the influence of distinct variables on interfacial area, superficial gas velocity being the most significant variable in the values obtained by the physical method. The values for area obtained by the chemical method were correlated by means of an equation based on dimensionless modules.

The presence of surfactants significantly decreases mass transfer and, therefore,

interfacial area values. The decrease in area was more pronounced for smaller pore size. In comparing the obtained area by the Danckwerts' method with respect to the photographic method, maximum deviations below 10 % result.

## NOTATION

A	Effective interfacial area, (m <sup>2</sup> )
d <sub>c</sub>	Column diameter, (m)
d <sub>p</sub>	Pore diameter, (m)
D	Gas diffusivity in the liquid phase, (m <sup>2</sup> s <sup>-1</sup> )
g	Gravitational constant, (m s <sup>-2</sup> )
K <sub>1</sub>	Parameter in Eq. (3), (m <sup>2</sup> )
k <sub>1</sub>	Pseudo-first order kinetics constant, (s <sup>-1</sup> )
k <sub>L</sub>	Mass transfer coefficient in the liquid phase, (m s <sup>-1</sup> )
k <sub>L</sub> a	Volumetric mass transfer coefficient, (s <sup>-1</sup> )
N	Absorption flux per volume, (mol l <sup>-1</sup> s)
q <sub>g</sub>	Gas flow rate, (mol s <sup>-1</sup> )
u <sub>G</sub>	Superficial gas velocity, (m s <sup>-1</sup> )

### *Dimensionless numbers*

Bo	Bond number, $\frac{g d^2 \rho}{\sigma}$
Fr	Froude number, $\frac{u_g^2 d}{g}$
Re	Reynolds number, $\frac{\rho u_g d}{\mu}$
Sc	Schmidt number, $\frac{\mu}{\rho D}$

### *Greek symbols*

μ	Viscosity of liquid phase, (kg m <sup>-1</sup> s)
ρ	Density of liquid phase, (kg m <sup>-3</sup> )
σ	Surface tension of liquid phase, (mN m <sup>-1</sup> )

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