

The Effect of Surfactants on Characteristics of Hydrodynamic and Mass Transfer Coefficient in Gas-Liquid Dispersion Column

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Abstract

This work studies the effect of liquid properties (water, solution 0.2-1.5 wt % for each of i-propanol, and n-butanol), temperature (26-55 °C) and superficial air velocity (1-15cm/s) on the gas holdup and mass transfer coefficient in air-liquid dispersion column. The experimental procedure was carried out by QVF column (10 cm i.d, 1.5 m height) and air bubbling by multi-orifice distributor (2 mm, 49 holes in square pitch). The experimental results are presented in two and three dimension graphs, these graphs indicate, increases in holdup and mass transfer coefficient with increasing weight percent of alcohols, temperature, and superficial velocity of air. The gas holdup and mass transfer coefficient are in the following order n-butanol > i-propanol > water. Also the experimental results were correlated by fitting empirical correlations.

Some of the results have been correlated on the basis of drift flux model in order to express the effect of surface active agent on radial uniformity flow and gas holdup profiles.

Keywords: Gas-liquid dispersion column, surface active agent, gas holdup, mass transfer coefficient.

تأثير المواد الخافضة للتوتر السطحي على الخصائص الهيدروديناميكية و معامل انتقال الكتلة الحجمي في برج التماس غاز - سائل

الخلاصة

كرس العمل لدراسة تأثير خواص السوائل على اداء برج التماس بين الغاز والسائل باستخدام موزع هواء متعدد الفتحات من خلال حساب كمية الغاز المحتجز و معامل انتقال الكتلة الحجمي. تم الخذ بالاعتبار ثلاثة متغيرات هي، تركيز الكحول في الماء، درجة حرارة السائل و سرعة الهواء الظاهرية. لغرض بحث طبيعة الاندماج للطور السائل تم استخدام عدة سوائل وهي ماء و ايزوبروبانول - ماء و بيوتان اعتيادي - ماء بالنسب الوزنية من ٠,٢ % الى ١,٥ % لكل محلول على حدة، بينما كان الطور الغازي هو الهواء. سرعة الهواء الظاهري و درجة حرارة المحلول المستخدمة في العملية كانت ضمن المدى (١ - ١٥) سم/ثانية، و (٢٦ - ٥٥) م° على التوالي. تم الاستنتاج من الدراسة ان تأثير سرعة الهواء الظاهرية اكثر بكثير من كل من درجة حرارة المحلول و تأثير الكحولات المضافة على كل من حساب كمية الغاز المحتجز و معامل انتقال الكتلة الحجمي. بعض النتائج تم ربطها بعلاقة رياضية باستخدام (Drift Flux Model) لتوضيح تأثير الكحولات المضافة على انتظام الجريان القطري وتوزيع كمية الغاز المحتجز.

Notation C = concentration of dissolved oxygen, kmol/m³ C_o = distribution coefficient, Eq.(3) K_{La} = volumetric liquid phase mass transfer coefficient, s⁻¹ t = time, s T = operating temperature, °C U_b = bubble rise velocity, cm/s U_g = superficial gas velocity, cm/s wt = weight percent of surfactant in water wt_i, wt_n = weight percent of surfactant (i-propanol, and n-butanol in water) Z_o, Z_1, Z_3 = liquid levels in the manometers, mm**Greek letters** ε_g = gas holdup**Subscripts** i = initial condition s = saturation condition**Introduction**

Gas-liquid dispersion columns are widely used in industrial gas-liquid operations (e.g. gas-liquid reactions, agitation by gas injection, fermentations, etc.) in chemical and biochemical process industries, due to their simple construction, low operating cost and high-energy efficiency (Mouza et al. 2004)

Fractional gas holdup is an important parameter in the design and scale-up of gas-liquid dispersion. It has several direct and indirect influences on the column performance. The direct and obvious effects are on the column volume, this is because the fraction of the volume is occupied by the gas and the respective phase volume becomes important depending upon the phase in which the rate controlling step takes place in the mass transfer process (Thorat et al. 1998).

Bubble coalescence in aqueous solutions of alcohols and electrolytes was significantly hindered by increasing solute concentration. The

inhibitory effect of alcohols on bubble coalescence increases with the increasing length of the carbon chain (Zahradnik et al. 1999). Coalescence of bubbles in gas-liquid dispersions will be inhibited, when the liquid phase is not pure component, but a mixture, as has been reported by Keitel and Onken (1982).

The influence of liquid properties on gas holdup is considerable and is very dependent on the sparger type. The liquid properties which are most important are the coalescing or non-coalescing properties, surface tension and the viscosity of the medium [Akita and Yoshida (1973), and Hikital et al. (1980)]. The major difference between the behavior of a gas-liquid dispersion column operating with foaming liquids and that operating with non-foaming liquids is the bubble dynamics. The surface active impurities in the presence or absence of foam allow the formation of smaller non-coalescing bubbles (Shah et al. 1985).

Camarasa et al. (1999) studied experimentally the effect of liquid phase properties and gas distribution on bubble and hydrodynamic characteristics of gas-liquid dispersion columns. Various measuring techniques used for, systematic measurements of bubble size, velocity, frequency, and gas holdup. Three spargers are used, a single-orifice sparger (5 mm in diameter), a multi-orifice sparger (62 holes of 1 mm uniformly space) and a porous glass plate (10 to 16 μm mean pore diameter, 5 mm in height). Three systems were used, air-water, air-aqueous solution (butanol), and air-aqueous solution (pentanol).

Deckwer et al. (1974) indicated that the knowledge of the residence time distribution of the phase is necessary to determine the volumetric mass transfer coefficient. In tall and small diameter bubble columns, the determination of volumetric mass transfer coefficient should be based on the concentration profiles measured at co-current or counter current flow along the column and evaluated by means of the axial dispersion model.

Bouaifi et al. (2001) studied the gas holdup, volumetric mass transfer coefficient, interfacial area, the bubble size, and bubble distribution in two gas-liquid dispersion columns and a non-standard vessel equipped with various dual impeller combinations.

Zou et al. (1988) studied gas hold up in a bubble column operated at elevated temperature for air-water and air-alcohol systems. They found that the hold up increases with increasing operating temperature.

The aim of the present work is to study the effect of different parameters such as weight percent of

solution alcohols addition (i-propanol and n-butanol) (0.2-1.5 wt %), temperature (26-55 °C) and superficial gas velocity (1-15 cm/s), in water-air dispersion column. Gas holdup and mass transfer coefficient were also measured at these variables. It has been predicted that the effect of surfactant is great on the improving these parameters.

Experimental Work

The experiments were carried out with a QVF cylindrical semi batch column of 10 cm i.d. and 1.5 m height. A schematic diagram is shown in Fig.1. Sparger of multi-orifice type was employed with 2mm (49 holes) in square pitch. In all the experiments, liquid phase were (water with aqueous alcohol solutions (water-i-propanol), and (water-n-butanol)) at weight percent of 0.2, 0.6, 0.8, 1.2, and 1.5 wt % respectively.

A compressor supplied air at the bottom of column, and the air superficial velocities were 1, 5, 8, 12, and 15 cm/s. Nitrogen was supplied at the beginning of the experiment at the bottom of column to remove the oxygen from the solution. Ten samples valves were located on the left side of the column in order to take the samples. The system was heated by electric heated at desired temperature, and controlled by means of a controller. A thermocouple was used to measure the temperature of the liquid, and it fixed in the middle of the column. The clear liquid height equal 125cm, and the temperature of solutions were 26, 35, 40, 50, and 55 °C. The liquid was discharged from the bottom of the column using centrifugal pump.

Values of average gas holdup (ϵ_g) were determined from the difference

in hydrostatic pressure along the bed height at two points by using U tube-manometer (Jeng et al. 1986):

$$e_g = \frac{(Z_1 - Z_2)}{Z_o} \dots\dots\dots(1)$$

where Z_o , Z_1 , and Z_2 are the liquid levels in the manometers.

The overall volumetric mass transfer coefficient used in this work is defined with respect to the clear liquid. The liquid in the column was sparged with a sufficient amount of nitrogen from a cylinder until the concentration of dissolved oxygen in the liquid became negligible, then air supply to the column where the dissolved oxygen concentration increased with time. Sample was taken along the column (10 points), with time difference between each one 30s started from bottom to the top, this make each run took 300s to be completed.

Because of the constant agitation by the gas bubbles, it can be assumed that the column is well mixed, so only the liquid phase resistance is influential to the rate of mass transfer. A material balance of oxygen dissolved in the liquid is given in Eq.(2)(Jeng et al. 1986):

$$K_L a = \frac{-2.303(1 - e_g)}{\frac{t}{1} \log \frac{(C_s - C)}{(C_s - C_i)}} \dots\dots\dots(2)$$

where C_s and C_i are the saturated and initial dissolved oxygen concentration respectively. $K_L a$ is obtained from the slope of straight line of Eq.(2) on semi log of graph papers.

Empirical Correlations

The experimental results of gas holdup and mass transfer coefficient for three systems were correlated by computer program of multiple non-linear regressions to fit the relationships. These equations represent the effect of each variables as alone or interaction on holdup or mass transfer coefficient as shown in Table 1.

Results and Discussions

Analysis of The Two Dimension Graphs

Figs.(2, 3, 4, 5, 6, and 7) show the effect of aqueous of alcohol on air holdup and mass transfer coefficient. The air holdup and mass transfer coefficient increase with increasing superficial air velocity for all liquid phase used (water, water-i-propanol, water-n-butanol), but in different ratios, because the coalescence rate in pure water is higher than water-i-propanol, and water-n-butanol mixtures, because the coalescence rate is dependent on the liquid surface properties, so alcohol in water as a surfactant (surface active agents) which hinders bubble coalescence by accumulating at the gas-liquid interface and orienting their hydrophilic group into liquid film surrounding the gas bubble and thus creating repulsive electric forces when two bubbles come close to each other. The concentration of the hydrophilic molecules at the surface increases with surfactant concentration and results in a lower surface tension. Air bubbles in the presence of surface active agents are small, rigid, with lower rising velocity and have a high residence time in the column,

resulting in increase in gas holdup and mass transfer coefficient. These notes

were agreed with the results reported by Keitel and Onken (1982).

The effect of weight percent of i-propanol and n-butanol in water on gas holdup and mass transfer coefficient are shown in Figures (2, and 5), where gas holdup and mass transfer coefficient increase with increasing weight percent of i-propanol and n-butanol, due to increase the concentration of surfactant which cause by the increase of concentration of the hydrophilic molecules at the surface and results in a lower surface tension. This was reported by Keitel and Onken (1982).

Figures (3, and 6) respectively show the influence of superficial gas velocity on gas holdup and mass transfer coefficient. Gas holdup and mass transfer coefficient increase with increasing superficial gas velocity. At higher superficial gas velocity smaller bubbles are formed, with large interfacial areas and a lower rising velocities leading to a large residence time and then higher values of gas holdup and mass transfer coefficient. These indications agree with that shown by Zahradnik et al. (1999).

Experimental results are verified from Figures 4, and 7, the effect of operating temperature on gas holdup and mass transfer coefficient. Gas holdup and mass transfer coefficient increase with increasing operating temperature. This can be attributed to the change of physical properties of the liquid phase with the temperature, for example when the temperature increases, the liquid phase viscosity, and surface tension decrease which lead to a lower the rate of coalescence, this decrease generates

smaller bubbles with a lower rising velocities and large interfacial areas, which gives higher values for gas holdup and mass transfer coefficient. This was reported by Zou et al. (1988) and Levich (1962).

Figures (8, and 9), show a comparison between the experimental and the predicted gas hold up and mass transfer coefficient, the empirical correlation are used to predict the theoretical data for the gas hold up and mass transfer coefficient, there is a good approximation between the two results with error less than 6 %.

Analysis of The Three Dimension Graphs

The experimental results are represented by three dimension graphs as shown in Figures 10 to 23, it is useful to show the optimum surface and which one of the variables more interaction than others.

Figs.(11 and 12) show that at high superficial gas velocity (12 to 16 cm/s), there is negligible effect of both i-propanol wt_i % and operating temperature on the gas hold-up, also in Figure (9), it is clear that the effect of i-propanol wt_i % can be neglected at high operating temperature (52 - 55 °C) on the gas hold-up, because at high velocity the terminal rise velocity can be neglected leading the ϵ_g in equation (3) to approach 1.0, at high temperature, the decrease in the viscosity by the effect of temperature is much greater than the effect of the surfactant which leads to neglect its effect compared to the effect of temperature.

Figure (15), shows that there is a negligible effect of operating temperature of the solution on the mass transfer coefficient at high

superficial gas velocity (12 - 16 cm/s), this because of the high mixing at high gas velocity leads to increase in the mass transfer much greater than the effect of temperature, at high wt_i % of i-propanol as shown in Figure (14), the effect of gas velocity has little effect compared to the effect of the wt_i % of i-propanol, also negligible effect of wt_i % at high gas velocity, this leads to a conclusion that it can be increased the mass transfer coefficient by controlling one of the variables U_g or wt_i % and keep the other constant at the desired value.

Figure (13), on the other hand shows that, to attend the increase in mass transfer coefficient it needs to increase both the operating temperature and wt_i % of i-propanol, this means that the effect of these two variables at high levels can reach the effect of superficial gas velocity.

The same results are predicted for n-butanol water solution [Figure (16 to 21)] and water alone [Figure (22 to 23)] on the three dimension graph.

Analysis of Average Gas Holdup Data Using Drift Flux Model

The Zuber and Findlay (1965) drift-flux model has been commonly recommended for fitting gas holdup data from gas -liquid dispersion column. At zero liquid flow rates, the model yields equation for gas holdup as a function of superficial air velocity [Zuber and Findlay (1965)]:

$$e_g = \frac{U_g}{(C_o U_g + U_b)} \dots \dots \dots (9)$$

where C_o is a distribution parameter accounting for non-uniformity of

flow and nature of the gas holdup profiles, and U_b the terminal rise velocity of a single bubble. By rearranging Eq.(9), it can be written [Zuber and Findlay (1965)]:

$$\frac{U_g}{e_g} = C_o U_g + U_b \dots \dots \dots (10)$$

And hence, if experimental data (U_g/e_g) are plotted against U_g, C_o can be obtained from the slope and U_b from the y-axis intercept. In Figure (24) the Zuber and Findlay (1965) drift -flux, illustrating the effect of i-propanol and n-butanol addition on gas holdup profile (C_o and U_b). Table 2 shows the values of C_o and U_b for water (0 wt% alcohol), and aqueous solutions of i-propanol and n-butanol at weight percent of 0.2 wt%, and 0.6 wt% for operating temperature of 26°C and 40°C. Eq.(10) shows the decrease in the values of C_o, significantly with increasing operating temperature, and increasing weight percent of i-propanol and n-butanol. These results indicate extremely high flow non-uniformity as well as strongly favourable effect of the surface active additives on radial uniformity of flow and gas holdup profiles. Also Eq.(10) shows the decrease in the U_b values with increasing operating temperature, and increasing weight percent of i-propanol and n-butanol reflects an increasing proportion of small bubbles in the solution at increase of operating temperature, and increasing weight percent of i-propanol and n-butanol.

Conclusions

Large effect of liquid phase properties, are very small concentration of surfactant in water, cause a large deviation in gas holdup

and mass transfer coefficient from pure water.

Gas holdup and mass transfer coefficient values for aqueous solution of alcohol (i.e. i-propanol and n-butanol) increase with increasing length of their carbon chain as follow; n-butanol > i-propanol > water.

Fitting well of gas holdup data with the Zuber and Findlay drift flux model indicates extremely high flow non-uniformity as well as strongly favourable effect of the surface active additives on radial uniformity of flow and voidage profiles.

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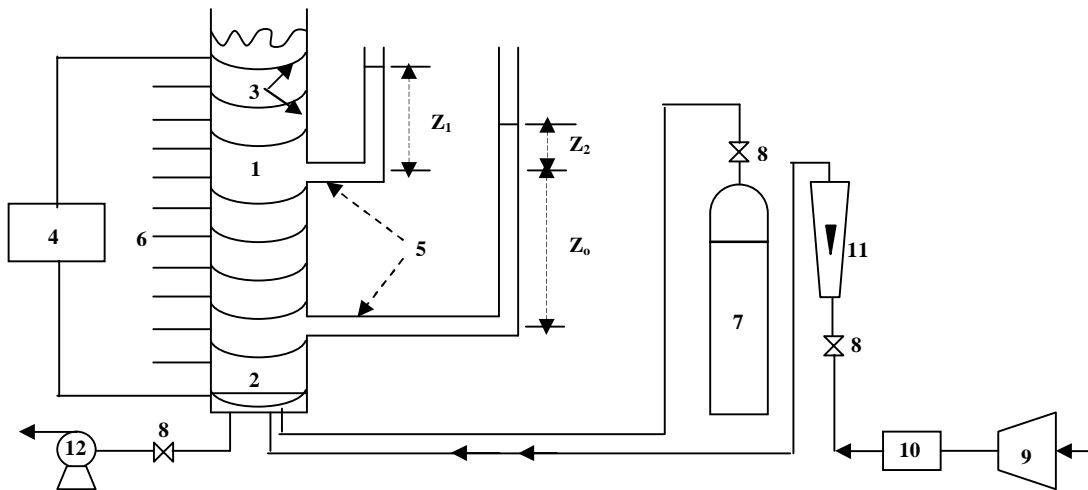


Fig.(1): Experimental set-up: (1) column: (2) sparger: (3) heating tape: (4) temperature controller: (5) manometers: (6) pressure taps: (7) nitrogen cylinder: (8) regulating valves: (9) air compressor: (10) air filter: (11) rotameter: (12) centrifugal pump.

Table 1. Results of regression analysis of gas holdup, and mass transfer coefficient for three different systems.

Air-water system

$$e_g = -0.329266 + 0.023379U_g + 0.020305T + 0.000487U_g^2 - 0.000168T^2 - 0.00039U_gT \dots\dots\dots(3)$$

Correlation coefficient = 0.9471

% Average error = 5.4561%

Standard deviation = 0.021503

$$K_L a = -3.35619 + 0.41314U_g + 0.09002T + 0.000167U_g^2 - 0.000079T^2 - 0.01032U_g T \dots\dots\dots(4)$$

Correlation coefficient = 0.9354

% Average error = 3.7575%

Standard deviation = 0.006057

Air-aqueous i-propanol solution system

$$e_g = -0.45208 - 0.34683wt_i + 0.046265U_g + 0.029237T + 0.026582wt_i^2 + 0.00041U_g^2 - 0.000215T^2 + 0.023459wt_i U_g + 0.003753wt_i T - 0.001362U_g T \dots(5)$$

Correlation coefficient = 0.9468

% Average error = 1.7377%

Standard deviation = 0.013081

$$K_L a = -0.66179 - 0.516256wt_i + 0.09115U_g + 0.029717T - 0.010964wt_i^2 + 0.000261U_g^2 - 0.00007T^2 + 0.049249wt_i U_g + 0.004157wt_i T - 0.003282U_g T \dots(6)$$

Correlation coefficient = 0.9364

% Average error = 2.01755%

Standard deviation = 0.005037

Air-aqueous n-butanol solution system

$$e_g = 0.277498 + 0.242936wt_n - 0.065493U_g + 0.004107T + 0.029132wt_n^2 + 0.000434U_g^2 - 0.000235T^2 - 0.027771wt_n U_g - 0.000774wt_n T + 0.002485U_g T \dots(7)$$

Correlation coefficient = 0.9468

% Average error = 1.7413%

Standard deviation = 0.01426

$$K_L a = -0.441839 - 0.338451wt_n + 0.058033U_g + 0.022016T - 0.011227wt_n^2 + 0.000273U_g^2 - 0.000072T^2 + 0.034249wt_n U_g + 0.002746wt_n T - 0.002154U_g T \dots(8)$$

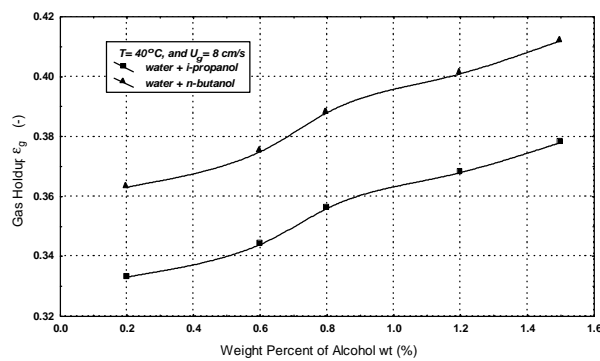
Correlation coefficient =0.9352

% Average error = 2.06213%

Standard deviation= 0.005314

**Table (2): Drift Flux Model Parameters for Water and
alcohol solution.**

Weight Percent of Alcohol (%)	Temperature (°C)	C_o	U_b (cm/s)
0	26	3.28	7.979
0	40	2.088	4.93
0.2	26	2.98	7.249
0.6	40	1.898	4.489
0.2	26	2.74	6.65
0.6	40	1.744	4.112

**Fig.2: Gas holdup versus weight percent of alcohol for three systems.**

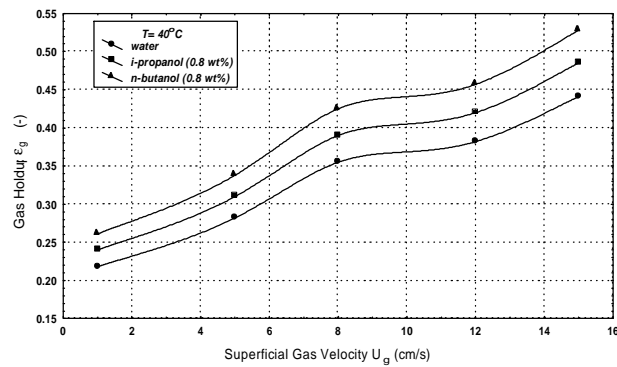


Fig.3: Gas holdup versus superficial gas velocity for three systems.

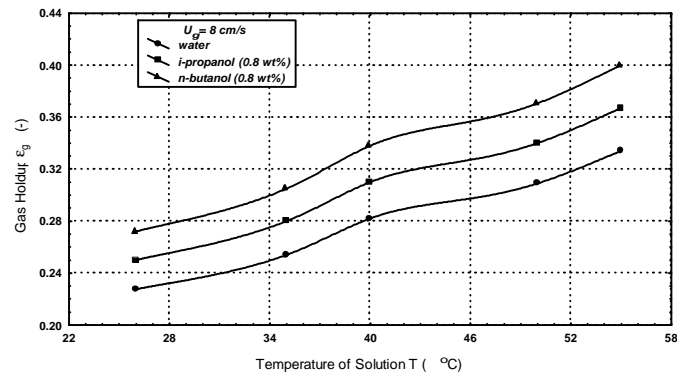


Fig.4: Gas holdup versus temperature of solution for three systems.

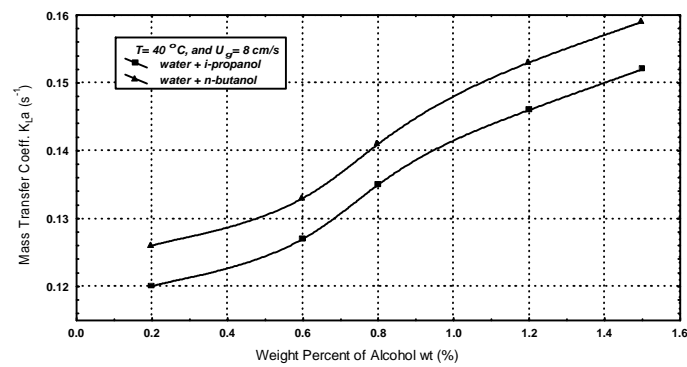


Fig.5: Mass transfer coeff. versus weight percent of alcohol for three systems.

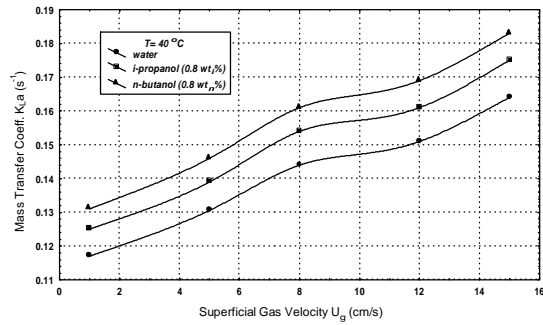


Fig.6: Mass transfer coeff. versus superficial gas velocity for three systems.

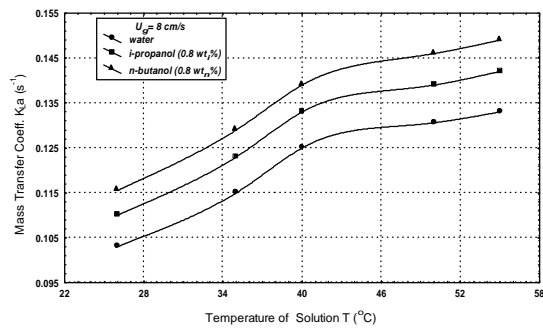


Fig.7: Mass transfer coeff. versus temperature of solution for three systems.

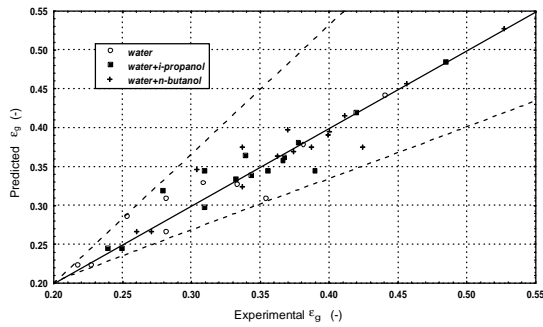


Fig.8: Show the relation between exp. and pred. gas holdup values .

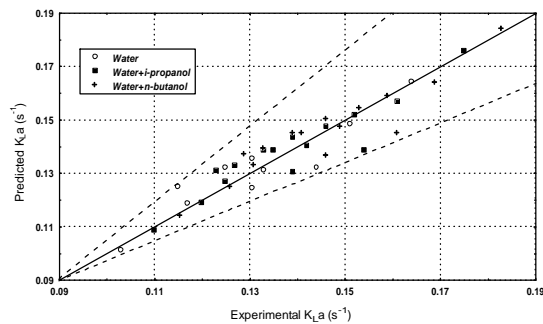


Fig.9: Show the relation between exp. and pred. mass transf. coeff. values.

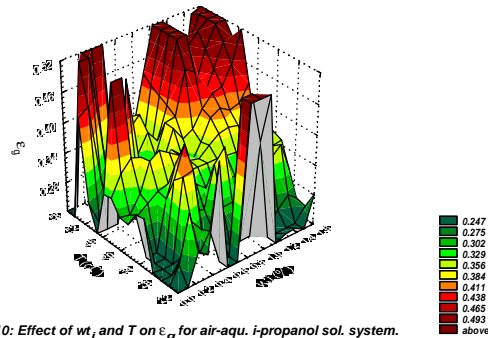


Fig.10: Effect of w_t and T on ϵ_g for air-aqu. i-propanol sol. system.

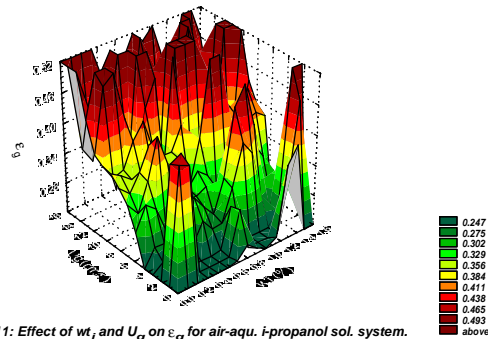


Fig.11: Effect of w_t and U_g on ϵ_g for air-aqu. i-propanol sol. system.

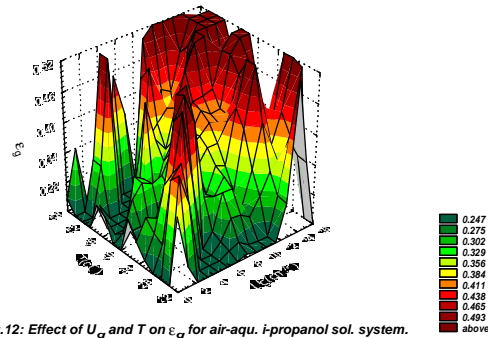


Fig.12: Effect of U_g and T on ϵ_g for air-aqu. i-propanol sol. system.

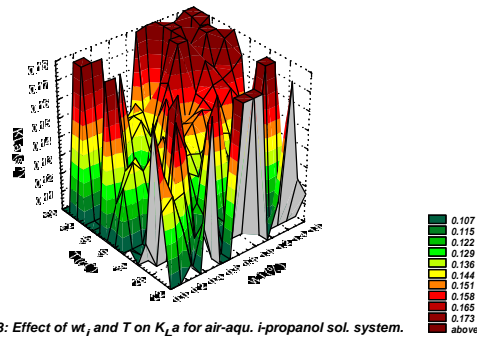


Fig.13: Effect of w_{t_i} and T on K_La for air-aqu. i-propanol sol. system.

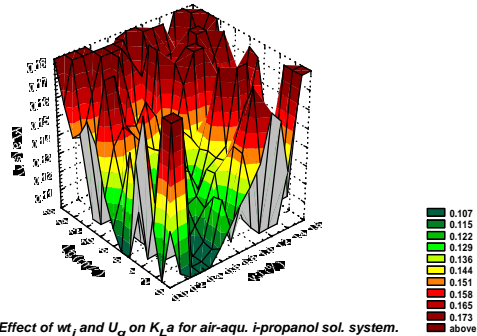


Fig.14: Effect of w_{t_i} and U_g on K_La for air-aqu. i-propanol sol. system.

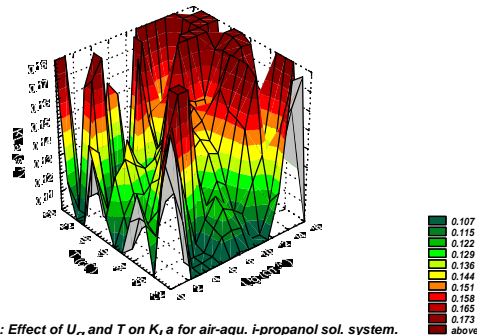


Fig.15: Effect of U_g and T on K_La for air-aqu. i-propanol sol. system.

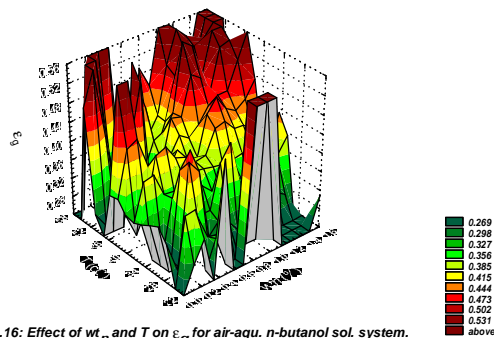


Fig.16: Effect of w_{t_n} and T on ϵ_g for air-aqu. n-butanol sol. system.

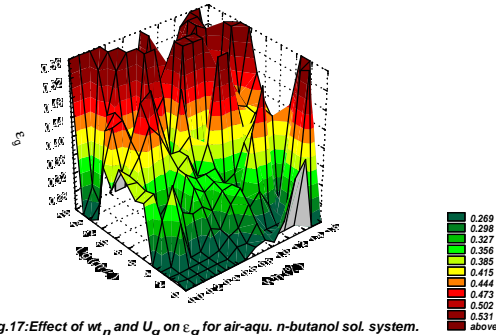


Fig.17: Effect of $w_{t,n}$ and U_g on ϵ_g for air-aqu. n-butanol sol. system.

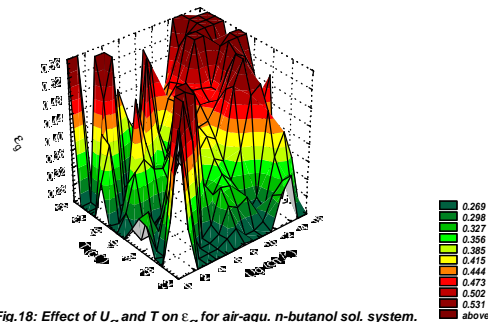


Fig.18: Effect of U_g and T on ϵ_g for air-aqu. n-butanol sol. system.

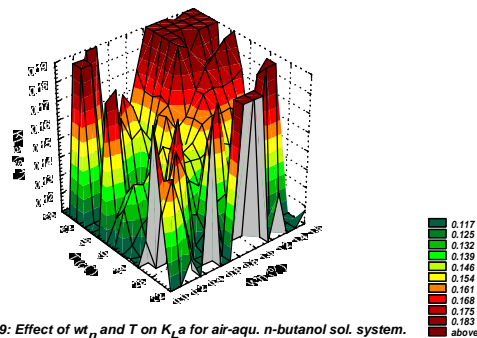


Fig.19: Effect of $w_{t,n}$ and T on K_La for air-aqu. n-butanol sol. system.

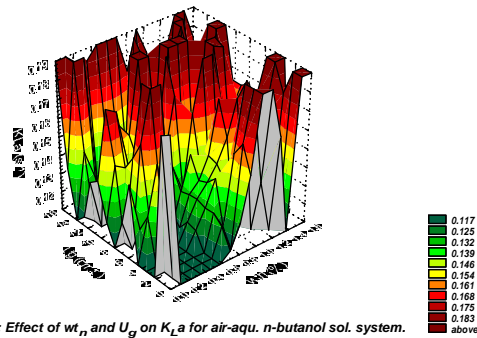


Fig.20: Effect of wt_n and U_g on K_La for air-aqu. n-butanol sol. system.

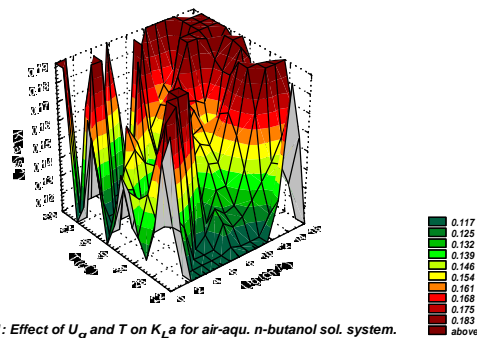


Fig.21: Effect of U_g and T on K_La for air-aqu. n-butanol sol. system.

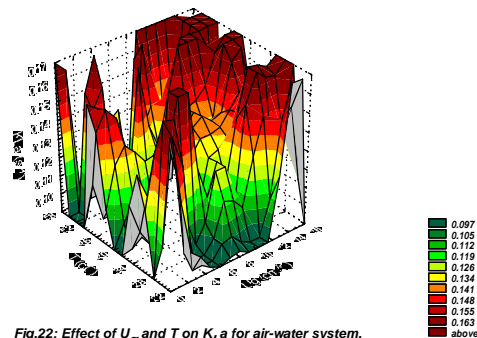


Fig.22: Effect of U_g and T on K_La for air-water system.

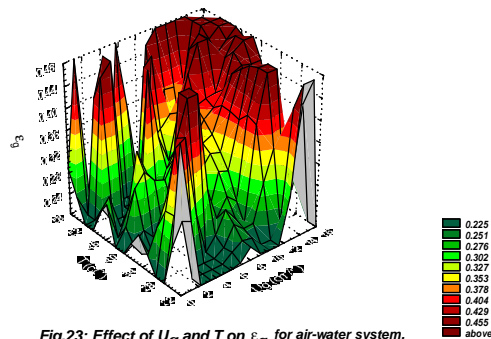


Fig.23: Effect of U_g and T on ϵ_g for air-water system.

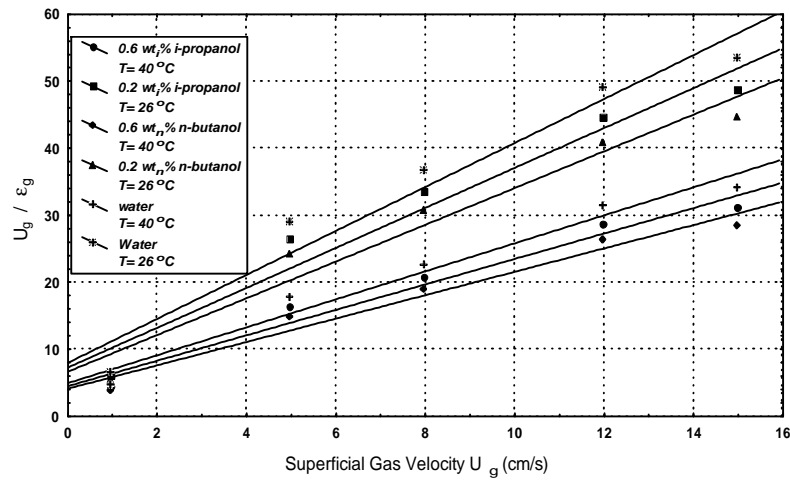


Fig.24: Zuber and Findlay Drift Flux Plot, Gas Holdup data For Three System.