INFLUENCE OF LIQUID PROPERTIES ON MASS TRANSFER COEFFICIENTS IN BUBBLE COLUMNS

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The gas phase holdup, the specific interfacial area a, and the liquid-side volumetric mass transfer coefficient $k_L a$ have been determined for two selected binary systems water–butanol and butanol––2-ethylhexanol in a short bubble column of 150 mm i.d. at gas phase velocities (air) in the range of $u_G = (0.31 - 2.52) . 10^{-2}$ m s^{-1}. The $k_L a$–u_G dependences show characteristic maxima which only partially can be explained by molecular and surface force interactions, induced by the liquid phase. The main parameter, influencing the $k_L a$-values, is a. At small gas velocities some irregularities in the Sauter diameter d_{32} and in the values of separated k_L are observed.

The holdup of the phases, the specific interfacial area, and the volumetric mass transfer coefficient (gas to liquid) are important parameters especially in the case, when chemical reactions occur. If the reaction kinetics are comparable with the time-dependent behaviour of the hydrodynamics the models used for column design based only on $k_L a$ can not longer be assumed. Therefore it becomes necessary to separate both the values and determine them by at least two independent methods.

It was shown in several review articles on this subject[1-6], that liquid phase properties (regardless the gas phase, properties like density, viscosity, and interfacial area) are included into correlation usually as values measured under static conditions. As geometrical parameters, only the main dimensions of a bubble column, the diameter, and the height are mostly included. Additionally the different flow regimes, like the homogeneous and the turbulent bubble flow in such apparatus, are often not distinguished. Because the conditions for the altered flow regimes cannot be easily found in the literature, the accuracy and the validity of such correlations concerning to other systems is limited.

The gas holdup and the interfacial area as size-distributed values depend considerably on impurities, which are in the case of alcohols in water[7-9] tensid-like. To avoid errors in the scale-up and design of bubble columns, the experimental check-up of the holdup in small bubble columns is recommended[1]. Especially measurements of volumetric mass transfer coefficients in nonaqueous systems are demanded because they are found out in the literature only sporadically with no systematic efforts.

The aim of this work was to interpret the measured values of ε_G, $k_L a$ and, separated from them, of a in binary alcohol–water and alcohol–alcohol solutions in a bubble layer with air as the gas phase. The main task was to connect the mass transfer behaviour with the distribution of the bubbles, which specific interfacial areas show characteristic maxima, depending on the mixture composition.

Our special interest was focussed on the bubble distribution under conditions of smaller gas phase velocities u_G which are common under industrial conditions. The critical Weber number We ≥ 2 providing the fully bubbling of the distributor cannot be established, it is usually of the order 0·05 at $u_G \leq 1 . 10^{-2}$ ms^{-1}. Therefore the gas throughput in single holes which are just bubble is generally raised, as well as the bubble volume[10]. Moreover, on such conditions an influence of the liquid phase parameters on the mechanism of buble formation could be expected[11-15].

EXPERIMENTAL

While the integral gas holdup can easily be measured by simple level measurement of the clear and bubble flow heights[16], the physical and chemical methods for determination of the specific interfacial area give different values up to 40% (ref.[1]). For the simplicity, at small gas velocities ($u_G < 0.04$ m s^{-1}) we decided to use informative photographic method[11 16] and determined the mean volume to surface (Sauter) diameter ratio, although some errors can occur by wall influence and the not measurable small bubble parts. But these errors are only systematical and do not influence in principle the concentration dependences, when the systems are investigated under identical hydrodynamic conditions.

TABLE I

Parameters of the measured systems

System	$\varrho . 10^{-2}$, kg m^{-3}	$\eta . 10^2$, Pa s	$\sigma . 10^2$, N m^{-1}
Butanol	8·017	2·271	2·28
Butanol (75 vol. %)–2-ethylhexanol	8·123	3·837	2·31
Butanol (50 vol. %)–2-ethylhexanol	8·152	5·103	2·33
Butanol (25 vol. %)–2-ethylhexanol	8·214	6·517	2·35
2-Ethylhexanol	8·263	8·732	2·39
Butanol (5 vol. %)–water	9·988	10·2	3·23
Butanol (2 vol. %)–water	9·924	9·22	4·41
Butanol (1 vol. %)–water	9·936	8·95	5·30
Butanol (0·6 vol. %)–water	9·940	8·79	5·99
Butanol (0·1 vol. %)–water	9·945	8·76	6·93
Water	9·953	8·75	7·20

The experiments were carried out in a bubble column shown in Fig. 1. The binary liquid systems were n-butanol–2-ethylhexanol (both p.a.) and water–n-butanol (p.a.) at the temperature 25°C (Table I).

The photographs of the bubble spectra were interpreted by a computer-supported analyzer. Errors caused by mutual covering of different bubbles are reduced by the choice of a front picture cut-out on a screen dividing the column into two segment parts (Fig. 1). The bubble size was determined on the basis of the two diameters of a rotation ellipsoid. The average diameters and the central moments up to the forth order did agree with other work[17] and gave logarithmic bubble size distributions.

The volumetric liquid-side mass transfer coefficients were determined by absorption of oxygen from air into the liquid phase, preliminarily saturated with nitrogen, under batch conditions[18]. The oxygen concentrations were measured with Clark-electrodes[19]. A separate liquid circuit was filled to provide a constant flow and therefore a stable diffusion regime at the electrode membrane, which was independent on the flow regimes in the column. Care was taken to separate the small remaining bubbles before the probe liquid was measured. The necessary corrections of the signals were carried out with the aid of the electrode function, considering additionally the diffusion through the membrane.
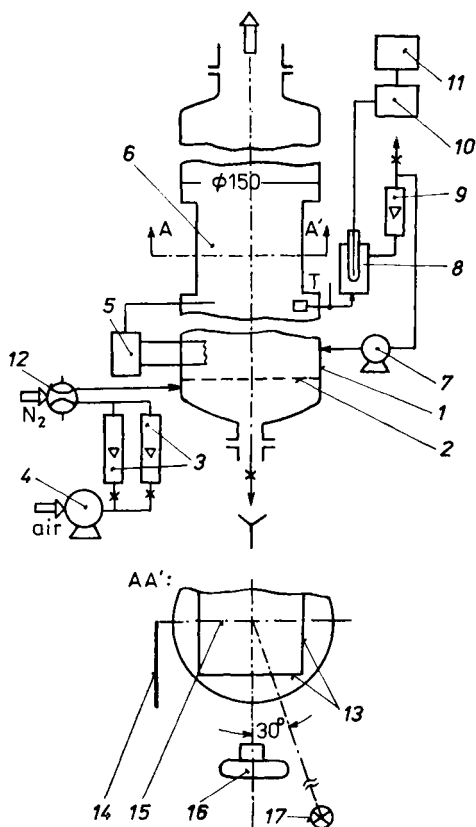

FIG. 1

Experimental bubble column. 1 Reactor, 2 sieve tray ($d_1 = 0.5$ mm, $\varphi = 0.2\%$), 3 flow meter, 4 gas blower, 5 temperature regulator, 6 bubble measuring section, 7 circuit pump, 8 Clarc-sensor, 9 circuit controller, 10 O_2--meter, 11 voltmeter, 12 value, 13 optical window, 14 reflector, 15 wall, 16 camera, 17 flash, measurement of holdup by stand tubes (not shown). T temperature measurement

RESULTS

In first experiments we stated that the mean bubble diameter depends on the mixture composition for both systems at small gas phase velocities $\left(u_G \leqq 6 . 10^{-2} \text{ m s}^{-1}\right)$ and offer a small maximum (Fig. 2). The explanation of this phenomenon is possible perhaps considering the specific mechanism of bubble formation at such conditions. The reason may be the larger gas velocity in one holes while the other are blocked and the distributor does not bubble fully. Moreover, small concentration of the additive component to the main liquid promote the coalescence in the neighbourhood of the holes even at the stadium of bubble creation. As can be seen at higher gas phase velocities this effect is diminished in the developed turbulent two-phase flow and suppressed due to the mutual interactions of bubbles.

Further experimental results are represented in the Figs 3 to 6, where the dependences of ε_G, a, $k_L a$, and k_L on the composition and u_G for both systems are shown.

The gas-holdup and the specific interfacial area show characteristic maxima, too. In the well known case of water–alcohol mixtures a first maximum occurs[18,19], which is often undefined in its intensity. It is mostly influenced by impurities such as traces of tensides. Hysteresis effects are supressed (Fig. 2a).

On the other hand the larger maximum at $u_G = 1.26 . 10^{-2} \text{ m s}^{-1}$ indidates the transition region between homogeneous and turbulent bubbling, which was stated also visually. The very small concentration range of the second component causing the first maximum demonstates the influence of orientation of molecules at the interface and its stabilization effects against bubble coalescence[7-9]. There is a second and broader maximum in ε_G and a, which still has not been interpreted in the litera-
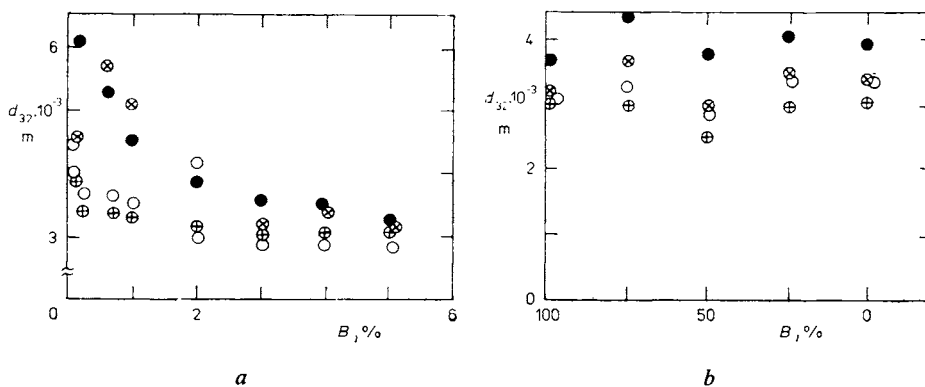

FIG. 2

The mean (Sauter) diameter of the gas bubbles d_{32} depending on the liquid composition of the systems. a Water–1-butanol (B), b 1-butanol (B)–2-ethylhexanol at 25°C and the superfacial gas phase velocity $u_G . 10^2$, m s^{-1}: ● 0·31, ⊗ 0·63, ○ 1·26, and ⊕ 2·52

ture[7,11] and which occurs at concentrations in the few per cent range. This pheno-
menon has been so far reported only in one paper[17]. The explanation of the maxima
in ε_G and a seems up to now more speculative as in the case of water–alcohol mix-
tures, because the interfacial area does not alter significantly with the composition,
even at higher concentrations (Table I).

From Fig. 4 we can conclude that the volumetric mass transfer coefficient $k_L a$ in
the case of water–alcohol solutions is not significantly influenced by a. Although
ε_G and a slightly increase, the values of $k_L a$ decrease rapidly in the range of small
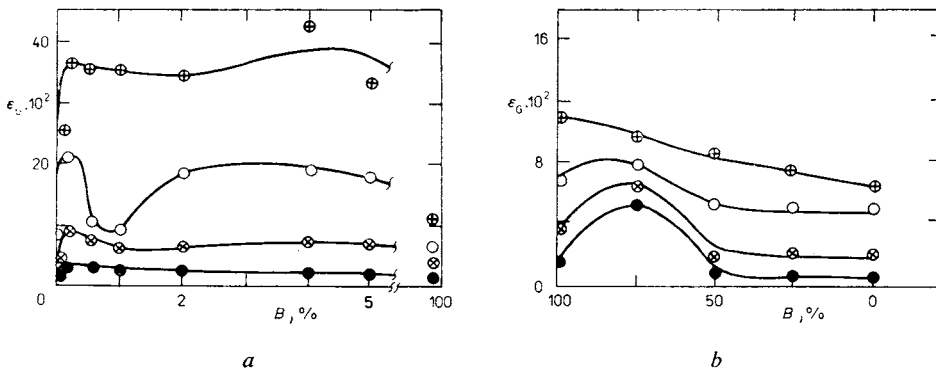alcohol concentratins at higher gas velocities. The reason is obviously a partial

a   *b*

Fig. 3

The gas holdup ε_G depending on the liquid composition and on u_G. Denotation as in Fig. 2

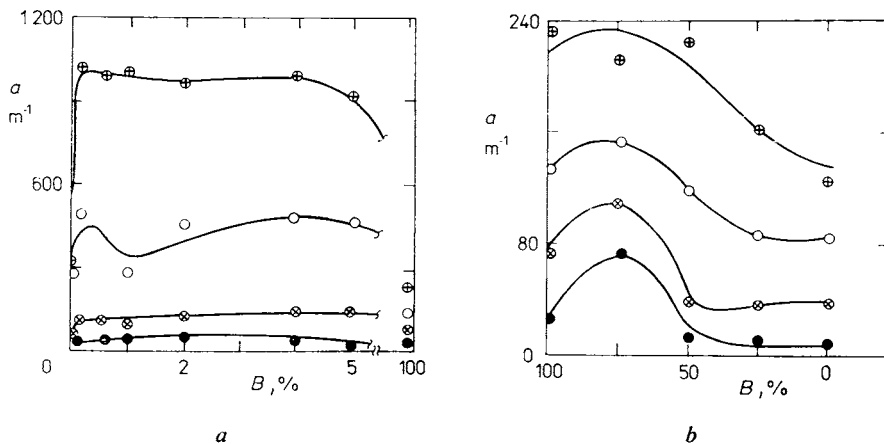*a*   *b*

Fig. 4

The specific interfacial area, a, m^{-1} depending on u_G. Denotation as in Fig. 2

immobilization of the interfacial area, which decreases bubble coalescence and mass transfer rate[20,21]. At higher alcohol concentrations again, the $k_L a$ increases regularly with the concentration, due to the increasing a. While for water–butanol solutions at small alcohol concentrations the correlations of Hikita[22] for $k_L a$ and Akita and Yoshida[12] for a show the true dependences on σ (with a negative exponent), this cannot be verified for alcoholic mixtures. As it is shown in literature[7] for ε_G
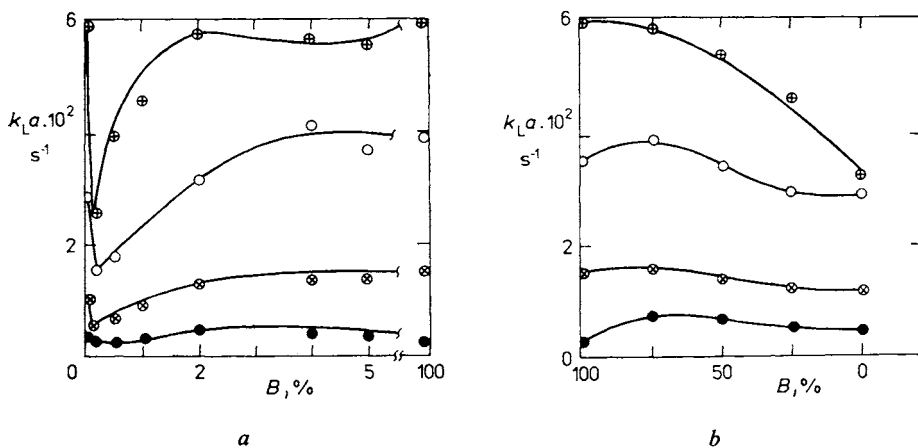

FIG. 5

The volumetric liquid-side mass transfer coefficients, $k_L a$, s^{-1}, depending on u_G. Denotation as in Fig. 2
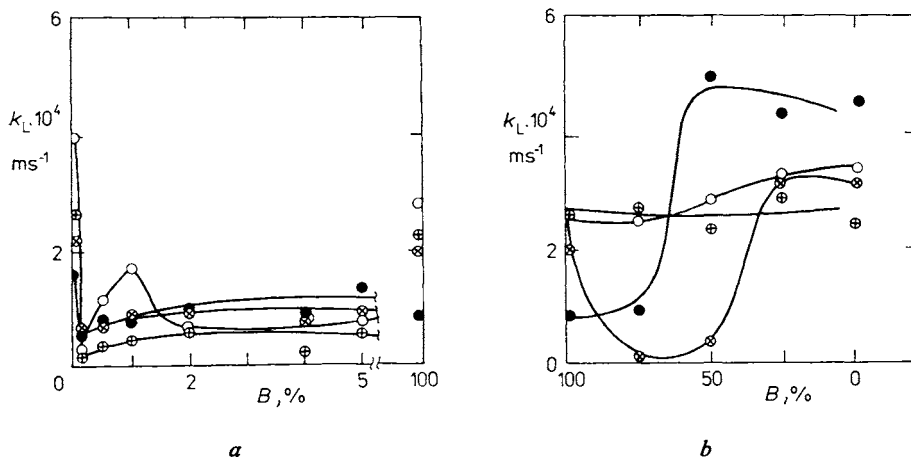

FIG. 6

The mass transfer coefficients k_L, $m\ s^{-1}$, depending on u_G. Denotation as in Fig. 2

and in Fig. 2 for the here investigated systems, only equations involving terms of $\varepsilon_G/(1 - \varepsilon_G)$ (Akita and Yoshida[23], Bach and Pilhofer[24]) are able to describe quite right the ε_G-curves. Analogous structures of relations for a and $k_L a$ have not yet been published.

Fig. 5 shows the values k_L, separated from the previous by mentioned dependences. We conclude that at medium concentrations and at least of the system butanol–2-ethylhexanol at higher gas velocities $\left(u_G \geqq 6 . 10^{-3} \text{ m s}^{-1}\right)$ the obtained k_L agree with Deckwer's results[25] and are of the order $k_L = (1-3) . 10^{-4} \text{ m s}^{-1}$. They are nearly independent on the mixture composition. Obviously there exists divergent k_L only in concentration ranges, where the compositions of the solutions are close to those of the pure components.

This confirms the conclusion that the volumetric mass transfer coefficient of the liquid phase alters justly and mostly by the influence of the altered a, which is on its part influenced by the hydrodynamics and the system properties. In pure organic systems the k_L tends to be higher than in aqueous one.

On the contrary we found out that at small gas velocities the k_L-values are no more constant and show e.g. S-shaped dependences.

CONCLUSIONS

The separation of $k_L a$ and a based on photographic methods is useful way to clear up mechanisms of mass transfer in bubble layers at relatively small gas velocities. Maximum $k_L a$ can only be expected under such system conditions, where maximum specific interfacial areas do occur, too.

Deviations in the mass transfer behaviour at smallest gas velocities, especially concerning nonaqueous systems, have to be considered in more details in further investigations.

LIST OF SYMBOLS

a	specific interfacial area, m^{-1}
d	bubble diameter, m
k	mass transfer coefficient, m s^{-1}
t	temperature, °C
u	superfacial velocity, m s^{-1}
ε	holdup,
ϱ	density, kg m^{-3}
η	dynamic viscosity, Pa s
σ	interfacial tension, N m^{-1}

Subscripts

G	gas
L	liquid

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