Gas-Liquid and Solid-Liquid Mass Transfer in Three-Phase Sparged Reactors With and Without Ultrasound

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Gas-liquid and solid-liquid mass transfer in three-phase sparged reactor has been studied with and without superimposed ultrasound. The results show marginal enhancement in mass transfer coefficient due to ultrasound. Thus it has been concluded that the reported enhancement in the rates of heterogeneous reactions due to ultrasound is likely due to other, e.g. chemical, effects of ultrasound.

The chemical effects of ultrasound have been known for a long time (1). In the last decade there has been a resurgence of interest in enhancing the rates of various chemical reactions through the application of ultrasound. Suslick (2) has presented an excellent review of the use of ultrasound in chemical synthesis. A number of industrially important catalytic reactions, e.g., hydrogenations, hydrosilations, Friedel-Crafts acylations and reduction of nitro to amino compounds, have been known to be speeded up by ultrasound (2–5). Moulton et al. (6,7) have studied ultrasonic hydrogenation of soybean oil. These authors have reported a significant (twentyfold) increase in the rate of hydrogenation with copper and nickel catalysts when the reactor was irradiated with ultrasound waves. Further, tests with the hydrogenated product confirmed that it has the desired flavor and stability. It was thus concluded that ultrasonic hydrogenation of soybean oil offers a method to produce good quality products at potentially lower cost than existing methods.

In order to develop the technique of ultrasonic hydrogenation and apply it on industrial scale, considerably more information will be required. Catalytic hydrogenation is a complex process involving four major steps, mass transfer from gas to liquid and from liquid to catalyst particle, diffusion inside catalyst particle, and chemical reaction on the catalyst surface. Suslick (2) has indicated that in heterogeneous reactions involving solid catalysts, ultrasound may have a number of effects ranging from activating the catalyst to improvement in mass transfer. There is, however, no study reported in the literature which definitely can establish the contribution of these effects independently. Recently, Zanwar and Pangarkar (8) have shown that considerable enhancement in the rates of particle-liquid mass transfer in packed beds can be realized by the application of ultrasound. There is no similar study available in the literature for three-phase sparged reactors which are likely to be used for hydrogenations with ultrasound. In our program to understand catalytic hydrogenation with ultrasound and develop this technique, it is planned to study the effect of ultrasound on the various steps given above. In the first part of the study the effects of ultrasound on gas-liquid and particle-liquid mass transfer coefficients, $k_L a$ and K_{SL}, respectively, and gas hold-up, ε_g, are investigated.

EXPERIMENTAL

The experimental set-up consisted of a stainless steel jar of 65 mm diameter and 75 mm in height. This jar was bonded directly to the ultrasonic transducer. A glass cylinder 65 mm in diameter and 180 mm in height was joined to the jar as an extension. The ultrasonic transducer used had four ring-shaped lead-zirconate-titanate (PZT) crystals bolted together. The transducer was bonded to the bottom of the jar by araldite and had an output of 140 W at 18 kHz frequency. Electric power was supplied by a power supply unit which was connected to the mains through a dimmerstat. An A.C. ammeter was connected in series between the power supply unit and the dimmerstat.

For the $k_L a$ measurement the system used was lean carbon dioxide (in air) aqueous sodium carbonate/bicarbonate buffer. Both air and carbon dioxide were metered through precalibrated rotameters in such a way that the volume percentage of carbon dioxide in the air-carbon dioxide mixture was approximately 8–10%. A known volume of the buffer solution was poured into the column. The gas mixture was sparged in the solution through a ring sparger having six one-mm diameter holes. The clear liquid height and dispersion height after gas sparging were noted. The experiments were conducted in the semibatch manner. The batch time was eight min, at the end of which liquid sample was withdrawn to get the final concentrations of sodium bicarbonate and carbonate. This procedure was repeated in the presence of ultrasonic vibrations. The effect of gas velocity was studied by varying the same from 0.06 to 0.25 m/s.

For K_{SL} measurement the system used was dissolution of benzoic acid granules in water. Benzoic acid granules were prepared by the method of Evans and Gerald (10). The surface area and shape factors of the granules were measured using the method of Carman (11). A known volume of water was charged into the reactor and the gas flow was started. A predetermined amount of benzoic acid granules was added to the reactor, and the stop watch was started. The duration of each experiment was four min, at the end of which a solution sample was removed, filtered and titrated against 0.01N sodium hydroxide to obtain the amount of benzoic acid dissolved. This procedure was repeated in the presence of ultrasonic vibrations. The effect of gas velocity was studied by varying the same from 0.01 to 0.2 m/s.

A detailed procedure for calculation of $k_L a$ is given by Sharma and Mashelkar (9). The calculation procedure for K_{SL} (solid-liquid mass transfer coefficient) for an experiment in the batch mode is given by:

$$K_{SL} \cdot t \cdot A_P = V \cdot \ln [(C^* - C_f)/C^*]$$

where A_P is the cumulative surface area of the particles calculated by the following equation:

$$A_P = 6W/(\phi \varrho_P d_P)$$

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A_P = surface area of particles, m^2
d_P = average screen size of particle, m
C_f = final concentration, $kmol/m^3$
C^* = saturation concentration, $kmol/m^3$
t = batch time, s
V = volume of liquid, m^3
W = weight of the particles, kg
ϕ = Carman's surface factor
ϱ_P = density of solid, kg/m^3.

Gas hold-up was calculated from the following equation, where H_D is the height of the dispersed liquid, m, and H_S is the initial height of the liquid, m.

$$\varepsilon_g = (H_D - H_S)/H_D$$

RESULTS

Figures 1 and 2 show the variation of ε_g with gas velocity, V_g, with and without ultrasound. It is evident that there is very little difference in ε_g values for the two cases. The gas hold-up in a dispersed gas-liquid system is an indication of the effective gas-liquid interfacial area it affords. It can thus be concluded that the effective interfacial area is unlikely to be different when ultrasound is used.

Liquid side mass transfer coefficient, $k_L a$. Typical $k_L a$ data are shown in Figure 1 for both cases studied. The data for experiments without ultrasound show excellent agreement with those of Sharma and Mashelkar (9). Here also there is only a marginal difference in the $k_L a$ values for the two situations, implying that ultrasound has no effect on gas-liquid mass transfer in a sparged reactor.

Particle-liquid mass transfer coefficient, K_{SL}. Figure 2 shows typical variation of K_{SL} with V_g, from which it can be concluded that the particle liquid mass transfer coefficient also is not significantly affected by the application of ultrasound.

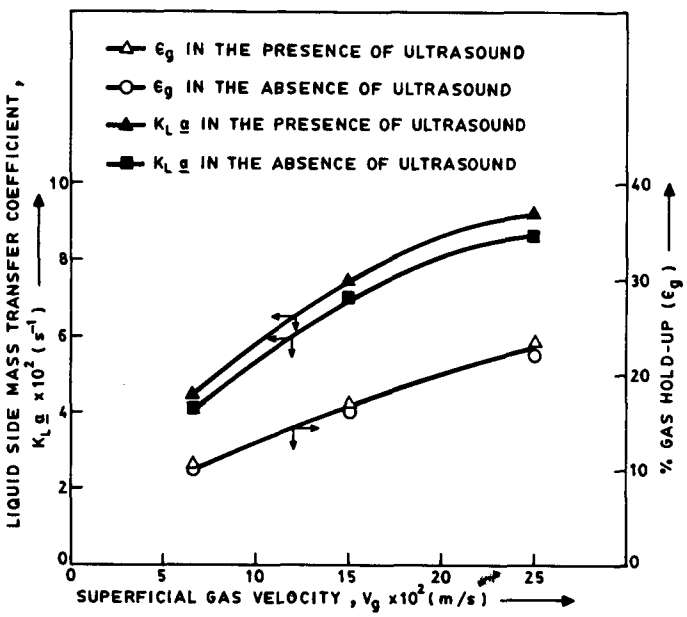

FIG. 1. Variation of ε_g and $k_L a$ with V_g.

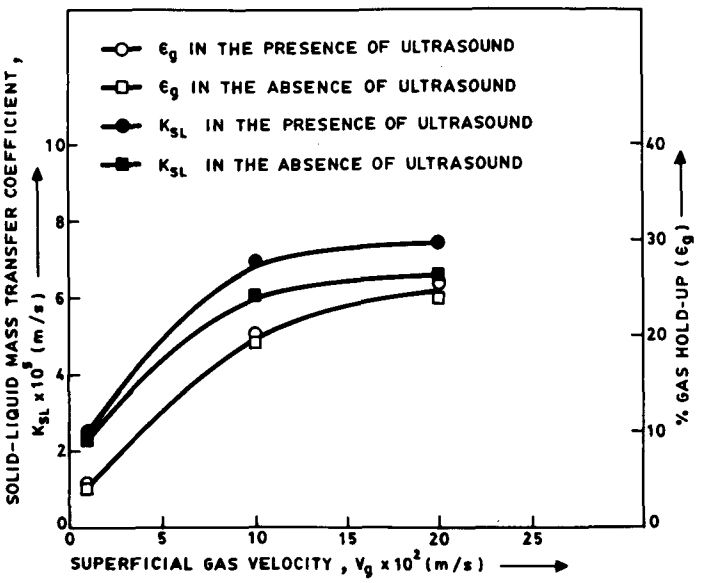

FIG. 2. Variation of ε_g and K_{SL} with V_g.

DISCUSSION

Any enhancement in mass transfer caused by ultrasound can be attributed to the phenomena of cavitation or acoustic streaming.

Zanwar and Pangarkar's (8) results for a relatively less turbulent packed bed situation indicated much higher levels of enhancement in the rate of mass transfer with ultrasound. By contrast, for the three-phase sparged reactor used in the present study, there is only marginal enhancement in the rate of mass transfer. It is known that a very high degree of turbulence is obtained in a bubble column (9). It is likely that the additional turbulence created by ultrasound is not significant compared to the underlying turbulence in a bubble column and, therefore, the effect is not pronounced. On the other hand, there is strong evidence in the literature which indicates chemical effects of ultrasound. For instance, Lindley et al. (12) obtained a 64-fold increase in the Ullmann coupling of activated aryl halides in dimethyl formamide with ultrasound. These workers could explain this high reactivity only through the ultrasonic breakdown of reactive intermediates and/or the desorption of products. Ultrasound was also shown to cause an interesting change in alumina catalyzed reaction of benzyl bromide with toluene and potassium cyanide (13). Mechanical stirring gave 83% Friedel-Crafts product, whereas sonication resulted in 76% of the substitution product. Thus, there are definite chemical effects of ultrasound which are probably the main sources of the high observed rates of reaction rather than the effect of ultrasound on mass transfer.

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