

GAS-AGITATED LIQUID-LIQUID EXTRACTION IN A SPRAY COLUMN

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The hydrodynamic characteristics of the air-water-toluene three-phase system in a spray extraction column at 20 °C were examined. The average and local hold-up data of the dispersed phase were determined in dependence on the flow rates of the continuous, dispersed and gaseous phases. The average gas phase hold-up was also measured and analyzed. A comparison was made of the hydrodynamic characteristics of the two-phase (water-toluene) and three-phase (air-water-toluene) systems.

With respect to the dispersed phase hold-up, a spray extraction column can operate in three modes of packing of the dispersed phase drops^{1,2}: dispersed, restrained, and dense. The efficiency is relatively low when the column is operated in the dispersed or restrained drop packing mode, mainly due to a high axial mixing in the continuous phase. An increased efficiency of the spray extraction column is attained with the dense packing of the dispersed phase drops, where the axial mixing in the two phases is significantly decreased.

The efficiency of non-mechanically agitated extraction columns with different packings can be considerably increased by introducing an inert gas as a mixing agent in the two-phase system³⁻⁵. The energy thus introduced increases the turbulence within the now three-phase, gas-liquid-liquid (G-L-L) system, which brings about improved dispersion of the droplets and, consequently, a higher hold-up and a larger mass transfer area³. Priestley and Ellis⁵ observed that the dispersed phase droplets were smaller and there was more backmixing in the continuous phase when a gas was introduced into the packed extraction column. Galkin et al.⁶ found the largest reduction of the height equivalent to a theoretical stage (HETS) in plate extraction columns with superficial gas velocities of 0.02 – 0.03 m s⁻¹. The authors concluded that this way of energy introduction was more efficient than by stirring or pulsation of the column.

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Although the positive effects of gas agitation in various types of non-mechanically agitated extractors have been known for many years, results of hydrodynamics and mass transfer studies in the G-L-L systems were only published after 1978.

Experimental hydrodynamic characteristics and mass transfer data have been reported for packed columns^{3-5,7}, for a spray column⁸ and for bubble columns⁹⁻¹¹. Ziebland and Hackl⁸ made first attempts to describe the phenomena associated with the introduction of an inert gas into the spray column. However, there are no literature data on the local hold-up of the dispersed phase in gas-agitated spray extraction columns.

As part of program undertaken to investigate the behavior of a G-L-L system in different columns^{12,13}, the present paper is concerned with the measurement of hydrodynamic characteristics of the air-water-toluene (A-W-T) three-phase system in a spray column. The results are also compared with those obtained for the operation of the same column with the two-phase water-toluene (W-T) system.

EXPERIMENTAL

A schematic diagram of the experimental set-up is shown in Fig. 1.

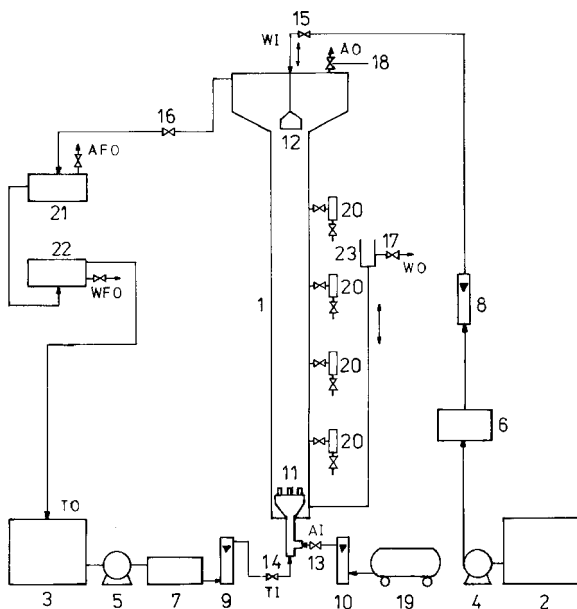


FIG. 1

Schematic diagram of the experimental set-up: 1 spray column, 2,3 water and toluene reservoirs, 4,5 piston pumps, 6,7 vessels for flow stabilization, 8 - 10 rotameters, 11 dispersed phase distribution, 12 continuous phase distributor, 13 - 18 valves, 19 air compressor, 20 sampling test tubes, 21,22 separators, 23 adjustable overflow tube; AI air inlet, AO air outlet, AFO air outlet from foam, TI toluene inlet, TO toluene outlet, WI water inlet, WO water outlet, WFO water outlet from foam

The cylindrical Borax glass column was 250 cm long, with a diameter of 10 cm, the conical section at the top of the column was 45 cm long and its maximum diameter was 30 cm. This column represented a modified spray column as used in our previous investigations of two-phase liquid-liquid (L-L) systems¹⁴. The modification was necessary with regard to the foam forming in the three-phase column, because work with the G-L-L system is associated with the appearance of foam at the interface and its discharge with the dispersed phase from the extraction column. For this reason, the dispersed phase had to be passed through two separators before its recirculation. In the first separator (21) the foam was disintegrated, while the other separator (22) served for settling and separation of water and toluene. After disintegration of the foam, air comes out through a hole at the top of the separator (21).

Air, water and toluene were used as the gaseous, continuous and dispersed phases, respectively. Water and countercurrent toluene, saturated with each other, were continuously introduced into the column by means of two piston pumps (4, 5). Air saturated with the two liquids was introduced at the column base by using a compressor (19). Water was introduced at the top of the column through a perforated plate-type distributor with holes 2 mm in diameter (12). The gas and toluene were fed to the column base through a common gas-liquid distributor (11), which was a plate 9 cm in diameter with 79 nozzles in the equilateral triangular arrangement. The nozzles were stainless steel tubes, 10 mm long and 1 mm in diameter, with a 6 mm spacing. As pointed out by Billet and Braun³, mixing of the air and toluene takes place in the distributor.

During the gas agitation, two-phase droplets are formed at the distributor nozzles or inside the column.

Temperature was held at 20 ± 1 °C. The flow rate ranges in terms of superficial velocities were: $0.03 - 0.46$ cm s⁻¹ for the dispersed phase (U_d), $0.03 - 0.14$ cm s⁻¹ for the continuous phase (U_c), and $0.24 - 0.68$ cm s⁻¹ for the gas phase (U_g).

In order to attain a clearer distinction of the interface at higher air flow rates, when the amount of foam was considerable, one drop of the yellow organic color SUDAN III per 20 dm³ of toluene was added.

The determination of the average dispersed phase and gas phase hold-ups in the three-phase G-L-L system is illustrated in Fig. 2.

At the beginning of each run, the column was filled to about half its volume with water. The toluene and water flow rates were increased gradually to a chosen level. When the volume of the two-phase dispersion became constant, the height of the interface above the toluene and air inlet, i.e. the height of the two-phase dispersion H_a , was recorded (see Fig. 2a). At that moment, air was introduced into the column at a chosen flow rate, and the interface level in the column increased. The new position of the interface (H_b) corresponds to the height of the three-phase dispersion above the toluene and air inlet (see Fig. 2b). The position of the interface was maintained in the cylindrical section of the column by the adjustable overflow tube (23). The average gas hold-up in the A-W-T three-phase system was calculated as the ratio of the gas phase volume V_g to the total volume of the three-phase dispersion V_b :

$$\varepsilon_g = \frac{V_g}{V_b} \quad (1)$$

Since the interface was maintained in the cylindrical section of the column, the heights of the three- and two-phase dispersions as well as the height of the gas phase were used to calculate the average gas hold-up in the three-phase dispersion:

$$\epsilon_g = \frac{H_g}{H_b} = \frac{H_b - H_a}{H_b} \quad (2)$$

The uncertainty of the average gas hold-up measurements in the A-W-T three-phase system is estimated to $\pm 5\%$.

The average dispersed phase hold-up in the A-W-T three-phase system was determined by the interruption method. After the column was left in operation in steady-state conditions for 30 min, the water and toluene inflow and outflow were stopped by disconnecting the piston pumps (4, 5) and closing the valves (14 – 17). Simultaneously the air compressor was turned off and the valves (13, 18) were closed. The decrease of the interface level was determined after water and toluene settled. The interface level (H_c) corresponds to the continuous phase height above the toluene and air inlet (see Fig. 2c). The average dispersed phase hold-up in the three-phase system was calculated as the ratio of the settled volume of the dispersed phase V_d to the total volume of the three-phase dispersion:

$$\epsilon_d = \frac{V_d}{V_b} = \frac{H_d}{H_b} = \frac{H_b - (H_c + H_g)}{H_b} \quad (3)$$

The heights H_b and H_g are the same as in Eq. (2).

The mean value of the average dispersed phase hold-up in the three-phase system was determined with an uncertainty of $\pm 3\%$.

The local hold-up of the dispersed phase in the A-W-T system was determined by the sampling method. For this, test samples were taken during operation from the column at different distances

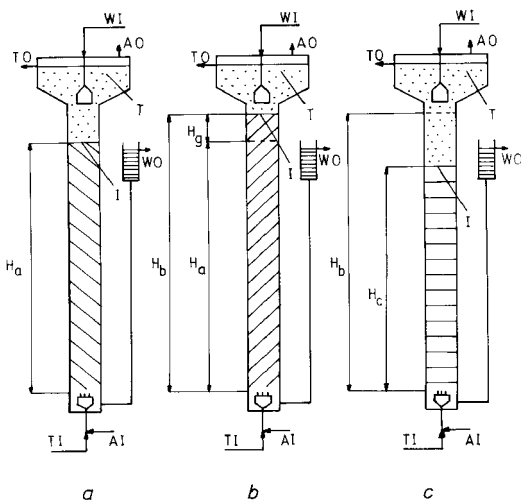


FIG. 2

Experimental set-up for measurement of the average dispersed phase and gas phase hold-up data: **a** situation before gas introduction, **b** situation before stopping the phase flows, **c** situation after stopping the phase flows. Symbols as in Fig. 1; I interface, T toluene

above the dispersed phase inlet (55, 105, 155 and 180 cm) using small cylindrical graduated test tubes (5 cm long and 2 cm in diameter) connected to the column (20).

During the operation of the spray column with the three-phase system, the A-W-T mixture was quickly introduced into the tube by opening a release valve. After closing the valve, the gas bubbles in the tube went upwards and the continuous and dispersed phases settled. Since the average gas hold-up was always below 1%, the volume of gas in the sample was neglected. The local dispersed phase hold-up was determined from the ratio of the settled volume of the dispersed phase to the total volume of sample. The average error of measurement of the local dispersed phase hold-up was $\pm 4\%$. Visual inspection indicated that the gas hold-up was homogeneously distributed throughout the column.

In order to compare the effect of the gas phase flow on the hydrodynamic parameters of the W-T two-phase system in the spray column, the average and local dispersed phase hold-ups were determined by the same experimental methods and for the same dispersed to continuous phase ratio as in the A-W-T three-phase system.

RESULTS AND DISCUSSION

The dependence of the average dispersed phase hold-up for the three-phase system in the spray column on the superficial velocities of the gaseous and dispersed phases, at a constant superficial velocity of the continuous phase, is shown in Fig. 3.

The average dispersed phase hold-up exhibits a relatively small increase with increasing superficial velocity of the gas phase. Moreover, Billet and Braun³ showed that an initial sinking of the dispersed phase hold-up takes place at gas superficial velocities below 0.2 cm s^{-1} . They concluded that within that gas flow rate range, the energy input is insufficient to produce intensive turbulence and, consequently, to form a large number of small droplets. On the other hand, the increase in the dispersed phase hold-up observed by them was below 20% for gas superficial velocities within the range of $0.2 - 0.6 \text{ cm s}^{-1}$ at a constant superficial dispersed phase velocity of 0.4 cm s^{-1} . Such a relationship between the gas phase flow and the dispersed phase hold-up may also be

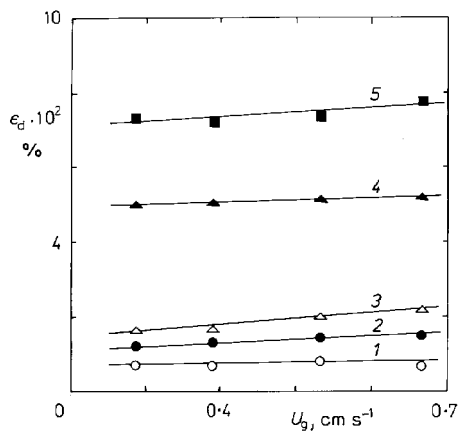


FIG. 3

Dependence of the average hold-up of the dispersed phase on the superficial velocity of the gas and dispersed phase in the air-water-toluene three-phase system: $U_c = 0.132 \text{ cm s}^{-1}$; U_d (cm s^{-1}): 1 0.032, 2 0.149, 3 0.255, 4 0.384, 5 0.452

due a negative effect of the dispersed phase flow on the gas phase hold-up (see Fig. 7 below). However, as Fig. 3 demonstrates the average dispersed phase hold-up exhibits, higher increase at increased superficial velocities of the dispersed phase.

A comparison of the average dispersed phase hold-up data for the A-W-T three-phase system and the W-T two-phase system at the same ratios of superficial velocities of the continuous and dispersed phases is presented in Fig. 4.

Regression analysis of the experimental data showed that the average hold-up of dispersed phase in the spray column is about 15% higher in the three-phase system than in the two-phase system under the same operating conditions. On the other hand, Ziebland and Hackl⁸ observed a 50 to 300% increase in the dispersed phase hold-up if the gas was introduced into the spray column together with the two-phase system. This increase, however, occurred when the gas phase superficial velocity was 1 – 5 cm s⁻¹.

The local dispersed phase hold-up, determined at constant superficial velocities of the continuous, dispersed and gaseous phases, increased with increasing distance above the inlet of the dispersed and gas phase, as shown in Fig. 5.

A comparison of the local dispersed phase hold-ups for the A-W-T and the W-T systems at the same superficial velocity ratios of the continuous and dispersed phases is presented in Fig. 6.

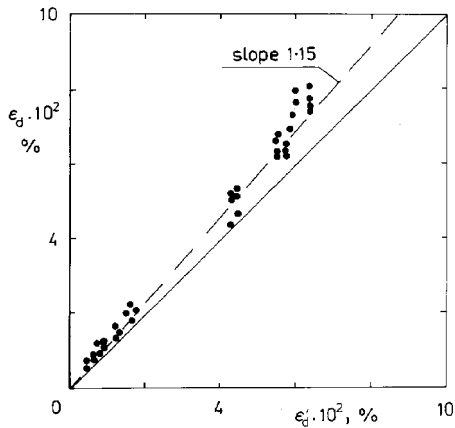


FIG. 4

Comparison of the average dispersed phase hold-up values in the air-water-toluene three-phase system and the water-toluene two-phase system: $U_d = 0.032 - 0.452$ cm s⁻¹; $U_c = 0.033 - 0.132$ cm s⁻¹; $U_g = 0.248 - 0.672$ cm s⁻¹. Broken line represents the mean value of increase in the average dispersed phase hold-up in the three-phase system

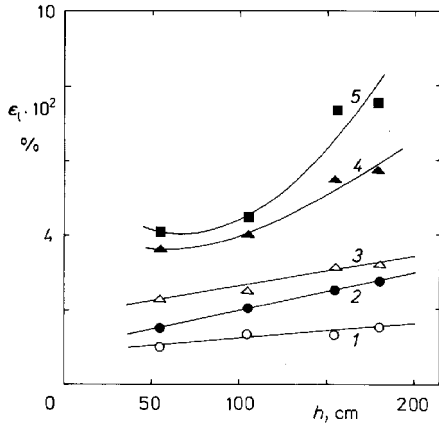


FIG. 5

Dependence of the local dispersed phase hold-up value on the distance above the dispersed and gas phase inlet in the air-water-toluene system: $U_g = 0.248$ cm s⁻¹; $U_c = 0.089$ cm s⁻¹; U_d (cm s⁻¹): 1 0.032, 2 0.149, 3 0.255, 4 0.384, 5 0.452

A comparison of the experimental data gave evidence that the local dispersed phase hold-up in the spray column is higher in the three-phase than in the two-phase system under the same operating conditions. The observed increase of approximately 25% can be explained in the same way as in the case of the average values of the dispersed phase hold-ups.

The average gas hold-up increases with increasing superficial gas phase velocity. On the other hand, the gas hold-up decreases with increasing superficial dispersed phase velocity (see Fig. 7). Bandyopadhyay et al.¹¹ also observed a decrease in the gas hold-up with increasing fraction of the organic phase in two G-L-L systems: air-water-kerosene and air-water-dibutyl phthalate. This effect was observed in the former system with kerosene below 20% (v/v) and in the latter system with dibutyl phthalate below 49% (v/v). The authors explained this behavior in terms of the nature of the spreading coefficient. In fact, an organic liquid having a positive spreading coefficient forms a thin film on the water surface and acts as a surface active agent. In the presence of a surfactant in water, the gas hold-up increases markedly because of the consequent retardation of the rising gas bubbles. Since the liquids kerosene and dibutyl phthalate possess negative spreading coefficients on water (-2.1 and -4.0 mN m⁻¹, respectively)¹¹, the gas bubbles in the systems rise faster at the interface.

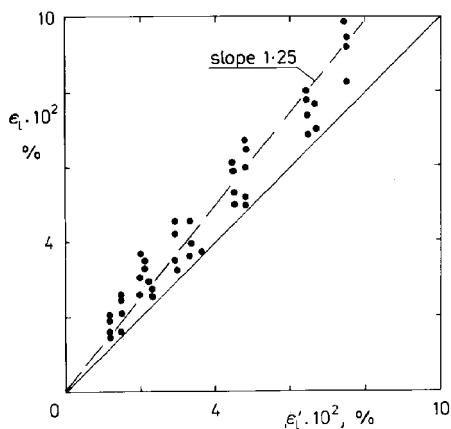


FIG. 6

Comparison of the local dispersed phase hold-up values for the air-water-toluene and water-toluene systems: $U_d = 0.032 - 0.452$ cm s⁻¹; $U_c = 0.033 - 0.132$ cm s⁻¹; $U_g = 0.248 - 0.672$ cm s⁻¹. Broken line represents the mean value of increase of the local dispersed phase hold-up in the three-phase system

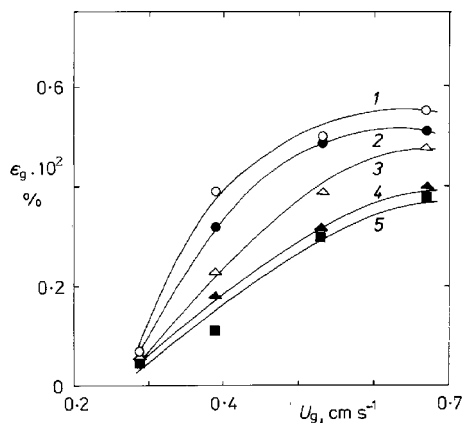


FIG. 7

Dependence of the average gas phase hold-up values on the superficial velocity of the dispersed and gas phase in the air-water-toluene system: $U_c = 0.132$ cm s⁻¹; U_d (cm s⁻¹): 1 0.032, 2 0.149, 3 0.255, 4 0.384, 5 0.425

Although the spreading coefficient of toluene on water is positive (8.7 mN m^{-1}), the oxygen–water–toluene three-phase system exhibits a sharp decrease in the gas hold-up in an agitated vessel if the dispersed phase content is lower than 5% (v/v) (ref.¹⁵). This implies that the spreading coefficient of the organic liquid on water is not the single factor affecting the gas hold-up. The lower interfacial tension and the higher apparent viscosity in the G–L–L system as compared to the G–L system may contribute as well: Kato et al.⁹ found that the gas hold-up is 10 – 15% lower in G–L–L systems than in G–L systems. This reduction in the gas hold-up has been attributed to an increase in the apparent viscosity of the bulk liquid resulting from the presence of the dispersed phase. In fact, in the A–W–T system investigated, larger bubbles are formed, bringing about a wider bubble size distribution. Hence, the larger bubbles rise much faster through the column, and consequently, the gas hold-up displays a sharp decrease.

CONCLUSIONS

The hydrodynamic characteristics of the air–water–toluene three-phase system in a spray column gave evidence that the average hold-up of the dispersed phase increases with increasing superficial velocity of the gas phase at constant superficial velocities of the dispersed and continuous phases. The average dispersed phase hold-up values in this three-phase system were found higher than in the water–toluene two-phase system in the same operating conditions.

At constant superficial velocities of the dispersed, continuous and gas phases, the local hold-up of the dispersed phase exhibits a nearly linear increase with increasing distance above the inlet of the dispersed and gaseous phases. The dispersed phase local hold-up values are higher in the three-phase system than in the two-phase system.

The average gas hold-up increases with increasing superficial velocity of the gas phase, whereas the gas hold-up determined at constant superficial velocities of the continuous and gas phases decreases with increasing superficial velocity of the dispersed phase.

SYMBOLS

h	distance above the toluene and air inlet, cm
H_a, H_b	heights of the two-phase and three-phase dispersions above the toluene and air inlet, cm
H_c, H_d, H_g	heights of the continuous, dispersed and gas phases, respectively, cm
U_c, U_d, U_g	superficial velocities of the continuous, dispersed and gas phases, respectively, cm s^{-1}
V_b	volume of the three-phase dispersion, cm^3
V_d, V_g	volumes of the dispersed and gas phases, respectively, cm^3
ϵ_d, ϵ'_d	average dispersed phase hold-up values in the three-phase and two-phase systems, respectively
ϵ_g	average gas phase hold-up in the three-phase system
ϵ_b, ϵ'_1	local dispersed phase hold-up values in the three-phase and two-phase systems, respectively

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