AXIAL DISPERSION IN A THREE-PHASE GAS-AGITATED SPRAY EXTRACTION COLUMN

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The continuous-phase axial dispersion coefficients of the three-phase gas–liquid–liquid system in a gas-agitated spray extraction column 10 cm i.d. at 20 °C were examined. The system used was water as continuous phase, toluene as dispersed phase, and air as gaseous phase. The rise in the gas phase superficial velocity increased the continuous-phase axial dispersion coefficient. A non-linear dependence between the continuous-phase axial dispersion coefficient and the continuous phase superficial velocity was observed. No correlation was found between the continuous-phase axial dispersion coefficient and dispersed phase superficial velocity. The increase in the gas phase hold-up corresponded to a slight increase in the continuous-phase axial dispersion coefficient. The increase in the dispersed phase hold-up generated a growth of the continuous-phase axial dispersion coefficient. A comparison was made of the continuous-phase axial dispersion coefficients of the three-phase (air–water–toluene) and two-phase (water–toluene) systems.
Key Words: Gas-agitated extraction column; Continuous-phase axial dispersion coefficient; Two- and three-phase systems.

The spray columns have usually been operated at relatively low dispersed phase hold-ups with a dispersed packing of drops. Although they can handle high flow rates and are simple and cheap, there have been relatively few applications of these columns because they suffer from serious backmixing of the continuous phase. However, if drops are produced at a rate slightly higher than that at which they coalesce at the main interface, a dense packing of drops extends from top toward the distributor of drops. It was shown that the backmixing reduced when the spray column operated with a dense packing of drops[1]. In spite of the fact that there has been relatively little experience in operation with dense packing of drops, such operation should probably be considered if the use of spray extraction column seems attractive. Another way of increasing the efficiency of a spray column is introduction of an inert gas as a mixing agent in the two-phase system[2–4]. This method of energy introduction increases the turbulence within the three-phase gas–liquid–liquid (G–L–L) system, which causes an improved dispersion of droplets and, consequently, a higher dispersed phase hold-up and therefore a greater mass transfer area[2].

An important factor in understanding the fundamental mass transfer mechanisms in countercurrent liquid–liquid extraction spray columns is the effect of backmixing. Both stagewise and differential (diffusion) models may be used[5,6]. For comparison purposes it is better to express the amount of backmixing as the axial dispersion coefficient (E) related to the diffusion model rather than by the backflow coefficient used in the stage-wise model.

A large quantity of experimental data[7–14] has been published on the axial dispersion of two-phase liquid–liquid systems, but only scarce data on three-phase gas–liquid–liquid systems are available in the literature[3,15]. Diaz et al.[3] showed that high dispersion coefficients are deduced in both the liquid phases for the three-phase air–water–kerosene system, in which water was the continuous phase and kerosene the dispersed phase. They also found that the values of the axial dispersion Peclet numbers of the water phase (Pe_{ct}) are 0.1 to 1.2, decreasing when the flow rates of air or kerosene grow, or when the flow water rate is reduced. Diaz et al.[3] concluded that the Peclet number for the kerosene phase (Pe_{dt}) decreases to the values between 0.4 and 0.1 when the air flow rate increases. Kato et al.[15] investigated the axial dispersion in multi-stage bubble columns for the air–water–kerosene system. They found that the dispersed-phase axial dispersion coefficient (E_{dt}) increased with increasing the gas velocity and column diameter, and was independent of the total liquid velocity in the range of 0.05–1.0 cm s^{-1}. Kato et al.[15] derived an empirical equation in which E_{dt} depends on the gas superficial velocity, column diameter and gravitational acceleration.

The present paper is concerned with the measurements of the continuous-phase axial dispersion of the three-phase air–water–toluene system in a spray extraction column. A comparison is made of the continuous-phase axial dispersion coefficients for the air–water–toluene (A–W–T) and the two-phase water–toluene (W–T) systems. The experimental data for the three-phase A–W–T system are used to develop an empirical equation for the prediction of continuous-phase axial dispersion coefficient in the three-phase system.

EXPERIMENTAL

The spray extraction column used in this investigations is similar to that described in detail in our preceding paper[4]. The only difference was in an apparatus for introducing the tracer solution into the column. This apparatus consists of a reservoir for the tracer solution, piston pump and tracer distributor. The column was of 10 cm i.d. and 280 cm length.

Air, water and toluene were used as the gaseous, continuous and dispersed phase, respectively. Water saturated with toluene was continuously introduced at the top of the column by means of the piston pump. Toluene saturated with water was brought at the bottom of the column by means of another piston pump. Air saturated with the two liquids was introduced by means of a compressor at the bottom of the column. The gas and toluene were fed at the bottom of the column through a common gas–liquid distributor, which was described in detail in our previous paper[4]. During the gas agitation, dispersed phase droplets and air bubbles are formed at the distributor nozzles or inside the column.

The temperature was held at 20 ± 1 °C. The flow rate ranges in terms of superficial velocities were: 0.064–0.212 cm s^{-1} for the continuous phase (U_c), 0.106–0.318 cm s^{-1} for the dispersed phase (U_d), and 0.071–0.248 cm s^{-1} for the gaseous phase (U_g). The density of water, toluene and air at the operating temperature were: 0.9982 g cm^{-3} (ρ_c), 0.8660 g cm^{-3} (ρ_d) and 1.025 . 10^{-2} g cm^{-3} (ρ_g), respectively. The viscosity of water, toluene and air were: 1.002 . 10^{-2} g cm^{-1} s^{-1} (μ_c), 0.611 . 10^{-2} g cm^{-1} s^{-1} (μ_d) and 1.8 . 10^{-4} g cm^{-1} s^{-1} (μ_g), respectively. Interfacial tension of the two-phase water–toluene system was 36.2 g s^{-2} (σ).

The average dispersed phase and gas hold-ups were determined by the interruption method, as described in detail in the paper[4]. The uncertainties of the average dispersed phase hold-up and average gas hold-up measurements in the three-phase A–W–T system are ±3% and ±5%, respectively. The volume/surface or Sauter mean drop diameter of the dispersed phase (d_{32}) was calculated by empirical correlation due to Vedaiyan *et al.*[16]:

$$d_{32} = 1.59 \left(\frac{\sigma}{\Delta \rho g} \right)^{0.5} \left(\frac{U_{d0}^2}{2 g d_0} \right)^{-0.067} . \tag{1}$$

After the column operation reached steady state for the chosen level of air, toluene and water flow rates, the tracer flow (5% of the continuous phase flow rate) was started. The tracer feed was a 3 g dm^{-3} solution of potassium chromate in water. The tracer solution, insoluble in toluene, was injected into the column at a place 5 cm above outlet of the continuous phase through four radially directed 1.5 mm holes on a 16 mm o.d. glass distributor. The tracer solution was brought into the column by means of radially directed streams with the aim of producing a uniform concentration of tracer in the continuous phase at the elevation of the tracer distributor.

The continuous phase samples were taken at seven different positions above the tracer flow inlet ($z = 0$, 50, 100, 125, 150, 175 and 200 cm) using small cylindrical graduated test tubes connected to the column, and analyzed by a spectrophotometer SPEKOL (Zeiss, Jena) at 460 nm. The numerical symbol zero denotes a sample position at the tracer inlet. The tracer concentrations in the continuous phase samples were obtained from the calibration curve plotting the sample absorbance against the tracer concentrations, and used to evaluation of the continuous-phase axial dispersion coefficients.

During the operation of the column, some of the traces was carried upstream in the continuous phase by the backmixing action of the drops. The column height upstream from the plane of injection of the tracer was indicated as the test section, and here the continuous phase tracer concentration was determined as a function of the column height. Since potassium chromate is practically insoluble in the dispersed phase used, the transport of the tracer in one direction by the flow of the continuous phase in the test section is just balanced by the transport in the opposite direction, according to Fick's law[9].

$$U_c C = -E_{ct} \frac{dC}{dz} \tag{2}$$

which is integrated to

$$\frac{U_c z}{E_{ct}} = \ln \frac{C_0}{C_z} . \tag{3}$$

Therefore, if the diffusion model applies, a value of the continuous-phase axial dispersion coefficient (E_{ct}) can be determined from the slope of the plot ln (C_0/C_z) against z. Two examples of evalu-

ation of the continuous-phase axial dispersion coefficient in the three-phase A–W–T system are shown in Fig. 1.

The value of the continuous-phase axial dispersion coefficient in the three-phase A–W–T system was determined with an uncertainty of ±5%.

RESULTS AND DISCUSSION

Figure 2 shows, for a gas-agitated spray extraction column, typical results of plotting the continuous-phase axial dispersion coefficient (E_{ct}) for the three-phase A–W–T system against gaseous phase superficial velocity, with the dispersed phase superficial velocity as a parameter, and at a constant continuous phase superficial velocity.

An increase in the gas phase superficial velocity, at a constant dispersed phase superficial velocity, corresponds to an increase in the continuous-phase axial dispersion coefficient. Diaz *et al.*[3] concluded that the hydrodynamic characteristics of the three-phase air–water–kerosene system depended mainly on the magnitude of gas flow. This means that increasing the gas phase velocity causes intensive turbulence, more eddies and back trapping, which increase the axial mixing of the continuous phase.

The dependence of the continuous-phase axial dispersion coefficient for the three-phase A–W–T system on the continuous phase superficial velocity, with the dispersed phase superficial velocity as a parameter, and at a constant gas phase superficial velocity, is shown in Fig. 3.
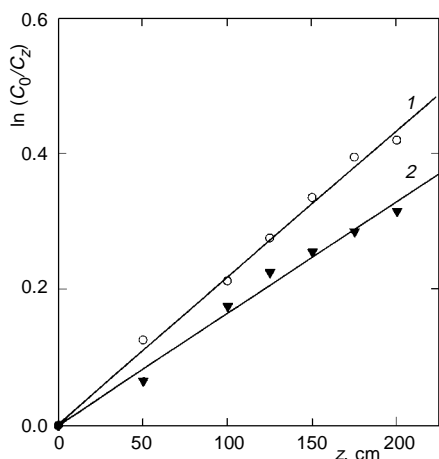

FIG. 1

Examples of evaluation of the continuous-phase axial dispersion coefficient in the three-phase air–water–toluene system: *1* $U_c = 0.106$ cm s^{-1}, $U_d = 0.318$ cm s^{-1}, $U_g = 0.071$ cm s^{-1}, $E_{ct} = 50.0$ cm^2 s^{-1}; *2* $U_c = 0.149$ cm s^{-1}, $U_d = 0.318$ cm s^{-1}, $U_g = 0.124$ cm s^{-1}, $E_{ct} = 91.1$ cm^2 s^{-1}
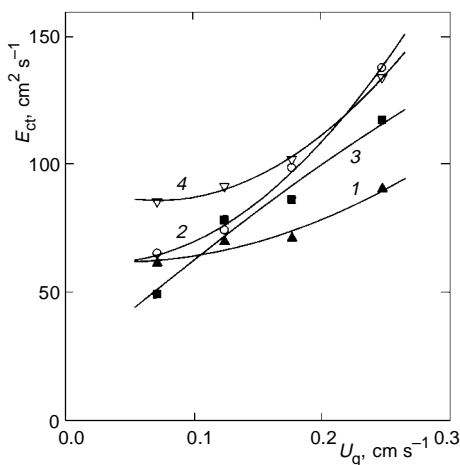
FIG. 2

Dependence of the continuous-phase axial dispersion coefficient values on the gas phase superficial velocity in the three-phase air–water–toluene system: $U_c = 0.149$ cm s^{-1}; U_d (cm s^{-1}): *1* 0.106, *2* 0.149, *3* 0.212, *4* 0.318

Figure 3 shows a non-linear dependence of the continuous-phase axial dispersion coefficient on the continuous phase superficial velocity. Statistical analysis of the experimental data presented in Fig. 3 indicated that this dependence is of the second-order polynomial type. Correlation coefficients for that type of relationship, in all cases shown in Fig. 3, were between 0.79 and 0.98.

Figure 4 illustrates the effect of the dispersed phase superficial velocity on the continuous-phase axial dispersion coefficient, with the water superficial velocity as a parameter, and a constant air superficial velocity.

It is apparent that continuous-phase axial dispersion coefficient is not clearly dependent on the dispersed phase superficial velocity over the range of toluene flows investigated. This independence of the flow rate of toluene is somewhat expected since the drop size is relatively constant ($d_{32} = 0.73–0.84$ cm). It means that the drop rise velocity does not vary much at all flow rates of toluene. Hazlebeck and Geankoplis[8] gave the same explanation for the two-phase water–kerosene system.

Figure 5 shows the dependence of the continuous-phase axial dispersion coefficient in the three-phase A–W–T system on the gas phase hold-up (ε_g). It can be seen from Fig. 5 that an increase in ε_g corresponds to a slight increase in the continuous-phase
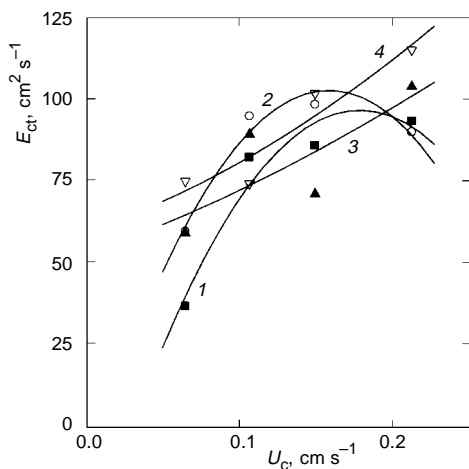

FIG. 3

Dependence of the continuous-phase axial dispersion coefficient values on the continuous phase superficial velocity in the three-phase air–water–toluene system: $U_g = 0.177$ cm s^{-1}; U_d (cm s^{-1}): *1* 0.106, *2* 0.149, *3* 0.212, *4* 0.318. Full lines represent the equations of the second-order polynomial type (*1* $R = 0.97$, $SD_e = 11.50$; *2* $R = 0.98$, $SD_e = 6.51$; *3* $R = 0.79$, $SD_e = 21.16$; *4* $R = 0.95$, $SD_e = 11.11$)
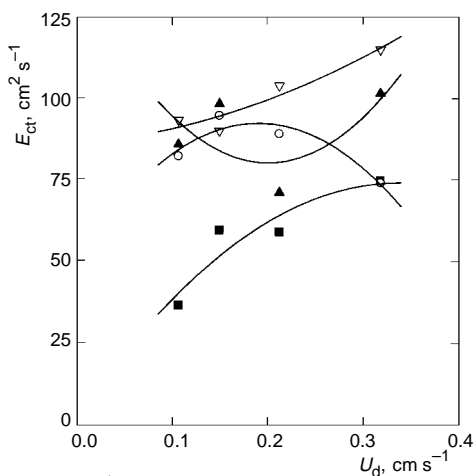
FIG. 4

Dependence of the continuous-phase axial dispersion coefficient values on the dispersed superficial velocity in the three-phase air–water–toluene system: $U_g = 0.177$ cm s^{-1}; U_c (cm s^{-1}): ■ 0.064, ○ 0.106, ▲ 0.149, ▽ 0.212

axial dispersion coefficient. The explanation of the effect of the gas phase hold-up on the continuous-phase axial dispersion coefficient is the same as in case of the effect of the gas phase superficial velocity on the E_{ct}.

The effect of the dispersed phase hold-up (ε_{dt}) on the continuous-phase axial dispersion coefficient is presented in Fig. 6. An increase in the dispersed phase hold-up causes a growth of the continuous-phase axial dispersion coefficient. It is in accordance with the result of Ugar ic[17] for the two-phase water–o-xylene system in the case when the spray extraction column operated with ε_{db} less than 8%.

A comparison of the continuous-phase axial dispersion coefficient data for the three-phase A–W–T system and the two-phase W–T system[14] at the same ratios of the superficial velocities of the dispersed and continuous phases ($U_d/U_c = 0.5$–5.0) is presented in Fig. 7.

Figure 7 shows that the continuous-phase axial dispersion coefficients in the spray extraction column are higher for the three-phase system than those obtained for the two-phase system[14] under the same operating conditions. Regression analysis showed that the mean increase in the E_{ct} was approximately 90%. This is in accordance with the conclusion about the effect of the gas phase superficial velocity on the continuous-phase axial dispersion coefficient (Fig. 2). On the other hand, Perrut et al.[1] found that for the dense packing of drops in the two-phase heptane–DMSO–benzene and heptane–water–acetone ternary systems, the continuous-phase axial dispersion coefficient was
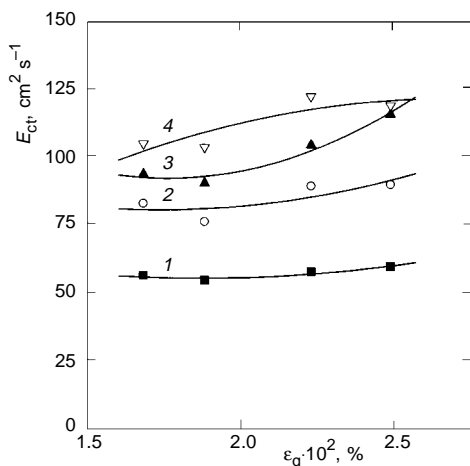

FIG. 5

Dependence of the continuous-phase axial dispersion coefficient values on the gas phase hold-up in the three-phase air–water–toluene system: $U_c = 0.212$ cm s^{-1}; $U_d = 0.106$–0.318 cm s^{-1}; U_g (cm s^{-1}): *1* 0.071, *2* 0.124, *3* 0.177, *4* 0.248
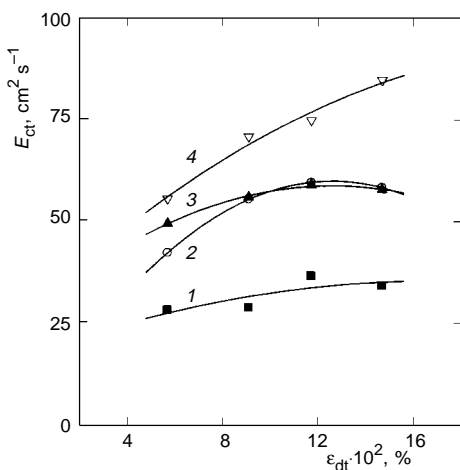
FIG. 6

Dependence of the continuous-phase axial dispersion coefficient values on the dispersed phase hold-up in the three-phase air–water–toluene system: $U_c = 0.064$ cm s^{-1}; $U_g = 0.071$–0.248 cm s^{-1}; U_d (cm s^{-1}): *1* 0.106, *2* 0.149, *3* 0.212, *4* 0.318

considerably reduced in comparison with the loose packing of drops. As indicated in our paper[4], the average and local dispersed phase hold-up values in the three-phase A–W–T system were found higher (15 and 25%, respectively) than those obtained in the two-phase W–T system under the same operating conditions. However, for the three-phase air–water–toluene system[4], the maximum value of the average dispersed phase hold-up was less than 10%. Thus, the mode of packaging of the dispersed phase drops is the dispersed mode. Since the maximum value of ε_{dt} is less than 15%, the same conclusion is valid for the three-phase A–W–T system investigated.

In the present paper, the equation for the prediction of the continuous-phase axial dispersion coefficient developed for the two-phase system[18] was extended by adding an average gas hold-up term, as is given below:

$$\frac{U_c \, d_{32}}{E_{ct}} = 0.124 \left(\frac{U_c^2 \, d_0 \, \rho_c}{\sigma}\right)^{0.77} \left(\frac{U_c \, d_0 \, \rho_c}{\mu_c}\right)^{-0.87} \varepsilon_{dt}^{-0.24} \, \varepsilon_g^{-0.23} \ . \qquad (4)$$
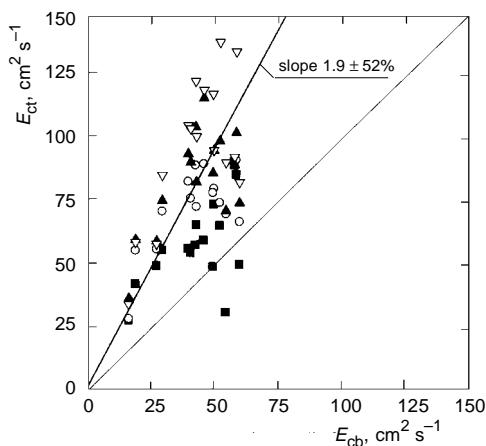

FIG. 7

Comparison of the continuous-phase axial dispersion coefficient values in the three-phase air–water–toluene system and the two-phase water–toluene system: $U_d = 0.106–0.318$ cm s^{-1}; $U_c = 0.064–0.212$ cm s^{-1}; $U_d/U_c = 0.5–5.0$; U_g (cm s^{-1}): ■ 0.071, ○ 0.124, ▲ 0.177, ▽ 0.248. (Line with slope = 1.9 represents the mean value of the increase in the continuous-phase axial dispersion coefficient in the three-phase system)
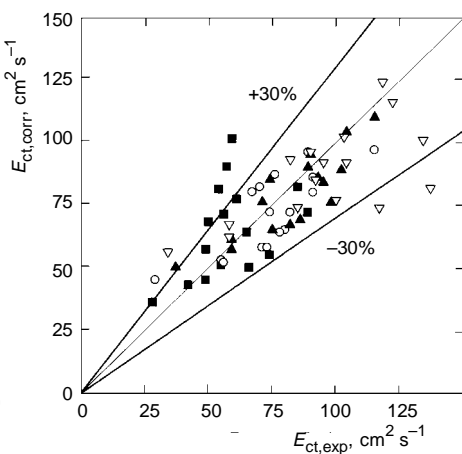
FIG. 8

Comparison of the continuous-phase axial dispersion coefficient calculated from Eq. (4) with experimental data for the three-phase air–water–toluene system: $U_d = 0.106–0.318$ cm s^{-1}; $U_c = 0.064–0.212$ cm s^{-1}; U_g (cm s^{-1}): ■ 0.071, ○ 0.124, ▲ 0.177, ▽ 0.248

Marquardt's algorithm[19] was used to calculate the constants in new correlation Eq. (*4*) for the data of the three-phase A–W–T system investigated.

The average deviation (δ) for the new correlation, Eq. (*4*), is 17.7%. The number of experimental data for the variables included in Eq. (*4*) was $N = 64$. Since the gas phase superficial velocity affects the continuous-phase axial dispersion coefficient in the three-phase A–W–T system, Fig. 2, an attempt was made to use U_g instead of U_c in the first dimensionless group on the right-side of Eq. (*4*). Unfortunately, a satisfactory agreement between the predicted and experimental values of E_{ct} was not obtained. The analytical equations for the prediction of the continuous-phase axial dispersion coefficient in the three-phase G–L–L system were not found in the literature. On the other side, Geankoplis *et al.*[13] showed that the continuous-phase axial dispersion coefficient in the two-phase water–MIBK system varied directly with the continuous phase velocity raised to the 0.42 power.

The comparison of the continuous-phase axial dispersion coefficient calculated from Eq. (*4*) with experimental data for the three-phase A–W–T system investigated is shown in Fig. 8. Seventy-one percent of the predicted continuous-phase axial dispersion coefficients lie within the ±20% limits and 84% within the ±30% limits. This is in accordance with the results for the two-phase system[14,18] and with the conclusion of Horvath *et al.*[20]. They concluded that an average deviation within 30% was sufficient for use with the backmixing models.

SYMBOLS

C	concentration of tracer, g dm^{-3}
DMSO	dimethyl sulfoxide
d_0	nozzle diameter of the distributor of dispersed phase, cm
d_{32}	volume/surface or Sauter mean drop diameter, cm
E	axial dispersion coefficient, cm^2 s^{-1}
g	gravitational acceleration, cm s^{-2}
MIBK	methyl isobutyl ketone
N	number of experimental points
Pe	Peclet number, $Pe = U\, d_{32}/E$
R	correlation coefficient,

$$R = |\sum_{1}^{N} (U - U_a)\, (E_{ct} - E_{ct,a})|/[(N-1)SD_u SD_e]$$

SD_e standard deviation of the axial dispersion coefficient, cm^2 s^{-1},

$$SD_e = \left[\frac{1}{(N-1)} \sum_{1}^{N} (E_{ct} - E_{ct,a})^2 \right]^{1/2}$$

SD_u standard deviation of the superficial velocity, cm s^{-1},

$$SD_u = \left[\frac{1}{(N-1)} \sum_1^N (U - U_a)^2 \right]^{1/2}$$

U superficial velocity, cm s^{-1}

z distance of the sampling points in axial direction, cm ($z = 0$ at the inlet of the tracer solution)

δ average deviation, %,

$$\delta = \frac{100}{N} \left[\sum_1^N \frac{(E_{ct,exp} - E_{ct,corr})}{E_{ct,exp}} \right]$$

ε average hold-up

μ viscosity, g cm^{-1} s^{-1}

ρ density, g cm^{-3}

$\Delta\rho$ $= \rho_c - \rho_d$, density difference between phases, g cm^{-3}

σ interficial tension, g s^{-2}

Subscripts

a   average value

b   two-phase system

c   continuous phase

corr   predicted value

d   dispersed phase

exp   experimental value

g   gas phase

t   three-phase system

0   value where $z = 0$

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