



# Entrainment and holdup of gas-liquid-liquid dispersion in a downflow gas-liquid-liquid contactor



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## ARTICLE INFO

### Keywords:

Three-phase  
Downflow  
Gas-liquid-liquid  
Entrainment  
Holdup  
Dispersion

## ABSTRACT

The gas-liquid-liquid downflow three-phase contactor has the advantage by dispersing gas and lighter liquid into the contactor in a continuous denser phase without any requirement of external power for gas-liquid-liquid processes like reactions and removal of organic compound dissolved in liquid. Present study focuses on entrainment rate of gas and lighter liquid and gas holdup in gas-liquid-liquid downflow contactor. Gas holdup and entrainment are studied using Paraffin liquid-water and Kerosene-water, in the gas-liquid-liquid downflow contactor. The gas holdup variation with fixed lighter liquid volume at different gas flow rates is enunciated based on different operating conditions. Models are developed for gas entrainment and gas holdup in terms of different dimensionless groups consisting of operating variables which can be used to interpret the gas-liquid-liquid downflow contactor.

## 1. Introduction

Gas-liquid-liquid mass transfer and reactions are important unit operations in chemical and biochemical processes. There are many conventional contactors or column reactors used for liquid-liquid operations in presence of gas. The removal of organic pollutant from waste water by a liquid through liquid-liquid interface, gas-liquid-liquid reactions (as shown in Table 1) are some important applications.

The removal efficiency can be increased by increasing the contact time between the two liquid phases by downward movement against its buoyancy by relatively higher downward movement of another liquid. Introduction of inert gas (air, nitrogen, oxygen) in the contactor may increase the further efficiency of the mass transfer. The inert gas acts as a mixing agent to increase the energy in the liquid-liquid (L-L) system. This increased energy causes an increase in dispersion of droplets and hence more separation between phases [14–18]. In continuous large scale operation some of the examples for gas-liquid-liquid are carbonylation, hydro formylation in the petrochemical industry, and gas-aided mass transfer (e.g., extraction). The performance of gas-aided mass transfer is approximately three times higher than liquid-liquid mass transfer without gas [16]. Multiphase catalytic reactions have numerous applications in chemical industries including newly emerging applications in fine chemicals and pharmaceuticals. The analysis of three-phase catalytic reactions are reported including both theoretical and experimental studies by different investigators [17,18]. Multiphase catalytic reactions involving gas, liquid and liquid reactants are

commonly encountered in a variety of chemical transformations. These reactions are generally classified as homogeneous and heterogeneous catalysis depending on the nature of the catalyst. It is well known that homogeneous catalysts consisting of soluble metal complexes have high activity and selectivity compared to the heterogeneous catalysts [13]. The development of biphasic catalysis involving two immiscible liquids has a major advantage in this context [19] and has already indicated its potential for wide range of applications. Catalysis in two phase (liquid-liquid) systems is generally referred as biphasic catalysis, however, many variations of such systems are practised depending on the objectives in specific reactions/processes. This concept involves selection of two liquid phases, such that the catalyst is soluble only in one phase, while the product exists in the other phase. The recent development of water soluble metal complexes has made it feasible to conduct gas-liquid-liquid catalytic reactions in the biphasic mode to achieve easy separation of the catalyst and products [13]. Many useful reactants and products in industrial processes consist of water immiscible organic compounds and hence, the concept of water soluble catalysis provides an important tool for separation of homogeneous catalysts and products.

The gas-liquid-liquid operations in the downflow contactors have many advantages over other conventional contactors, since it has the capability of fluid dispersing into the column without any external power requirement and it made attention to the scientific community for process intensification for achieving higher mass transfer and multiphase reaction. With the increasing energy costs and emission of

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**Nomenclature**

$A_r$	Area ratio (–)
$C$	Constant defined in model Eq. (2) (–)
$C_o$	Distribution parameter (–)
$d_c$	Column diameter (m)
$d_b$	Bubble diameter (m)
$E_s$	Kinetic energy (W)
$E_{um}$	Energy used (W)
$E_{min}$	Minimum energy (W)
$Fr$	Froude number ( $=u_M^2/gd_c$ ) (–)
$g$	Gravitational acceleration ( $m/s^2$ )
$h_c$	Height of column (m)
$h_m$	Mixing height (m)
$H_p$	Penetration depth
$H_{cl}$	Height of continuous liquid (m)
$H_r$	Ratio of height of three phase mixture to diameter of column ( $h_m/d_c$ ) (–)
$H_{ll}$	Height of lighter liquid (m)
$H_{ll-cl}$	Height of mixture in column (m)
$H_{g-ll-cl}$	Height of interface of the mixture (m)
$i.d$	Inner diameter (m)
$k_m$	Mixing loss coefficient (–)
$L_j$	Jet length (m)
$L_c$	Length of the column (m)
$m_g$	Mass flow rate of gas (kg/s)
$m_m$	Mass flow rate of mixture (kg/s)
$M_o$	Morton number ( $=g\mu_{sl}^4/\rho_{sl}\sigma_{sl}^3$ ) (–)
$N$	Number of repeated experiments
$Q_g$	Gas entrainment rate ( $m^3/s$ )
$Q_{cl-ll}$	Continuous and lighter liquid mixture flowrate ( $m^3/s$ )
$R^2$	Correlation coefficient (–)
$Re_j$	Jet Reynolds number ( $=\rho_l d_c u_j/\mu_l$ )
$Re_g$	Gas Reynolds number ( $=\rho_g d_c u_{sg}/\mu_g$ )
$SE$	Standard error (–)
$STDEV$	Standard deviation
$u_b$	Rise velocity of bubble (m/s)
$u_c$	Velocity of continuous phase (m/s)
$u_d$	Drift velocity (m/s)
$u_j$	Jet velocity (m/s)

$u_s$	Slip velocity (m/s)
$u_{sg}$	Superficial gas velocity (m/s)
$u_{sl}$	Superficial liquid velocity (m/s)
$U$	Standard uncertainty
$u_{e,min}$	Minimum entrainment velocity (m/s)
$V_c$	Volume of continuous phase (ml)
$V_s$	Volume of solvent (ml)
$We$	Weber number ( $=\rho_{sl} u_j^2 d_n/\sigma$ ) (–)
$x$	Mass quality (–)
$x_i$	Experimental value at $i$
$\bar{x}$	Mean of repeated experiments
$X$	Lockhart-Martinelli parameter (–)

**Greek letters**

$\epsilon_{cl}$	Continuous liquid hold up (–)
$\epsilon_{ll}$	Lighter liquid hold up (–)
$\epsilon_g$	Gas phase hold up (–)
$\rho_{cl}$	Continuous liquid density ( $kg/m^3$ )
$\rho_{ll-cl}$	Lighter and continuous liquid mixture density ( $kg/m^3$ )
$\rho_g$	Gas phase density ( $kg/m^3$ )
$\mu_{cl}$	Continuous liquid viscosity ( $kg/m.s$ )
$\mu_{ll}$	Lighter liquid viscosity ( $kg/m.s$ )
$\mu_{ll-cl}$	Lighter liquid and continuous liquid mixture viscosity ( $kg/m.s$ )
$\sigma_l$	Surface tension of liquid (N/m)
$\bar{\epsilon}$	Average dissipation energy per unit volume (N m/s)
$\eta_m$	Energy efficiency (–)

**Subscripts**

$cl$	Continuous liquid
$ll$	Lighter liquid
$g$	Gas
$g-ll-cl$	Gas-lighter liquid-continuous liquid mixture
$j$	Jet velocity
$m$	Mixture
$ll-cl$	Lighter liquid-continuous liquid mixture
$sg$	Superficial gas
$s$	Superficial liquid

**Table 1**  
Examples of gas-liquid-liquid catalytic reactions.

Reaction	Catalyst	Category	Author
Oligomerization of ethylene to $\alpha$ -olefins (SHOP process)	Ni-(C <sub>6</sub> H <sub>5</sub> ) <sub>2</sub> PCH <sub>2</sub> COOH in butanediol	G-L-L/organic phase catalysis	Bauer et al. [1]
Oligomerization of ethylene to $\alpha$ -olefins (SHOP process)	Ni-(C <sub>6</sub> H <sub>5</sub> ) <sub>2</sub> PCH <sub>2</sub> COOH in butanediol	G-L-L/organic phase catalysis	Mason [2]
Hydroformylation of propylene to $n$ -butyraldehyde	Rh-TPPTS or Rh-BISBIS in aq. phase	G-L-L/water soluble catalysis	Kuntz [3]
Carbonylation of benzyl chloride to phenyl acetic acid	NaCo(CO) <sub>4</sub> /Bu <sub>4</sub> NBr; aq. NaOH	G-L-L/organo metallic PTC	Cassar and Foa [4]
Hydroformylation of propylene to $n$ -butyraldehyde	Rh-TPPTS or Rh-BISBIS in aq. phase	G-L-L/water soluble catalysis	Kuntz [5]
Carbonylation of isobutyl phenyl ethyl alcohol to ibuprofen (Boots-Hoechst process)	Pd(PPh <sub>3</sub> ) <sub>2</sub> Cl <sub>2</sub> – aq. HCl	G-L-L/organic phase catalysis	Elango et al. [6]
Selective hydrogenation of $\alpha$ -f3 unsaturated aldehyde	Rh-TPPTS or Ru-TPPTS	G-L-L/water soluble catalysis	Grosselin et al. [7]
Hydroformylation of propylene to $n$ -butyraldehyde	Rh-TPPTS or Rh-BISBIS in aq. phase	G-L-L/water soluble catalysis	Herrmann et al. [8]
Hydroformylation of 1-hexene to heptaldehydes	Rh-BISBIS	G-L-L/water soluble catalysis	Herrmann et al. [9]
Hydroformylation of allyl alcohol	HRh(CO)(PPh <sub>3</sub> ) <sub>3</sub> $n$ -heptanol-water	G-L-L/organic phase catalysis	Deshpande et al. [10]
Oligomerization of terminal alkynes	Rh/water soluble phosphines	G-L-L/water soluble catalysis	Baidossi [11]
Carbonylation of allyl chloride to vinyl acetic acid/crotonic acid	Pd/PPh <sub>3</sub> /aq. NaOH	G-L-L-S	Prasad [12]
Hydroformylation of 1-octene	(Rh(COD)Cl) <sub>2</sub> ; Rh:TPPTS:	G-L-L/organic phase catalysis	Chaudhari et al. [13]

G-L-L: gas-liquid-liquid; G-L-L-S: gas-liquid-liquid-solid.

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