

# Comparison of hydrodynamic and mass transfer performances of an emulsion loop-venturi reactor in cocurrent downflow and upflow configurations

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## Abstract

Hydrodynamic parameters (gas-induced flow rate and gas hold-up) and mass transfer characteristics ( $k_L a$ ,  $k_L$  and  $a$ ) have been investigated in a gas–liquid reactor denoted “Emulsair” in which the distributor is an emulsion-venturi and the gas phase is self-aspirated by action of the kinetic energy of the liquid phase at the venturi throat. Two configurations, respectively cocurrent downflow and cocurrent upflow were compared. A chemical method involving the dispersion of a CO<sub>2</sub>–air mixture in a monoethanolamine (MEA) aqueous solution was used to measure mass transfer parameters. Experimental results showed that only the homogeneous bubbling regime prevailed in the upward configuration, while an annular regime could also be observed for cocurrent downflow at low liquid flow rate. Gas-induced flow rate and gas hold-up were usually smaller for cocurrent upflow, both at constant liquid flow rate and specific power input. The same stood for mass transfer properties. Conversely, specific power requirements were lower at constant liquid flow rate and mass transfer characteristics were enhanced at constant gas-induced flow rate for cocurrent upflow. A comparison with other gas–liquid contacting devices showed that the Emulsair reactor is a versatile tool avoiding the presence of mechanically moving parts when high and quickly adaptable dissolved gas supply is required. The cocurrent upflow configuration can be preferred when high gas flow rates are desired because the evolutions of gas-induced flow rate and mass transfer characteristics exhibit a stronger dependence on specific power input in the homogeneous bubbling regime for this configuration.

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## 1. Introduction

In gas–liquid and gas–liquid–solid reactions, the overall production rate is often limited by interphase mass transfer. Relatively large interfacial areas are particularly desirable when absorption is accompanied by a rapid chemical or biochemical reaction. This is particularly true for absorption, such as the treatment of industrial gaseous waste including the absorption of CO<sub>2</sub>, H<sub>2</sub>S, SO<sub>2</sub>, NO<sub>x</sub>, or VOCs, but also for aerobic fermentation and biological wastewater treatments that require high

and quickly adaptable oxygen supply for enhancing the growth of micro-organisms. Many multiphase contacting devices have been described [1]. These differ mainly by the way the dispersion is achieved, the range of the overall gas fraction in the reactor, the interfacial area between the phases and the power requirements for gas dispersion. Three types can be distinguished:

- mechanically stirred tanks in which the driving force of gas dispersion is the power supplied by impellers;
- gas-driven reactors in which dispersion is induced by the gas phase, such as bubble columns with a continuous liquid phase, or packed columns with a continuous gas phase;

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- liquid-driven reactors in which the kinetic energy of the liquid phase is responsible for gas dispersion.

Venturi devices constitute a special type of gas–liquid contactors that belong both to the second and the third class of gas–liquid reactors, as a function of the gas-to-liquid flow rate ratio ( $Q_G/Q_L$ ). Their first advantage is that dispersion is achieved through the kinetic energy of the continuous phase that can be either the liquid (emulsion venturi:  $Q_G/Q_L < 1$ ) or the gas (jet venturi:  $Q_G/Q_L > 10$ ), which may result in either bubbly flows or sprays, respectively. Another advantage is that venturi devices can be used as gas distributors when the reaction is slow, but also as chemical reactors when the kinetics is fast. Additionally, liquid-driven venturis can be easily used as gas-inducing devices; this may reduce the cost of gas supply or constitute a simple and cheap way for recycling the gas phase when this is pure, which is generally the case, for example, for hydrogenations and oxidations. However, there is still a lack of experimental data and modeling procedures to account for the hydrodynamic and mass transfer characteristics of gas–liquid venturi devices, especially when the liquid is the continuous phase, whereas there is an abundant literature on conventional bubble columns, aerated stirred tanks and on the comparison of their respective performance [2]. In the literature, liquid-driven venturis were used first as gas distributors in gas–liquid and gas–liquid–solid reactors without mechanical agitation, such as upward and downward bubble columns with an imposed gas flow rate [3,4]. For gas-inducing systems, Cramers et al. [5], Cramers and Beenackers [6] investigated ejectors, which constitute a particular class of venturi devices in which there is no converging section. Similarly, Cramers et al. [7] studied a particular ejector geometry in which the divergent was replaced by a straight tube. Hydrodynamic and  $k_L a$  data from an emulsion loop-venturi reactor equipped with a conventional venturi with a cocurrent downflow configuration were reported by Gourich et al. [8]. A comparison of this “Emulsair” reactor with other gas–liquid contacting devices was described by Gourich et al. [9] as a function of specific power input. This reactor was shown to be a versatile tool for biochemical and wastewater treatments for which operating conditions have to be modified quickly without the need for mechanically moving parts.

The aim of this work is therefore to compare the hydrodynamic and mass transfer characteristics of cocurrent downflow and cocurrent upflow configurations of the “Emulsair” reactor as a function of liquid flow rate and specific power input. This comparison will include gas-induced flow rate, gas-hold-up,  $k_L a$ , but also  $k_L$  and  $a$  parameters. A comparison with the performances of other conventional gas–liquid contactors, such as bubble column reactors or aerated stirred tanks will also be provided.

## 2. Materials and methods

Experiments were carried out in an “Emulsair” reactor that consisted of a transparent cylindrical tank (30 cm diameter and 0.03 m<sup>3</sup> liquid volume) with a conical bottom topped by an emulsion-venturi made of plexiglas to allow visual observation

of flow regimes. The venturi consisted of a convergent (0.096 m length, 20° angle), a throat (0.02 m length) and a divergent (0.225 m length, 5° angle). The divergent is prolonged by an immersed gas–liquid distributor placed in the center of the tank. The liquid phase was totally recirculated in a “liquid loop”. The main components of this liquid recirculation loop included a valve for flow control, a calibrated flowmeter, a heat exchanger to maintain constant liquid temperature ( $20 \pm 1$  °C) and a recirculation pump. The liquid flow rate  $Q_L$  was varied between  $3.33 \times 10^{-4}$  and  $27.8 \times 10^{-4}$  m<sup>3</sup> s<sup>-1</sup>. The gas phase (CO<sub>2</sub> + air) was self-aspired into the venturi by action of the kinetic energy of the liquid recirculation at the throat through four symmetrically radial orifices (1 cm diameter). The gas flow rate  $Q_G$  was dispersed in the divergent (0.24 L volume) and measured using a volumetric flowmeter. The gas–liquid emulsion was formed through three series of eight orifices laid out regularly on the surface of the distributor. The cocurrent downflow configuration has already been described in detail [8,9]. In this case, the liquid phase was recirculated from the bottom of the reactor and both phases were completely separated in the tank, which allowed the total recirculation of the liquid phase without bubbles. In the cocurrent upflow configuration (Fig. 1), the liquid loop was more complex because the liquid phase was recovered by overflow, which required an additional gas–liquid separation step with a constant liquid level separator to avoid bubble recirculation. Consequently, the feed tank of the downward configuration was used as the gas–liquid separator, which required an additional feed system for the upward configuration (Fig. 1). In both configurations, all the experiments were carried out at atmospheric pressure.

Gas hold-up  $\varepsilon_G$  in both configurations was measured using the volume expansion method. This was shown to be nearly as effective as a more accurate dynamic tracer technique in the Emulsair reactor, even when liquid level fluctuations occurred at high liquid flow rate [8]. The mass transfer parameters  $k_L a$ ,  $k_L$  and  $a$  were measured using a chemical method based on CO<sub>2</sub> absorption from a CO<sub>2</sub>–air mixture in an aqueous solution of monoethanolamine (MEA). The modeling assumptions can be summarized as follows:

- both phases are perfectly mixed;
- the reaction takes place only in the divergent of the venturi, the pipe downstream from the venturi (corresponding to the distributor) and the reaction tank ( $V_{L1}$ : volume of the active zone which is the same in case of upflow and downflow);
- the gas-side resistance to mass transfer is negligible;
- mass transfer may be described by the film-penetration model;
- Henry’s law applies at the gas–liquid interface.

The first assumption is in agreement with those proposed in the literature for similar systems [10–13]. Although it is clear that hydrodynamics and mass transfer differ between the venturi, the distributor and the tank, local measurements cannot be carried out directly in the divergent and the distributor. Furthermore, the gas dispersion coupled to the high liquid recirculation rate favors mixing of both phases in the tank [8]. However, this has been checked using RTD measurements in the gas phase

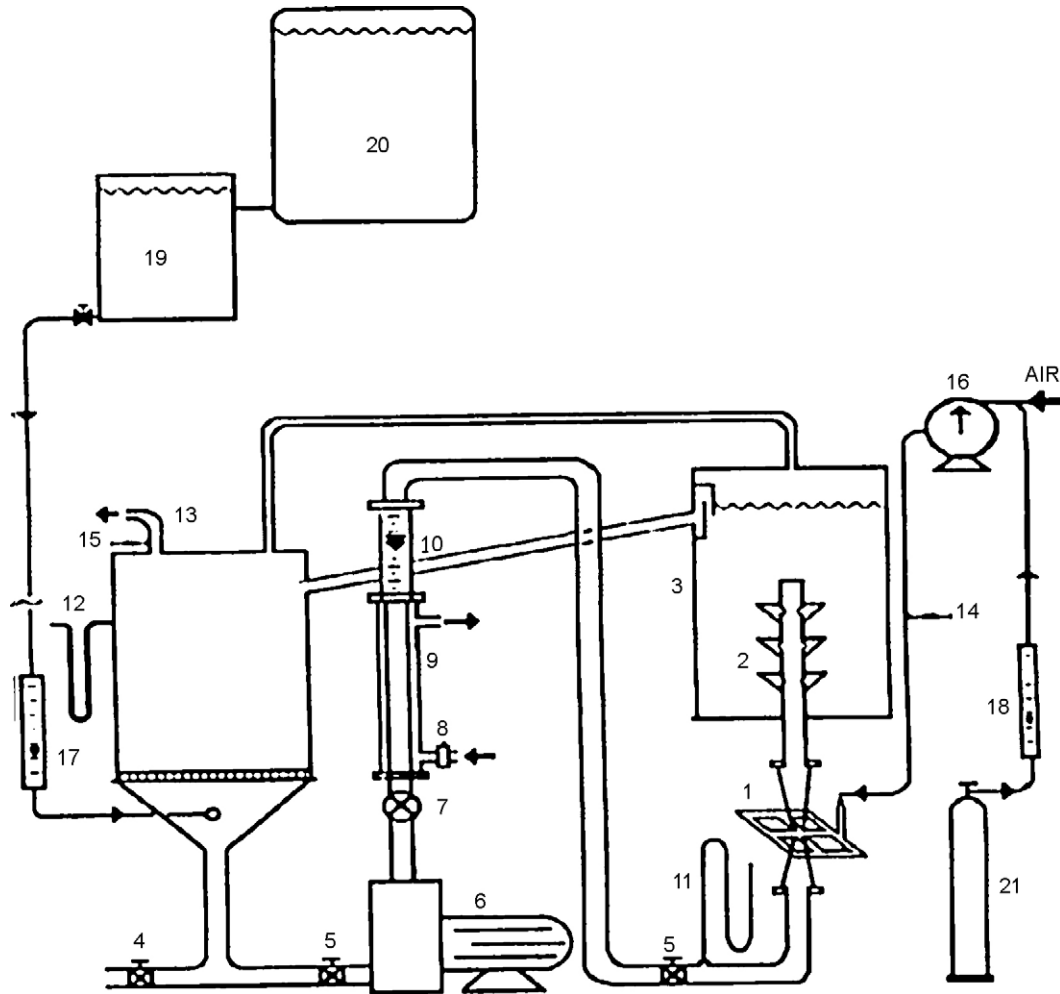


Fig. 1. Experimental set-up for the cocurrent upflow configuration: 1, emulsion venturi; 2, gas distributor; 3, reaction tank; 4, drain; 5, quarter-turn valve; 6, centrifugal pump; 7, liquid flow regulation valve; 8, electrovalve for temperature control; 9, heat exchanger; 10, liquid flowmeter; 11 and 12, overflow; 13, gas outlet; 14, gas sampling system (inlet) to TCD; 15, gas sampling system (outlet) to TCD; 16, gas volumetric flowmeter; 17, flowmeter; 18, CO<sub>2</sub> mass flow controller; 19 and 20, 40 L and 200 L feed tanks; 21, CO<sub>2</sub> bottle.

with helium as a low-solubility gas and we have also varied the inlet concentration of CO<sub>2</sub> over a wide range and shown that the estimates of  $k_L$  and  $a$  remain the same. These experiments have confirmed that the first zone is close to a perfectly mixed tank, although it includes the venturi and the distributor.

The overall volumetric mass transfer coefficient  $k_L a$  was obtained when reaction rate was slow in the mass boundary layer around the bubbles, but rapid enough in the bulk so as to have no dissolved CO<sub>2</sub> in the liquid phase at the outlet of the reactor. This required that Hatta number ( $Ha$ ) was lower than 0.3 (Eq. (1)).

$$Ha = \frac{\sqrt{D_{CO_2} k_2 [MEA]}}{k_L} \quad (1)$$

Eq. (1) assumes a first-order reaction in CO<sub>2</sub> and MEA, in agreement with literature data [14].  $k_2$  the kinetic constant of the acid–base reaction, [MEA] the concentration of MEA in the reactor and  $D_{CO_2} = 1.9 \times 10^{-9} \sqrt{1 - 4.11x}$  is the diffusion coefficient of CO<sub>2</sub> in the liquid phase that is a function MEA

molar fraction  $x$  [15]. The Danckwerts plot technique was used to estimate simultaneously the mass transfer parameters,  $k_L$  and  $a$ , from mass transfer experiments when  $0.3 < Ha < 3$ . The mass transfer properties were obtained from a mass balance on the gas phase using gas chromatography (Porapak Q column) and a thermal conductivity detector (TCD) for the measurement of CO<sub>2</sub> concentration both in the inlet and the outlet gas streams of the reactor. Further details on the gas sampling system involving a pneumatically-controlled six-port valve are reported in Gourich et al. [8]. This method was faster than the direct measurements of MEA concentration in the liquid phase and allowed data acquisition using a RTI 815 A/D acquisition card; it gave directly access to the mass transfer rate  $\Phi_{CO_2}$ . The prevailing absorption regime depended on the MEA concentration in the inlet stream that ranged between 0.02 and 2.0 mol/L, while CO<sub>2</sub> mole fraction in the inlet stream ranged between 1 and 5%. For  $Ha^2 \ll 1$ ,  $k_L a$  was directly estimated from  $\Phi_{CO_2}$  and CO<sub>2</sub> solubility in the liquid phase  $C_{CO_2,L}^*$  (Eq. (2)) that was obtained using Henry's constant  $H_{CO_2} = 26.4 \text{ bar m}^3 \text{ kg mol}^{-1}$  at 20 °C [15]:

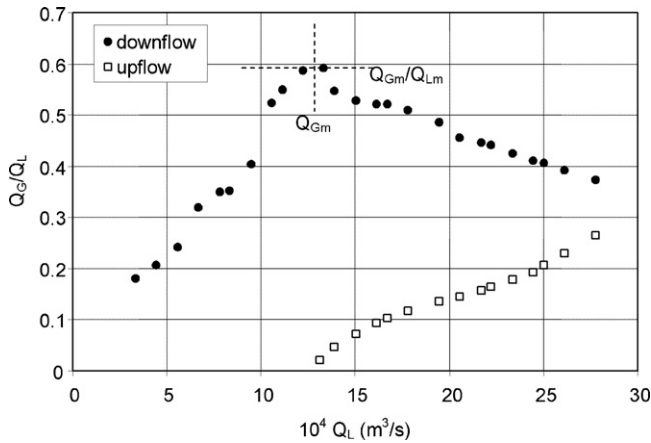


Fig. 2. Evolution of the  $Q_G/Q_L$  ratio as a function of  $Q_L$ .

$$\Phi_{CO_2} = k_L a V_{L1} C_{CO_2,L}^* \quad (2)$$

$Ha$  calculation required the estimation of [MEA] that could be easily deduced from a mass balance on  $CO_2$  on both phases. For  $k_L$  and  $a$  estimation, the Danckwerts method derived the following model when  $0.3 < Ha < 3$ :

$$\Phi_{CO_2} = a V_{L1} C_{CO_2,L}^* \sqrt{k_L^2 + D_{CO_2} k_2 [MEA]} \quad (3)$$

in which  $V_{L1}$  is the liquid volume of the reaction tank. This method consisted in plotting the  $(\Phi_{CO_2}/C_{CO_2,L}^*)^2$  ratio vs. [MEA]. The curve should be a straight line with  $(a V_{L1} C_{CO_2,L}^*)^2 D_{CO_2} k_2$  as the slope and  $(k_L a V_{L1} C_{CO_2,L}^*)^2$  as the intercept, which gave access to  $k_L a$ ,  $a$  and consequently to  $k_L$ . Kinetic data ( $k_2$ ) was measured in a falling film reactor in which the interfacial area and the mass transfer coefficient were known in advance. Experiments gave  $k_2 = 4340 \text{ m}^3/\text{kmol}\cdot\text{s}$  at  $20^\circ\text{C}$  and atmospheric pressure, which is in accordance with the literature [15]. The results obtained for  $k_L a$  measurements for the downward configuration were in agreement with those obtained previously using the oxygenation dynamic method and a dynamic tracer technique [8]. For the emulsion loop reactors, the operating costs essentially depend on the kinetic energy of the liquid phase dissipated between the venturi throat and the liquid free surface in the reaction tank [16]. The specific power input for gas dispersion  $E_L$  ( $\text{W}/\text{m}^3$ ) was estimated from the Eq. (4) by measuring the pressure at the upstream of the venturi using bourdon manometer and the relative pressure at the throat ( $P_C$ ).  $E_L$  can be deduced from the Bernoulli balance

$$E_L = \left( P_C + \frac{1}{2} \rho_L U_C^2 + \rho_L g \Delta z \right) \frac{Q_L}{V_{L1}} \quad (4)$$

in which  $U_C$  is the liquid velocity at the venturi throat (deduced from  $Q_L$ ),  $\rho_L$  the liquid density and  $\Delta z$  the algebraic distance between the venturi throat and the liquid free surface in the reaction tank. The sign of the gravitational potential energy in Eq. (4) depended on the configuration:  $\Delta z$  was negative for cocurrent upflow and positive for cocurrent downflow.

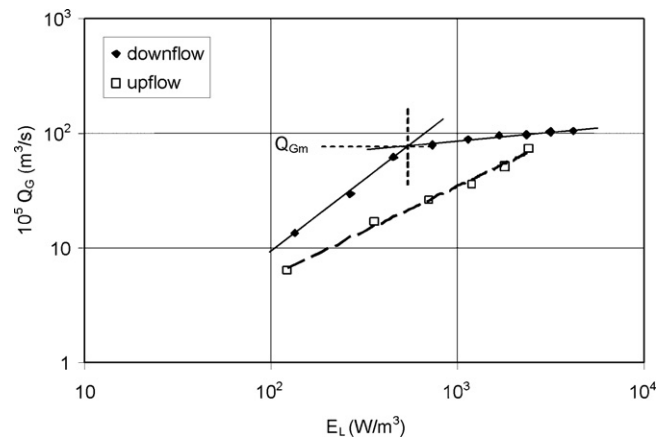


Fig. 3. Evolution of  $Q_G$  as a function of specific power input  $E_L$ .

### 3. Results and discussion

#### 3.1. Hydrodynamics

For the emulsion loop-venturi reactors, a certain minimum liquid velocity at the venturi throat is needed for gas induction to start. In this case, according to the Bernoulli's principle, when the liquid is pumped at a high velocity, a low pressure is created in the throat of the venturi. The analysis of gas-induced flow rate  $Q_G$  showed that self-aspiration started when  $Q_L$  was higher than  $2.78 \times 10^{-4}$  and  $13.1 \times 10^{-4} \text{ m}^3/\text{s}$  for cocurrent downflow and cocurrent upflow configurations, respectively (Fig. 2). In both cases,  $Q_G$  was always an increasing function of  $Q_L$ , even when  $Q_G/Q_L$  decreased in the downward configuration at higher liquid flow rate. However, a crossover point could probably exist between the two  $Q_G/Q_L$  curves at higher liquid flow rate. As a result, gas-induced flow rates remained in the following ranges in this work, respectively  $6.0 \times 10^{-5} \leq Q_G \leq 10.39 \times 10^{-4} \text{ m}^3/\text{s}$  and  $2.89 \times 10^{-5} \leq Q_G \leq 7.40 \times 10^{-4} \text{ m}^3/\text{s}$  for downward and upward configurations. Actually, gas-induction started approximately at the same specific power input, about  $100 \text{ W}/\text{m}^3$ , as shown by Fig. 3, which means that  $E_L$  was lower for cocurrent upflow at constant liquid flow rate. This is confirmed by Fig. 4 that plots the evolution of  $E_L$  vs.  $Q_L$  for both configurations. This behavior was indeed a direct consequence of Eq. (4). The lower

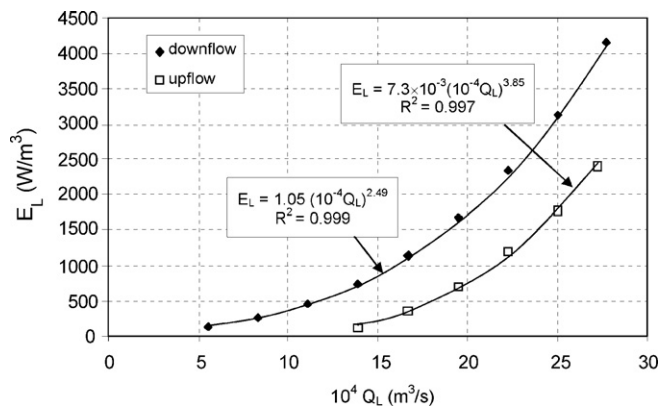


Fig. 4. Evolution of  $E_L$  as a function of  $Q_L$ .

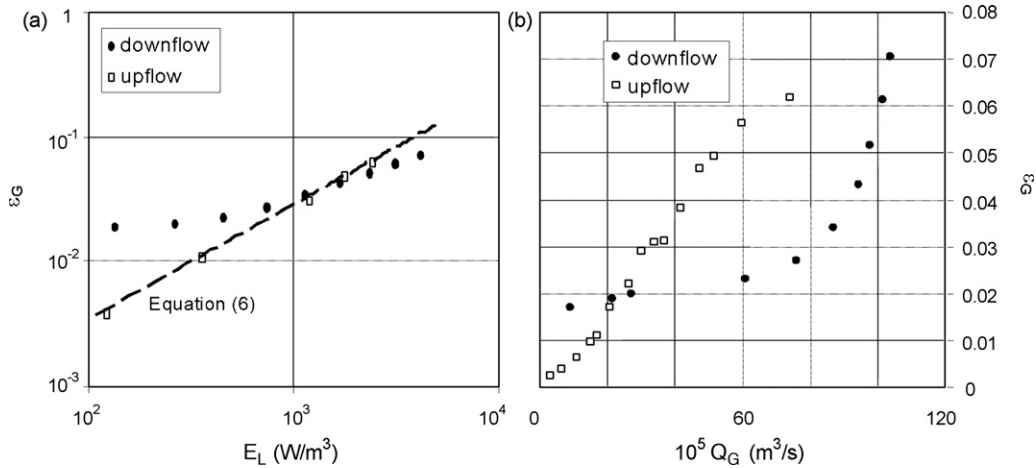


Fig. 5. Evolutions of  $\epsilon_G$  as a function of  $E_L$  (a) and  $Q_G$  (b).

$E_L$  values measured for the upward configuration at constant  $Q_L$  stemmed first from the gravitational potential energy term that was never negligible in comparison to the kinetic energy of the liquid phase, even when  $U_C$  at the throat was maximum (about 9 m/s in this work). However, this should lead to a nearly constant gap between the two  $E_L$  curves at constant  $Q_L$ , while Fig. 4 showed that this gap increased with  $Q_L$ . As a result, frictional effects due to the differences between the liquid loops in both configurations seemed also to play a role and to be lower for cocurrent upflow. Nevertheless,  $Q_G$  remained always higher for cocurrent downflow at constant  $E_L$  in this study.

In the downward configuration, two flow regimes had been reported in the *emulsion venturi* [8], depending on the gas–liquid flow ratio:

- the annular flow pattern in which the gas formed a central core with water flowing down through a peripheral annulus along the wall of the divergent at low gas–liquid flow ratio (i.e. the two-phase gas and liquid flow coaxially in the divergent section);

- the homogeneous bubbling regime in which very small bubbles (i.e. the gas–liquid emulsion) occupied all the divergent section at high gas–liquid flow ratio.

This transition was characterized by a maximum in the  $Q_G/Q_L$  vs.  $Q_L$  plot observed for  $Q_G/Q_L$  about 0.59, which corresponded to  $Q_{Gm} = 7.9 \times 10^{-4} m^3/s$  (Fig. 2). Similarly, it corresponded to a sudden change of slope in the log–log plot of  $Q_G$  vs.  $E_L$ , around  $600 W/m^3$  (Fig. 3). Conversely, these behaviors were never observed for cocurrent upflow: visually, only the homogeneous bubbling conditions were reported. This was in accordance with Figs. 2 and 3 that exhibited neither maximum nor change of slope. However, the  $Q_G/Q_L$  ratio was always an increasing function of  $Q_L$  in the upward configuration, which differed widely from the behavior observed for the downward configuration in the same flow pattern. These differences resulted mainly from the buoyancy forces that opposed to gas dispersion and favored the formation of a gas core in the divergent in the downward configuration, while they enhanced gas dispersion for cocurrent upflow. Fig. 3 highlights the strong

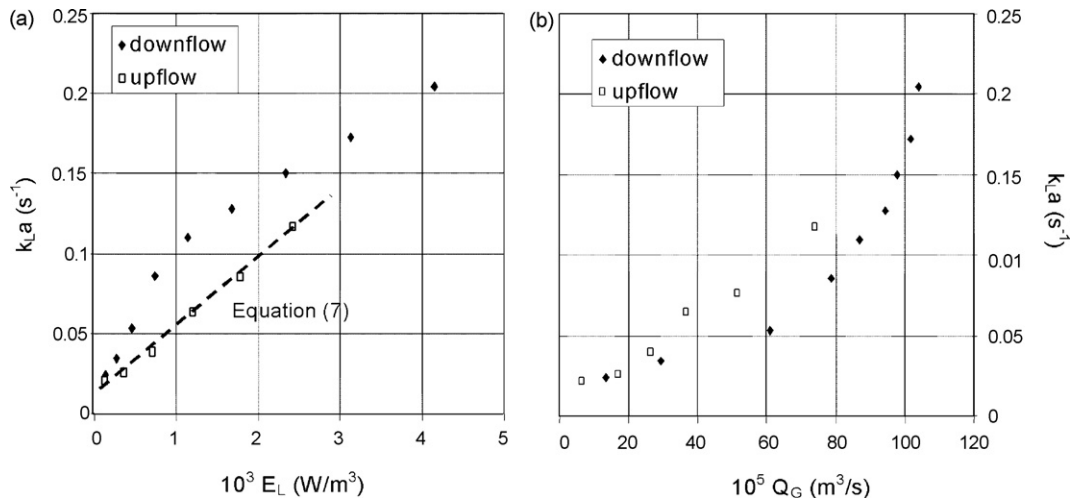


Fig. 6. Evolutions of  $k_L a$  as a function of  $E_L$  (a) and  $Q_G$  (b).

influence of flow regime on gas-induced flow rate.  $Q_G$  rose steeply in the annular regime, while it increased only slightly in the homogenous flow pattern for cocurrent downflow. Gourich et al. [8] had shown that  $Q_G$  varied successively as  $Q_L^2$  and  $Q_L^{0.5}$  when  $Q_L$  was increased. Conversely,  $Q_G$  exhibited a monotonous increase in Fig. 2 in cocurrent upflow and was proportional to  $Q_L^3$ , but the correlation coefficient of this expression was poor ( $R^2 = 0.95$ ). A better agreement was obtained when the evolution of  $Q_G$  was expressed as a function of  $E_L$ :

$$Q_G = 0.161 E_L^{0.78} (R^2 = 0.994) \quad (5)$$

As a comparison,  $Q_G$  varied roughly as  $E_L^{1.05}$  ( $R^2 = 0.98$ ) in the annular regime and  $E_L^{0.16}$  in the homogeneous flow pattern ( $R^2 = 0.97$ ) for cocurrent downflow. This gave an estimation of the crossover point between the two  $Q_G$  curves for  $E_L \approx 4000 \text{ W/m}^3$  and  $Q_G \approx 11 \times 10^{-4} \text{ m}^3/\text{s}$ . As a result, the downward configuration could become rather expensive from an energetic point of view if the desired gas flow rate was far higher than  $Q_{Gm}$ ; cocurrent upflow should therefore be preferred in this case.

The evolution of gas hold-up in the reactor is reported in Fig. 5. This shows that  $\varepsilon_G$  became higher for cocurrent upflow only when  $E_L$  was higher than  $1600 \text{ W/m}^3$ . As mentioned previously for  $Q_G$ ,  $\varepsilon_G$  was adequately correlated to  $Q_L$  for cocurrent downflow using power-law models by Gourich et al. [8], but a relationship between  $\varepsilon_G$  and  $E_L$  provided more accurate results for cocurrent upflow (Fig. 5a). In this case,  $\varepsilon_G$  could therefore be modeled using the following correlation:

$$\varepsilon_G = 4.51 \times 10^{-5} E_L^{0.929} (R^2 = 0.999) \quad (6)$$

An interesting point is that plotting  $\varepsilon_G$  vs.  $Q_L$  (which can be obtained by combining Figs. 4 and 5) would demonstrate that the downward configuration provides always the highest  $\varepsilon_G$  values at constant  $Q_L$ . This confirms that cocurrent downflow enhanced gas dispersion at fixed  $Q_L$ , mainly because gas-induced flow rate was higher (Fig. 2). Conversely, the  $\varepsilon_G$  vs.  $Q_G$  plot showed the opposite behavior for  $Q_G$  values between  $2 \times 10^{-4}$  and  $7.9 \times 10^{-4} \text{ m}^3/\text{s}$  (Fig. 5b). This was not expected because constant  $Q_G$  means higher  $Q_L$  for the upward configuration, which gives a lower  $Q_G/Q_L$  ratio. Consequently, the  $\varepsilon_G$  vs.  $Q_G$  plot can only be explained by the increase of kinetic energy with  $Q_L^2$  that promotes the formation of smaller bubbles and by the fact that bubble coalescence is reduced when  $Q_G$  is lower. These effects compensate apparently the negative impact expected for  $Q_L$  increase at constant  $Q_G$  on  $\varepsilon_G$ .

As a conclusion, the Emulsair reactor with cocurrent downflow provided both higher  $Q_G$  and  $\varepsilon_G$  values when  $Q_L$  was kept constant. The respective performances of both configurations were closer when  $E_L$  was kept constant. On the contrary, the upward configuration provided better results at high-energy input when large  $Q_G$  values were desired because it was not sensitive to the flow regime transition observed in cocurrent downflow. Indeed,  $Q_G$  seemed to be limited at about  $11 \times 10^{-4} \text{ m}^3/\text{s}$  in this configuration, as illustrated by Fig. 5b.

### 3.2. Mass transfer

Experiments showed that  $k_L a$  increased continuously when  $E_L$  was increased, regardless of reactor configuration (Fig. 6a). In the downward configuration, the influence of flow transition was observed and corresponded to a sudden break of the  $k_L a$  slope in the  $k_L a$  vs.  $E_L$  curve. Although small bubbles were formed in the homogeneous bubbling regime, the increase of  $k_L a$  with  $E_L$  was slower than in the annular regime, probably because the slope of the  $Q_G$  vs.  $Q_L$  curve was lower in the homogeneous flow pattern (Fig. 2). Such behavior was not observed in the upward configuration, as expected. Fig. 6a shows clearly that  $k_L a$  was always higher for the downward than for the upward configuration at constant  $E_L$ . However, the increase of  $k_L a$  vs.  $E_L$  was steep both in the annular regime for the downward configuration ( $k_L a \sim E_L^{0.74}$ ) and for the upward configuration ( $k_L a \sim E_L^{0.77}$ ), but less rapid in the homogeneous regime of the downward configuration ( $k_L a \sim E_L^{0.48}$ ). As a result, a crossover point between the two  $k_L a$  curves could be expected for  $E_L \approx 4 \text{ kW/m}^3$ , which corresponds roughly to the crossover point expected for  $Q_G$  curves in Fig. 3. While  $k_L a$  was adequately correlated to  $Q_L$  in the downward configuration [8], a better agreement was obtained with  $E_L$  for cocurrent upflow, in accordance with previous results:

$$k_L a = 2.65 \times 10^{-4} E_L^{0.77} (R^2 = 0.982) \quad (7)$$

This relationship applied in the whole range of  $E_L$  values, as no regime transition was observed for this configuration. As already mentioned for  $\varepsilon_G$ , the gap between the two  $k_L a$  curves would be amplified in a  $k_L a$  vs.  $Q_L$  plot, but the  $k_L a$  vs.  $Q_G$  plot shows that the upward configuration was more efficient per  $\text{m}^3$  gas sucked at the venturi throat (Fig. 6b). This confirms the key role of the kinetic energy of the liquid phase on gas dispersion.

Contrary to previous works [8,9],  $a$  and  $k_L$  values were measured and they are reported in Figs. 7 and 8 respectively, which allows a better understanding of  $k_L a$  evolution. The evolution of  $a$  in Fig. 7a showed that  $a$  values were lower, but only slightly in cocurrent upflow at constant  $E_L$ , although  $Q_G$  and  $\varepsilon_G$  were lower in this configuration (Figs. 3 and 5). As a result,  $a$  was higher at constant  $Q_G$  for cocurrent upflow (Fig. 7b). This behavior could be expected on the basis of the above-mentioned results on  $\varepsilon_G$ : indeed, the higher kinetic energy of the liquid phase transmitted to the gas in the upward configuration should enhance bubble break-up and reduce coalescence rate, both when either  $Q_L$  or  $E_L$  were kept constant between both systems (Figs. 2 and 4). It was however not possible in this work to confirm experimentally that bubble diameters were smaller at constant  $Q_G$  for the upward configuration. Using the fact that  $a = 6\varepsilon_G/d_b$ , the average bubble diameters  $d_b$  were estimated, but all the values remained around  $0.7 \pm 0.1 \text{ mm}$  and both configurations could not be distinguished, mainly because of the statistical error on  $\varepsilon_G$  and  $a$  measurements. Surprisingly,  $a$  did not seem affected by the change in flow regime in the divergent for the downflow configuration. This means that the contribution of the gas core in the annular regime on the interfacial area was negligible, which could however be expected. Thus, the differences in  $k_L a$  values

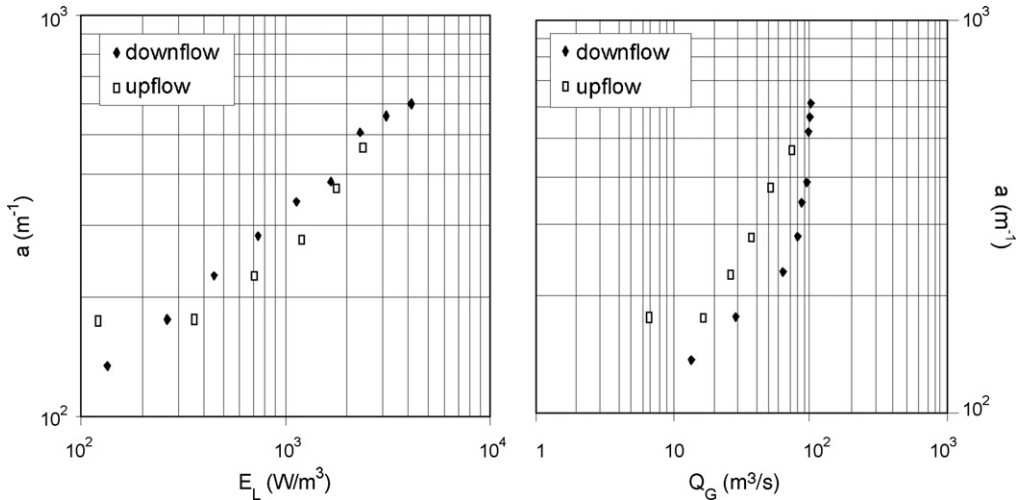


Fig. 7. Evolutions of  $a$  as a function of  $E_L$  (a) and  $Q_G$  (b).

should mainly result from  $k_L$ . This conclusion is confirmed by Fig. 8. In the downward configuration, the transition appeared clearly:  $k_L$  increased with  $E_L$  in the annular regime:

$$k_L = 0.0129 Q_L^{0.595} (R^2 = 0.98) \tag{8}$$

while  $k_L$  remained nearly constant in the homogeneous bubbling regime, about  $3.8 \times 10^{-4}$  m/s. This behavior demonstrates surprisingly that  $k_L$  depended mainly on  $Q_G$ . Indeed,  $k_L$  varied steeply when  $Q_G$  did the same ( $Q_G \sim Q_L^2$ ), but it became nearly independent of  $E_L$  and also  $Q_L$  when  $Q_G$  varied as  $Q_L^{0.5}$ . This interpretation is confirmed by Fig. 8b that represents  $k_L$  vs.  $Q_G$ . As mass transfer takes essentially place in the divergent of the venturi, it seems that  $Q_G$  plays a key role in the complex gas–liquid flow rate that occurs in the throat and the divergent and that the relation between  $k_L$  cannot be simply related to the turbulent kinetic energy of the liquid phase. This means that bubble-induced turbulence, that depends mainly on  $Q_G$ , may prevail, at least when  $Q_L$  is high enough. These assumptions are confirmed by the analysis of the results obtained with the upward

configuration. First,  $k_L$  increased continuously as a function of  $E_L$  for cocurrent upflow and followed Eq. (9) as a function of  $E_L$ :

$$k_L = 3.26 \times 10^{-5} E_L^{2.68} (R^2 = 0.99) \tag{9}$$

$k_L$  values were always lower than in the downward configuration at constant  $E_L$  (Fig. 8) and the same stood at constant  $Q_L$ , as expected. Conversely, they were rather close to those of the downward configuration at constant  $Q_G$ , as shown in Fig. 8b, which demonstrates the key importance of  $Q_G$  on  $k_L$ .

As a conclusion,  $k_L a$ ,  $k_L$  and  $a$  increased with  $E_L$ ,  $Q_L$  and  $Q_G$  in both configurations, despite the flow transition observed in cocurrent upflow. While  $a$  values were close at constant specific energy input, which highlights the key role of kinetic energy of the liquid phase on bubble size,  $k_L$  values depended mainly on  $Q_G$ . As a result,  $k_L a$  was higher in the downward configuration both at constant  $Q_L$  and  $E_L$  values, but the opposite behavior was observed at constant  $Q_G$ . This shows clearly that the choice between downflow and upflow depends widely on the objec-

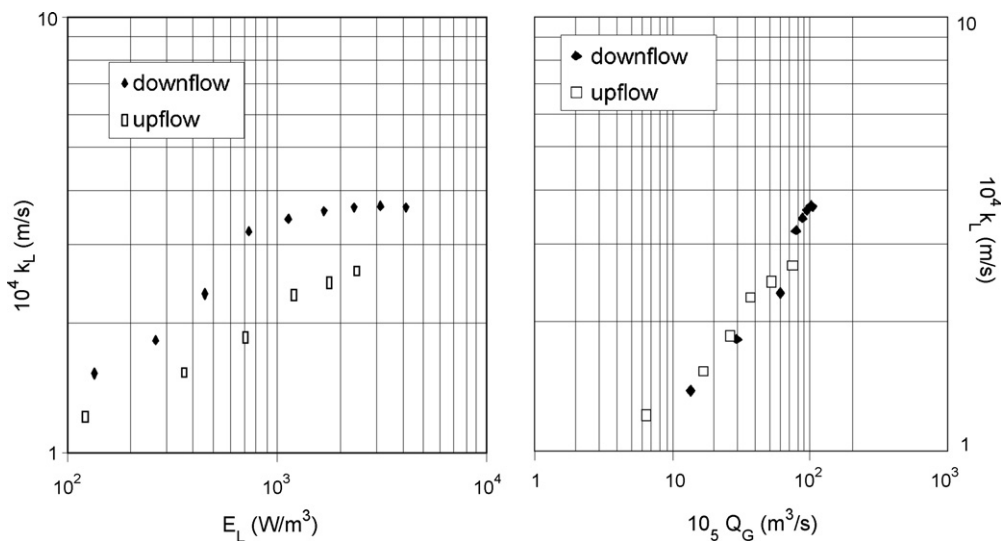


Fig. 8. Evolutions of  $k_L$  as a function of  $E_L$  (a) and  $Q_G$  (b).

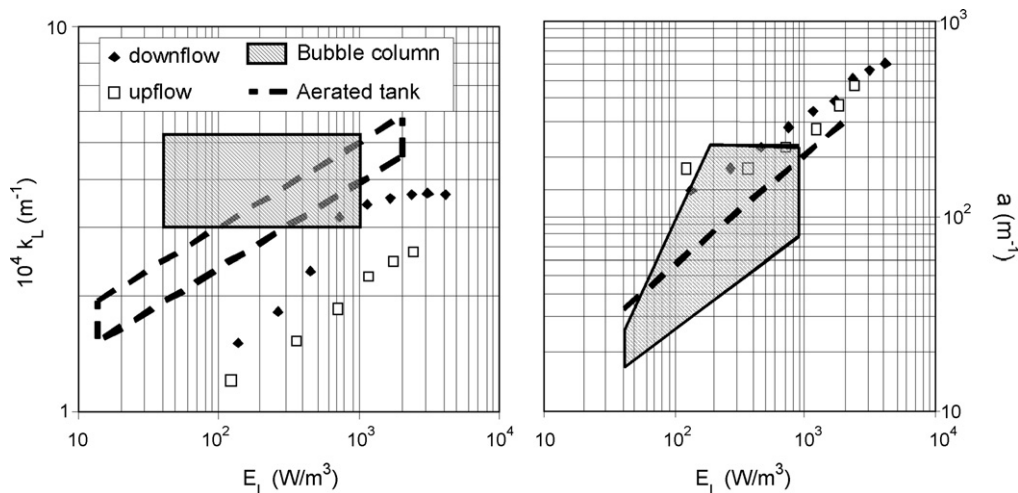


Fig. 9. Comparison of  $k_L$  and  $a$  values with those of other contacting devices.

tives that have to be achieved: cocurrent upflow can be preferred when high power input or gas flow rates are required because it is not sensitive to the bubbling regime transition, i.e. when they are higher than  $4 \text{ kW/m}^3$  and  $11 \times 10^{-4} \text{ m}^3/\text{s}$  in this work, respectively.

### 3.3. Comparison with other gas–liquid contacting devices

A comparison between bubble columns, airlift reactors, aerated stirred tanks with gas-induction and the cocurrent downflow Emulsair reactor in terms of  $\varepsilon_G$ ,  $k_L a$  and  $k_L a/\varepsilon_G$  has already been published by Gourich et al. [9]. Figs. 5 and 7 finalize this comparison for the cocurrent upward configuration. The Emulsair reactor presents always the lowest  $\varepsilon_G$  and  $k_L a$  values as a function of  $E_L$ , regardless of configuration. However,  $k_L a/\varepsilon_G$  was maximum for  $E_L > 1 \text{ kW/m}^3$ , which denoted good mass transfer performance per volume of gas when gas–liquid mass transfer became the limiting step. In this region,  $k_L a/\varepsilon_G$  values were between 2 and 3 for the downward configuration, but only between 1 and 2 for cocurrent upflow. For  $k_L$  and  $a$ , a quantitative comparison with bubble columns equipped with several types of spargers (perforated plate, porous plate, membrane. . .) and aerated stirred tanks equipped with several types of impellers is presented in Fig. 9. This comparison is supported by literature data on bubble column reactors [2,17–19]. The results for airlift reactors from [20–23] have not been directly reported in Fig. 9 because they correspond to the top right of the region of bubble columns [24]. For stirred reactors with gas-induction, Fig. 9 has also been established both using general data on aerated stirred tanks [2,25–27], but also with specific results on gas-induced devices [28].

As expected, Fig. 9 shows that the Emulsair reactor provides the higher interfacial area, but lower  $k_L$  values than bubbles columns and aerated stirred tanks because of the low  $\varepsilon_G$  (i.e. pneumatically-induced mixing is low) and the lack of mechanical agitation in the reactor respectively. It remains therefore a very versatile tool when the presence of mechanically moving parts has to be avoided and when high and quickly adaptable

dissolved gas supply is required as the only operating parameter is the liquid flow rate when the geometry is fixed. This is particularly suitable for fast chemical reactions, but also for biochemical reactions that require high oxygen throughputs during micro-organism growth and for which high oxygen throughputs are not necessary or even unfavorable in the other phases. Similarly, for shear-sensitive micro-organisms, the downflow configuration constitutes an alternative to stirred tanks and airlift reactors, especially when low cost bubble columns are not suitable. This configuration can also be used in the presence of a solid phase and demonstrates therefore a larger applicability than the upward configuration. Additionally to Fig. 9, it has already been shown that the Emulsair reactor presents similar performances as other loop-venturi reactors or ejector-loop reactors [8], but also as reciprocating plate columns [29,30] that do not usually present the same flexibility as the Emulsair reactor. Conversely, the performance of the Emulsair reactor is probably lower than centrifugal field reactors [24], such as rotating packed beds [31], but power requirements are far lower for the Emulsair reactor than for reactors in which gravity is replaced by centrifugal force.

Although this comparison between gas–liquid contacting devices is all but complete because gas dispersion in liquids is still an evolving field, especially with upflow monolith reactors with more and more efficient packings [32], it confirms that the Emulsair reactor (especially the downward configuration), constitutes a versatile gas-inducing device for carrying out complex gas–liquid reactions, for example in the fields of biochemical engineering and environmental engineering, when the gas flow rate must be adapted to requirements that may evolve with time.

## 4. Conclusions

In this work, a quantitative comparison of hydrodynamic parameters and mass transfer properties of an emulsion-venturi reactor, the Emulsair, has been described between a cocurrent upflow and a cocurrent downflow configuration, both behaving as a gas-inducing device. Quantitative correlations have been



derived, especially for the upward configuration that had not been studied previously. Gas flow rate and specific power input were always lower for cocurrent upflow; the same stood for gas hold-up. Conversely, specific power requirements were lower at constant liquid flow rate and mass transfer properties were enhanced at constant gas-induced flow rate for cocurrent upflow. A comparison with other gas–liquid contacting devices showed that the Emulsair reactor is a versatile tool avoiding the presence of mechanically moving parts when high and quickly adaptable dissolved gas supply is required because it generates high interfacial area at the expense of lower  $k_L$  values. The cocurrent upflow configuration can be preferred when high gas flow rates are desired because the evolutions of gas-induced flow rate and mass transfer properties exhibit a stronger dependence on specific power input in the homogeneous bubbling regime for this configuration.

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