



# Process Intensification in Gas-Liquid Mass Transfer by the Introduction of Additives: A Review

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## ABSTRACT

To overcome the challenges of the increasing global energy and to solve the global energy & environment problems, process intensification is one way to develop new efficient production pathways for the chemical industry. Process intensification plays an important role in the gas-liquid mass transfer processes. This review provides an overview of the developments in gas-liquid mass transfer enhancement. A major enhancement method, namely introducing additives (including nanoparticles, oil, electrolyte, and surfactant) summarized and discussed here, includes the most recent accomplishments in gas-liquid mass transfer engineering. This review is expected to inspire new research for future developments and potential applications in scientific research and industry regarding gas-liquid mass transfer engineering. Finally, it presents conclusions and perspectives on enhancing gas-liquid mass transfer.

## INTRODUCTION

The rapid development of the chemical and energy industries leads to an increase in the emissions of greenhouse gas, then resulting in global warming. It is well-known that global warming is a critical challenge for the international community. Greenhouse gases, including carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrogen oxides (NO<sub>x</sub>), are believed to be the source of global warming (Abeydeera et al. 2019). H<sub>2</sub>-liquid mass transfer limits the rate of bi-methanation reactions due to the low distribution of H<sub>2</sub> in solution, limiting its solubility and thus its availability to methanogens. Besides, methane is the main component of natural and shale gas, accounting for 90% of the total abundance (Radler 2011, Kuuskraa et al. 2013). Thus, methane is considered the next generation of carbon feedstock as one of the earth's most widely used high-energy resources.

Nevertheless, methane is a non-polar gas, and its low solubility in solution hinders efficient mass transfer. Oxygen is widely used in microbial growth and product synthesis as a low-cost green oxidant (Garcia-Ochoa et al. 2010). However,

oxygen is less soluble in aqueous solution. Ozonation is one of the most widely used AOP technologies, which has been applied to treat various types of organic wastewater. However, ozone's equilibrium concentration and mass transfer efficiency in water are limited, leading to poor ozone utilization (Gome & Upadhyay 2012). As deduced in the above finding, new approaches are needed to address these limitations.

Process reinforcement is important in various chemical science and engineering aspects (Ramshaw 1983). It allows for resource maximization and does more with less (Luo 2013). As shown in Table 1, the more and the less are summarized (Li et al. 2019). The mass transfer between gas and liquid is widely used in chemical engineering. The rate and efficiency of mass transfer determine the cost and potential profit of gas-liquid mass transfer equipment. Therefore, process reinforcement in the gas-liquid mass transfer has been advocated worldwide (Garcia et al. 2017). Fundamentally, process reinforcement of gas-liquid mass transfer is always related to the total driving force, mass transfer coefficient, and gas-liquid interface area (Fig. 1) (Jensen et al. 2021)

Table 1: The interpretation of process reinforcement. Adapted from ref. (Li et al. 2019).

Types of process reinforcement	“More”	“Less”	Examples
Structural intensification	Production or quality	Size of plants	Microreactor
Energetic intensification	Energy efficiency	Energy consumption	Rotating packed bed
Temporal intensification	Productive rate	Time	Microwave heating
Functional intensification	Functionality	Number of unit operations	Reactive distillation

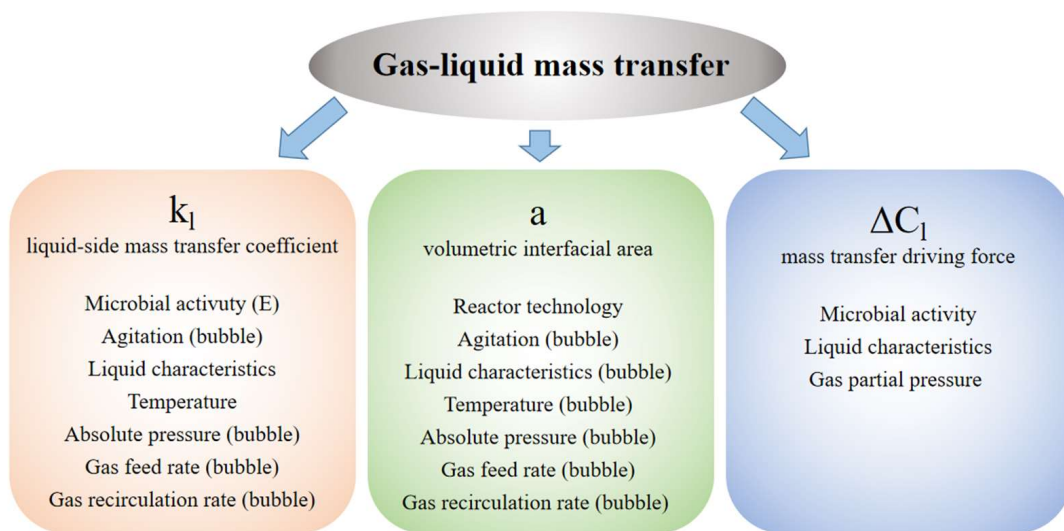


Fig. 1: Overview of the relation between the discussed process parameters and gas-liquid mass transfer. E denotes mass transfer enhancement (Jensen et al. 2021).

This paper summarizes effective reinforcement approaches in the gas-liquid mass transfer process. This technology includes introducing additives such as nanoparticles, oil, electrolyte, and surfactant. It is investigated the recent progress of this technology for enhancing gas-liquid mass transfer performance. More importantly, the mechanisms of different additives are summarized. Finally, the potentials and challenges of this technology of improving gas-liquid mass transfer performance are prospected.

## INTRODUCTION OF ADDITIVES

### Nanoparticles

When small particles are added to a system, a number of possible reactions, such as predation effects, hydrodynamic effects, or inhibition of bubble coalescence, can occur (Fig. 2) (Beenackers & Swaaij 1993, Zhang et al. 2022). This method increases the gas-liquid interface area  $a$  and affects the gas-liquid mass transfer coefficient  $k_L$ , which is greatly affected by the particle size, hydrophobicity, hydrophilicity, and other surface characteristics. Additives are not consumed in the reactors and can be reused by appropriate methods, such as filtering or centrifuge.

Mehdipour et al. (2021) studied  $\text{CO}_2$  absorption enhancement by  $\text{SiO}_2$  and  $\text{ZnO}$  nanoparticles in the rotating liquid sheet contactor. The effect of the tube rotation rate, nanoparticles concentration, gas flow rate, and  $\text{CO}_2$  concentration on the absorption performance was investigated. Also, the adsorption flux increased with an increase in gas flow rate, and increased first and then decreased with an increase in  $\text{CO}_2$  inlet concentration.  $\text{CO}_2$  absorption measurements confirmed that nanoparticles significantly enhanced the separation effect. In this work,  $\text{ZnO}$  nanofluids were more efficient than  $\text{SiO}_2$  nanofluids. Lee et al. (2021) used  $\text{SiO}_2$  to increase  $\text{CO}_2$  absorption performance. In this study, to analyze the enhancement of  $\text{CO}_2$  absorption performance by a nanofluid, the visualization experiment of  $\text{CO}_2$  diffusion was carried out using the shadowgraph method. They observed that the absorption performance of the nanofluid enhanced up to 23.05% by adding 0.05 vol% nanoparticles. The visualization results showed that the hydrodynamic effect was the main factor in improving the mass transfer of nanofluids.

Lakhdiessi et al. (2020) examined the influence of inert nonporous hydrophilic glass beads on the volumetric gas-liquid mass transfer coefficient  $k_L a_L$ . Under constant particle

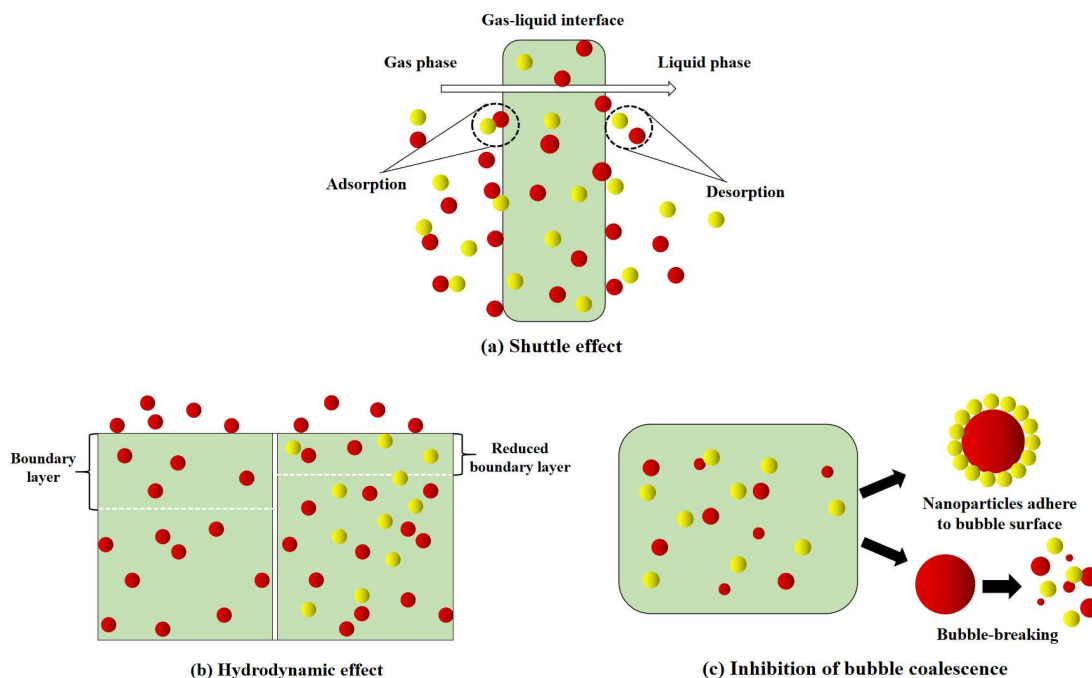


Fig. 2: Shuttle effect (a), hydrodynamic effect (b), and inhibition of bubble coalescence (c). (Beenackers & Swaaij 1993, Zhang et al. 2022).

size, the increase of solid concentration from 1% (v/v) to 5% (v/v) did not affect  $k_L a_L$ . In addition, increasing particle size in the micron range had little effect on  $k_L a_L$  at a constant concentration. In this work, with the increase of solid concentration, the bubble coalescence enhanced, resulting in the increase of bubble size and the decrease of oxygen-liquid interface area  $a_L$ . Secondly, the other particles moved towards the bubble surface due to the collision phenomenon, resulting in local turbulence and increased the liquid side mass transfer coefficient  $k_L$ . And finally, it was concluded that the two-phase approach does not predict the low variability of the experimental three phases  $k_L a_L$  very well. Esmaeili-Faraj et al. (2016) studied  $H_2S$  and  $CO_2$  absorption enhancement by exfoliated graphene oxide (EGO) water and synthesized silica (SS). Based on the adsorption of  $H_2S$  by these functions in EGO and SS nanoparticles, the mass transfer coefficients were 5- and 2-fold for the base fluid, respectively. They concluded that the grazing effect increased mass transfer coefficients in nanofluids (adsorption of gas molecules on the surface of nanoparticles).

## Oil

This part introduces the effect of the addition of the second dispersive liquid phase on the enhancement of gas-liquid mass transfer. Physical properties of the liquid mixture, such as density, viscosity, gas solubility, and gas diffusivity, and the gas-liquid properties, such as droplet distribution

in the boundary layer, possible mass transfer pathways, mass transfer coefficient, and gas-liquid interface area, were changed depending on the interface properties of the dispersed liquid (Dumont & Delmas 2003). In the process of mass transfer, there were two possible path ways in the boundary layer (Dumont & Delmas 2003):

Mass transfer in series: there was a gas→liquid mass transfer, and direct gas→oil contact is impossible (Fig. 3(a-b)).

Mass transfer in parallel: gas→oil contact was possible, and the gas→water mass transfer and the gas→oil mass transfer took place (Fig. 3c).

To examine the effect of the non-aqueous phase on the mass transfer of styrene vapor, Parnian et al. (2016) used silicone oil and experimentally analyzed the parameters that could affect the mass transfer process. Mass transfer of styrene was very sensitive to  $\phi$  when  $\phi < 10\%$ ; however, at  $\phi > 10\%$ , the introduction of silicone oil was insensitive to the process. It was concluded that the mass transfer mechanism is changed to the gas→water→oil pathway by increasing  $\phi$  value from 2% to 20%. Thus, the  $K_L a_L$  increased obviously. Ultimately, the  $K_L a_L$  stayed unchanged regardless of the volume fraction of silicone oil because of the dominance of the gas-water distribution coefficient. This was in agreement with the results of other researchers (Dumont et al. 2014), who showed that the decrease in  $K_L a_L$

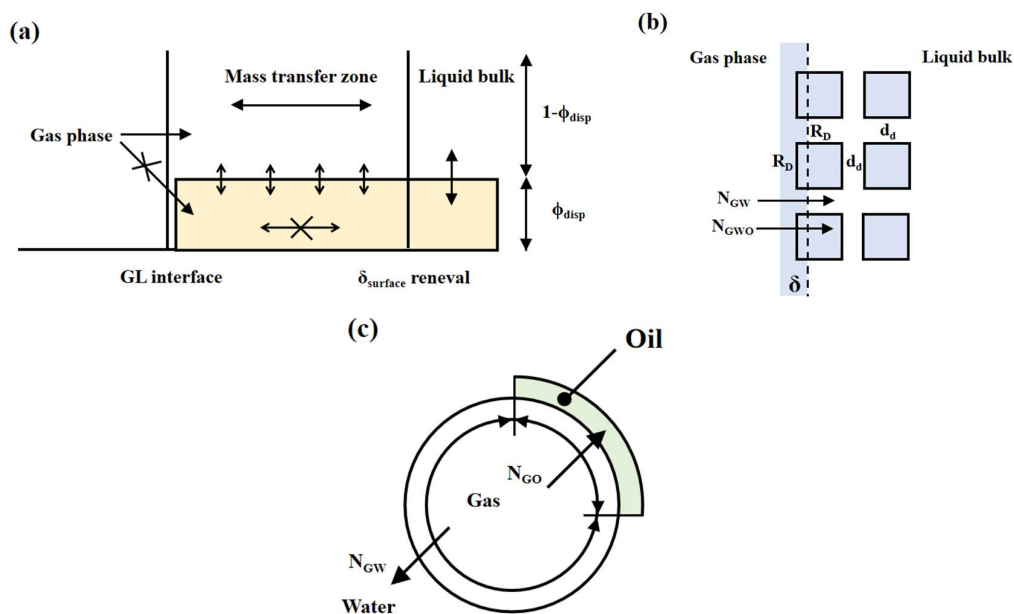


Fig. 3: Mass transfer pathway in series: (a) example of pseudo-homogeneous model representation (no direct gas→oil contact (Brilman et al. 2000); (b) example of heterogeneous model representation (combination of flux to gas→water and gas→water→oil (Zamir et al. 2015)), and (c) mass transfer pathway in parallel (Dumont & Delmas 2003).

value might be related to the change in the distribution coefficient ratio. In addition, Dumont et al. (2014) pointed out that adding silicone oil slightly retarded the mass transfer rate of styrene compared to the air/water system, and  $K_L a$  decreased. Li et al. (2010) explained the  $\text{CO}_2$  mass transfer enhancement mechanism by choosing  $\text{CO}_2$ -benzene or octane/water as a gas/liquid absorption system. The experimental results indicated that increased energy input increased the mass transfer enhancement factor. Small droplets from the dispersed phase enhanced the gas-liquid mass transfer. Small droplets of oil acted like catalysts in chemical reactions. They analyzed the shuttle mechanism and summarized the essence of enhancement of gas-liquid mass transfer was that the dispersed liquid phase increased the components concentration gradient of solute near the gas-liquid interface by absorption. Rols et al. (1990) proposed that the main mechanism of oxygen transfer enhancement was forming a thin film at the gas-liquid interface. This mechanism required gas bubble-oil droplet coalescence followed by oil droplet-oil film coalescence. When bubbles are submerged in the emulsion, the organic liquid covers the surface of the bubbles, and the film formed is loaded with oxygen as long as the bubbles do not explode. Sauid et al. (2013) found that adding palm oil as the organic phase increased the volumetric mass transfer coefficient ( $k_L a$ ). The effect of palm oil on the viscosity and rheology,  $k_L a$ , and gas holdup in the xanthan solution was investigated.

The results showed that introducing palm oil up to 10% vol increased  $k_L a$  by 1.5 to 3 folds, and the maximum  $k_L a$  value was  $84.44 \text{ h}^{-1}$  (Fig. 4). High oxygen transfer was obtained without providing additional energy. The increase was due to increased liquid turbulence caused by rigid organic phase droplets (Rols et al. 1990). The spreading coefficient played a significant role in changing the oxygen transfer capability in the system (Yoshida et al. 1970). When the diffusion coefficient was positive, the organic encounter on the water surface propagated like a surfactant to reduce the surface tension, thereby increasing the interface area and eventually increasing (Yoshida et al. 1970). This favorable effect promoted the formation of small bubbles due to the properties of palm oil.

However, other researchers have drawn different conclusions. Boltes et al. (2008) have examined the effect of airflow rate and organic fractions of dodecane on the gas-liquid mass transfer in oil-water emulsions. For the same organic fraction, the airflow rate enhancement has witnessed an increase in the  $K_L a$ . In agreement with the previous result, it was confirmed that the  $K_L a$  values obtained for the organic liquids were higher than the corresponding values for water (Mehrnia et al. 2005, Shariati et al. 2007). For the same airflow, the  $K_L a$  value decreased with an increase in the organic fraction, and then with a further increase in the organic fraction, the  $K_L a$  value was increased. Shariati et al. (2007) also concluded that the mass transfer decreases

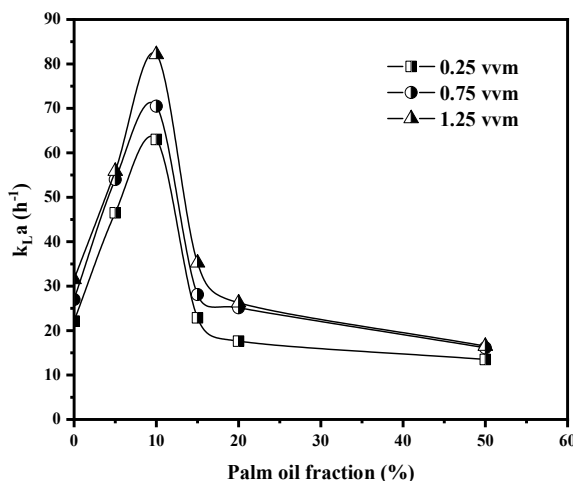


Fig. 4: Influence of palm oil on  $k_{L,a}$ . (Saud et al. 2013).

with an increased water-to-oil phase volume ratio. Kundu et al. (2002) have investigated the gas-liquid mass transfer enhancement by adding different second immiscible organic phases (toluene, 2-ethyl-1-hexanol, decyl alcohol, anisole, dodecane, n-decane, and n-heptane). The results showed that adding 1% of n-decane, dodecane, and n-heptane in liquid increased the mass transfer. However, adding toluene, anisole, and 2-ethyl-1-hexanol limited the mass transfer. Non-linear variations of the enhancement factor were to be noticed. This may be due to the formation of an oil-water complex (several small droplets trapped in a large oil droplet as multiple droplets by Sajjadi et al. (2002)) that occurs at a higher concentration of dispersed phase and lower gas velocity. In addition, the main effect of adding oil appears to be a change in bubble size. Larger bubble diameters and lower gas fractions mean lower surface area and, thus,

reduced mass transfer. However, these results cannot be explained by the modest change in the properties of the gas-oil saturated water interface due to oil addition.

### Electrolyte

Kim et al. (2016) employed various electrolytes such as  $MgSO_4$ ,  $Na_2SO_4$ ,  $K_2SO_4$ ,  $MgCl_2$ ,  $NaCl$ ,  $KCl$ ,  $MgBr_2$ ,  $NaBr$ ,  $KBr$ ,  $Mg(NO_3)_2$ ,  $NaNO_3$ , and  $KNO_3$  to investigate the effect of the electrolytes on methane-water volumetric mass transfer coefficient ( $k_{L,a}$ ). In this work, an increase in electrolyte concentration could enhance the  $k_{L,a}$ . Anions had a greater effect on  $k_{L,a}$  enhancement than that of cations (Fig. 5). For electrolytes with the same cations, the  $k_{L,a}$  value of sulfates, chlorides, bromides, and nitrates were 613-711, 381-488, 141-290, and 233-266  $h^{-1}$ , respectively. For electrolytes with the same anions, magnesium-containing electrolytes

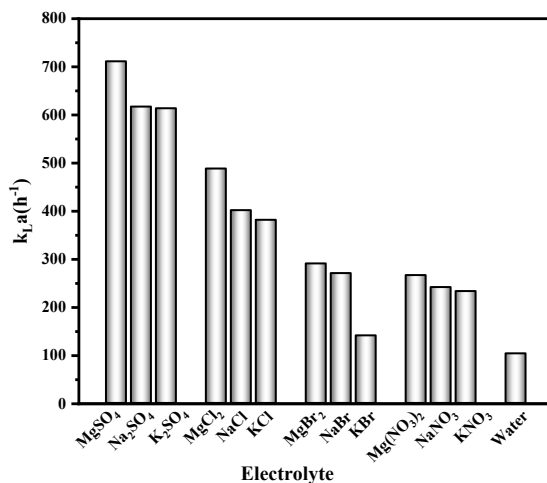


Fig. 5: Methane water volumetric mass transfer coefficient for various electrolytes at 5 wt% concentration. (Kim et al. 2016).



exhibited larger  $k_L a$  values than those containing sodium and potassium. Magnesium sulfate had the greatest strengthening effect on  $k_L a$  at the concentration of 5 wt%, up to  $711 \text{ h}^{-1}$ . This was the highest  $k_L a$  value of methane water reported in the published literature. Inhibition of bubble coalescence in electrolyte solution favored the increase of the  $k_L a$  value of methane water. The enhanced effect of electrolytes on methane water mass transfer could be attributed to increased methane water interfacial area. Anions have a stronger effect on  $k_L a$  enhancement than cations. For all electrolytes, ions with high charge density greatly enhanced methane water's  $k_L a$  value.

Góngora García et al. (2021) studied the combined effects of mixed electrolyte species ( $\text{KH}_2\text{PO}_4$ ,  $\text{K}_2\text{HPO}_4$ ,  $(\text{NH}_4)_2\text{SO}_4$ , and  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ ) on the oxygen transfer in a bubble column. Electrolytes containing ions are prone to solute interactions or crystallization. Volume mass transfer coefficient ( $k_L a$ ) and specific interface area ( $a$ ) are enhanced due to the inhibition of bubble coalescence as the electrolyte concentration increases to a critical value. In saline solution, the  $k_L a$  tended to increase (between 58 and 72%) as the nominal ionic strength increased from 0 to 0.085. The electrolyte inhibits bubble coalescence (resulting in smaller bubbles forming), thus maintaining a higher interface area. In addition, the small bubble and the low slip velocity appeared due to the low oxygen diffusion rate in the salt solution. Thus, the local mass transfer decreased (Taweel et al. 2013, Baz-Rodríguez et al. 2014). Overall, the influence on  $a$  seems to control the  $k_L a$  value. Jang et al. (2018) employed various electrolytes such as  $\text{CaCl}_2$ ,  $\text{K}_2\text{HPO}_4$ ,  $\text{MgSO}_4$ ,  $\text{NaCl}$ , and  $\text{NH}_4\text{Cl}$  to investigate the effects of the electrolytes on the enhancement of  $\text{CO}$  mass transfer in hollow fiber membrane bioreactor. The lowest value of  $k_L a$  for  $\text{CO}$  achieved in water was  $137.2 \text{ h}^{-1}$ . However,  $k_L a$  values of  $\text{CO}$  increased to  $568.8 \text{ h}^{-1}$  (5%  $\text{NH}_4\text{Cl}$ ),  $340.2 \text{ h}^{-1}$  (5%  $\text{MgSO}_4$ ),  $279.4 \text{ h}^{-1}$  (3%  $\text{NaCl}$ ),  $410.0 \text{ h}^{-1}$  (5%  $\text{CaCl}_2$ ), and  $465.8 \text{ h}^{-1}$  (5%  $\text{K}_2\text{HPO}_4$ ). Electrolytes inhibited  $\text{CO}$  bubble coalescence, enhancing

the maximum  $k_L a$  by a factor of 4.14. Dense electrical layers that formed near the gas bubbles inhibited bubble coalescence (Fig. 6) (Craig et al. 1993), thus increasing the mass transfer area.

Various electrolytes, including  $\text{NaNO}_3$ ,  $\text{NaCl}$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{Na}_2\text{SO}_4$ ,  $\text{KCl}$ ,  $\text{LiCl}$ ,  $\text{CsCl}$ ,  $\text{BaCl}_2$ , and  $\text{MgCl}_2$ , were employed by Kazakov et al. (2014) to investigate the influence on the  $\text{O}_2$  mass transfer in the presence of quartz particles in a poly (vinyl chloride) shell ( $\text{SiO}_2/\text{PVC}$ ). Positive hydrating ions significantly reduced the  $\text{O}_2$  mass transfer enhancement factor and increased by negative hydrating ions. In electrolytes containing different cations with the same anion ( $\text{KCl}$ ,  $\text{LiCl}$ ,  $\text{CsCl}$ ,  $\text{BaCl}_2$ , and  $\text{MgCl}_2$ ), the dependences of the  $\text{O}_2$  mass transfer enhancement factor  $E$  on the salt concentration were various. For  $\text{KCl}$ ,  $\text{LiCl}$ ,  $\text{CsCl}$ ,  $\text{BaCl}_2$ , and  $E$ , it was increased with an increase in electrolyte concentration. The introduction of these salts into the aqueous phase strengthened  $\text{O}_2$  mass transfer. However, the addition of  $\text{MgCl}_2$  seemed to not affect  $E$ . For electrolytes containing different anions with the same cations ( $\text{NaNO}_3$ ,  $\text{NaCl}$ ,  $\text{Na}_2\text{CO}_3$ , and  $\text{Na}_2\text{SO}_4$ ), the dependences of the coefficient  $E$  on the salt concentration strongly vary between electrolytes. For salts containing singly charged anions ( $\text{NaNO}_3$  and  $\text{NaCl}$ ),  $E$  increased with electrolyte concentrations reaching 2.5-2.6. Nevertheless, the concentrations of salts with doubly charged anions increased, and the  $E$  decreased to 0.6-0.7. It was concluded that introducing these electrolytes was detrimental to the  $\text{O}_2$  absorption rate. Baz-Rodríguez et al. (2014) have examined the effect of inorganic electrolytes ( $\text{NaCl}$ ,  $\text{MgCl}_2$ ,  $\text{CaCl}_2$ ) on oxygen-liquid mass transfer in a bubble column. The volumetric mass transfer coefficient ( $k_L a$ ), mass transfer coefficients ( $k_L$ ), and specific interfacial area ( $a$ ) were determined. Using a dimensionless concentration ( $c_i = c/c_{ic}$ ) as the independent variable, the  $k_L a$  value was the same regardless of the kind of electrolytes at constant superficial gas velocities ( $v_{sg}$ ); the same happened for  $k_L$ .

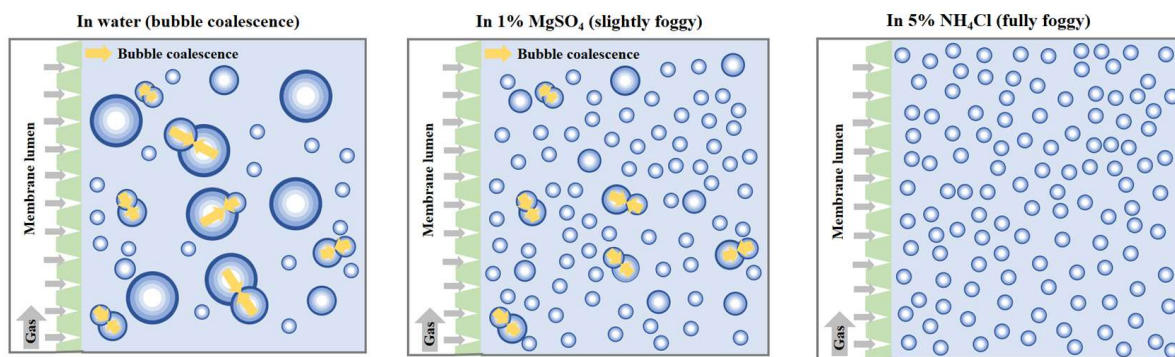


Fig. 6: Comparisons of the gas bubbling in water and electrolyte solutions. (Craig et al. 1993).

The critical concentrations for bubble coalescence ( $c_{ic}$ ) were the key to characterizing the hydrodynamic behavior (Ribeiro & Mewes 2007, Syeda & Reza 2011). The presence of electrolytes strongly changed the hydrodynamics and gas-liquid mass transfer, mainly due to the decrease of bubble size and the corresponding increase  $a$ . The bubble coalescence inhibition played an important role in changing the hydrodynamic behavior. As the slip velocity decreased with the increase of  $c_r$ ,  $k_L$  decreased. The presence of electrolytes reduced the  $k_L$ , their effect on a large enough to lead to a net increase in  $k_L a$ . A 40-50% increase in  $k_L a$  compared with air-pure water. Nevertheless,  $k_L$  was reduced by about 60% compared to air-pure water. In agreement with the results reported previously, it was confirmed that  $k_L$  decreased in the presence of electrolytes (Taweel et al. 2013).

### Surfactant

Surfactants are compounds that reduce the surface tension (or interfacial tension) between multiphase. The different types of surfactants are displayed in Fig. 7. (Bakthavatchalam et al. 2020). Adding surfactants in the gas and liquid phases to adjust the surface tension can affect the gas-liquid mass transfer process. We know that surfactants are amphiphilic organic compounds containing hydrophobic and hydrophilic groups. This property makes them accumulate at the gas-liquid interface, which may positively or negatively affect gas-liquid mass transfer. Hence, it's worth studying the influence of surfactants on the mass transfer process.

Pichetwanit et al. (2021) investigated the influence of nonionic surfactants with a small hydrophilic (Tergitol TMN-6, Triton X-100, Tergitol 15-S-9 and Tween-80) and cationic surfactants with a short linear chain (dodecyl trimethyl ammonium bromide (DTAB) and cetyl trimethyl

ammonium bromide (CTAB)) on the mass transfer of carbon dioxide ( $CO_2$ ) from the gas phase to monoethanolamine (MEA) solution. The addition of surfactant reduced the surface tension of the MEA and increased the contact area for the absorption of  $CO_2$ . Generally, mass transfer interfacial area increased with the addition of surfactant (Chaumat et al. 2007, Hebrard et al. 2009, García-Abuín et al. 2012, McClure et al. 2015). The results showed that the  $CO_2$  equilibrium loading with nonionic and cationic surfactants slightly increased with surfactant concentration. Excluding tween-80 and Triton X-100, other surfactants improved the absorption rate of  $CO_2$  in MEA solution. These results indicated that the initial  $CO_2$  absorption rate of MEA was improved by using a small hydrophilic head non-ionic surfactant or a short linear chain cationic surfactant, such as Tergitol TMN-6 or DTAB. In addition, an increase in the longer hydrocarbon chain in nonionic or cationic surfactants did not significantly promote the absorption rate of  $CO_2$  in the MEA solution. It was worth noting that the surfactant had no significant effect on the MEA solution's kinematic viscosity but slightly increased amine viscosity. Dang et al. (2022) conducted experiments with surfactants (Tween 80 with different surface tensions) to study possible enhancement in oxygen transfer. It had been observed that with increased surfactant concentration, the gas-liquid mass transfer performance first deteriorated and then improved. The addition of surfactant changed the surface tension. Surfactants work in two ways: (1) The addition of surfactant reduced the surface tension of bubbles, resulting in the decrease of bubbles coalescence, fine and uniform dispersion, and increased gas-liquid interface area. (2) Surfactant molecules attached themselves to the gas-liquid interface and formed a single-molecule film with certain mechanical strength, which hindered gas-liquid mass

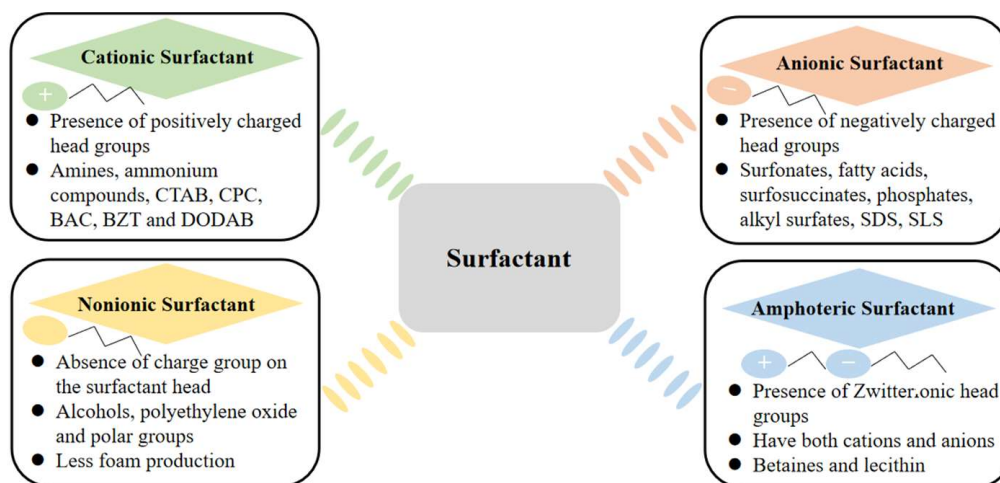


Fig. 7: Types of surfactants. Reproduced from ref. (Bakthavatchalam et al. 2020).

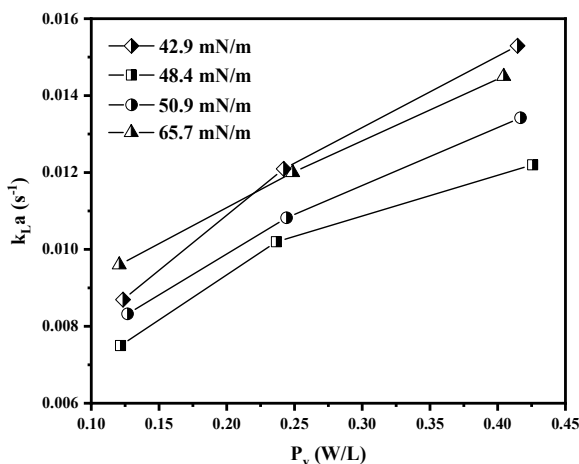


Fig. 8: Effect of surface tension on the gas-liquid mass transfer coefficient  $k_{L,a}$  (Dang et al. 2022).

transfer. As could be seen from Fig. 8, with a decrease in the surface tension from  $65.7 \text{ mN}\cdot\text{m}^{-1}$  to  $48.4 \text{ mN}\cdot\text{m}^{-1}$ , the single-molecule film formed by surfactant molecules at the gas-liquid interface had inhibitory oxygen mass transfer than promotion. An increase in surfactant concentration was expected to correspond with an increase in the inhibitory effect. It was observed that the enhancement in gas-liquid mass transfer gradually increased with a decrease in surface tension. At low speeds, however, the smaller mass transfer coefficient of  $42.9 \text{ mN}\cdot\text{m}^{-1}$  than that of  $65.7 \text{ mN}\cdot\text{m}^{-1}$ .

Yang et al. (2020) studied the influence of surfactant (oleic acid) on the nitrogen-liquid mass transfer. When the oleic acid concentration was increased, the mass transfer coefficient decreased, or the interface resistance increased. It was observed that oleic acid decreased the gas-liquid mass transfer by a factor of 4. This indicated that the excess surface concentration of oleic acid at the gas-liquid interface increased with the aqueous phase concentration until the aqueous phase concentration reached the critical micelle concentration. Since the solubility of nitrogen in pure oleic

acid was very small, the presence of oleic acid molecules at the interface increased the possibility that nitrogen transport across the interface would be impeded (Fig. 9). However, when oleic acid concentration was  $2 \text{ mg L}^{-1}$ , the interface resistance was about four times that for pure water (Birgand et al. 2007). In addition, the low concentration of oleic acid had a little effect on the diffusion coefficient of nitrogen in water

Lebrun et al. (2022) investigated the effect of the surfactants' nature on oxygen mass transfer. The mass transfer coefficient of oxygen ( $k_L$ ) values were determined for each of the aqueous solutions with the presence of three cationic surfactants with different hydrophobic chain lengths (including hexadecyltrimethyl ammonium chloride, dodecyltrimethyl ammonium chloride trimethyl octyl ammonium chloride), and four nonionic surfactants with different hydrophilic chain lengths (including Triton X-100, Triton X-102, Triton X-165, Triton X-305). It could be seen that with an increase in the bulk concentration of surfactant, the liquid-side mass transfer coefficient decreased, ranging

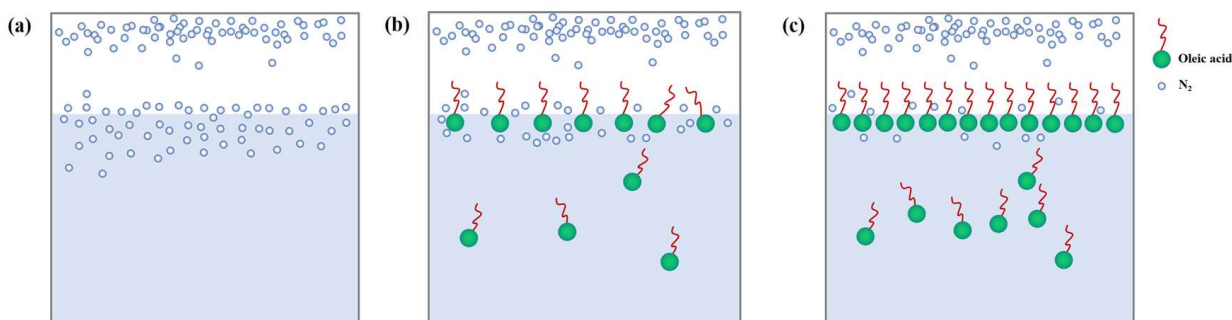


Fig. 9: Schematic of nitrogen transferring into aqueous solution with resistance at the interface due to accumulation of oleic acid: (a) control (no oleic acid); (b) small surface concentration of oleic acid; and (c) large surface concentration of oleic acid (Yang et al. 2020).



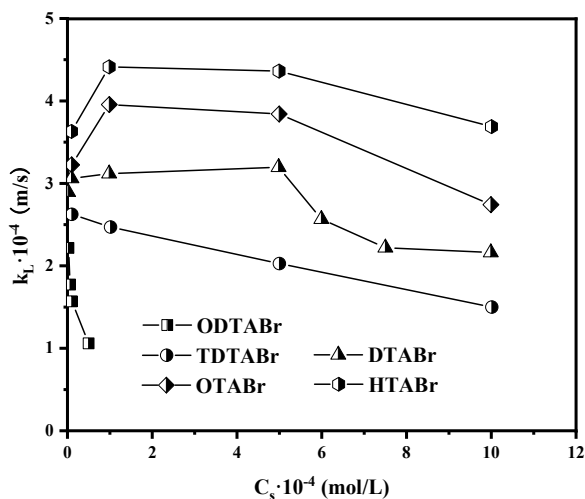


Fig. 10: Influence of different concentrations for several surfactants on the mass transfer coefficient (Gómez-Díaz et al. 2009, García-Abuín et al. 2010).

from  $5.6 \times 10^{-4}$  to  $0.4 \times 10^{-4}$   $\text{m}\cdot\text{s}^{-1}$ . However, increasing the length of the hydrophilic chain of nonionic surfactants does not affect the oxygen mass transfer, even though it changes the interface density. Increasing the hydrophobic chain length of cationic surfactants led to a decrease in the mass transfer coefficient of oxygen. And finally, the Sherwood number was calculated in each medium and classical correlations for gas-liquid mass transfer prediction.

The effect of the hydrocarbonate chain length of surfactant on the gas-liquid mass transfer process was investigated by García-Abuín et al. (2010). The surfactants with the long hydrocarbonate chain (hexyl-(HTABr)), surfactants with the short hydrocarbonate chain (octyl-(OTABr), decyltrimethylammonium bromide (DTABr)) and surfactants with an intermediate hydrocarbonate chain (tetradecyl-(TDTABr), octadecyl-trimethyl ammonium bromide (ODTABr)) were used in this work. When the hydrocarbon chain length of the surface agent was increased, the carbon dioxide ( $\text{CO}_2$ ) absorption rate decreased. At low concentrations of surfactant (HTABr, OTABr, and DTABr), gas-liquid mass transfer enhancement was observed. However, for surfactants TDTABr and ODTABr, the enhancement was not observed due to the reduction in the driving force (Fig. 10). In addition, the effect of various surfactants on the interface area depends on the surfactant with different bicarbonate chain lengths. Generally, mass transfer interfacial area increased with the addition of surfactant (Chaumat et al. 2007, Hebrard et al. 2009, García-Abuín et al. 2012, McClure et al. 2015). However, surfactants with a long chain length did not affect the interfacial area, obtaining values for the gas-liquid interfacial area similar to pure water.

## CONCLUSION

In this paper, many studies on gas-liquid mass transfer enhancement are reviewed. Gas-liquid mass transfer can be enhanced by increasing one or more of the overall driving force, mass transfer coefficient, and interface area. Research on the intensification of gas-liquid mass transfer by introducing additives must be further strengthened. For further research directions, the following observations are provided.

- (1) Study the effect of additives on the overall system performance and downstream processing, including separation and recovery/reuse of additives before the practical application of various mass transfer enhancement systems, which support their application's economic feasibility before additives application becomes a reality.
- (2) Please select a suitable liquid solvent and/or gas carrier and study the effect of physical properties of the phase, such as viscosity, interfacial tension, etc., on process design, which help us achieve better process designs.
- (3) Mass transfer research should not be limited to determining macroscopic properties; it encourages researchers to explore the underlying mechanisms.

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## REFERENCES

- Abeydeera, U., Jayantha, W.M. and Samarasinghalage, T. 2019. Global research on carbon emissions: A scientometric review. *Sustainability*, 11: 3972.
- Bakthavatchalam, B., Habib, K., Saidur, R., Saha, B.B. and Irshad, K. 2020. A comprehensive study on nanofluid and ionanofluid for heat transfer enhancement: A review on current and future perspective. *J. Mol. Liq.*, 305: 112787.
- Baz-Rodríguez, S.A., Botello-Alvarez, J.E., Estrada-Baltazar, A., Vilchiz-Bravo, L.E., Padilla-Medina, J.A. and Miranda-López, R. 2014. Effect of electrolytes in aqueous solutions on oxygen transfer in gas-liquid bubble columns. *Chem. Eng. Res. Des.*, 92: 2352-2360.
- Beenackers, A.A.C.M. and Swaaij, W.P.M.V. 1993. Mass transfer in gas-liquid slurry reactors. *Chem. Eng. Sci.*, 48: 3109-3139.
- Birgand, F., Skaggs, R.W., Chescheir, G.M. and Gilliam, J.W. 2007. Nitrogen removal in streams of agricultural catchments: A literature review. *Crit. Rev. Env. Sci. Technol.*, 37: 381-487.
- Boltes, K., Caro, A., Leton, P., Rodríguez, A. and García-Calvo, E. 2008. Gas-liquid mass transfer in oil-water emulsions with an airlift bioreactor. *Chem. Eng. Process. Process Intensif.*, 47: 2408-2412.
- Brilman, D.W.F., Goldschmidt, M.J.V., Versteeg, G.F. and Swaaij, W.P.M.V. 2000. Heterogeneous mass transfer models for gas absorption in multiphase systems. *Chem. Eng. Sci.*, 55: 2793-2812.
- Chaumat, H., Billet, A.M. and Delmas, H. 2007. Hydrodynamics and mass transfer in bubble column: Influence of liquid phase surface tension. *Chem. Eng. Sci.*, 62: 7378-7390.
- Craig, V.S.J., Ninham, B.W. and Pashley, R.M. 1993. The effect of electrolytes on bubble coalescence in water. *Journal of Physical Chemistry*, 97: 10192-10197.
- Dang, X.H., Guo, J.H., Yang, L., Xue, S., Ai, B.Y., Li, X.N., Chen, L.Y., Li, W., Qin, H.Y. and Zhang, J.L. 2022. Effects of blade structures on the dissolution and gas-liquid mass transfer performance of cup-shaped blade mixers. *J. Taiwan Inst. Chem. E.*, 131: 104149.
- Dumont, E., André, Y. and Cloirec, P.L. 2014. Mass transfer coefficients of styrene into water/silicone oil mixtures: New interpretation using the "equivalent absorption capacity" concept. *Chem. Eng. J.*, 237: 236-241.
- Dumont, E. and Delmas, H. 2003. Mass transfer enhancement of gas absorption in oil-in-water systems: A review. *Chem. Eng. Process. Process Intensif.*, 42: 419-438.
- Esmaili-Faraj, S.H. and Esfahany, M.N. 2016. Absorption of hydrogen sulfide and carbon dioxide in water-based nanofluids. *Ind. Eng. Chem. Res.*, 55: 4682-4690.
- García, G.E.C., Schaaf, J.V.D. and Kiss, A.A. 2017. A review on process intensification in HiGee distillation. *J. Chem. Technol. Biot.*, 92: 1136-1156.
- García-Abuín, A., Gómez-Díaz, D., Losada, M. and Navaza, J.M. 2012. Bubble column gas-liquid interfacial area in a polymer+surfactant+water system. *Chem. Eng. Sci.*, 75: 334-341.
- García-Abuín, A., Gómez-Díaz, D., Navaza, J.M. and Sanjurjo, B. 2010. Effect of surfactant nature upon absorption in a bubble column. *Chem. Eng. Sci.*, 65: 4484-4490.
- García-Ochoa, F., Gomez, E., Santos, V.E. and Merchuk, J.C. 2010. Oxygen uptake rate in microbial processes: An overview. *Biochem. Eng. J.*, 49: 289-307.
- Gome, A. and Upadhyay, K. 2012. Chemical kinetics of ozonation and other processes used for the treatment of wastewater containing pharmaceuticals: A review. *Int. J. Curr. Res. Rev.*, 4: 157-168.
- Gómez-Díaz, D., Navaza, J.M. and Sanjurjo, B. 2009. Mass-transfer enhancement or reduction by surfactant presence at a gas-liquid interface. *Ind. Eng. Chem. Res.*, 48: 2671-2677.
- Góngora-García, O.R., Aca-Aca, G. and Baz-Rodríguez, S.A. 2021. Mass transfer in aerated culture media combining mixed electrolytes and glucose. *Bioprocess Biosyst. Eng.*, 44: 81-92.
- Hebrard, G., Zeng, J. and Loubiere, K. 2009. Effect of surfactants on liquid side mass transfer coefficients: A new insight. *Chem. Eng. J. Lausanne*, 148: 132-138.
- Jang, N., Yasin, M., Kang, H., Lee, Y., Park, G.W., Park, S. and Chang, I.S. 2018. Bubble coalescence suppression driven carbon monoxide (CO)-water mass transfer increase by electrolyte addition in a hollow fiber membrane bioreactor (HFMBR) for microbial CO conversion to ethanol. *Bioresour. Technol.*, 263: 375-384.
- Jensen, M.B., Ottosen, L.D.M. and Kofoed, M.V.W. 2021. H<sub>2</sub> gas-liquid mass transfer: A key element in biological Power-to-Gas methanation. *Renew. Sustain. Energy Rev.*, 147: 111209.
- Kazakov, D.A., Vol Khin, V.V., Borovkova, I.S. and Popova, N.P. 2014. Kinetics of oxygen absorption by aqueous electrolyte solutions in the presence of microencapsulated quartz particles activating the mass transfer in the liquid phase. *Russ. J. Appl. Chem.*, 87: 88-94.
- Kim, K., Lee, J., Seo, K., Kim, M.G., Ha, K.S. and Kim, C. 2016. Enhancement of methane-water volumetric mass transfer coefficient by inhibiting bubble coalescence with electrolyte. *J. Ind. Eng. Chem.*, 33: 326-329.
- Kundu, A., Dumont, E., Duquenne, A.M. and Delmas, H. 2002. Mass transfer characteristics in gas-liquid system. *Canad. J. Chem. Eng.*, 81: 640-646.
- Kuuskräa, V., Stevens, S.H. and Moodhe, K.D. 2013. Technically Recoverable Shale Oil and Shale Gas Resources: An Assessment of 137 Shale Formations in 41 Countries Outside the United States. US Energy Information Administration, US Department of Energy, US, pp. 1-730.
- Lakhdiessi, E.M., Fallahi, A., Guy, C. and Chaouki, J. 2020. Effect of solid particles on the volumetric gas-liquid mass transfer coefficient in slurry bubble column reactors. *Chem. Eng. Sci.*, 227: 115912.
- Lebrun, G., Benaissa, S., Men, C.L., Pimienta, V., Hébrard, G. and Dietrich, N. 2022. Effect of surfactant lengths on gas-liquid oxygen mass transfer from a single rising bubble. *Chem. Eng. Sci.*, 247: 117102.
- Lee, W., Xu, R.H., Kim, S., Park, J.H. and Kang, Y.T. 2021. Nanofluid and nanoemulsion absorbents for the enhancement of CO<sub>2</sub> absorption performance. *J. Clean. Prod.*, 291: 125848.
- Li, H., Wu, C.H., Hao, Z.Q., Li, X.G. and Gao, X. 2019. Process intensification in vapor-liquid mass transfer: The state-of-the-art. *Chin. J. Chem. Eng.*, 27: 1236-1246.
- Li, W.X., Peng, H., Zhang, Z.G. and Chen, K. 2010. Gas-liquid mass transfer in oil-water emulsions in a stirrer. *Chem. Eng.*, 11: 5-9.
- Luo, L.G. 2013. Heat And Mass Transfer Intensification and Shape Optimization: A Multi-Scale Approach. Springer Science & Business Media, London.
- McClure, D.D., Lee, A.C., Kavanagh, J.M., Fletcher, D.F. and Barton, G.W. 2015. Impact of surfactant addition on oxygen mass transfer in a bubble column. *Chem. Eng. Technol.*, 38: 44-52.
- Mehdipour, M., Keshavarz, P. and Rahimpour, M.R. 2021. Rotating liquid sheet contactor: A new gas-liquid contactor system in CO<sub>2</sub> absorption by nanofluids. *Chem. Eng. Process.*, 165: 108447.
- Mehrmia, M.R., Towfighi, J., Bonakdarpor, B. and Akbarnejad, M.M. 2005. Gas hold-up and oxygen transfer in a draft-tube airlift bioreactor with petroleum-based liquids. *Biochem. Eng. J.*, 22: 105-110.
- Parnian, P., Zamir, S.M. and Shojaosadati, S.A. 2016. Styrene vapor mass transfer in a biotrickling filter: Effects of silicone oil volume fraction, gas-to-liquid flow ratio, and operating temperature. *Chem. Eng. J.*, 284: 926-933.
- Pichetwanit, P., Kungsanant, S. and Supap, T. 2021. Effects of surfactant type and structure on properties of amines for carbon dioxide capture. *Coll. Surf. A Physicochem. Eng. Asp.*, 622: 126602.
- Radler, M. 2011. Worldwide oil production steady in 2011: Reported reserves grow. *Oil Gas J.*, 109: 26-27.
- Ramshaw, C. 1983. Hige distillation-An example of process intensification. *Chem. Eng.*, 1: 13-14.
- Ribeiro, C.P. and Mewes, D. 2007. The influence of electrolytes on gas

- hold-up and regime transition in bubble columns. *Chem. Eng. Sci.*, 62: 4501-4509.
- Rols, J.L., Condoret, J.S., Fonade, C. and Goma, G. 1990. Mechanism of enhanced oxygen transfer in fermentation using emulsified oxygen-vectors. *Biotechnol. Bioeng.*, 35: 427-435.
- Sajjadi, S., Zerfa, M. and Brooks, B.W. 2002. Dynamic behavior of drops in oil/water/oil dispersions. *Chem. Eng. Sci.*, 57: 663-675.
- Saud, S.M., Krishnan, J., Ling, T.H. and Veluri, M.V.P.S. 2013. Enhancement of oxygen mass transfer and gas holdup using palm oil in stirred tank bioreactors with xanthan solutions as simulated viscous fermentation broths. *Biomed. Res. Int.*, 13: 409675.
- Shariati, F.P., Bonakdarpour, B. and Mehrnia, M.R. 2007. Hydrodynamics and oxygen transfer behavior of water in diesel microemulsions in a draft tube airlift bioreactor. *Chem. Eng. Process. Intensif.*, 46: 334-342.
- Syeda, S.R. and Reza, M.J. 2011. Effect of surface tension gradient on gas hold-up enhancement in aqueous solutions of electrolytes. *Chem. Eng. Res. Des.*, 89: 2552-2559.
- Taweel, A.M.A., Idhbeaa, A.O. and Ghanem, A. 2013. Effect of electrolytes on interphase mass transfer in microbubble-sparged airlift reactors. *Chem. Eng. Sci.*, 100: 474-485.
- Yang, Z.H., Telmadarreie, A., Dong, M.Z. and Bryant, S. 2020. A pressure-decay method to determine the influence of a surface-active agent on the interface and internal resistances to gas-liquid mass transfer. *Chem. Eng. J.*, 387: 124108.
- Yoshida, F., Yamane, T. and Miyamoto, Y. 1970. Oxygen absorption into oil-in-water emulsions. A study on hydrocarbon fermentors. *Ind. Eng. Chem. Process Des. Dev.*, 9: 570-577.
- Zamir, S.M., Babatabar, S. and Shojaosadati, S.A., 2015. Styrene vapor biodegradation in single- and two-liquid phase biotrickling filters using *Ralstonia eutropha*. *Chem. Eng. J.*, 268: 21-27.
- Zhang, H., Wang, B., Xiong, M.Y., Gao, C.Y., Ren, H.Y. and Ma, L. 2022. Process intensification in gas-liquid mass transfer by nanofluids: Mechanism and current status. *J. Mol. Liq.*, 346: 118268.