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Effect of electrolytes in aqueous solutions on oxygen transfer in gas–liquid bubble columns

Sergio A. Baz-Rodríguez [a,c,*], José E. Botello-Alvarez [b],
Alejandro Estrada-Baltazar [c], Luis E. Vilchiz-Bravo [a],
José A. Padilla-Medina [d], Rita Miranda-López [b]

[a] Facultad de Ingeniería Química, Universidad Autonoma de Yucatan, Campus de Ciencias Exactas e Ingenierias,
Periferico Norte Kilometro 33.5, Tablaje Catastral 13615, Col. Chuburna de Hidalgo Inn, Merida, Yucatán C.P. 97203,
Mexico
[b] Departamento de Ingeniería Bioquímica, Instituto Tecnológico de Celaya, Av. Tecnológico y García Cubas s/n,
Celaya, Guanajuato C.P. 38010, Mexico
[c] Departamento de Ingeniería Química, Instituto Tecnológico de Celaya, Av. Tecnológico y García Cubas s/n, Celaya,
Guanajuato C.P. 38010, Mexico
[d] Departamento de Ingeniería Electrónica, Instituto Tecnológico de Celaya, Av. Tecnológico y García Cubas s/n,
Celaya, Guanajuato C.P. 38010, Mexico

A B S T R A C T

The effects of inorganic electrolytes (NaCl, $MgCl_2$, $CaCl_2$) in aqueous solutions on oxygen transfer in a bubble column were studied. Electrolyte concentrations (c) below and above the critical concentrations for bubble coalescence (c_{tc}), and six superficial gas velocities (v_{sg}), were evaluated. The volumetric mass transfer ($k_L a$) and the mass transfer (k_L) coefficients were experimentally determined. It was found that the concentration of electrolytes reduced the k_L, but the interfacial area (a) increased enough to result in a net increase of $k_L a$. Using as independent variable a normalizing variable ($c_r = c/c_{tc}$), and maintaining fixed v_{sg}, similar values of $k_L a$ were observed regardless the kind of electrolyte; the same happened for k_L. This suggests that c_r quantifies the structural effects that these solutes exert on mass transfer. Also, once $c_r = 1$ was reached, no significant variations were found in $k_L a$ and k_L for constant v_{sg}. It is concluded that the gradual inhibition of bubble coalescence ($c_r < 1$) governs the significant changes in hydrodynamics and mass transfer via the reduction of bubble size and the consequent increment of a and gas holdup (ε_g). Finally, regarding the effects of v_{sg} on mass transfer, transition behaviors between those expected for isolated bubbles and bubble swarms were observed.

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Keywords: Bubble columns; Hydrodynamics; Oxygen transfer; Aqueous electrolyte solutions; Bubble coalescence

1. Introduction

The study of mass transfer phenomena through gas–liquid interfaces is fundamental to understanding the behavior of chemical and biochemical processes. For instance, microorganisms' growth and survival in aerobic biological systems require oxygen transfer through an air–water interface; hence, oxygen transfer is one of the main issues on bioreactor design due to the low solubility of oxygen in aqueous solutions (Jackson, 1991). Interfacial gas–liquid mass transfer is a complex phenomenon, since it depends on transport properties at mobile bubble interfaces, physicochemical properties of the

Nomenclature

a	specific interfacial area based on liquid volume (m^2/m^3)
a_w	specific interfacial area in air–pure water systems based on liquid volume (m^2/m^3)
b'	parameter in Hughmark's correlation
c	concentration of electrolyte in aqueous solution (mol/m^3)
c_{O_2}	concentration of dissolved oxygen in the liquid phase (mol/m^3)
c_r	reduced concentration of electrolyte in aqueous solution ($c_r = c/c_{tc}$)
c_{tc}	critical electrolyte concentration for bubble coalescence (mol/m^3)
d_{32}	Sauter mean diameter (m)
D_L	oxygen diffusivity in aqueous media (m^2/s)
Eo	Eötvos number
g	acceleration of gravity (m/s^2)
H	Henry constant for oxygen in aqueous media (m^3Pa/mol)
$k_L a$	volumetric mass transfer coefficient (1/s)
$k_L a_w$	volumetric mass transfer coefficient in air–pure water systems (1/s)
k_L	mass transfer coefficient (m/s)
k_{Lw}	mass transfer coefficient in air–pure water systems (m/s)
L	level of mixture in the column (m)
p_{O_2}	partial pressure of oxygen in the gas phase (Pa)
$p_{O_2,IN}$	partial pressure of oxygen at inlet of the column (Pa)
R	universal gas constant (m^3Pa/mol K)
Re	particle Reynolds number
Sc	Schmidt number for liquid phase
T	temperature, K
v_s	slip velocity of a bubble relative to liquid (m/s)
v_{sg}	superficial air velocity (m/s)
v_{sl}	superficial velocity of liquid (m/s)

Greek symbols

ε_g	global fractional gas holdup
ε_l	global volumetric fraction of liquid phase
ρ_L	density of liquid phase (kg/m^3)
μ_L	dynamic viscosity (Pa s)
σ	surface tension (N/m)

liquid phase, local velocity fields and geometry of interfaces. In practice, these combined effects are included on volumetric mass transfer coefficients ($k_L a$) (Lewis and Withman, 1924), so that considerable efforts have been made in order to determine qualitative and quantitative relationships to the associated variables (Shah et al., 1982; Kantarci et al., 2005). However, regarding aqueous media, most of the expressions commonly used to estimate the volumetric coefficients were determined by using an air–water system; then, if such correlations are applied in general, significant interfacial effects would be neglected due to the potential complexity of the liquid systems involved. Among the several components that increase the complexity of liquid systems (*i.e.*, culture media) inorganic salts are found, also identified as electrolytes.

Literature reports significant research efforts aimed to understand the hydrodynamic and mass transfer behavior of gas–liquid systems with aqueous electrolyte solutions (AES). Early works observed that bubble size decreases in air–AES bubbly flows with respect to air–water systems (Marrucci and Nicodemo, 1967; Lessard and Zieminski, 1971; Zieminski and Whittemore, 1971). It is possible that the liquid surface tension increment, due to the presence of inorganic electrolytes, causes the reduction in the bubble volume when they are generated in the diffuser (Martín et al., 2006). However, it seems that the inhibitory effect on bubble coalescence is the most relevant contribution to mean bubble size reduction (Zieminski and Whittemore, 1971; Jamialahmadi and Muller-Steinhagen, 1990; Prince and Blanch, 1990; Zahradník et al., 1995; Kluytmans et al., 2001; Botello-Álvarez et al., 2011). This allows the maintenance of bubbles as single elements during the approach or collision with other bubbles. In this sense, a transition concentration of coalescence (c_{tc}) has been defined as the concentration of electrolyte in aqueous solution from which the air bubbles reduce their coalescence above 50%, in respect to that observed in pure water (Marrucci and Nicodemo, 1967; Lessard and Zieminski, 1971).

Several studies address the way electrolytes induce bubble coalescence inhibition. Some authors claim that coalescence occurs because the liquid is drained from the space between two approaching bubbles until a minimum film thickness is reached; then the liquid film ruptures and the bubbles coalesce. If the formation of ordered structures among ions and water molecules in AES (Jungwirth and Tobias, 2006) prevents the critical liquid draining, then the coalescence is inhibited (Lessard and Zieminski, 1971; Tsang et al., 2004; Chan and Tsang, 2005). Other authors have proposed that the inhibition of bubble coalescence above a minimum film thickness can be explained in terms of the change of surface tension with concentration (Marrucci and Nicodemo, 1967; Prince and Blanch, 1990; Christenson and Yaminsky, 1995; Syeda and Reza, 2011). The amount of dissolved air that is affected by the electrolyte concentration has also been suggested as an important factor in the bubble coalescence inhibition (Weissenborn and Pugh, 1995, 1996). However, there is no consensus about the preeminence of one of the proposing causes for coalescence inhibition behavior. An interrelation of those structural changes would be a possible explanation, along with effects of hydrodynamic variables such as local gas velocity (Ribeiro and Mewes, 2007b; Nguyen et al., 2012).

Bubble size significantly affects the oxygen transfer. Its size reduction in AES causes low slip velocities since smaller bubbles rise slowly. This causes a reduction in the mass transfer coefficient (k_L). Moreover, the overall mass transfer is affected. Residence time of small bubbles is larger than that of large bubbles. Higher residence time also causes the fractional gas holdup to increase and, since there are more small bubbles in the system, the available interfacial area will also be larger (Pohorecki and Nowosielski, 1986; Jamialahmadi and Muller-Steinhagen, 1990; Zahradník et al., 1995; Kluytmans et al., 2001; Ruthiya et al., 2006). The above causes an increment in $k_L a$.

Regarding physicochemical and transport properties of phases, electrolytes in aqueous solutions cause an increase on the liquid phase viscosity, since the ion–solvent interactions make the fluid motion harder; as a consequence, slip velocity decreases. Also, the molecular diffusivity of oxygen on the liquid phase is reduced as the electrolyte concentration increases. A feasible explanation is that the oxygen migrating from air has to pass through the ionic film representing a mass transfer resistance (Ju and Ho, 1985). Since both the high