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Impact of surfactants and cellulose on the hydrodynamic behavior and liquid-side mass transfer coefficient of a bubble column

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ABSTRACT

The aim of this paper is to present and understand the effect of surfactants, cellulose and their combination on the hydrodynamic behavior and the liquid side mass transfer coefficient of a bubble column. For that purpose, the effect of liquid properties on the interfacial area and the liquid-side mass transfer coefficient was investigated. Bubbles were generated in a small-scale bubble column having an elastic membrane with a single orifice as the gas sparger. Different aqueous solutions containing surfactants (SDS) and cellulose (MCC) were investigated and characterized concerning their surface tension and viscosity. The interfacial areas (a) were calculated from the bubble diameters (D_B), the bubble frequencies (f_B) and the terminal bubble rising velocities (U_B). The liquid-side mass transfer coefficients (k_L) were calculated from the volumetric mass transfer coefficients (k_{La}) measured by the dynamic method. In the range of concentration under test, the experimental results showed that the addition of MCC to the studied liquid phases did not affect the mass transfer coefficient. However, the addition of SDS to water and to MCC in water system decreases the mass transfer coefficient.

Keywords: Surfactant; Microcrystalline cellulose; Bubble generation; Interfacial area; Volumetric mass transfer coefficient; Liquid-side mass transfer coefficient; Superficial gas velocity.

1. INTRODUCTION

29 Bubble Columns are usually encountered in chemical, pharmaceutical, petrochemical
30 and metallurgical industries as multiphase reactors due to their simple construction, low cost
31 and ease of operation [1]. In the majority of wastewater treatment plants (WWTPs), the
32 activated sludge (biological treatment in the aerated tank) is employed for the degradation of
33 organic pollutants present in wastewaters. A source of dissolved oxygen by injecting air
34 bubbles is then required to ensure the metabolism of the microorganisms which use the
35 organic matter as a source of nutrition, and then degrade it [2]. In the gas-liquid reactors,
36 mass transfer must be maximum for a given power consumption [3]. In conventional
37 wastewater treatment plants, air supply used in activated sludge treatment can represent up to
38 70% in terms of energy expenditure [4][5]. Therefore, accurate determination of mass transfer
39 parameters is of prime interest to avoid over- or under-estimating of the required oxygen
40 supply to the process [4]. The process optimization involves a good understanding of how the
41 different operational parameters influence the aeration efficiency of the system [5].

42 Most part of investigations in bubble columns have been performed for aqueous
43 systems. Based on a large number of studies on oxygen transfer in clean water, the American
44 Society of Civil Engineers (ASCE) standard was established to measure the oxygenation
45 capacity of aeration devices in clean water [6]. The impacts of physical variables such as
46 temperature, reactor geometry, pressure, surface tension, mixing and viscosity on oxygen
47 transfer in clean water are well documented and understood. However, the presence of real
48 contaminants into raw wastewater is not yet well evaluated thereby it strongly impacts the
49 efficiency of the process. To improve the productivity of the reactors, it is required to well
50 study and understand the relationship between the mass transfer rate and the liquid phase
51 contents.

52 The most commonly occurring contaminants accumulating at the environmental
53 gas/liquid interfaces are surfactant compounds. Because of their wetting, dispersing,
54 solubilizing and foaming properties, they enter in the formulation of pharmaceuticals,
55 cosmetics, pesticides, and household products [2][7]. Surfactant world production was 1.7
56 million tons in 1984. It increased from 9.3 million tons in 1995 to over 13 million tons in
57 2008. Their compound annual growth rate (CAGR) is increasing until now [2]. Thus, their
58 presence in the aeration tank of WWTPs must be absolutely considered. Surfactants are
59 amphiphilic compounds having both hydrophilic and hydrophobic groups [2]. Due to their
60 nature, surfactants accumulate at the interface of liquid-gas systems and lower the interfacial
61 tension. This last decreases with increasing the surfactant concentration until the critical

62 micelle concentration (CMC). The ability of surfactant to alter oxygen mass transfer has been
63 vastly considered in literature.

64 Loubière and Hébrard (2004) explored the effect of liquid surface tension on the bubble
65 formation from both rigid and flexible orifice. Results indicated that the effect of surface
66 tension on the generated bubbles depends on whether the bubbles are produced from a rigid or
67 a flexible orifice [8]. Alves et al. (2004) investigated the effect of Polyethylene glycol
68 surfactant on the average mass transfer coefficient in an aerated stirred tank. Results indicated
69 that bubbles in surfactant solution behave as rigid bubbles, while bubbles in tap water behave
70 closer to having a mobile interface [9]. Painmanakul et al. (2005) studied the effect of
71 surfactants on the mass transfer parameters in aqueous solutions with Sodium dodecyl sulfate
72 anionic surfactant and Lauryl dimethyl benzyl ammonium bromine cationic surfactants in a
73 small scale bubble column. Results showed that the presence of surfactants affects the bubble
74 generation phenomenon [10]. Rosso et al. (2006) studied the effects of interfacial surfactant
75 contamination on bubble gas transfer. A gas transfer reduction of 30–70% of pure water
76 values in surfactant solutions was shown [11]. Sardeing et al. (2006) examined the effect of
77 anionic, cationic and non-ionic surfactants on the mass transfer rate of bubbles in a small-
78 scale bubble column. Results revealed that whatever the liquid phase, three zones are found
79 on the liquid-side mass transfer coefficient variation with the bubble diameter [12]. Hébrard et
80 al. (2009) studied the effect of high concentration of surfactants on liquid side mass transfer
81 coefficient in free gas–liquid interface. A reduction in the mass transfer coefficient with an
82 increase of surfactant concentrations was detected as well as a plateau when the concentration
83 reaches critical micelle concentration [13]. Jamnongwong et al. (2010) inspected the effect of
84 presence of various substances commonly encountered in biological media. Results showed
85 that in presence of surfactants, for all liquid phases under test, oxygen diffusion coefficients
86 decreased when compared to clean water [14]. Takagi and Matsumoto (2011) reviewed the
87 recent investigations associated with subsequent variation of bubble behavior due to the
88 surfactant adsorption/desorption on the bubble surface. They concluded that the presence of
89 surfactants influences the small-scale behavior of each bubble [15]. Kotti et al. (2013)
90 considered the effects of the presence of anionic and cationic surfactants in the liquid phase
91 and the hydrodynamic regime of the bubble flow on the oxygen transfer rate in an
92 electroflotation process. They revealed that the specific interfacial area tends to increase by
93 the addition of cationic surfactant and decreases with the anionic surfactant [16]. Jimenez et
94 al. (2014) used a powerful technique, based on the Planar Laser Induced Fluorescence (PLIF)

95 technique to locally visualize and quantify the impact of surfactants in wastewaters on
96 hydrodynamics and oxygen mass transfer. Bubbles have been found to be more spherical with
97 a reduced rise velocity in the presence of surfactants up to the CMC. Hydrodynamic
98 characteristics were found to be practically constant above the CMC, even if the oxygen mass
99 transfer decreased [4]. McClure et al. (2015) compared the influence of surfactant addition
100 and sparger design on the mass transfer, the gas holdup and bubble size distribution in a
101 bubble column. Results showed that addition of surfactants cause an approximately threefold
102 decrease in mass transfer coefficient [17]. Aoki et al. (2015) investigated the effect of
103 concentration of surfactant on the rate of mass transfer for single rising bubbles. They used
104 Triton X-100 as surfactant and showed that for small bubbles the mass transfer rate decreases
105 with increase of surfactant concentration. Furthermore, they proved that the surfactant adsorbs
106 only in the bubble tail region [18]. Haghnegahdar et al. (2016) investigated the effect of
107 surfactant on the bubble shape and mass transfer in a milli-channel using high-resolution
108 microfocus X-ray imaging. Results showed that the presence of surfactants causes a change of
109 the bubble shape and leads to a slight increase of the liquid film thickness around the bubble.
110 They also concluded that the presence of surfactant has a more significant impact on the
111 dissolution rate of small bubbles [19].

112 Hydrodynamic behavior and mass transfer parameters of other types of medium such as
113 non-Newtonian media have been widely investigated. The microcrystalline cellulose is a
114 significant fraction of the solid organic matters [20]. Its presence as a non-Newtonian fluid in
115 WWTPs is believed to be mainly from toilet paper. This last is one of the mostly used
116 ubiquitous personal hygiene products, particularly in Northern Americas and European
117 countries. The first packaged toilet paper was commercialized in New-Jersey by Joseph
118 Gayetty in 1857. Paper wastes are mostly originated from wood pulp or cotton which consists
119 of 90–99% cellulose fibers [21]. A part of paper wastes is precipitated in the primary
120 clarification tank implanted to separate suspended solids from sewage. Then, cellulose could
121 be a potential resource removed easily from wastewater by for instance sieving [20]. It can be
122 used as a renewable and biodegradable resource for the production of biofuels and others
123 functional chemicals such as cellulose acetate and cellulose ether [21]. The second part of
124 paper wastes is transported to the aerated tank where it is hardly degraded because it needs
125 first to be hydrolysed before it can be metabolized. The biological hydrolysis of cellulose may
126 take much more time than the usual hydraulic retention time ($HRT < 12$ h) for conventional
127 biological treatment process and it strongly depends on temperature [20]. The ability of

128 cellulose present in the aerated tank to alter oxygen mass transfer has not been much
129 considered in literature compared to surfactant.

130 Popovic and Robinson (1987) studied the specific interfacial area in an external
131 circulation-loop airlifts and a bubble column using a carboxymethyl cellulose/sulphite
132 solution. Results showed that the specific interfacial areas for the studied systems was found
133 to be two- to three fold smaller than in the non-viscous Newtonian systems previously
134 investigated by the authors [22]. Guo-Qing et al. (1995) [23] and Vasconcelos et al. (2003)
135 [24] showed that systems with higher viscosity such as carboxymethyl cellulose exhibited a
136 lower k_{La} than those running with lower viscosity mediums. Roberts et al. (2011) investigated
137 the effects of water interactions in cellulose suspensions on mass transfer and saccharification
138 efficiency at high solids loadings. Results showed that increasing water constraint is related to
139 the increased viscosity of the fluid in the suspensions [25].

140 Given that the presence of surfactant compounds and MCC is simultaneously
141 unavoidable in the aerated tanks of WWTPs, the study of their combination on the
142 hydrodynamic behavior and liquid-side mass transfer coefficient in a bubble column should
143 be important to well estimate the required oxygen supply to the aeration process. In keeping
144 with this context, and for the first time, this paper study the effect of surfactants and MCC
145 combination on the hydrodynamic behavior and the mass transfer parameters. The
146 investigation is based on the determination of the effect of liquid properties on bubble
147 generation phenomenon, interfacial area and liquid-side mass transfer coefficient. Bubble
148 column having an elastic membrane with a single orifice as gas sparger was used in this work.
149 It offers the possibility to well master the interfacial area provided by the bubble train
150 allowing an accurate estimation of the liquid side mass transfer coefficient.

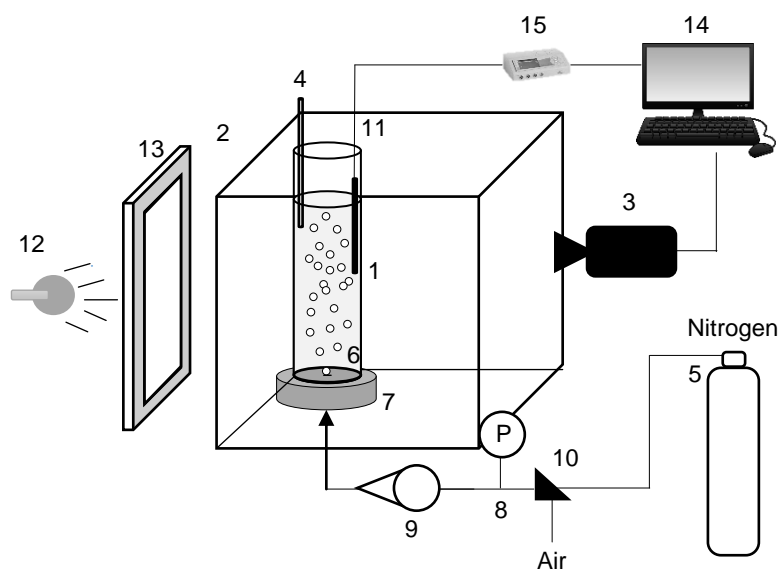
151

152 2. MATERIAL AND METHODS

153

154 A schematic overview of the experimental device is represented in Figure 1. The
155 experiments are carried out in a glass bubble column (1) of 0.05 m in diameter and with a
156 liquid height (H_L) of 0.20 m. The column is immovable into a doublewall glass vessel (2) of
157 0.40 m in width, 0.40 m in length and 0.30 m in height. The bubble diameters and their
158 terminal rising velocities used to calculate the specific interfacial area have been determined
159 thanks to image analysis and high speed camera (3). The temperature in the bubble column is

160 measured by means of a thermometer (4). Nitrogen (5) is employed for oxygen elimination in
161 the liquid phase. Piece of 0.60 m diameter of an industrial rubber membrane sparger (6) is
162 used as gas sparger. Bubbles are generated by a single puncture located at the membrane
163 center. The membrane is assembled on a circular clamping ring (7) composed of two jaws;
164 this fixing system coupled with the use of a dynamometric spanner enables the same initial
165 tension to be applied, thus giving reproducible results [10]. The gas flow is monitored by a
166 pressure gauge (8) and regulated by a gas flow meter (9). A three-way valve is used to inject
167 either air or nitrogen (10). An oxygen microsensor (11) is used to measure the change in
168 dissolved oxygen concentration. The double wall vessel is filled with water and a white
169 plywood plate (13) is introduced to improve the image acquisition quality. Acquisition
170 computer is connected to visualize and treat the results (14). SDS, MCC and SDS/MCC
171 solutions are introduced at the top of the bubble column.



184 **Figure 1.** Schematic overview of the experimental device

185
186 To understand the effect of cellulose, surfactants and their combination on the liquid-
187 side mass transfer coefficient, several liquid phases have been prepared and characterized.
188 Compressed air and nitrogen from laboratory lines were the gas phases used in our work.
189 Note that, a great care was taken for the cleaning and rinsing procedure of vessels to avoid
190 contamination of surfactant. To prepare synthetic liquid phases, distilled water was combined
191 with two types of compounds: (i) a microcrystalline cellulose (MCC); (ii) an anionic
192 surfactant named sodium dodecyl sulfate (SDS), synonymously sodium lauryl sulfate (SLS).
193 They were selected as commonly found in the biological tanks of WWTPs. The SDS used

194 (CAS Number 151-21-3) and the MCC (CAS Number: 9004-34-6) were purchased from
 195 Sigma Aldrich. Three solutions of SDS, four solutions of MCC and four solutions of
 196 SDS/MCC combination were tested. SDS1, SDS2 and SDS3 represent the solutions
 197 containing 1 g/L, 2 g/L and 2.59 g/L of SDS respectively. MCC0.5, MCC1, MCC2 and
 198 MCC5 represent the solutions containing 0.5 g/L, 1 g/L, 2 g/L and 5 g/L of MCC
 199 respectively. SDS/MCC 0.5, SDS/MCC1, SDS/MCC2 and SDS/MCC5 represent the
 200 solutions containing 1 g/L of SDS and 0.5 g/L, 1 g/L, 2 g/L and 5 g/L of MCC respectively.
 201 Given that the liquid phases under study are dilute aqueous solutions, their density can be
 202 assumed to be equal to that of tap water (997 kg/m³). The viscosity of SDS solutions also can
 203 be assumed to be equal to that of tap water (10⁻⁵ Pas) [14]. Therefore, the liquid-phases
 204 characterization will consist on the determination of the static surface tensions, the critical
 205 micelle concentration (CMC) and the adsorption parameters for SDS solutions; the rheology
 206 for MCC solutions; and the dissolved oxygen concentration at saturation (C*) which depends
 207 on the temperature for both SDS and MCC solutions.

208 At the contrary of the dynamic surface tension measurement, the surface age is not
 209 taken into account in the static surface tension measurement. This last can be accomplished by
 210 two methods: (i) the static method of Du Noüy, using a KRÜSS K6 Force Tensiometer. This
 211 method is based on slowly lifting a ring, often made of platinum, from the surface of a liquid;
 212 (ii) the static method based on the pendant drop using KRÜSS DSA25 Drop Shape Analyzer.
 213 Both methods gave the same results.

214 To calculate the critical micelle concentration (CMC) for surfactant aqueous solutions,
 215 static surface tension has to be determined for a number of different concentrations. When the
 216 surfactant concentration increases, the surface tension tends to decrease until levelling off.
 217 Here, the solution is saturated in surfactants compounds and there is micelles formation. The
 218 CMC is then reached. Deduced from the curve linked the surfactant concentrations and the
 219 surface tensions, the CMC is reported.

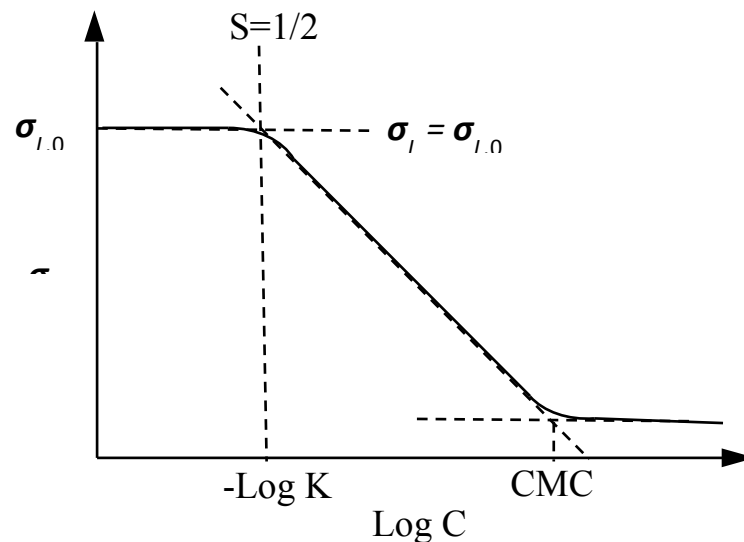
220 To characterize the adsorption parameters at a gas–liquid interface, the Langmuir theory
 221 is used [12][8]. The kinetics of adsorption and diffusion of the surfactant molecules towards
 222 the bubble interface can be defined by using the equations (1) and (2):

$$223 \quad S_e = \frac{\Gamma_e}{\Gamma_\infty} = K \frac{C}{1+KC} \quad (1)$$

$$224 \quad \sigma_{L,O} - \sigma_L \approx RT_a \Gamma_\infty \text{Log}(K) + RT_a \Gamma_\infty \text{Log}(C) \quad (2)$$

225 Where S_e is the surface coverage ratio at equilibrium, C is the solute concentration in
 226 the liquid phase, K is the adsorption constant at equilibrium, Γ_∞ is the surface concentration at
 227 saturation, $\sigma_{L,0}$ is the surface tension when the solvent is pure and Ta the adsorption
 228 temperature. The surface concentration at saturation Γ_∞ is determined using the slope of the
 229 curve linked the surface tension and $\text{Log}(C)$ described in Figure 2. From the values of C , K
 230 and Γ_∞ , the surface coverage ratio S_e is deduced. Given its importance, S_e is a parameter
 231 which can be chosen to classify the bubbles according to their interface nature [12]:

- 232 · $S_e = 0$ corresponds to an interface free of surfactants,
- 233 · $S_e = 1$ corresponds to an interface saturated with surfactants,
- 234 · $0 < S_e < 1$ corresponds to a non-saturated interface.

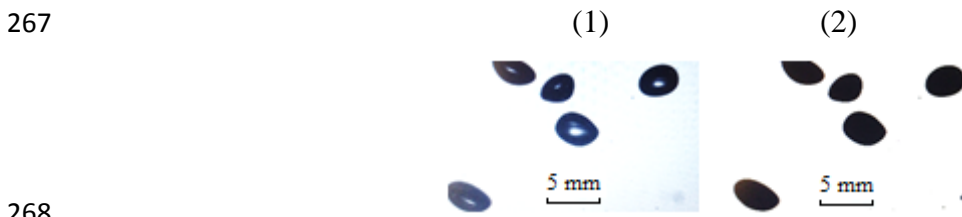


246 Figure 2. Diagram for determining the characteristic adsorption parameters

248 Because of the Newtonian behavior of surfactant solutions, the viscosity of their liquid
 249 phases is not modified. However, the microcrystalline cellulose solutions have a non-
 250 Newtonian fluid behavior, the viscosity of the liquid-phases changes with concentration
 251 changes. That brings us to study the rheology of MCC solutions. A rheological measurement
 252 consists on determining the longitudinal pressure loss associated to the liquid flow rate
 253 through a capillary tube of known geometry [5]. To determine the rheological behavior of
 254 MCC solutions under study, a Thermo Fisher Scientific HAAKE MARS rheometer-system
 255 equipped with a plate-plate geometry was used. The Ostwald-de Waele rheological
 256 parameters were determined using (3), where K represents the flow consistency index and n
 257 the flow behavior index.

258 $\eta = K\gamma^{n-1}$ (3)

259 As shown in Figure 1, bubbles are photographed with a 64-bit Pylon Viewer fast camera
 260 [acA1920-155um] that can shoot up to 390 frames per second. The lighting is accomplished
 261 by a LED lamp. The introduction of a white plywood plate into the reactor increases the
 262 contrast of the images. Images are visualized on the acquisition computer and are treated with
 263 the ImageJ software. Figure 3 presents a typical sequence of image treatment which is based
 264 on a transformation of the acquired image into a binary image. Different arithmetical and
 265 geometrical operations as filling the holes followed the binarization to give a uniform surface
 266 treatment and to remove superfluous images.



268
 269 Figure 3. Typical sequence of image treatment: (1) image acquisition; (2) image binarization
 270

271 Afterward the images acquisition and treatment, the effect of liquid phase
 272 concentrations on hydrodynamic and mass transfer parameters can be determined.

273 At low gas flow rates, bubbles are spherical and Eq. (4) is considered to calculate the
 274 diameter. At high gas flow rates, bubbles become ellipsoidal and Eq. (5) is considered to
 275 calculate the diameter after the measurement of the geometrical characteristics [10]. An
 276 average bubble diameter is deduced from the measurement of 70 to 100 bubbles diameter.

277 $D_B = \sqrt{\frac{4 \times A_B}{\pi}}$ (4)

278 $D_B = \sqrt[3]{(l^2 \times h)}$ (5)

279 The bubble frequency (f_B) can be calculated using two methods: (i) the ratio of air flow
 280 rate to bubble volume; (ii) the direct counting of bubbles on the image sequence.
 281 Jamnongwong et al. [3] found a good agreement between both methods. The first method was
 282 used in this work. The bubble formation frequency is determined as (6):

283 $f_B = \frac{q}{V_B}$ (6)

284 Where V_B is the average detached bubble volume and q is the gas flow rate. Note that a
 285 gas flow rate of 1.16 ml/s is fixed whatever the experiments. This low value hinders any
 286 surface deformation.

287 The terminal rising bubble velocity (U_B) can be estimated via two methods: (i) by
 288 measuring the distance between two frames; (ii) by following the variation in the bubble
 289 extremity coordinates with time. In this work, the first method was used. The terminal rising
 290 bubble velocity is determined as (7), where ΔD is the bubble spatial displacement between $t =$
 291 0 and $t = T_{frames} = 1/100$ s.

$$292 \quad U_B = \frac{\Delta D}{T_{frames}} \quad (7)$$

293 The specific interfacial area (a) is defined as the ratio between the bubble surfaces (S_B)
 294 and the total volume in the reactor (V_{Total}) [10] [12] as in (8), where H_L and A are respectively
 295 the liquid height ($H_L = 0.20$ m) and the cross-sectional area ($A = 1.5 \cdot 10^{-3}$ m²). The number of
 296 bubbles (N_B) is deduced from the terminal rising bubble velocities (U_B) and the bubble
 297 formation frequency (f_B) previously determined.

$$298 \quad a = N_B \frac{S_B}{V_{Total}} = f_B \frac{H_L}{U_B} \frac{S_B}{AH_L + N_B V_B} \quad (8)$$

299 Depending on the bubble shape, the bubble surfaces are calculated:

300 - For spherical bubble (9):

$$301 \quad S_B = \pi \cdot d_B^2 \quad (9)$$

302 - For ellipsoidal bubble (10), where e is the ratio between h and l .

$$303 \quad S_B = 2\pi \left[\frac{l^2}{4} + \left(\frac{l^2}{4} \times \frac{l}{2e} \ln \frac{(1+e)}{(1-e)} \right) \right] \quad (10)$$

304 The superficial gas velocity (U_g) is defined as the average velocity of the gas that is
 305 sparged into the column. In calculations, superficial gas velocity is simply expressed as the
 306 volumetric flow rate (q) divided by the cross-sectional area of the column (A). Flow rates of
 307 0.55 ml/s, 1.16 ml/s, 2 ml/s and 2.5 ml/s were chosen to study the influence of superficial gas
 308 velocity on mass transfer parameters.

309 The volumetric mass transfer coefficient (K_{La}) can be appreciated through two methods:
 310 (i) the sulfite static method based on a mass balance on sulfite sodium (Na_2SO_3) concentration

311 during aeration time [10][3][12]; (ii) the classical method named the non-stationary or the
 312 dynamic method [10][14][13]. In this work, the classical method was used. It consists in
 313 passing nitrogen through the liquid phase in order to remove the oxygen content and to
 314 substitute it with air at the beginning of the experiment. The oxygen concentration in the
 315 liquid phase is measured with an oxygen microsensor. In our experiments, UNISENSE
 316 microsensor which is a miniaturized Clark-type oxygen sensor was used. The sensor is related
 317 to a high-sensitivity picoammeter, allowing the resulting reduction current from the oxygen
 318 penetration to be converted into a signal. The conversion of this signal into an equivalent
 319 concentration of oxygen (C_L) is assured by considering a linear conversion and by multiplying
 320 with the atmospheric level solubility. During the experimental route, sufficient time is
 321 available to reach the oxygen saturation (C_L^*) in the liquid. The response time of the used
 322 probe is fixed to be as low as 50 ms, which corresponds to the experimental error estimated to
 323 $\pm 2\%$ for the volumetric mass transfer coefficient determination. To apply this method,
 324 several assumptions are made: the liquid phase is perfectly mixed, the response time of the
 325 probe is negligible and the oxygen depletion from gas bubbles is negligible. As the oxygen
 326 concentration increases, the mass transfer rate is given by the equation (11):

$$327 \quad \ln(C_L^* - C_L) = \ln(C_L^*) - k_{Lat}t \quad (11)$$

328 The volumetric mass transfer coefficient is then deduced from the slope of the curve
 329 linking the variation of $\ln(C_L^* - C_L)$ with time (t).

330 However, the signal (S) of the polarographic probe can be directly used for calculating
 331 the volumetric mass transfer coefficient [14]. Indeed, the volumetric mass transfer coefficient
 332 in the liquid phase is deduced from the slope of the curve described by (12):

$$333 \quad \ln \frac{S^* - S}{S^* - S_0} = -k_{Lat}t \quad (12)$$

334 The volumetric mass transfer coefficient is the product of the liquid-side mass transfer
 335 coefficient (K_L) and the specific interfacial area. The most commonly method used to estimate
 336 the liquid-side mass transfer coefficient [10][14][12][13] is by using the equation (13):

$$337 \quad k_L = \frac{k_{La}}{a} \quad (13)$$

338

339 **3. RESULTS AND DISCUSSION**

340

341 Concentrations of the liquid phases and their characterization are reported in Table 1.
 342 According to the experimental values of surface tension and taking account for experimental
 343 uncertainties, the surface tensions of MCC solutions do not vary from the one of tap water (σ_L
 344 = 72.2 mNm⁻¹). On the contrary of inorganic substances addition [14][8], adding surfactants
 345 to tap water strongly lowers the surface tensions (σ_L = 39.0–43.5 mNm⁻¹). When the
 346 surfactant concentration increases, the surface tension tends to decrease until saturation of the
 347 solution with surfactants compounds. Here, the CMC is reached and the surface tension
 348 remains constant above this concentration. Change in surface tension of SDS and SDS/MCC
 349 solutions is believed to affect hydrodynamics and oxygen mass transfer. The critical micelle
 350 concentration of SDS found (2.01 g/L) is slightly higher to these found in literature (1.9 g/L)
 351 [10][14][12]. The value of the surface coverage ratio se is assumed to be equal to 0 in the case
 352 of water. However, se increases with the surfactant concentration in the liquid phase.
 353 Furthermore, the surface coverage ratio se is equal to 1 when the concentration is equal or
 354 higher than the critical micelle concentration. Here, the interface is saturated.

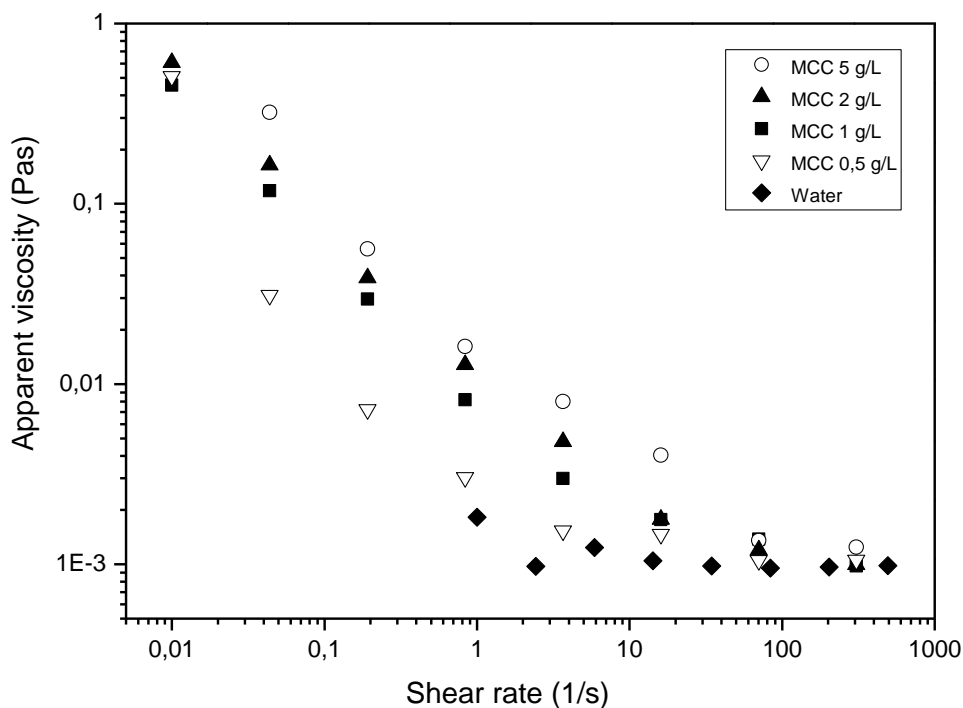
355 Table 1. Properties of liquid phases under test

Liquid phase	C (mol/L)		C (g/L)		M (g/mol)	σ_L (mN/m)	CMC (g/L)	C* (mg/L)	K (m ³ /mol)	Γ_{∞} (mol/m ²)	Se
	SDS	MCC	SDS	MCC							
Tap water	0	0	0	0	18	72.2	-	8.81	-	-	0
SDS1	3.5 x 10 ⁻³		1.00			43.5		9.18			0.9
SDS2	7 x 10 ⁻³	0	2.01	0	288.37	39	2.01	9.13	6.25	3.50 x 10 ⁻⁶	1
SDS2.59	9 x 10 ⁻³		2.59			39		9.12			1
MCC0.5		3 10 ⁻³		0.50		72		8.98			
MCC1		6 10 ⁻³		1.00		72.2		9.01			
MCC2	0	1.2 10 ⁻²	0	2.00	162.14	72	-	8.89	-	-	-
MCC5		3 10 ⁻²		5.00		72.1		8.86			
SDS/MCC0.5		3 10 ⁻³		0.5		43.7		9.06			
SDS/MCC1		6 10 ⁻³		1		43.8		9.15			
SDS/MCC2	3.5 10 ⁻³	1.2 10 ⁻²	1	2	-	43.6	-	9.18	-	-	-
SDS/MCC5		3 10 ⁻²		5		43.7		9.20			

356

357 To more understand the microcrystalline cellulose rheology, concentrations ranging from
 358 0.5 to 5 g/L of MCC solutions are studied. Variation of the apparent liquid viscosity as a
 359 function of the applied shear rate is shown in Figure 4. The obtained Ostwald-de Waele
 360 rheological parameters are presented in Table 2. Results show that the liquid apparent
 361 viscosity decreases as the applied shear stress increases. It can also be shown that the

362 microcrystalline cellulose liquid apparent viscosity and the consistency index (K) increase
 363 simultaneously with the MCC concentration. This result can be explained by a higher number
 364 of interactions between cellulose particles that consequently move less freely and exert more
 365 resistance to flow. On the opposing, it is observed that the flux index (n) decreases when the
 366 MCC concentration increases, thereby highlighting the shear-thinning character of the MCC
 367 liquid (decrease in viscosity with an increase in shear rate). From the values of n obtained for
 368 all concentrations ($0 < n < 1$), it can be concluded that the MCC liquid phases are a highly
 369 viscous pseudo-plastic solutions.



370

371 Figure 4. Rheology of microcrystalline cellulose liquids

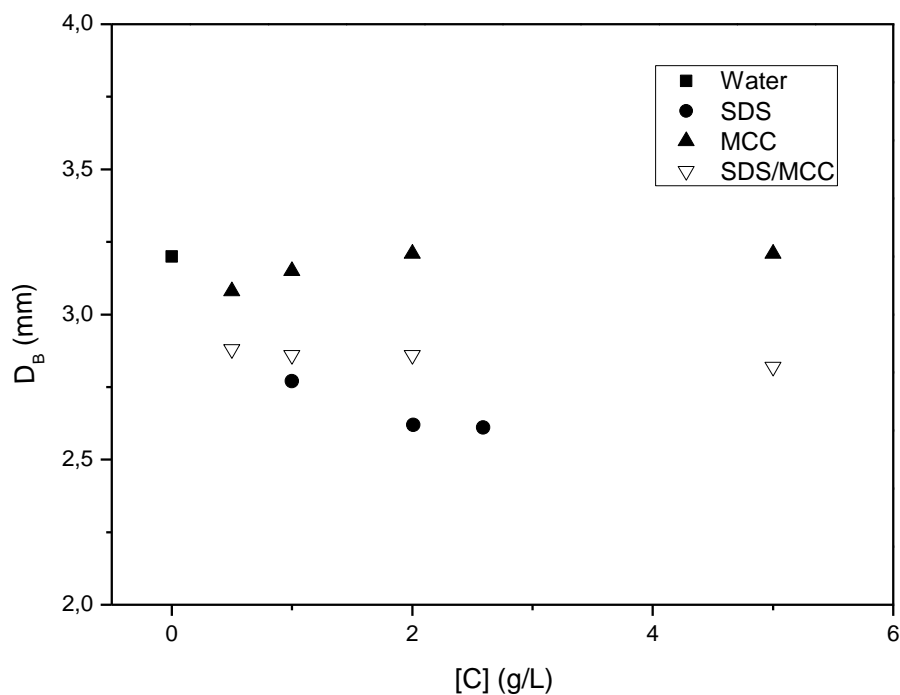
372 Table 2. Results of the multiple regression analysis on the rheology experimental data

Parameter	Liquid phase			
	MCC0.5	MCC1	MCC2	MCC5
K (Pa s)	0.013	0.018	0.024	0.029
n	0.451	0.405	0.381	0.171

373

374 In the part below, the effect of liquid phase concentrations on hydrodynamic and mass
 375 transfer parameters is considered. All the data reported and commented have been obtained
 376 for a constant superficial gas velocity of $7.73 \times 10^{-4} \text{ m s}^{-1}$.

377 The relation between the detached average bubble diameters and the liquid phase
 378 concentrations for each solution are shown in Figure 5. The result shows that the average
 379 bubble diameters obtained with SDS, MCC and SDS/MCC solutions were lower than that
 380 obtained with the tap water, except for SDS/MCC at 2 and 5 g/L where the average diameter
 381 was greater than that of water. This result can be explained by the coalescence between
 382 bubbles due to the high viscosity of the solutions. For SDS solutions, it was observed that the
 383 average bubble diameter decreased as the concentration increased due to the differences in
 384 term of dynamic surface tensions. The effect was also observed by Loubière and Hébrard [8]
 385 and Painmanakul et al [10] reporting that the bubble diameter of SDS solution with the
 386 surface coverage ratio (Se) equal to 1 was smaller than those with low Se , which clearly
 387 demonstrates the modification of the bubble diameters due to the surfactant concentration. For
 388 SDS concentrations above the CMC (SDS2.59), the bubble diameter remained practically
 389 constant. For MCC solutions, firstly the average diameter initially decreased but then increase
 390 with the concentration remained constant after the concentration was above 2 g/L. For
 391 SDS/MCC solutions, the bubbles diameter seems to be independent of the concentration of
 392 MCC as the MCC presenting in the liquid phase could adsorbed a certain amount of SDS
 393 leading to a limitation of surfactants effect on the bubble sizes. Whatever the solutions under
 394 test, the change of bubbles diameter compared with that of water is certainly due to the
 395 change in surface tension for SDS solutions, change in viscosity for MCC solutions, and both
 396 surface tension and viscosity for SDS/MCC solutions.

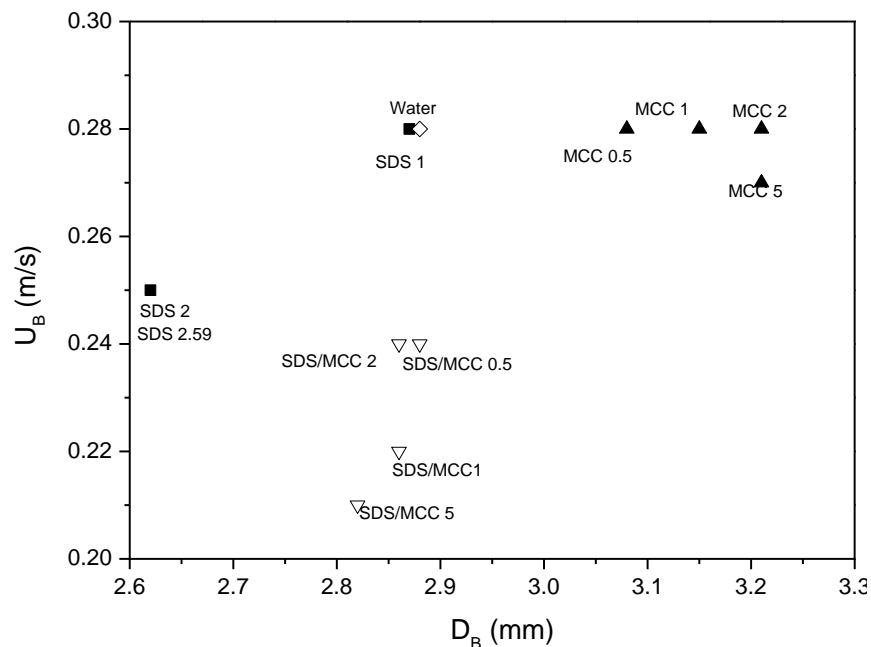


398

Figure 5. Bubble diameter versus liquid phase concentration

399

400 Variations of the rising bubble velocity with the bubble diameter generated for each
401 SDS, MCC and SDS/MCC solutions are plotted in Figure 6. Over the whole bubble diameter
402 range (2.62 – 3.2 mm), the terminal rising bubble velocities obtained varies between 0.21 and
403 0.28 m.s^{-1} and are included to those given by Grace and Wairegi [26]. These results show
404 that the terminal rising bubble velocity can be affected by the presence of SDS, MCC and
405 their combination; For SDS and SDS/MCC solutions, the terminal rising bubble velocities are
406 respectively reduced from 0.28 to 0.25 m.s^{-1} and from 0.28 to 0.21 m.s^{-1} compared to those of
407 tap water. This diminution is certainly due to the liquid film generated by surfactants
408 compounds around the bubbles which prevents bubbles from moving faster.



409

Figure 6. Terminal rising bubble velocity versus bubble diameter

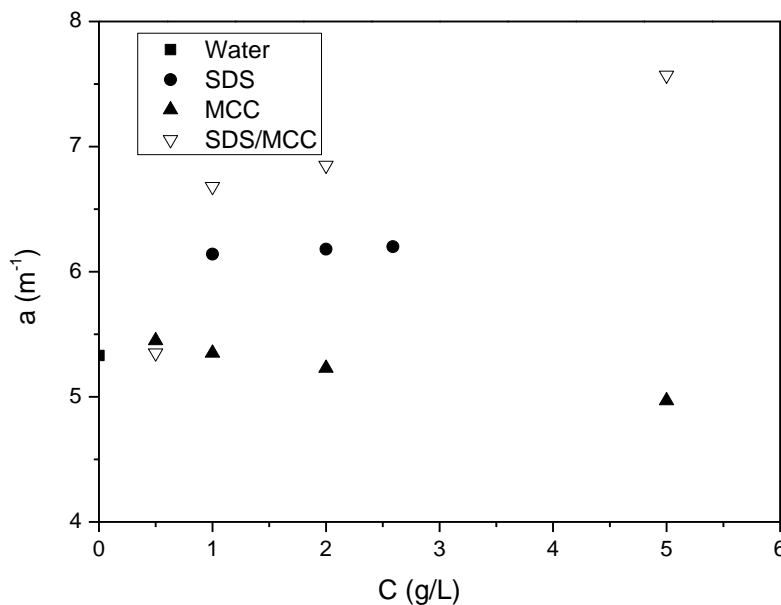
410

411

412 The relation between the interfacial area and the concentration for each SDS, MCC and
413 SDS/MCC solutions is shown in Figure 7. Results show that the values of the interfacial area
414 vary between 4.97 and 7.97 m^{-1} for the concentration varying between 0.5 and 5 g/L . Except
415 the MCC liquids, whatever the SDS and SDS/MCC concentration in the liquid phase, the
416 value of interfacial areas were higher than that of water. For MCC liquid phases, the
417 interfacial area decreases when the MCC concentration increase. This result is undoubtedly
418 due to the increase in the viscosity of the solution as reported in Figure 5 correlating to an

419 increase in bubble sizes as reported on Figure 6. The interfacial areas related to surfactant
 420 solutions are significantly higher than that found for tap water. This great influence on the
 421 interfacial area can be explained by the bubble size decrease in presence of surfactant
 422 compounds. The result consistent with that obtained by Painmanakul et al [10]. For the SDS
 423 and SDS/MCC solutions, the values of a obtained with Se equal to 1 were larger than that
 424 obtained with the lower Se value meaning that the interfacial area increased with SDS
 425 concentrations. With SDS/MCC solutions the interfacial area remains larger than that of water
 426 as bubble slip velocities and bubble sizes are lower, giving important gas holdup and
 427 interfacial area.

428 To better understand the effect of the microcrystalline cellulose and its combination
 429 with surfactants on the mass transfer phenomena, the volumetric mass transfer coefficient and
 430 the liquid-side mass transfer coefficient are considered separately in the next part.



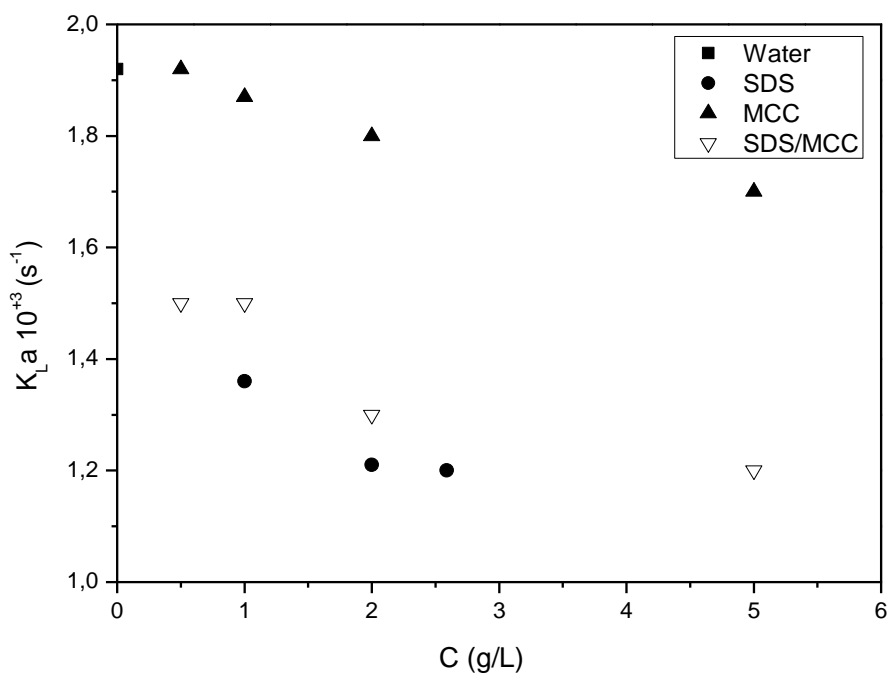
431

432 Figure 7. Interfacial area versus the liquid phase concentration

433

434 Variation of the volumetric mass transfer coefficient with the liquid phase concentration
 435 for the different solutions is plotted in Figure 8. The result indicates that the values of $k_L a$
 436 vary between 1.20×10^{-3} and $1.36 \times 10^{-3} \text{ s}^{-1}$ for solutions containing SDS, 1.70×10^{-3} and
 437 $1.92 \times 10^{-3} \text{ s}^{-1}$ for MCC solutions and 1.20×10^{-3} and $1.50 \times 10^{-3} \text{ s}^{-1}$ for SDS/MCC solutions. It
 438 is clearly shown that the volumetric mass transfer coefficients of all solutions were
 439 significantly smaller than those of tap water. For MCC and SDS/MCC solutions, the

440 volumetric mass transfer coefficient decreased as the concentration increases due to the
 441 viscosity change and to the presence of antifoam which promoted bubble coalescence and,
 442 therefore, reduced the $k_L a$. The similar results concerning the effect of viscosity on $k_L a$ were
 443 obtained by Ruen-ngam *et al* [27], and Duran *et al* [5]. In addition, for SDS solutions, the
 444 lowest $k_L a$ values are obtained with the surface coverage ratio at equilibrium equal to 1, which
 445 proved that the presence of surfactants, even in small quantities, has important effects on the
 446 oxygen mass transfer. In order to comprehensively understand these phenomenon, the liquid-
 447 side mass transfer coefficient (k_L) was then performed.



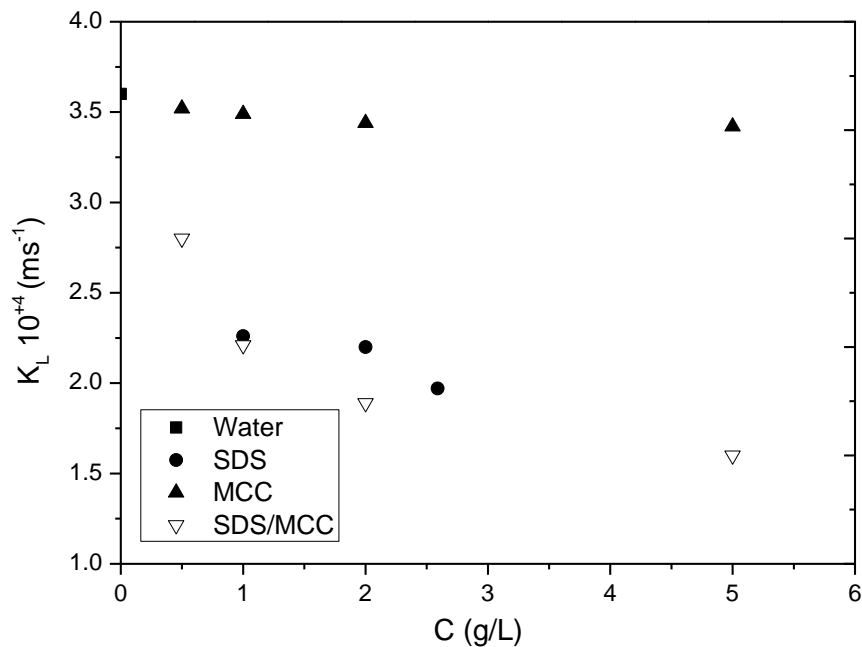
448

449 Figure 8. Volumetric mass transfer coefficient versus liquid phase concentration

450

451 Relation between the liquid-side mass transfer coefficient and liquid phase
 452 concentration for each SDS, MCC and SDS/MCC solutions is shown in Figure 9. The result
 453 indicates that the values of k_L varied between 1.97×10^{-4} and $2.21 \times 10^{-4} \text{ m.s}^{-1}$ for solutions
 454 containing SDS, 3.42×10^{-4} and $3.52 \times 10^{-3} \text{ m.s}^{-1}$ for MCC solutions and 1.60×10^{-4} and
 455 $2.80 \times 10^{-4} \text{ m.s}^{-1}$ for SDS/MCC solutions. It was observed that liquid-side mass transfer
 456 coefficients of solutions containing surfactant were significantly smaller than that of water
 457 which concord with various literatures [10][14][12]. This decrease can be explained by the
 458 fact that the surfactant was assembled at the gas-liquid interface of bubbles. This distribution
 459 of surfactants molecules interrupts the mass transfer by modifying the composition and

460 increasing the thickness of the liquid film around the bubbles. For MCC solutions, it seems
461 that the concentration of the liquid phase do not affect the liquid-side mass transfer
462 coefficient.



463

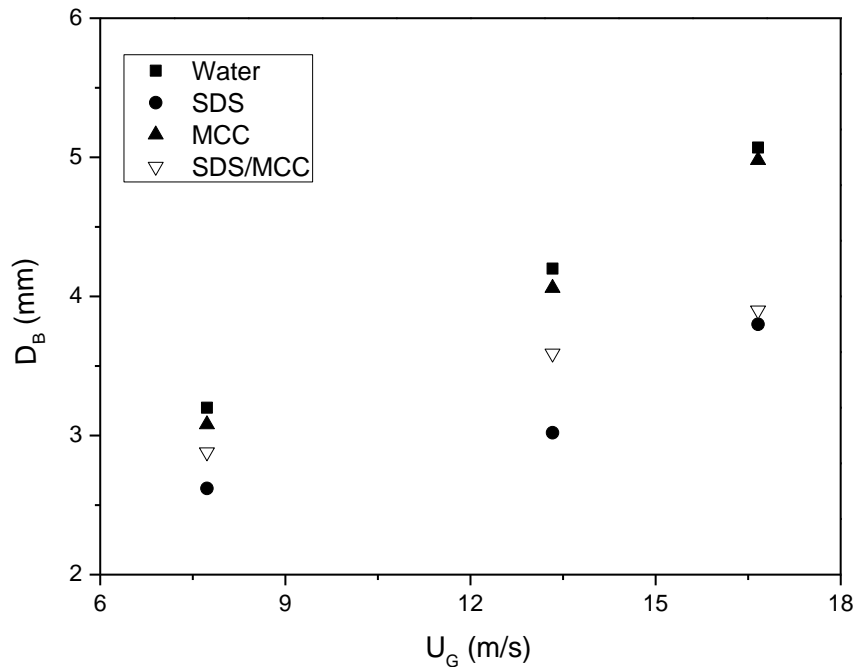
464 Figure 9. Liquid-side mass transfer coefficient versus liquid phase concentration

465

466 Below, superficial gas velocities of $3.66 \cdot 10^{-4}$ m/s, $7.73 \cdot 10^{-4}$ m/s, $13.33 \cdot 10^{-4}$ m/s and
467 $16.66 \cdot 10^{-4}$ m/s corresponding respectively to the flow rates of 0.55 ml/s, 1.16 ml/s, 2 ml/s and
468 2.5 ml/s were studied to understand the influence of superficial gas velocity on hydrodynamic
469 and mass transfer parameters with the presence of surfactants. Tap water, SDS2.59, MCC0.5
470 and SDS/MCC0.5 are liquid-phases chosen to be studied in this part.

471 The variation of the bubble diameters in the bubble column as a function of the
472 superficial gas velocity is shown in Figure 10. The values of D_B obtained in this work are in
473 the range of 2 to 5 mm. As it can be observed, whatever the liquid phases, the mean bubble
474 size increases with increasing superficial gas velocity. This result is in accordance with that
475 reported by Loubiere et al [10] for flexible membrane sparger where an increase in gas flow
476 rate lead to enlarge the flexible hole punctured into the membrane. That can also be explained
477 by increase in dispersion of small bubbles within the system with increasing aeration rate and
478 increasing the bubble collision frequency, that leads to higher coalescence rate and an
479 increase in bubble diameter [28]. Figure 12 also illustrates that the addition SDS and MCC to

480 water reduces the bubble size. This decrease can be attributed to the changes in surface
481 tension and viscosity for SDS and MCC solutions respectively as mentioned earlier in the
482 previous section.



483

484 Figure 10. Bubble diameter versus superficial gas velocity

485

486 Figure 11 presents the variation of the terminal rising bubble velocity as a function of
487 superficial gas velocity. Result shows that whatever the liquid composition, the terminal
488 rising bubble velocity increase with the superficial gas velocity. As found by Duran *et al* [5],
489 the multiple regression analysis did not show a statistically significant impact of the anionic
490 surfactant SDS and the suspended matter MCC studied on the oxygen transfer coefficient.
491 Nevertheless, the dissolved substances can participate to the depletion of the oxygen transfer
492 coefficient by reducing the bubbles terminal rise velocity [12] [9].

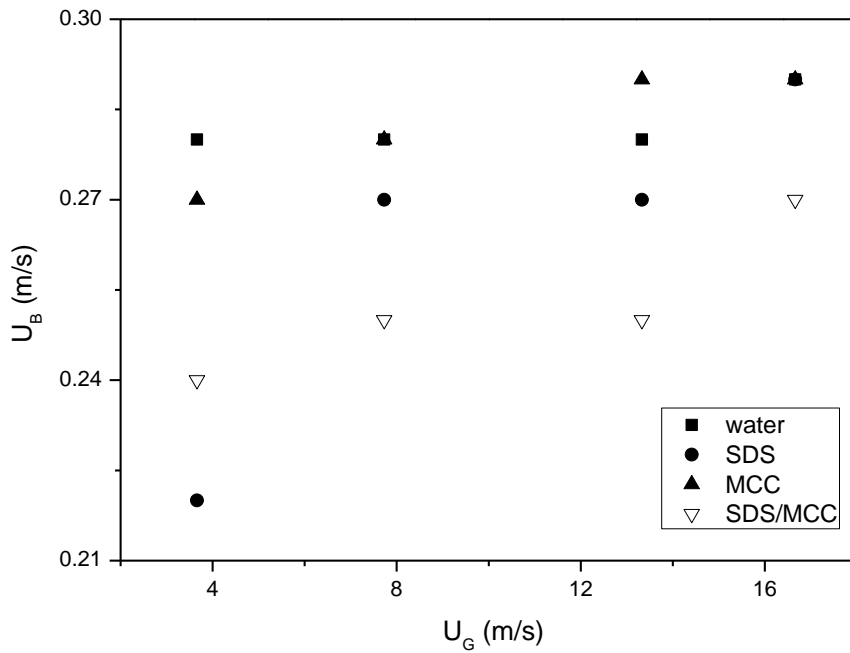
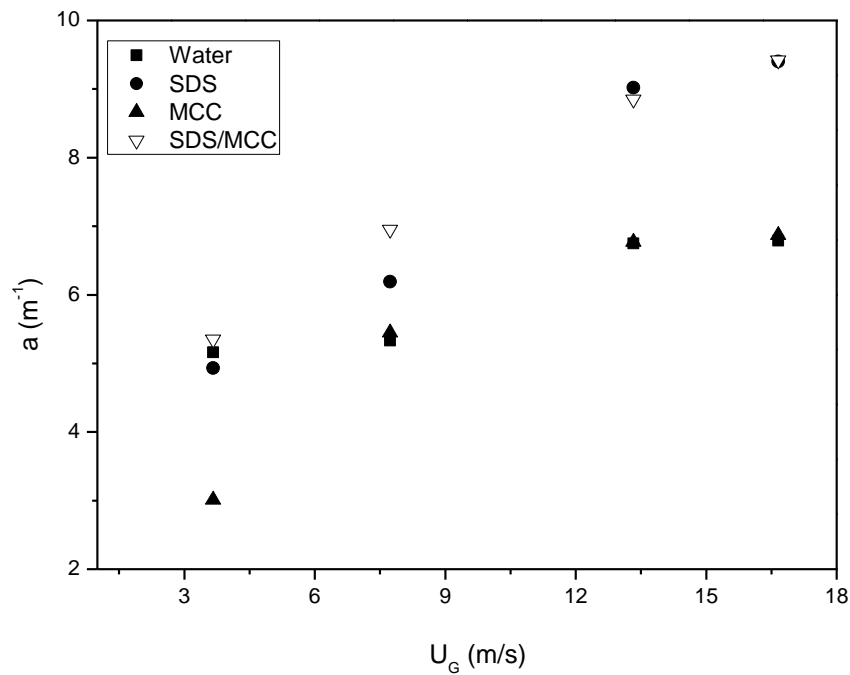


Figure 11. Terminal rising bubble velocity versus superficial gas velocity

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495

496 Variation of the experimental gas–liquid interfacial area with superficial gas velocity is
 497 illustrated in Figure 12. As it can be observed, the interfacial area increases with increasing
 498 the superficial gas velocity certainly by increasing in gas hold-up. Except for MCC and SDS
 499 solutions at low superficial gas velocity, Figure 13 reveals that the interfacial area of the SDS,
 500 MCC and their combination in aqueous solutions is larger than that of tap water. Increasing
 501 the MCC and SDS concentration reduces the bubble size due to decrease in surface tension.
 502 The surface tension has been related to the interfacial area through its effect on bubble size
 503 [28].



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Figure 12. Interfacial area versus superficial gas velocity

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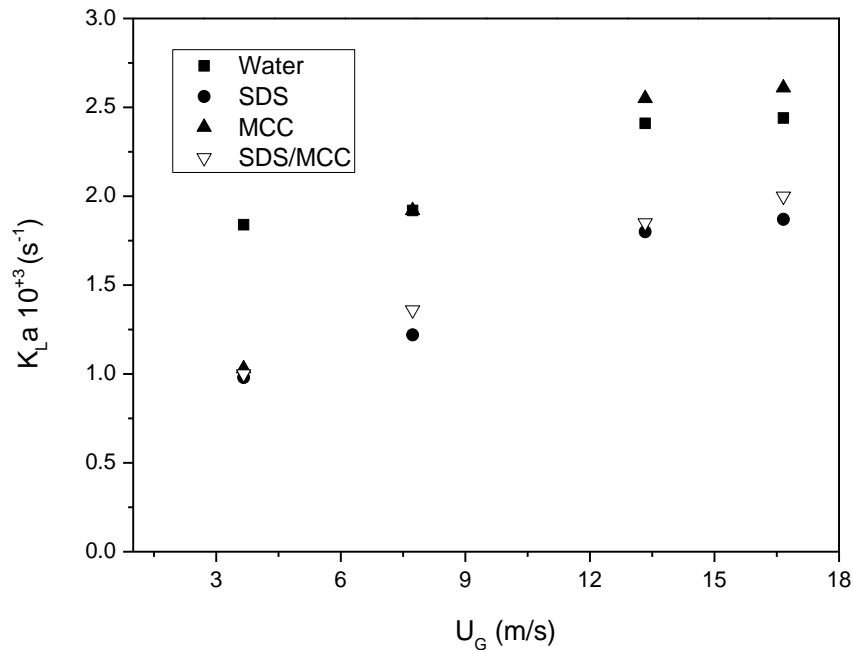
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Figure 13 presents the variation of volumetric oxygen transfer coefficient for the liquid phases under study as a function of superficial gas velocity. As it can be observed in this figure, except for MCC solutions at high superficial gas velocity, the volumetric oxygen transfer coefficients of the SDS, MCC and their combination in aqueous solutions are lower than that of tap water. It is also observed that the value of $k_L a$ for all of liquids increases with the superficial gas velocity which can be mainly explained by an increment in the gas hold up and therefore a larger specific interfacial area. This result corresponds to the homogeneous regime, where $k_L a$ increases with the gas velocity [29]. Knowing that the flow rate varies proportionally with the superficial gas velocity, the same results on the $k_L a$ were observed by De Jesus *et al* by increasing the flow rate [5]. Besides, the results showed that the volumetric oxygen transfer coefficient is mostly impacted by the MCC and SDS concentration and by superficial gas velocity.

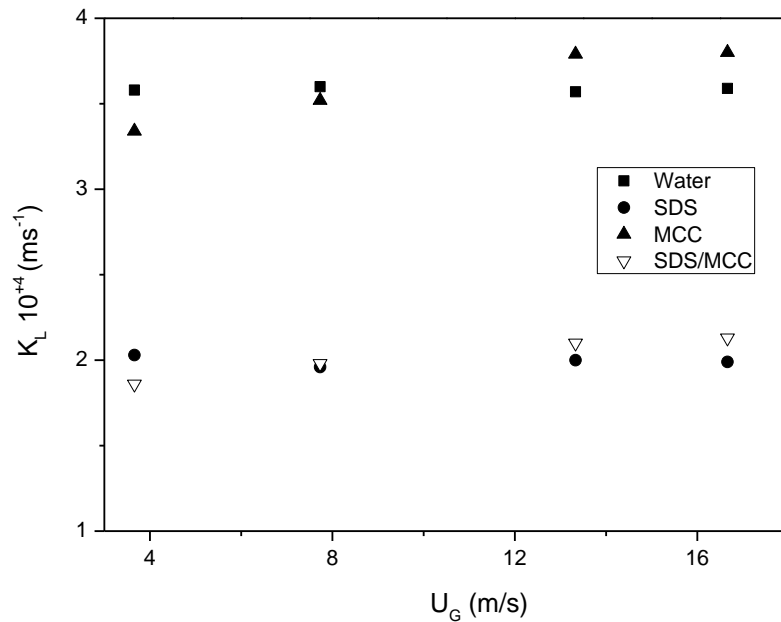


519

520 Figure 13. Volumetric mass transfer coefficient versus superficial gas velocity

521

522 Figure 14 demonstrates the effect of superficial gas velocity on liquid-side mass transfer
 523 coefficient. It seems that for a given liquid phase, the superficial gas velocity do not affect the
 524 liquid-side mass transfer coefficient. As shown in this figure, by addition of surfactant to
 525 water, the mass transfer coefficient decreases. The presence of surfactants over a gas–liquid
 526 interface increases the interfacial area by reducing the average bubble diameter, but decreases
 527 the mass transfer coefficient by increasing the liquid phase mass transfer resistance. Indeed,
 528 the presence of surface contaminants can bring to a lower degree of turbulence in the liquid
 529 film around bubbles and a reduction of liquid renewal at interface, therefore, increasing the
 530 liquid phase mass transfer resistance [13]. It can also bring to a reduction of diffusion
 531 coefficient at the film made at the gas liquid interface where the surfactants can be
 532 accumulated [15]. According to Figure 15, the addition of MCC to water and to SDS solution,
 533 doesn't modify the mass transfer coefficient. To better understand the effect of MCC on the
 534 liquid-side mass transfer coefficient, it should be important to study their presence in other
 535 liquid phases and follow by colorimetric technics their behavior at the interface.



536

537 Figure 14. Liquid-side mass transfer coefficient versus superficial gas velocity

538

539 **4. CONCLUSION**

540 The objective of this study was to investigate the effect of surfactants, microcrystalline
 541 cellulose and their combination on the hydrodynamic behavior and the mass transfer
 542 parameters of a bubble column. Rheological measures showed that an increase in MCC
 543 concentration causes an increment in apparent viscosity and accentuates the shear thinning
 544 behavior. However, it doesn't affect the liquid-side mass transfer coefficient. It was shown
 545 that the presence of surfactants affects the bubble generation process, hence the interfacial
 546 area and the different mass transfer parameters. Indeed, the addition of surfactant to water or
 547 to MCC in water system decreases the mass transfer coefficient. The k_{La} value for MCC
 548 solutions and the k_{La} and k_L value of surfactant solutions were shown to be significantly
 549 smaller than those of water. The volumetric mass transfer coefficient was shown to be
 550 increased with the superficial gas velocities whatever the liquid phases. However, it seems
 551 that for a given liquid phase, the superficial gas velocity doesn't affect the liquid-side mass
 552 transfer coefficient. In future work, it should be important to study the presence of surfactant
 553 and cellulose in other liquid phases to better understand their effect on the hydrodynamic
 554 behavior and liquid-side mass transfer coefficient.

555

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560

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