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# Influence of electrolyte concentration on holdup, flow regime transition and local flow properties in a large scale bubble column

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**Abstract.** We experimentally investigate the influence of the electrolyte concentration on holdup, flow regime transition and local flow properties in a large scale bubble column, with air and water as working fluids. The column is 0.24 m inner diameter, 5.3 m height and the air is introduced by a spider sparger up to a superficial gas velocity of 0.2 m/s. The influence of five NaCl concentrations are investigated by using gas holdup and optical probe measurements. The gas holdup measurements are used for analysing the flow regime transition between the homogeneous and the transition regime and the optical probe is used for studying the local flow characteristics at different radial positions. The presence of NaCl - up to a critical concentration - increases the gas holdup. The increase in the gas holdup is due to the inhibition of the coalescence between the bubbles and, thus, the extension of the homogeneous regime. The results are in agreement with the previous literature on smaller bubble columns.

## 1. Introduction

Bubble columns are widely used as multiphase reactors in the chemical, petrochemical, pharmaceutical and food industries. One of the main hydrodynamic parameters in bubble columns is the gas holdup,  $\epsilon_G$ , defined as the gas volume fraction in the mixture. The gas holdup - together with the mean bubble diameter,  $d_b$  - enables the computation of the interfacial area for the evaluation of the heat and mass transfer. The values of the gas holdup and the bubble size depend upon the superficial gas velocity,  $U_G$  (equal to the ratio of the volumetric flux to the area), and are related to the flow regime: mainly, the homogeneous and the heterogeneous regime. The former is associated with small superficial gas velocities and is characterised by the presence of small, uniform-sized bubbles with negligible interactions. The latter is associated with high superficial gas velocities and is characterised by a wide variety of bubble sizes and high coalescence and breakage phenomena. The transition from the homogeneous to the heterogeneous regime is a gradual process in which a transition flow regime occurs. This flow regime is characterized by large flow macro-structures with large eddies and widened bubble size distribution due to the onset of the bubble coalescence [1].

The correct prediction of the flow regime transition from the homogeneous to the transition regime is very important for the reactor design, effective operation and scale-up. For example, Krishna et al. [2] proved that non-coalescing systems can be used for predicting high-pressure systems. Among the different non-coalescing systems, we consider the electrolytes. It is well known that most electrolytes inhibit bubble coalescence in water [3-8] and, in this respect, a key concept is the transition

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concentration,  $c_t$ , defined as the concentration above which bubble coalescence is drastically reduced [6, 7]. Depending on the concentration of the electrolyte, we may define a “*coalescent regime*” ( $c/c_t \leq 1$ ) and a “*non-coalescent regime*” ( $c/c_t > 1$ ). In the case of NaCl – which is the subject of this study - the threshold is  $c_t = 0.145$  mol/l [9]. It is, therefore, obvious that the use of electrolytes can significantly alter the gas holdup in bubble columns [9-15]. In particular, the increase of  $\epsilon_G$  is considered a consequence of the homogeneous regime stabilisation (an increase in the superficial gas velocity of the regime transition onset,  $U_{trans}$ ) upon the addition of electrolytes into water [11].

Different studies investigated the influence of the electrolytes on the bubble column hydrodynamics and, in the following, we propose a brief literature survey. Thorat and Joshi [16] investigated a 0.385 m inner diameter (between 0.385 and 3.08 m height) bubble column with different gas spargers and using three media (water, 0.2 M NaCl, 1% CMC). The regime transition was investigated by using the Wallis plot and the homogeneous regime was stabilized by using NaCl: the coalescence suppression was seen to be the reason. Grover et al. [17] investigate a 0.1 m inner diameter (1.5 m height) bubble column using two electrolytes (NaCl, CuCl<sub>2</sub>). Using the Wallis plot, they reported a stabilisation of the homogeneous regime. Kelkar et al. [18] investigate a 0.154 m inner diameter (3.25 m height) bubble column using three electrolytes (NaCl, CaCl<sub>2</sub> and Na<sub>2</sub>SO<sub>4</sub>). The authors reported an increase in the gas holdup and a negligible effect of the electrolytes above the critical concentration. Zahradnik et al. [9] investigated a 0.14, 0.15 and 0.29 m inner diameter (2.6 m height) bubble columns and studied the influence of nine electrolytes. The gas holdup grew continuously for  $c \leq c_t$ , but little changes in  $\epsilon_G$  were observed for  $c > c_t$ . Moreover, a similar dependence of  $\epsilon_G$  vs.  $U_G$  was found for all electrolytes at  $c = c_t$ . Ribeiro and Mewes [11] studied a 0.12 m inner diameter (1.25 m height) bubble column using three electrolytes (NaCl, Na<sub>2</sub>SO<sub>4</sub>, NaI). Four values of the electrolyte concentration were tested in the “*coalescent regime*” and the gas holdup was found to increase till the critical concentration.

Despite the role of the electrolytes is quite well assessed, there is a lack of studies concerning large scale bubble columns. We contribute to the discussion investigating a 0.24 m inner diameter, 5.3 m height bubble column, which is larger than the columns typically studied. Air is introduced by using a spider sparger up to a superficial gas velocity up of 0.2 m/s and five NaCl concentrations ( $0 \leq c/c_t \leq 1.17$ ) are tested. We investigate the influence of electrolyte concentration over gas holdup, flow regime transition and local flow properties by using gas holdup and optical probe measurements. The gas holdup measurements are used for investigating the flow regime transition. The optical probe measurements were used to study the local flow characteristics at different radial positions. This experimental investigation mainly aim to provide further data for expanding the existing dataset.

## 2. The experimental setup

The bubble column, made of Plexiglas, is 0.24 m inner diameter and 5.3 m height (Figure 1). The authors have used the same facility for studying the bubble column hydrodynamic in the annular gap configuration [19-20]. The reader may also refer to the dissertation of Carrara [21] for a complete and detailed description of the facility. The compressed air passes through a filter and, then, a pressure valve regulator controls the pressure upstream the rotameters used to set-up the air flow rate (and, then, for computing  $U_G$ ). The air distributor is a spider sparger with holes ranging from 1 to 4 mm [21]. Due to the nature of the observed flow phenomena and their sensitivity to surface tension forces, clean filtered deionized water and filtered air were used. The air and water temperatures were maintained at room temperature ( $22 \pm 1$  °C) during the experiments. Five concentrations of NaCl were tested and are listed in Table 1. The values of gas density, used for computing  $U_G$  are based upon the operating conditions existing at the column mid-point. The mid-point column pressure was assumed to be equal to the column outlet pressure plus one-half the total experimental hydrostatic pressure head.

**Table 1.** NaCl concentrations tested.

$c$ [mol/l]	0	0.02	0.07	0.12	0.145 <sup>1</sup>	0.170
$c/c_t$ [-]	0	0.14	0.48	0.84	1	1.17

<sup>1</sup>Critical concentration,  $c_t = 0.145$  mol/l [9]

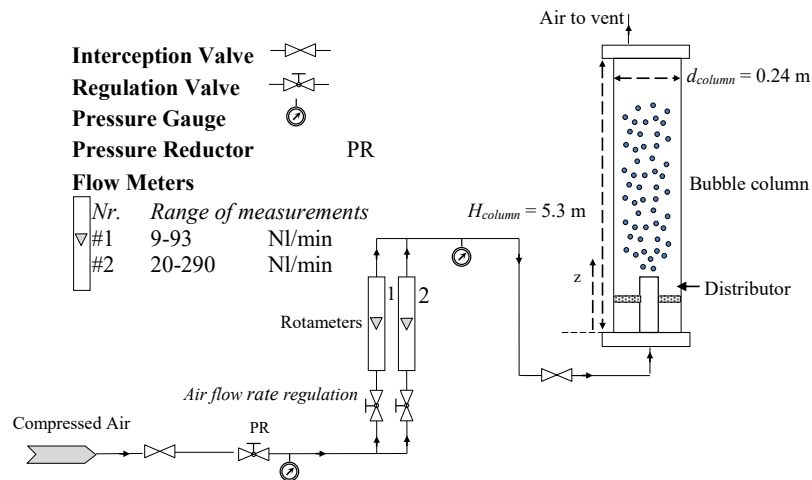


Figure 1. The experimental facility.

### 3. The measurement techniques

#### 3.1. Holdup measurements

Measurements of the bed expansion allowed the evaluation of the gas holdup  $\varepsilon_G$ . The procedure involves measuring the location (height) of liquid free surface when air flows in the column. The gas holdup is, then, obtained using the following relation:

$$\varepsilon_G = \frac{(H_D - H_0)}{H_D} \quad (1)$$

Where  $H_D$  and  $H_0$  are the heights (measured from the sparger) of the free surface after and before aeration, respectively. The error in the gas holdup measurements is estimated approximately 10%.

#### 3.2. Flow regime transition analysis

Although the flow transition from the homogeneous to the transition regime does not happen instantaneously [1], the definition of an approximate transition point is helpful for modelling the hydrodynamic of bubble columns [2]. In this study, we employ two methods from the literature for investigating the regime transition: (i) the swarm velocity and (ii) the drift flux method.

The swarm velocity method was developed by Zuber and Findlay [22] and is based on the evaluation of the swarm velocity,  $U_{swarm}$ :

$$U_{swarm} = U_G / \varepsilon_G \quad (2)$$

In this method, the swarm velocity is plotted against the superficial gas velocity:  $U_{swarm}$  is constant in the homogeneous regime and starts to increase - at a certain transition superficial velocity,  $U_{trans}$  - as the system enters the transition/heterogeneous regime. The appearance of the first large bubble is responsible for the increase in the swarm velocity and is an indication of flow regime transition. This method has been employed also by Krishna et al. [23], Letzel et al. [24] and Gourich et al. [25], Ribeiro and Mewes [11] and Besagni et al. [19-20].

The drift-flux method was proposed by Wallis [26] and has been widely applied in the literature [11, 26, 27]. This method is based on the drift flux (that represents the gas flux through a surface moving with the speed of the two-phase mixture) and it is experimentally obtained as follows:

$$J_T = U_G (1 - \varepsilon_G) \quad (3)$$

Theoretically, the drift flux is written in terms of a parameter,  $U_b$ , whose dependence upon  $\varepsilon_G$  varies with the prevailing regime:

$$J_E = U_b (1 - \varepsilon_G) \quad (4)$$

The idea is to employ a model for  $U_b$  valid for the homogeneous regime and to plot  $J_E$  and  $J_T$  in the same graph as a function of  $\varepsilon_G$ . In the homogeneous regime  $J_E$  is equal to  $J_T$  and the transition point is, thus, defined when:

$$J_T \neq J_E \quad (5)$$

The evaluation of  $U_b$  is a matter of discussion in the literature, as different models have been proposed and applied. In this study, we follow the approach of Krishna et al. [2, 28], which is based on the empirical model of Richardson and Zaki [29]:

$$U_b = u_\infty (1 - \varepsilon_G)^{n-1} \quad (6)$$

where  $n$  is fluid-dependent ( $n \cong 2$  for water) and  $u_\infty$  - the terminal velocity of an isolated bubble - should be fitted with the aid of the experimental data in the determination of the regime transition point. Combining Eq. (4) and Eq. (6) results:

$$J_E = u_\infty \varepsilon_G (1 - \varepsilon_G)^n \quad (7)$$

### 3.3. Local measurements

A double fiber optical probe system (RBI Instrumentation) measures the local flow properties. Similar optical probes have been used in previous studies [19, 20, 30-35] and Besagni et al. [20] proposed a discussion concerning the uncertainty of the optical probe measurements. An optoelectronic module emits the laser to the probe tip and converts the reflected optical signal into a digital signal. From the digital signal, the bubble frequency  $f$  (bubble number per unit time) and local void fraction  $\varepsilon_{G,Local}$  can be obtained. By cross-correlating the signals of the two tips, the bubble traveling time from one tip to the other can be estimated and bubble velocity  $u_b$  can be calculated. Assuming that bubbles are spherical, bubble Sauter mean diameter  $d_b$ , is calculated as follow [21, 34]:

$$d_b = 3\varepsilon_{G,Local} u_b / 2f \quad (8)$$

However, the spherical bubbles assumption (Eq. (8)) is approximately valid for small bubbles only ( $d_b < 3.5$  mm [34]). For accounting the non-sphericity of bubbles, the following equation is used [32]:

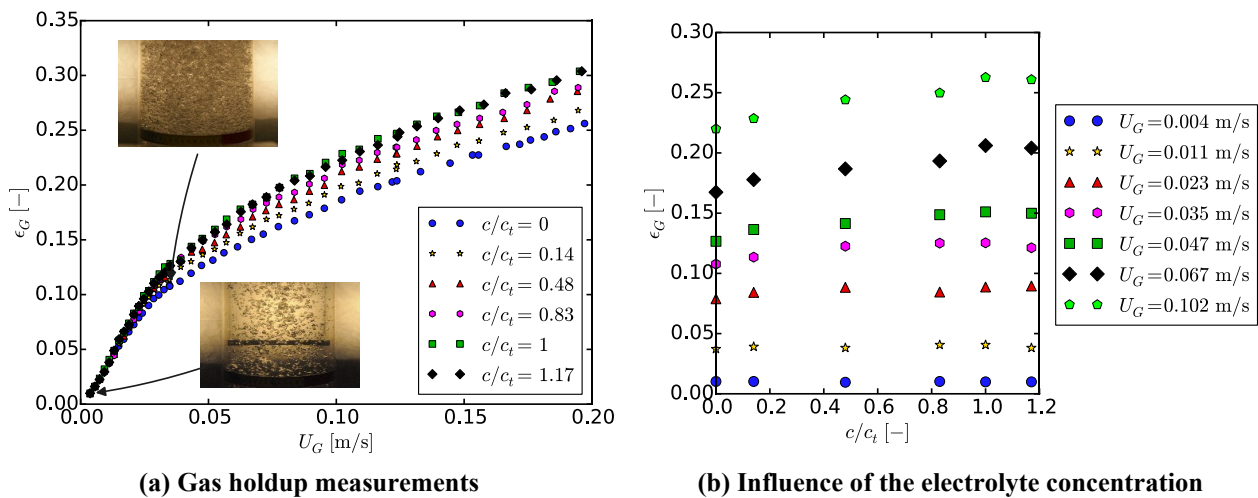
$$d_{b,corrected} = \alpha^{-2/3} 3\varepsilon_{G,Local} u_b / 2f \quad (9)$$

where  $\alpha$  is the aspect ratio. All the measurements have been obtained using a sampling period equal to 1000s, which is large enough to produce reliable time-averaged values and is far above the typical values of 1–5 min for similar optical probes [31, 33, 34, 36]. In this study, the optical probe was used for the following cases:  $U_G = 0.0037$  m/s and  $c/c_t = 0$ ,  $c/c_t = 0.48$  and  $c/c_t = 1.17$ .

## 4. The experimental results

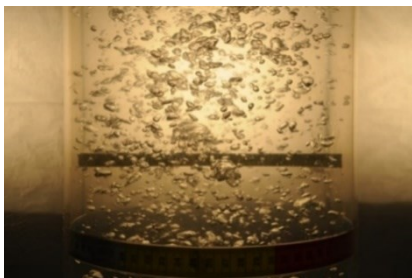
### 4.1. Holdup measurements

The gas holdup measurements are presented in Figure 2a. At low superficial gas velocities, the relationship between the gas holdup and the superficial gas velocity is linear, followed by a change in tendency at a transition superficial gas velocity,  $U_{trans}$ . The linear trend corresponds to the homogeneous/bubbly flow and the change in tendency is due to flow regime transition toward the transition regime. An example of flow visualisation for the bubbly flow is proposed in Figure 3, whereas the heterogeneous flow is presented in Figure 4. The gas holdup grows continuously while increasing the electrolyte concentration till the critical concentration, as reported in the previous literature [9, 11] (Figure 2a and b). The gas holdup increases also in the homogeneous regime, where the coalescence phenomena are limited, in agreement with Ribeiro and Mewes [11].

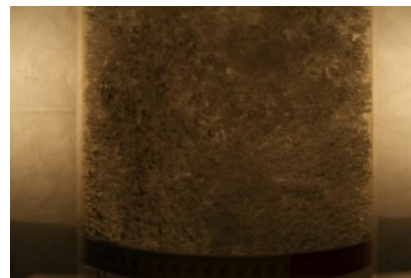


**Figure 2.** The gas holdup.

An interesting aspect is the non-linearity of the electrolytes effect upon the gas holdup (Figure 2a and b). The curve for  $c/c_t = 0.14$  and  $0.48$  is already shifted to considerable higher  $\epsilon_G$  values in comparison to the curve related to  $c/c_t = 0$ , while the relative distance between the curves associated with  $c/c_t = 0.48$ , a  $0.83$  and  $1$  is considerable lower. The non-linearity of the electrolytes effect upon the gas holdup was also concluded by Ribeiro and Mewes [11]. Above the critical concentration, there is no remarkable difference in the gas holdup, as expected from the literature [9].



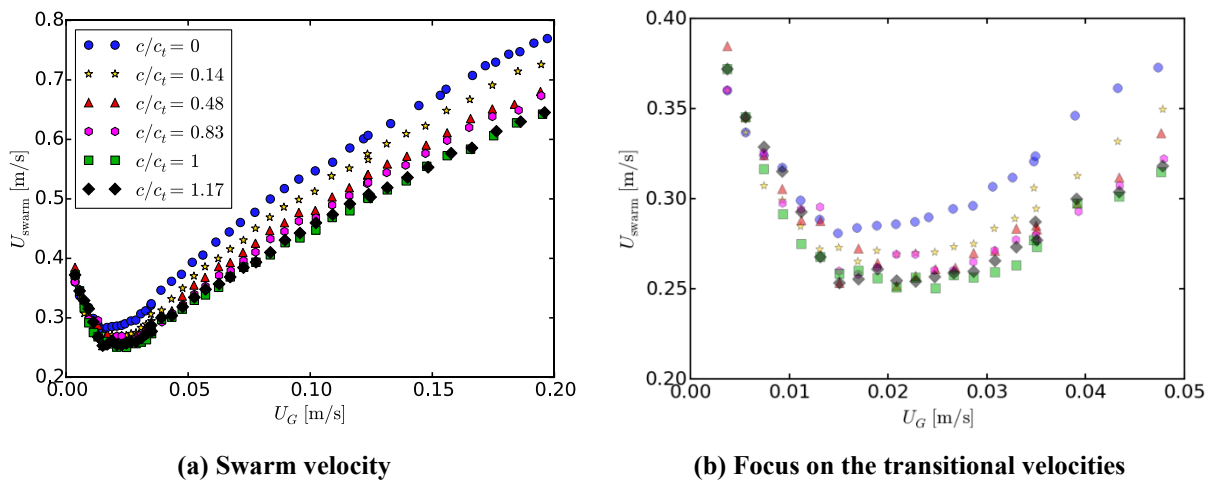
**Figure 3.**  $U_G = 0.0037$  m/s –  $c/c_t = 0$ .



**Figure 4.**  $U_G = 0.1665$  m/s –  $c/c_t = 0$ .

#### 4.2. Flow regime transition analysis

The gas holdup increases while increasing the electrolyte concentration till the critical value. Accordingly with the literature, the reason for the increase of the gas holdup lie in the inhibition of the coalescence phenomena; this hypothesis is verified also for our large bubble column by analyzing the flow regime transition using the two methods previously discussed (Section 3.2). The results of the swarm velocity method are presented in Figure 5a and the results of the drift flux method are presented in Figure 6. The values of the transitional gas velocity (Figure 5b and 6) are in agreement between the two methods and, following the proposal of Ribeiro and Mewes [11], the transition points have been evaluated as the mean of the two values and are presented in Figure 7 (where the dash lines are used for giving a better idea of the trend only). The transitional gas velocity,  $U_{trans}$  lies in the range between  $U_{trans} = 0.0264$  and  $0.0338$  m/s, depending on the NaCl concentration. Instead, the transitional holdup is between  $\epsilon_{G,trans} = 0.0831$  and  $0.1209$ . Both  $U_{trans}$  and  $\epsilon_{G,trans}$  increase while increasing the NaCl concentration, till the critical ratio:  $U_{trans}$  and  $\epsilon_{G,trans}$  increase up to 22 and 45%, respectively, if compared with  $c/c_t = 0$ . This supports the hypothesis that increasing the NaCl concentration, the homogeneous flow regime is stabilized and, thus,  $\epsilon_G$  increases by the inhibited coalescence. Beyond the critical concentration,  $U_{trans}$  and  $\epsilon_{G,trans}$  do not change anymore.



**Figure 5.** The swarm velocity method for the analysis of the flow regime transition.

The typical values of  $U_{trans}$  found in literature for air-water systems (considering  $c/c_t = 0$ ) in bubble columns of diameter higher than 0.15 m range between  $U_{trans} = 0.01$  and 0.08 m/s at ambient operating conditions, depending on the distributor geometry [9, 23, 37-42].

We may also compare our data ( $c/c_t = 0$ ) with literature correlations. One of the first correlations proposed is due to Wilkinson et al. [38]:

$$U_{trans} = 0.5U_{b,small} \exp(-193\rho_G^{-0.61}\mu_L^{0.5}\sigma^{0.11}) \quad (10)$$

where:

$$U_{b,small} = 2.25 \frac{\sigma}{\mu_L} \left( \frac{\sigma^3 \rho_L}{g \mu_L^4} \right)^{-0.273} \left( \frac{\rho_L}{\rho_G} \right)^{0.03} \quad (11)$$

Another correlation was proposed by Reilly et al. [39] and reads:

$$U_{trans} = \frac{\rho_L}{\rho_G} (1 - \varepsilon_{G,trans}) \left( \frac{B^*}{A^*} \right)^{1.5} \quad (12)$$

where:

$$\varepsilon_{G,trans} = 0.59 B^{*1.5} \left( \frac{\sigma^{0.12} \rho_G^{0.96}}{\rho_L} \right)^{0.5} \quad (13)$$

and:

$$A^* = 2.81 \rho_L \rho_G^{0.96} \sigma^{-0.12} \quad (14)$$

For water as the liquid phase,  $B^* = 4$ . The results of the comparison are presented in Table 3: Eq. (10) largely underestimate the transition gas velocity, whereas, Eq. (12) and (13) gives a value for  $U_{trans}$  and  $\varepsilon_{G,trans}$  in better agreement with the experimental data, as also observed in other studies [20, 40].

**Table 2:** Flow regime transition: comparison with the literature.

	Exp. $c/c_t = 0$	Wilkinson et al. [38]	Reilly et al. [39]
$U_{trans}$ [m/s]	0.0241	0.0029	0.0322
$\varepsilon_{G,trans}$ [-]	0.0831	0.01152 <sup>1</sup>	0.1484 <sup>1</sup>

<sup>1</sup> The value is given by  $U_{trans}/U_{b,small}$  Eq. (11)

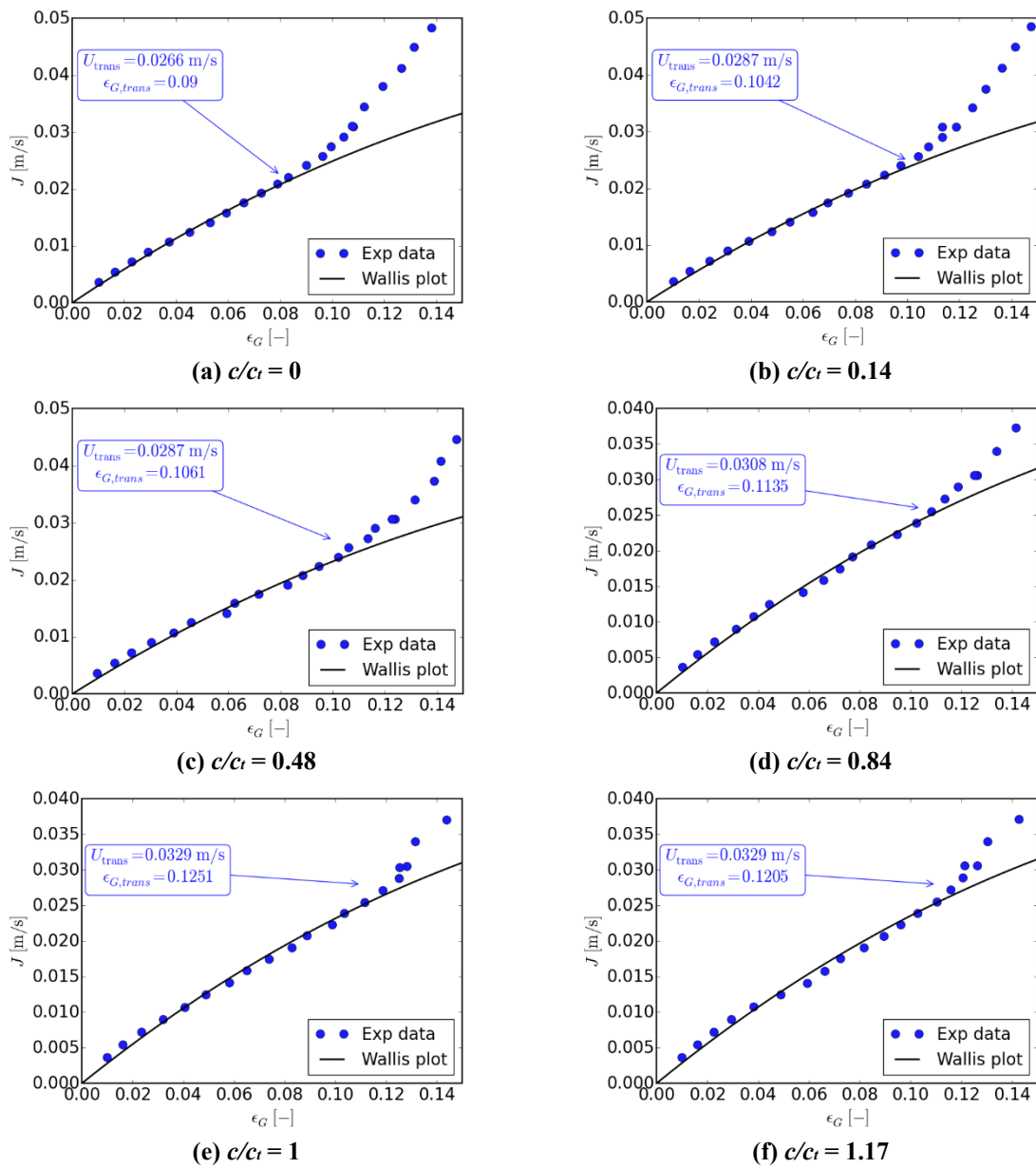


Figure 6: The drift flux method for the analysis of the flow regime transition.

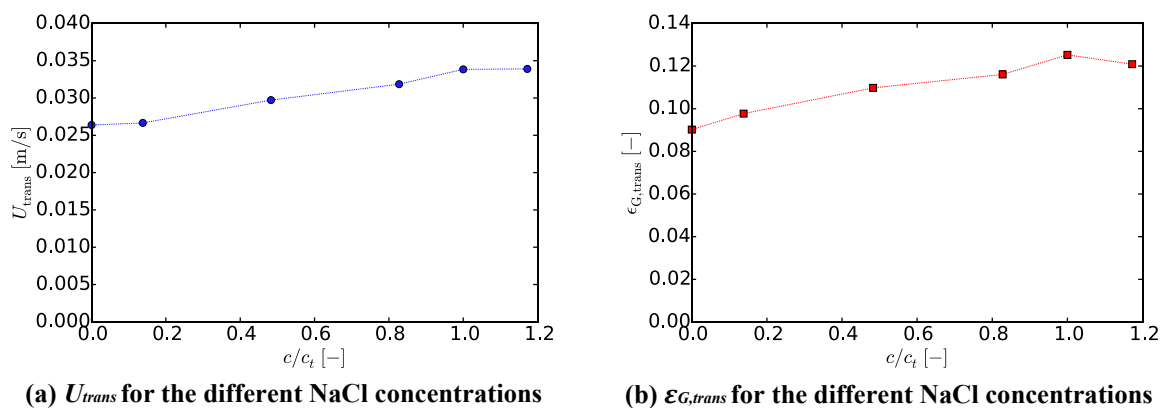
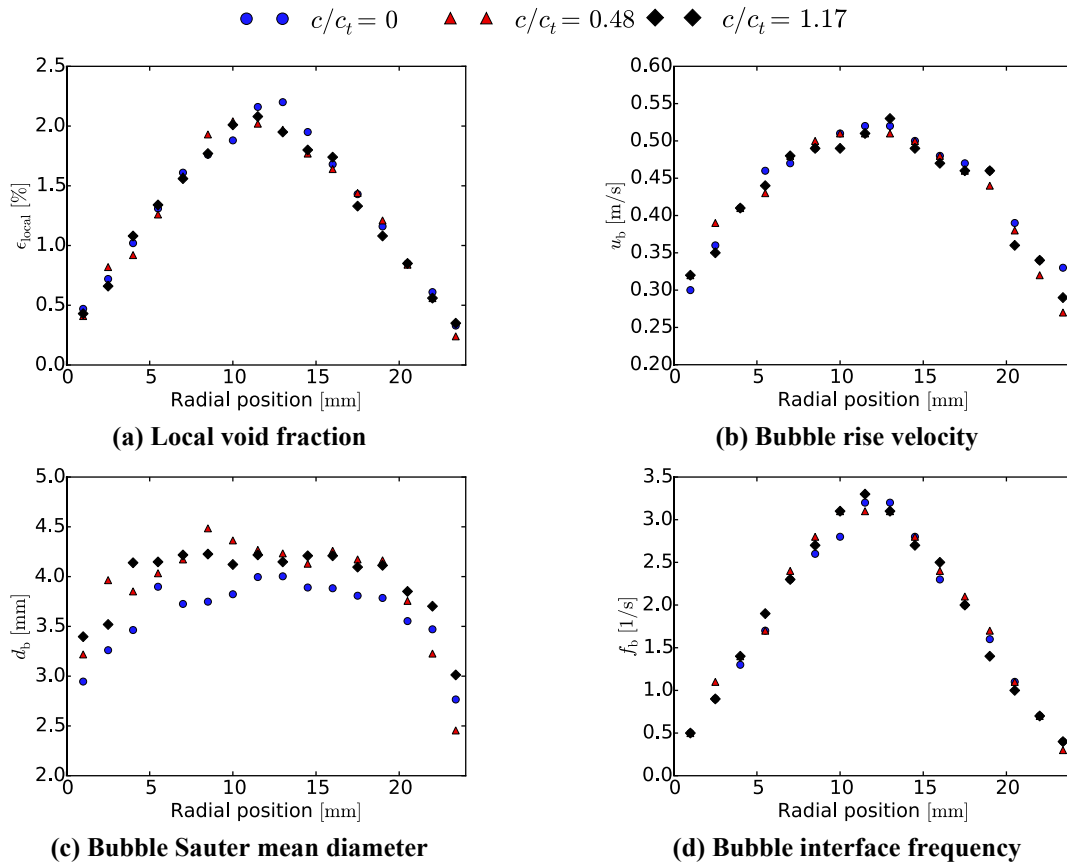


Figure 7: Transitional parameters:  $U_{trans}$  and  $\epsilon_{G,trans}$ .



#### 4.3. Local measurements

Figure 8 presents the radial optical probe measurements for  $c/c_t = 0$ ,  $c/c_t = 0.48$  and  $c/c_t = 1.17$ .



**Figure 8:** Optical probe measurements.

The local void fraction profiles are not flat, but are centre peaked. This is probably due to the distributor having large opening, which creates large bubbles in the centre of the column. Furthermore, there is no remarkable difference between the three salt concentrations (Figure 8a). This was expected because - for low  $U_g$  - these are no remarkable differences in the gas holdup (Figure 2a and 2b). Local void fraction  $\varepsilon_{G,Local}$  obtained with the optical probe were compared to global gas holdups by integrating the radial measurements over the column cross sectional area:

$$\langle \varepsilon_G \rangle = \frac{1}{\pi R_C^2} \int_0^{R_C} \varepsilon_{G,Local} 2\pi r dr \quad (15)$$

where  $R_C$ , is the radius of the column ( $R_C = 0.12$  m). Table 3 shows that the integrated local measurements were higher if compared to the global value, but they are very similar one another (as for the gas holdup values). The error may derive from the pressure gradient along the column (the volume of the bubbles increases while rising the column) as well as the developing region of the two-phase flow. Further analysis will be devoted to the uncertainty connected to the optical probes.

**Table 3.** Comparison between local (optical probe) and global gas holdup measurements.

$U_G$ [m/s]	$c/c_t$ [-]	$\langle \varepsilon_G \rangle$ [%]	$\varepsilon_G$ [%]
<b>0.0037</b>	0	1.68	1.02
<b>0.0037</b>	0.43	1.62	0.96
<b>0.0037</b>	1.17	1.61	0.99

Concerning the bubble vertical velocity  $u_b$  (Figure 8b), the values are between 0.3 (near wall) and 0.5 (at the center of the column) and there is no remarkable differences between the three curves. Figure 8c presents the bubble mean diameter obtained by using Eq. (9) using mean aspect ratio from the image analysis:  $\alpha = 0.649$  for  $c/c_t = 0$ ,  $\alpha = 0.808$  for  $c/c_t = 0.48$  and  $\alpha = 0.816$  for  $c/c_t = 1.17$ . The image analysis method is the one detailed by Besagni et al. [20] and the sampling was performed considering the bubbles near the wall of the column. The number of bubbles sampled and the operating conditions considered the are the followings:

- $c/c_t = 0$  - 3218 bubbles -  $U_G = 0.0037, 0.0074, 0.111, 0.0149$  and  $0.188$  m/s;
- $c/c_t = 0.48$  - 1481 bubbles -  $U_G = 0.0037$  and  $0.0074$  m/s;
- $c/c_t = 1.16$  - 1341 bubbles -  $U_G = 0.0037$  and  $0.0074$  m/s.

A complete discussion of the number of bubbles sampled, the results concerning the bubbles size distributions and the bubble shape is far beyond the scope of this paper, and detailed results for the image analysis will be presented elsewhere. It is interesting that the mean diameter for  $c/c_t = 0.48$  and for  $c/c_t = 1.17$  are higher: this is due to the change in the bubble shape, caused by the coalescence inhibition and the modified interaction between the bubbles. Figure 8d presents the bubbly interface frequency and the three curves are similar, as expected (Section 3.3).

## 5. Conclusions

We have experimentally investigated the influence of electrolyte concentration over holdup, flow regime transition and local flow properties in a large scale bubble column, with air and water as working fluids. The column is 0.24 m inner diameter, 5.3 m height and the air is introduced by using a spider sparger up to a superficial gas velocity up of 0.2 m/s. Five NaCl concentrations were tested and the experimental investigation consists in gas holdup and optical probe measurements. The gas holdup measurements were used for investigating the flow regime transition and the data obtained from the optical probe were used to study the local flow characteristics at different radial positions.

It is found that the presence of NaCl - up to the critical concentration - increases the gas holdup, even for the operation in the homogeneous regime, where bubble coalescence frequencies are not high. A non-linear increase in the gas holdup with the electrolyte concentration was verified, whose extent was greater for small concentrations and progressively levelled off as the electrolyte content in the liquid phase was raised. The gas holdup measurements are used for investigating the flow regime transition between the homogeneous and the transition regime. The transitional gas velocity lies in the range between  $U_{trans} = 0.0264$  and  $0.0338$  m/s, depending on the NaCl concentration. Instead, the transitional holdup is between  $\varepsilon_{G,trans} = 0.0831$  and  $0.1209$ . Both the transitional holdup and transitional gas velocity increase while increasing the NaCl concentration, till the critical ratio. This support the hypothesis that increasing the NaCl concentration, the homogeneous flow regime is stabilized and, thus, the gas holdup increase by the inhibited coalescence.

The data obtained from the optical probe were used to study the local flow characteristics at different radial positions. The optical probe data for  $c/c_t = 0$ ,  $c/c_t = 0.48$  and  $c/c_t = 1.17$  were compared. There is no remarkable difference between the local void fractions, bubbly frequencies and bubble rise velocities between the three cases. However, the bubble Sauter mean diameters increase when considering the cases with NaCl: this is due to the change in the bubble shape, caused by the coalescence inhibition and the modified interaction between the bubbles.

The results - in agreement with previous publication in smaller bubble columns - have extended the existing dataset concerning the effects of electrolytes in bubble columns. Further studies may concern higher NaCl concentrations as well as the study of other surfactants. It would be interesting to study the bubble size distribution and bubble shape for different operating conditions and different NaCl concentrations by means of the image analysis previously presented by the authors [20]. Finally, the present experimental facility may be operated in the counter-current operating condition [19, 20]: the influence of the electrolyte on the counter-current operating condition will be also investigated.

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