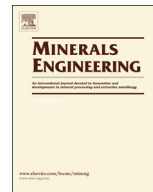




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## Salt effect on gas dispersion in flotation column – Bubble size as a function of turbulent intensity

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## ABSTRACT

The present work investigates the role of chloride salts on bubble coalescence properties by average bubble diameter measurement. The tested chloride salts (NaCl, MgCl<sub>2</sub> and CaCl<sub>2</sub>) were dissolved in fresh running water and used as feeding of a three-meter high pilot flotation column. The column was operated in close circuit while the superficial gas rate ( $J_g$ ) was varied. Gas holdup in the collection zone of the column was measured using pressure sensors. Drift flux model is employed to obtain the mean bubble diameter between two pressure-tapping points. The change of cations of chloride salts has an indisputable effect on bubble coalescence; hence the bubble size in the column varies as a function of chloride salt type. The hydrodynamics factors on bubble coalescence and difference in inhibition behavior of monovalent (NaCl) and divalent cations (CaCl<sub>2</sub> and MgCl<sub>2</sub>) based on the partition of the different hydrated ions at the gas-liquid interface were highlighted. The characterisation of the liquid phase used for flotation separation should not only focus on the concentration of salts but also on the ion-water, ion-ion and water-water properties as a function of hydrodynamic condition.

## 1. Introduction

The use of process water of industrial residual brines with high inorganic salt concentration is getting more frequent in flotation plants. The residual brines from soda-ash process has a high Mg:Ca ratio that is generally inferior or equivalent to 1:10 in liquid fraction. The carbonation of brine solutions containing fly ashes from coal combustion has been proved to have a great potential for CO<sub>2</sub> sequestration (Soong et al., 2004). Previous studies have already shown the possibility of using residues of the ammonia-soda process for CO<sub>2</sub> sequestration producing high reactive calcium carbonate (Filippova et al., 2013). Competition between ions present in solution can influence the nature and size distribution of precipitated carbonates: a high concentration in Ca<sup>2+</sup> favors calcite formation. Contrary to Ca, the ions of Mg stabilize amorphous, unstable and hydrated phases (Filippov et al., 2013) and then decrease the amount of well-crystallized carbonates. Magnesium incorporation also inhibits carbonate growth and seems to change their morphology, from rhombohedral to spindle-like shape (Davis et al., 2000). This phenomena will impact the efficiency of froth flotation intended before landfilling of neutralized liquid brine by affecting the bubble-particle collision/attachment phenomena, the bubble-bubble collision/coalescence, and consequently the hydrodynamic condition in

the flotation machine (Filippov et al., 2014, 2015).

The electrolytes in salt solutions can inhibit bubble coalescence resulting decrease in bubble size that determines the carrying capacity of flotation machines. Not every salt inhibit coalescence some has simply no effect on bubble interaction. Inhibition occurs only upon the matching combination of anion and cation. The mechanism by which a bubble is stabilized against coalescence is a dynamic process influenced by the location of both the anion and cations within the interface and subsurface that ultimately is strongly influenced by ion hydration (Parsons et al., 2010; Weissenborn and Pugh, 1995). Thus ion hydration (but not the hydration force) is critical to specific ion effects in bubble coalescence, as it strongly influences the positioning of ions in the subsurface region which ultimately determines whether coalescence is inhibited (Craig, 2011).

In the present work the effect of cations on bubble coalescence was studied in order to understand the change of bubble-bubble interactions in brines. The hypothesis of the study was that the relative size and hydration of the cations affect the structure of subsurface around the air bubbles that would change the properties of bubbles against hydrodynamic stresses and that would lead to differences in bubble coalescence.

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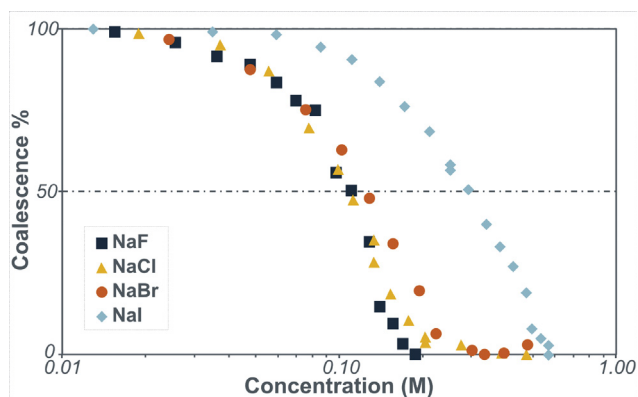


Fig. 1. Bubble coalescence inhibition in sodium halides [based on (Henry and Craig, 2010)].

## 2. Background theory

Craig et al. (1993a,b) grouped the anions and cations into two categories,  $\alpha$  and  $\beta$ , based on their effect on bubble coalescence. While  $\alpha\alpha$  and  $\beta\beta$  electrolytes inhibit coalescence, the mixed combination of the two type electrolytes,  $\alpha\beta$  and  $\beta\alpha$ , has no effect. The designations  $\alpha$  and  $\beta$  are empirical based on the results of bubble coalescence experiments and they have not been successfully associated with any physical parameter of the ion. They do not correlate with solution surface tension (Craig et al., 1993a,b), ion size or charge (Marčelja, 2006), ion polarizability or geometry (Henry and Craig, 2010), Gibbs-Marangoni elasticity (Henry et al., 2007; Marčelja, 2006; Pollock, 1912), or hydration force (Craig, 2011).

Even though the  $\alpha\alpha$  and  $\beta\beta$  electrolytes inhibit bubble coalescence, their effect on the extent of coalescence prevention differs. In pure water, 100% coalescence occurs and above a certain electrolyte concentration the coalescence is inhibited (0% coalescence). Fig. 1 shows the effect of  $\alpha\alpha$  alkali metal halides on bubble coalescence inhibition. With increasing size of halide ions the salt concentration required for coalescence inhibition increases too (Henry and Craig, 2010). The change from coalescence to non-coalescence is not happening at a certain concentration but during a wide concentration range. The change from non-coalescence to coalescence is called transition zone while the concentration where only 50% coalescence occurs is the transition concentration (Kracht and Rebolledo, 2013; Nguyen et al., 2012). Kracht and Rebolledo (2013) studied the bubble coalescence property in electrolyte solution as a function of gas flow rate. The higher the gas flow rates the greater the hydrodynamic force around the bubbles during the collision. The results showed that with increasing gas flow the required salt concentration for coalescence inhibition increases too and in case of certain salts the extend of transition zone increased too.

The presence of electrolyte affects the microscopic structure and consequently the dynamic of the air/liquid interface. Jungwirth and Tobias (Jungwirth and Tobias, 2002) used molecular dynamic (MD) simulation to predict structure, dynamic, and thermodynamic properties of air/liquid interface in the presence of  $\alpha\alpha$  alkali metal halides as a function of increasing anion radius on the molecular scale. With increasing size of halide ion the place occupied at the air/liquid interface increased and consequently more cation is attracted to the subsurface (Fig. 2). In the NaF electrolyte the fluoride concentration in the surface is reduced by 93% with respect to the subsurface, in NaCl solution the chloride concentration drops by only 29%, however in the NaBr and NaI electrolytes the concentration of anions at the interface is actually enhanced with respect to the subsurface especially in the NaI where the density profile indicates that the attracted cations are accumulated and pack in the upper layer of the subsurface. Fig. 2 highlights two really interesting ion radius related property: first – the bigger the radius of

halide ion the greater the number frequency of the ion at the air/liquid interface with respect to the subsurface and second – the smaller the radius of the halide ion the more equally distributed with the sodium at the subsurface.

The hydration level of the halide cations can explain the first property. Table 1 summarizes some physical properties of the halide ions such as: ionic radius, hydration radius, hydration number, Jones-Doyle B coefficient ( $B_{\eta}$ ), entropy change beyond the hydration shell ( $\Delta S_{\text{hydration}}$ ) and partial molal volume  $-\bar{V}^{\circ}$ , (the effect of one mole of ion on the volume of a large amount of water). The difference in hydration level is well represented by the hydration number that determines the number of water molecule associated with the ion. More indicative and sensitive measure of the hydration level is the Jones-Dole viscosity B coefficient ( $B_{\eta}$ ). For strongly hydrated ions the  $B_{\eta}$  is positive while for weakly hydrated ions it is negative. The  $B_{\eta}$  coefficient of  $F^{-}$  is positive, as it was expected base on Fig. 2, and as the  $B_{\eta}$  coefficient of halide ions decreased the closer the anions got to the air/liquid interface.

The explanation of the second property is a bit more complex and it cohere with the previously introduced hydration level of ions. Salts are build up of opposite charge of ions that relative radii size to each other can have a great effect on the hydration level of ion-pairs (Collins, 2004). It is considered that two strongly hydrated small ions (hard ions) of opposite charge or two weakly hydrated large ions (soft ions) of opposite charge would strongly attract to each other forming direct ion pairs that expelling the hydration water between them. Collins (2004) named these direct ion pairs to inner sphere ion pairs. However, if a salt dissolve to a hard and a soft ion these ions will not dehydrate to form an inner sphere ion pair.

The  $B_{\eta}$  coefficient is a good measure to identify which ion can be considered as hard or soft ion (Marcus, 2009). Hard ions have positive  $B_{\eta}$  coefficient and called kosmotropes while soft ions have negative  $B_{\eta}$  coefficient and it is called chaotropes. In more general language the kosmotropes and chaotropes has been replaced by structure-makers and structure-breakers respectively. These terms are referring for the effect of ion on the degree of order in the water solvent. The entropic changes ( $\Delta S_{\text{hydration}}$ ) is a good indicator to determine the degree of order in the system. The lower the  $\Delta S_{\text{hydration}}$  the more ordered the system and the “stiffer” the hydrogen-bonding network in the solvent so it is considered as structure-maker. While with higher  $\Delta S_{\text{hydration}}$  the system is less ordered so “looser” the hydrogen bonding system and it is called structure breaker (Shattuck et al., 2016).

Although the literature presents a consensus on the influence of ions (type, concentration and their combination) on bubble coalescence, there is no definitive agreement on the inhibition mechanism and the transition salt concentration. In addition to the role of classical DLVO forces (electrostatic repulsion and Wan der Waals attraction), the role of non-DLVO forces such as repulsive hydration pressure (Chan and Tsang, 2015) and attractive hydrophobic forces were analysed and modelled to define the interactions between two bubbles (Firouzi et al., 2015; Firouzi and Nguyen, 2017). It was found that the hydrophobic attraction was reduced with increased salt concentration because of the decreased solubility of gas, the change of surface tension and the rheology of the thin liquid film at the gas-water interface. These phenomena lead to a decrease in water drainage and delay the rupture of liquid films by affecting the contact time and bubble coalescence. A comparison of few models to define the transition salt concentration (Firouzi et al., 2015) and a new experimental approach (Firouzi and Nguyen, 2014) reveals that the model predictions failed when compared with experimental data. An approximation of the experimental data shows a linear correlation between the transition salt concentration (TC95) and bubble size ( $\sim R^{-0.5}$ ) in accordance with the model suggested by Prince (Prince and Blanch, 1990) in which the retarded Casimir-van der Waals attraction was used instead of the non-retarded London-van der Waals attraction. Moreover, the proposed model of

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