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INFLUENCE OF SOLUTION pH AND ELECTROLYTE PRESENCE ON BUBBLE VELOCITY IN ANIONIC SURFACTANT SOLUTIONS

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Influence of sodium n-dodecyl sulfate (SDDS) concentration, presence of electrolytes (NaCl and KCl) and the solution pH variation on profiles of the bubble local velocity and values of the bubble terminal velocity were studied. It was found that in SDDS solutions the bubble accelerated rapidly after the detachment and either a maximum was observed (low SDDS solution concentrations) followed by a period of monotonic decrease or the terminal velocity was attained immediately after the acceleration stage (high SDDS concentrations). Addition of the electrolyte and/or the pH variation caused significant diminishing the local velocity in low concentrations solutions and lowering the bubble terminal velocity to a value characteristic for SDDS solutions of high concentrations. This effect of electrolyte and pH on the bubble velocity variations was attributed to lowering surface tension of the SDDS solutions.

keywords: sodium n-dodecyl sulfate, SDDS, bubble; surface tension, adsorption coverage; velocity; fluidity of interface; adsorption kinetics; dynamic structure of adsorption layer.

INTRODUCTION

Bubble motion is an important issue in many industrial applications. As Kulkarni and Joshi (2005) underlined in their review, the gas bubbles and gas-liquid contacting are the most important and very common operations in the chemical process industry, petrochemical industry, and mineral processing. In applications such as absorption, distillation, and froth flotation, the interaction of two phases occurs through dispersing the gas into bubbles and their subsequent rise in the liquid pool. In froth flotation

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the bubble acts as a carrier of the attached particles having density greater than the continuous liquid medium (pulp). Probability of formation of the stable bubble-particle aggregate is considered (Derjaguin and Dukhin, 1960; Schimmoler et al., 1993; Ralston and Dukhin, 1999) to be a product of the probabilities of collision and attachment (formation of the three-phase contact) and the probability that detachment would not subsequently occur. The probability of the collision is determined mainly by hydrodynamic conditions of the bubbles and particles motion. As surfactant adsorption at the interface of the rising bubble can lower its terminal velocity over two-fold (Clift et al., 1978; Krzan and Małysa, 2002ab; Krzan et al., 2007), so an addition of the surfactant (frother) can affect significantly not only the collision probability but also the probability of the particle attachment due to prolongation of the contact time between the colliding bubble and particle. Various reagents are employed in mineral industry as frothers (Laskowski, 1998) to modify properties of the liquid/gas interfaces and to facilitate the flotation aggregate formation (Leja, 1982). The frother addition assures formation of the froth layer, facilitates the gas dispersion and increases the gas/liquid interfacial area, as a result of diminishing sizes of the bubbles formed, and lowers significantly velocity of the bubbles formed. The size of bubbles formed and their velocity are considered (Azgomi et al., 2007) to be the main factors determining the gas holdup in flotation column and the bubble velocity is related to its size. As smaller bubble size and velocity leads to a larger the gas holdup, so Azgomi et al. (2007) attempted to find a unique relationship among gas holdup, gas rate and bubble size, which would be independent of frother (surfactant) type. However, the attempt was unsuccessful and they found in their analysis of the bubble size and gas holdup vs. frothers concentration that for the same gas holdup, the bubble sizes for different frothers were different. A failure of this analysis is most probably related to the fact that the size of the bubbles formed and the bubble rising velocity is determined by the state of adsorption layer at the bubble surface. Identical bulk concentration does not mean that the adsorption coverage is similar.

The presence of adsorption layer and time of formation of the dynamic adsorption layer (DAL) are the parameters which affect, to a great extent, values of the local and terminal velocities of the rising bubbles (Clift et al., 1978; Sam et al., 1996; Krzan and Małysa, 2002ab; Krzan et al., 2007). In water devoid of surfactants, the bubble surface is fully mobile, and therefore, the bubble velocity is higher than that of solid sphere of identical diameter and density (Levich, 1962). Adsorption layer of surfactant molecules at the bubble surface retards fluidity of the gas/liquid interface and lowers the bubble rising velocity. Simultaneously, as a result of the viscous drag exerted by continuous medium on the bubble surface, an uneven distribution of the adsorbed surfactant molecules along the interface of the rising bubble is devel-

oped. Such adsorption layer, called the dynamic adsorption layer (DAL) (Dukhin et al., 1998), determines values of the bubble terminal velocity and profiles of the bubble local velocity (Krzan et al., 2007). Formation of the DAL means that the adsorption coverage (surface concentration) is at minimum at the upstream pole of the moving bubble, while at the rear pole is higher than the equilibrium coverage (Dukhin et al., 1995, 1998). This gradient of the surface concentration (surface tension) retards mobility of the bubble surface and consequently the bubble velocity is lowered.

Bubble formation is a dynamic process and in surfactant solutions is interrelated with adsorption of surfactant molecules at the expanding bubble surface. A degree of the adsorption coverage at the surface of the bubble formed is determined by the rate of bubble formation and kinetics of the surfactant molecules adsorption of at the expanding bubble surface (Warszynski et al., 1998b; Jachimska et al., 2001). The equilibrium adsorption coverage is attained at surface of the bubble formed and starting to rise only in the case when the adsorption is faster than the bubble surface expansion. The bubble formed accelerates immediately and at some distance from the point of its formation reaches the terminal velocity which is determined by the balance between all acting forces.

The paper presents results of studies on local and terminal velocities of the bubbles rising in n-dodecyl sulfate solutions of different concentrations, and on influence of solution pH and presence of various electrolytes on the bubble motion.

EXPERIMENTAL

A square glass column (40×40 mm) having at the bottom a capillary of inner diameter 0.075 mm was used. Bubbles were formed at the capillary orifice with the help of Cole-Parmer syringe pump enabling high precision control of the gas flow. Figure 1 presents schematically the set-up used in measurements of the bubble dimensions and velocities. A Moticam 2000 digital camera (with Nikkor f=60 objective and rings for magnification) was used to monitor and record the bubbles motion. The picture recorded was magnified approximately twenty-fold as a result of applying the rings between the objective and the camera body. To determine local velocities of the bubble at various distances from the capillary orifice a stroboscope lamp (Drelloscope) illumination with 100 flashes per second was applied. The data were next analyzed by the image analysis software SigmaScan Pro 5.0.

Sodium n-dodecyl sulfate and sodium and potassium hydroxides were used as received from Fluka and Merck. Sodium and potassium chlorides used were heated up to 550°C in order to get rid of any surfactant contaminations. The surfactant solutions were carefully prepared immediately before experiment to avoid any a long term hy-

drolysis. Distilled water (Millipore) was used for the solution preparation. The measurements were carried out at room temperature, 22 ± 1 °C.

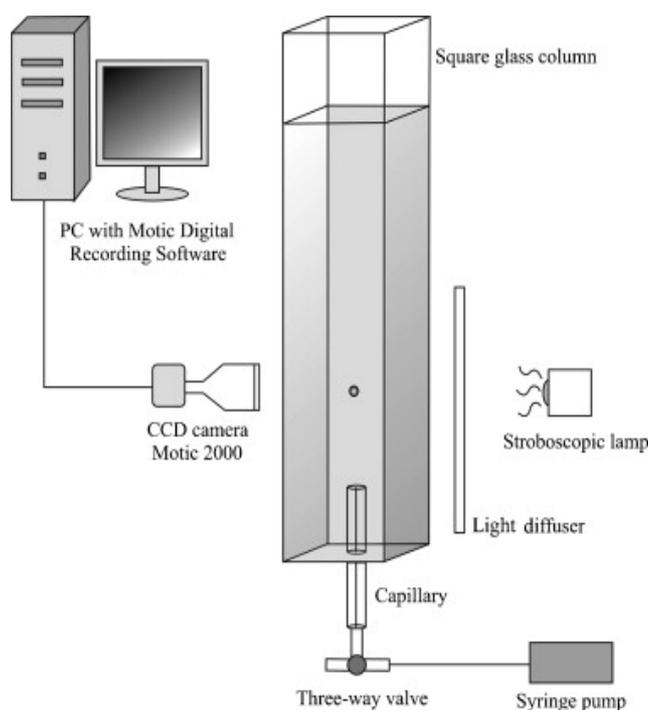


Fig. 1. The experimental set-up

RESULTS AND DISCUSSION

Local velocity profiles, i.e. variations of the bubble local velocity with distance from the point of the bubble formation, are different for pure water and surfactant solutions. Generally, in clean water there can be distinguished two stages: i) acceleration, and ii) steady state motion, when the terminal velocity is attained. In surfactant solutions an additional stage can be observed, i.e. a maximum followed by a deceleration until the terminal velocity is established (Krzan and Małysa, 2002ab; Krzan et al., 2004, Małysa et al., 2005, Krzan et al., 2007ab). The bubble terminal velocity in surfactant solutions is lower than that in clean water. The presence and the position of the maximum is a function of the surfactant concentration and both of them depend on type of the surfactant used.

Figure 2 present the images of bubbles departing from the capillary orifice at various concentrations of sodium n-dodecyl sulfate solutions. From the left, there are images showing the bubble detachment and acceleration in $1 \cdot 10^{-5}$ M, $1 \cdot 10^{-4}$ M and $3 \cdot 10^{-3}$ M SDDS solutions, respectively. As these experiments were carried out under identical stroboscopic illumination (100 flashes per second) so it can be noted immediately that the bubbles velocity was increasing with distance from the capillary and decreasing with increasing the SDDS concentration. It is seen also that the bubbles, which were spherical at the capillary orifice, underwent deformation immediately after departure and the deformation was the largest in lowest concentration. There were also observed differences in the bubble path motion at different concentrations. At concentration of $1 \cdot 10^{-5}$ M the bubbles moved up along a straight line. In $1 \cdot 10^{-4}$ M the presence of the zig-zag and later helical path of the rising bubble was observed from a distance of ca. 10 cm above the capillary. At high SDDS concentration ($3 \cdot 10^{-3}$ M) the path of the bubbles rise was again linear. These differences in the bubble path motion are related – in our opinion – to degree of immobilization of the bubble surface. The bubble interface in $1 \cdot 10^{-5}$ M SDDS solution is practically fully mobile, in $1 \cdot 10^{-4}$ M the surface is partially immobilized, and in $3 \cdot 10^{-3}$ M the bubble surface is fully immobilized – fluidity of the solution/gas interface is completely retarded.

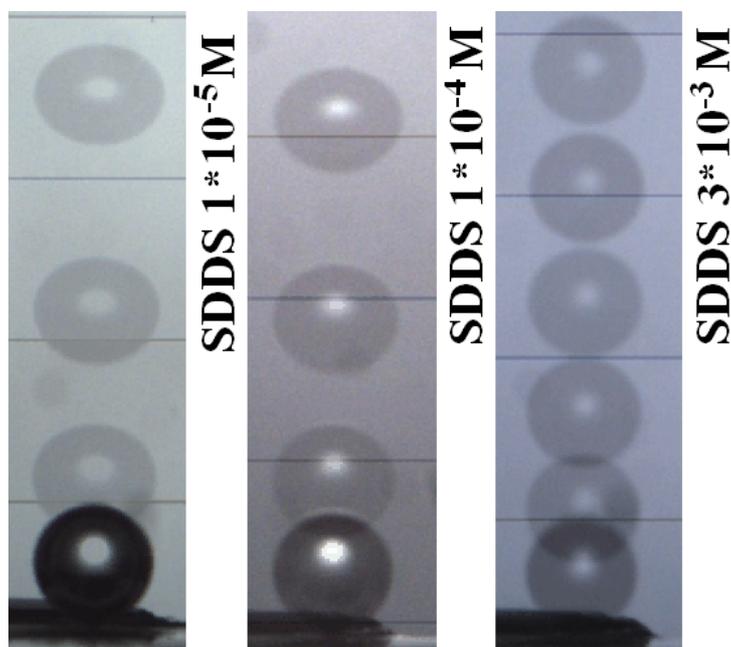


Fig. 2. Examples of images of the bubbles departing from the capillary orifice at SDDS solutions of concentration $1 \cdot 10^{-5}$, $1 \cdot 10^{-4}$ and $3 \cdot 10^{-3}$ mol dm⁻³. Strobe frequency-100 flashes per second

Figure 3 presents the results of the measurements of bubbles' local velocity as a function of the distance from the capillary in SDDS solutions. As a reference the values of bubbles' local velocities in distilled water are also presented. Generally, the variations (profiles) of the local velocities with distance show a similar feature like in the case of previously studied nonionic and cationic surfactants. Immediately after detachment, the bubble velocity increases rapidly in all the SDDS studied solutions and in distilled water. Having passed the period of rapid acceleration either a maximum was observed (low SDDS solution concentrations) followed by a period of monotonic decrease, which ended up (in the majority of cases) as a plateau (a constant value of the terminal velocity), or the terminal velocity was attained immediately after the acceleration stage (high SDDS concentrations). The height and the width of the maximum diminished with increasing concentration of the surfactants. The distance from the capillary at which the maximum velocity was observed, for a definite concentration of the surfactants, shifted closer to bubble's starting point with increasing solution concentration. As can be observed in Figure 3, bubbles' terminal velocity was decreasing steadily with increasing solution concentration, from 34.8 ± 0.3 cm/s in distilled water, down to a level of ca. 15 cm/s, which is almost identical like for all surfactants studied at their highest solution concentrations (Krzan and Malysa, 2002ab; Krzan et al., 2004, Malysa et al., 2005, Krzan et al., 2007ab).

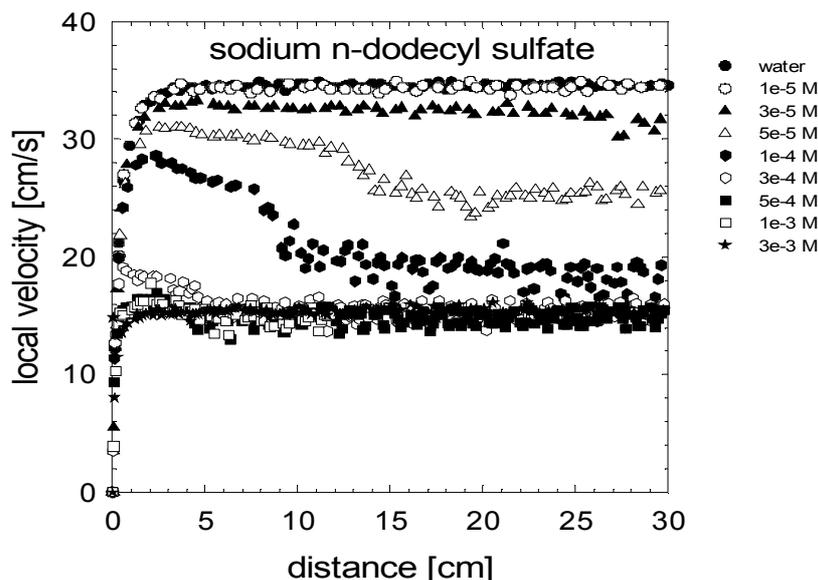


Fig. 3. Bubble local velocities in water and SDDS solutions of various concentrations

The degree of adsorption coverage and distribution of the adsorbed surfactants over surface of the rising bubble are of crucial importance for the bubble motion be-

cause there exist minimum adsorption coverage necessary for a complete immobilization of the bubble interface. The values of these minimum adsorption coverage needed to immobilize the bubble interface can be quite different and, as shown by Krzan et al. (2002ab; 2004, 2007ab), can vary from ca. 2% up to 25%, depending on the surfactant type. Figure 4 presents the dependence of the bubble terminal velocity on the degree of adsorption coverage at surface of the bubble detaching from the capillary in solutions of sodium n-dodecyl sulfate. The degree of adsorption was calculated using the adsorption kinetics model for the expanding bubble surface elaborated by Warszynski et al., (1998b) and described in details in (Jachimska et al., 2001). The model assumptions are as follows: (i) bubble grows uniformly with constant (velocity) up to its detachment from the capillary orifice, (ii) surfactant molecules are transferred to the interface by the convective-diffusion mechanism, and (iii) adsorption kinetics is described by the Frumkin-Hinshelwood model, which at equilibrium is consistent with the Frumkin adsorption isotherm. Values of parameters of the Frumkin adsorption isotherm used in the calculations were taken from the paper by Warszynski et al., (1998b). Knowing that in our experiments the time of the bubble surface expansion, i.e. the time available for the surfactant adsorption, was 1.6 s we could evaluate the degree of adsorption coverage. It was found that the adsorption coverage at the surface of the detaching bubble was closed to the equilibrium adsorption coverage in all SDDS solutions - only in lowest SDDS concentrations the coverage's at the surface of the detaching bubble were lower, but still over 80% of the equilibrium ones. As seen in Figure 4, the minimum adsorption coverage needed for a complete retardation of fluidity of the rising bubble interface was equal to ca. 10%.

The SDDS solution of concentration $1 \cdot 10^{-4}$ mole/dm³ was chosen for studies of the influence of the solution pH and the electrolyte presence on the bubble motion. At this concentration (see Figure 2) there was a distinct maximum, i.e. three different stages of the bubble motion, indicating that dynamic structure of the adsorption layer (DAL) was not established at the stage of the bubble acceleration. Moreover, at the adsorption coverage at surface of the detached bubble (ca. 2%) still did not assure a complete retardation of the bubble surface fluidity. Thus, in this solution an influence of pH and electrolyte presence on various stages of the bubble motion can be observed.

Figure 5 presents images of bubbles departing from the capillary orifice for the $1 \cdot 10^{-4}$ M SDDS solutions of different pH's; not adjusted (ca. 7), pH 2 (HCl added) and pH 11 (NaOH added), respectively. As these experiments were carried out under stroboscopic illumination of 100 flashes per second, it can be clearly seen that the pH variation had a strong impact on the bubble acceleration and velocity immediately after its detachment from the capillary – the bubble acceleration was much smaller. An addition of the electrolyte had a similar effect on the bubble acceleration and initial velocity.

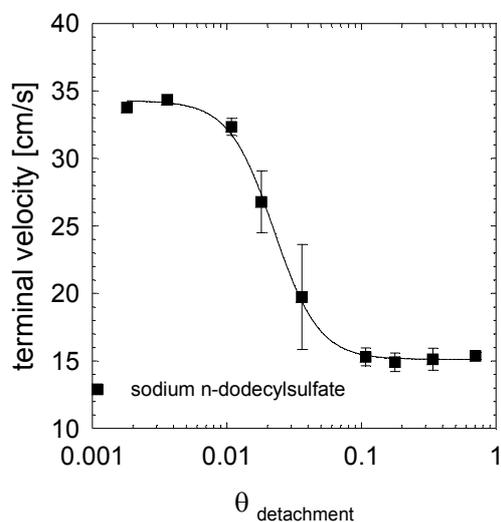


Fig. 4. Bubble terminal velocity as a function of the adsorption coverage degree over surface of the bubbles detaching from the capillary in sodium n-dodecyl sulfate solutions (adsorption time = 1.6 s)

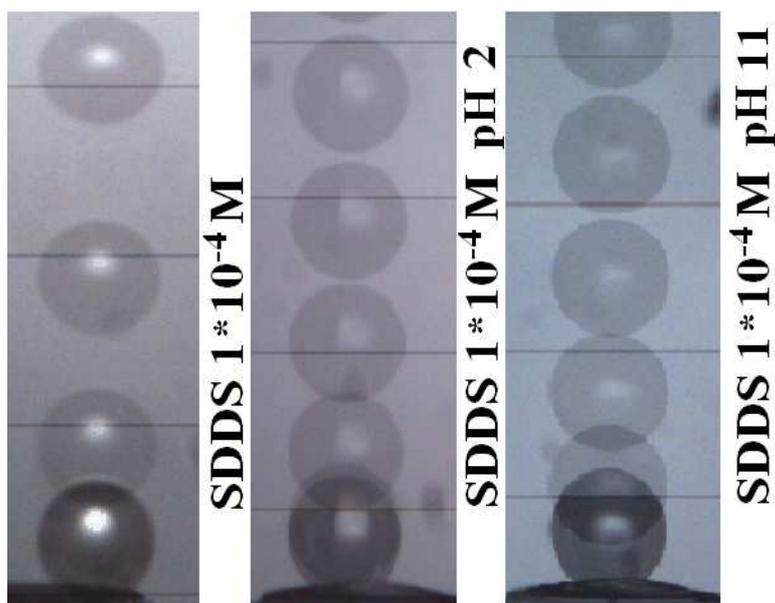


Fig. 5. Images of bubbles departing from the capillary orifice in $1 \cdot 10^{-4}$ mole/dm³ SDDS solutions of different pH's; not adjusted (ca. 7), pH 2 (HCl added) and pH 11 (NaOH added), respectively. Strobe frequency-100 flashes per second

Figures 6A–D present the quantitative data on influence of electrolyte (NaCl or KCl) and solution pH on profiles of the bubble local velocity and the terminal velocity values. To check if there is any specific effect of the type of the electrolyte cation, the potassium and sodium chlorides were used and the alkaline pH of the solutions was adjusted by applying either NaOH or KOH. As seen in Figures 6A–D, without any additives in $1 \cdot 10^{-4}$ M SDDS solutions, the period of rapid acceleration is followed by a stage of velocity decrease, preceding an attainment of the terminal velocity equal to 17–18 cm/s. An addition of the electrolyte and/or the pH variation had a drastic influence on the bubble velocity profiles. As can be observed in Figures 6A–D, the maxima disappeared and the terminal velocity stage was achieved immediately after the acceleration period. Moreover, the terminal velocity was lowered to ca. 14–15 cm/s and was attained already at distance of 2–3 cm from the capillary orifice. A small maximum can be noted only in solutions at pH=11 (Figures 6A and 6B) without any electrolyte added.

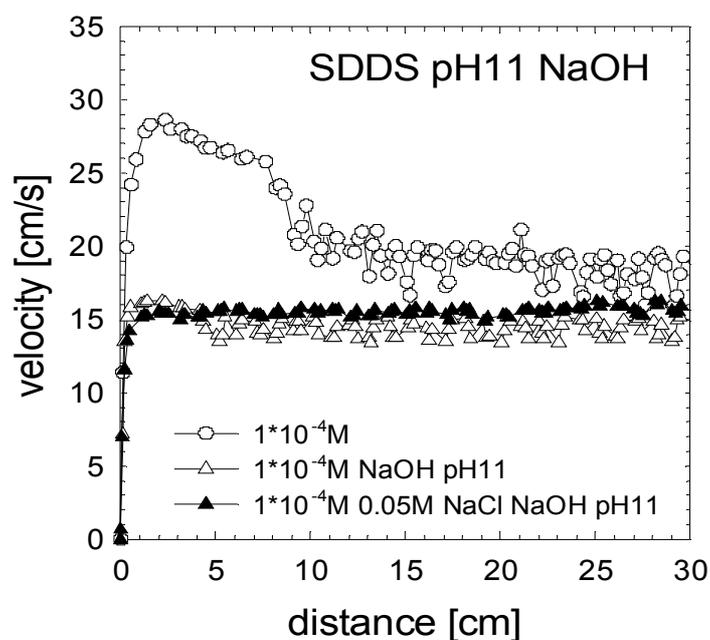


Fig. 6A. Profiles of the bubble local velocity in $1 \cdot 10^{-4}$ M SDDS solutions of pH=11 (NaOH adjusted), without and with 0.05 M NaCl added. The velocity profile in $1 \cdot 10^{-4}$ M SDDS solutions without any additives is given as the reference

In our opinion the influence of electrolyte and pH on the bubble velocity variations is related to the effect of electrolytes on surface tension of ionic surfactant solutions. When the ionic surfactant molecules are adsorbed, the electrical double layer is built and, as a result, an energetic barrier of electrostatic origin appears. It is known that

adsorption of ionic surfactants is strongly influenced by the electrical double layer arising due to surfactants ion adsorption and repulsive interactions between the adsorbed ionic molecules within the adsorption layer. Warszynski et al., (1998a) showed and developed a theoretical model to describe the influence of electrolyte on the surface tension of ionic surfactants. There are presented experimental data for a broad range of NaCl concentrations (0–1M NaCl) showing that addition of NaCl can shift the SDDS surface tension isotherm by more than order of magnitude towards lower concentrations. This effect of lowering of surface tension of ionic surfactant solutions is due to neutralization of the surface charge of adsorbed ionic surfactant molecules by counterions adsorbed in the Stern layer (Warszynski et al., 1998a). The counterion adsorption causes that interface is almost “neutral” for the adsorbing surfactant, and thus, the surface activity of the ionic surfactant almost approaches the surface activity of nonionic surfactant with the same carbon chain length. In other words the electrolyte presence means that at identical bulk concentration the SDDS solution surface tension is lowered. Lower surface tension means higher SDDS adsorption coverage over interface of the rising bubble and higher retardation of the bubble surface fluidity. As a result the bubble local and terminal velocities are lowered in electrolyte presence.

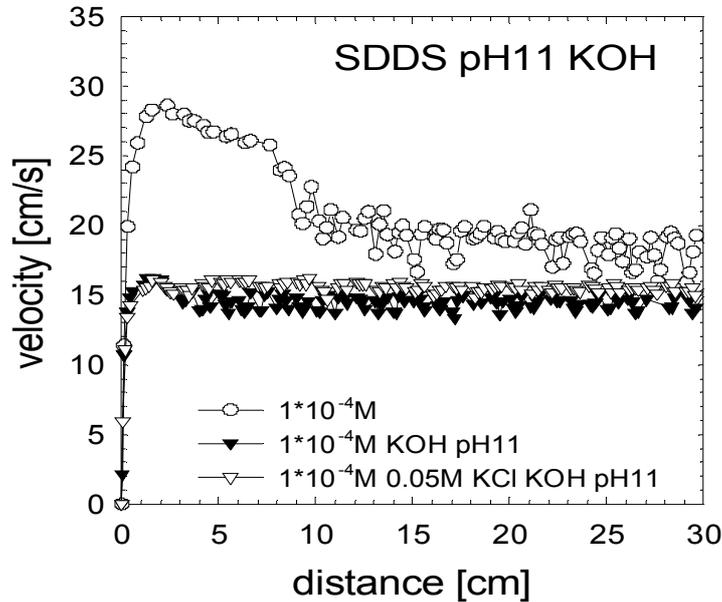


Fig. 6B. Profiles of the bubble local velocity in $1 \cdot 10^{-4}$ M SDDS solutions of pH=11 (KOH adjusted), without and with 0.05 M KCl added. The velocity profile in $1 \cdot 10^{-4}$ M SDDS solutions without any additives is given as reference

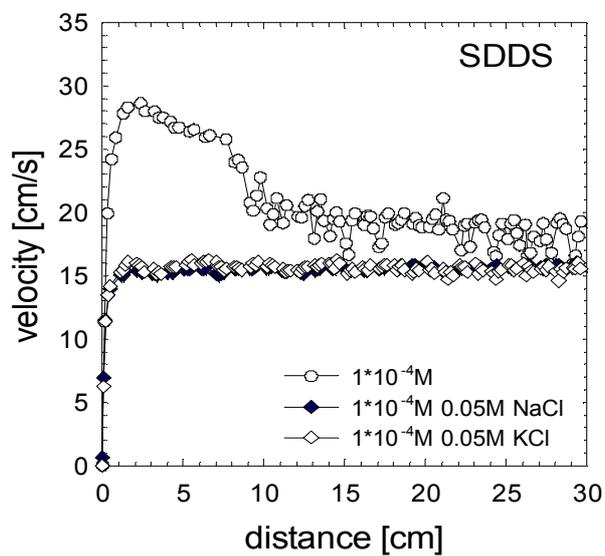


Fig. 6C. Profiles of the bubble local velocity in $1 \cdot 10^{-4} \text{ M}$ SDDS with 0.05 M NaCl or 0.05 M KOH added. The velocity profile in $1 \cdot 10^{-4} \text{ M}$ SDDS solutions without any additives is given as the reference

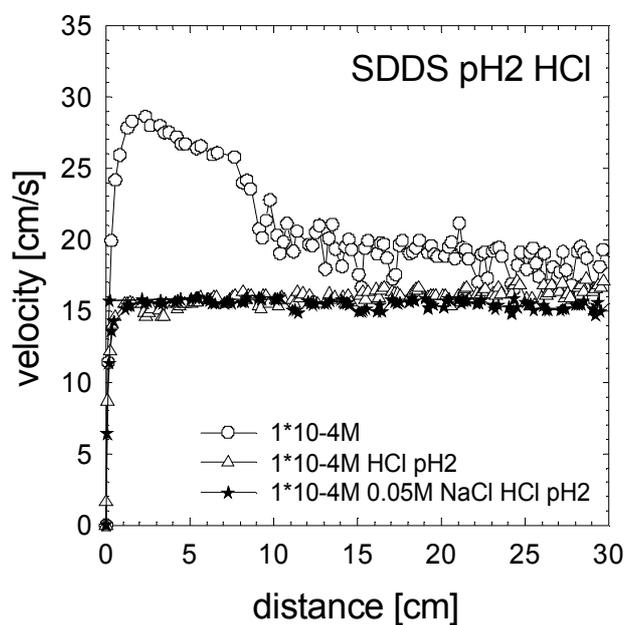


Fig. 6D. Profiles of the bubble local velocity in $1 \cdot 10^{-4} \text{ M}$ SDDS solutions of pH=2 (HCl adjusted), with and without 0.05 M NaCl added. The velocity profile in $1 \cdot 10^{-4} \text{ M}$ SDDS solutions without any additives is given as the reference

To confirm that the influence of pH and electrolyte on the bubble motion is due to lowering surface tension of the SDDS solutions, the sizes of bubbles formed were measured and collected in Table 1. Size of the bubble formed at the capillary orifice under equilibrium or quasi-equilibrium conditions is given by Tate law (Adamson, 1990):

$$r_b = \sqrt[3]{\frac{3 \sigma_k}{2 \Delta \rho g}}$$

where r_b is the bubble radius, d_0 is the capillary orifice diameter, σ is the solution surface tension, $\Delta\rho$ is the density difference between air and water and g is the gravity acceleration. As showed elsewhere (Krzan et al. 2002a) the sizes of the detaching bubbles were in good agreement with that calculated from the Tate law. Thus, from the measured size of the detaching bubbles the solution surface tension can be evaluated. As can be clearly seen in Table 1, the bubble diameter decreased from 1.45 mm in the SDDS solution without any additives, down to ca. 1.39–1.40 mm in the presence of NaCl, NaCl/NaOH, KCl, KCl/KOH or HCl, NaCl/HCl, KCl/HCl. In the case of experiments with addition of NaOH or KOH, the bubble diameters were 1.42–1.43 mm. Using the Tate law the surface tension values reflecting these variations of the bubble diameter were calculated and are listed in Table 1. When in $1 \cdot 10^{-4}$ M SDDS solution without any electrolyte the surface tension was 72 mN/m, then it decreased to ca. 62.5 mN/m for the bubble diameter of 1.39mm (solution pH=2). An addition of 0.05 M of NaCl or KCl caused the bubble diameter to diminished from 1.45 to 1.40 mm and its is equivalent to the surface tension decrease from 72 to ca. 64.5 mN/m. It is worthy to add here that these evaluations of the surface tension variations with electrolyte addition are in reasonable agreement with data published by Warszynski et al. (1998a).

Table 1. Variations of the bubble diameter in $1 \cdot 10^{-4}$ M SDDS solutions without and with electrolyte addition and at different pH's. Diameter of the capillary orifice – 0.075mm

SDDS $1 \cdot 10^{-4}$ M with addition of:	bubble diameter [mm]	surface tension [mN/m] (calculated from the Tate law)
-	1.45 ± 0.01	72.0 ± 0.5
0.05 M NaCl	1.40 ± 0.01	64.5 ± 1.0
0.05 M NaCl NaOH pH 11	1.41 ± 0.01	65.5 ± 1.0
0.05 M NaCl HCl pH 2	1.40 ± 0.01	64.5 ± 1.0
NaOH pH 11	1.43 ± 0.01	68.5 ± 0.8
HCl pH 2	1.39 ± 0.01	62.5 ± 1.0
0.05M KCl	1.40 ± 0.01	64.5 ± 1.0
0.05 M KCl KOH pH 11	1.40 ± 0.01	64.5 ± 1.0
0.05 M KCl HCl pH 2	1.40 ± 0.01	64.5 ± 1.0
NaOH pH 11	1.43 ± 0.01	68.5 ± 0.8

CONCLUSIONS

Concentration of sodium n-dodecyl sulfate (SDDS), presence of electrolyte and the solution pH variation affect significantly bubble motion parameters. The bubble terminal velocity was decreasing with increasing SDDS concentration, from 34.8 ± 0.3 cm/s in distilled water to ca. 15 cm/s at high SDDS concentrations. Lowering the bubble terminal velocity to 15 cm/s means that fluidity of the bubble surface is completely retarded due to formation of the SDDS dynamic adsorption layer. It was found that the minimum SDDS adsorption coverage needed for a complete retardation of fluidity of the rising bubble interface was equal to ca. 10%.

The bubbles detached from the capillary accelerated rapidly in the SDDS solutions and either a maximum was observed (at low SDDS solution concentrations) followed by a period of monotonic decrease or the terminal velocity was attained immediately after the acceleration stage (at high SDDS concentrations). The height and the width of the maximum diminished with increasing SDDS concentration.

An addition of the electrolyte and/or the pH variation caused significant diminishing of the bubble velocity in the SDDS solutions. It was shown that the influence of electrolyte and pH on the bubble velocity variation was caused by lowering the surface tension of the SDDS solutions. The electrolyte presence means that at an identical SDDS bulk concentration the solution surface tension is lowered. A lower surface tension means a greater SDDS adsorption coverage over the interface of rising bubble and a greater retardation of the bubble surface fluidity. Therefore, the bubble local and terminal velocities were lowered in the electrolyte presence.

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Krzan M., Małyśa K., *Wpływ pH roztworu oraz obecności elektrolitów na prędkości pęcherzyków gazowych w roztworach wodnych anionowych związków powierzchniowo aktywnych* Physicochemical Problems of Mineral Processing, 43 (2009), 43–58 (w jęz. ang)

Wyznaczono profile prędkości lokalnych oraz zmiany prędkości granicznych pęcherzyków powietrza w roztworach n-dodecylosiarczanu sodu (SDDS) o różnych stężeniach, w obecności różnych elektrolitów (NaCl i KCl) oraz przy różnych pH roztworów. Stwierdzono, że przy niskich stężeniach SDDS pęcherzyki po oderwaniu od kapilary zaczynają wypływać z dużym przyspieszeniem początkowym i osiągają maksimum, po którym występuje etap zmniejszenia prędkości, aż do osiągnięcia stałej wartości granicznej. W

wysokich stężeniach przyspieszenie było niższe, a prędkości graniczne ustalały się bezpośrednio po etapie przyspieszenia – nie występowało maksimum. Stwierdzono, że dodatek elektrolitów do badanych roztworów oraz zmiany pH roztworów powodują drastyczne zmniejszenie prędkości lokalnych przy niskich stężeniach SDDS i obniżenie prędkości granicznej aż do wartości charakterystycznej dla wysokich stężeń SDDS. Zjawisko to związane było jest ze znacznym obniżeniem napięcia powierzchniowego w roztworach po dodaniu elektrolitu oraz po zmianie pH.

słowa kluczowe: n-dodecylosiarczan sodowy SDDS, pęcherzyk gazowy, napięcie powierzchniowe, pokrycie adsorpcyjne, prędkość, ruchliwość powierzchni, kinetyka adsorpcji, dynamiczna struktura warstwy adsorpcyjnej.

