



## Dynamics of rear stagnant cap formation at the surface of rising bubbles in surfactant solutions at large Reynolds and Marangoni numbers and for slow sorption kinetics



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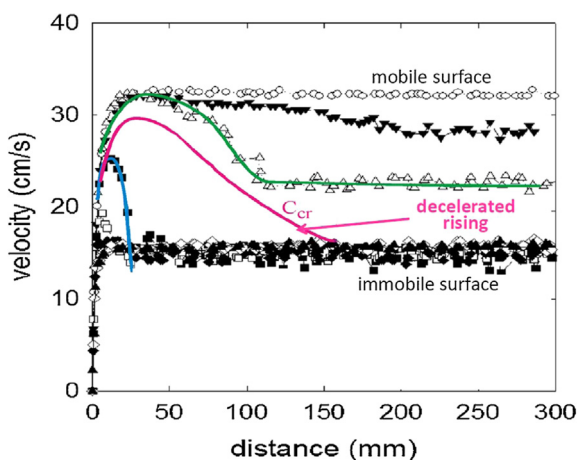
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### HIGHLIGHTS

- Based on a quasi-steady approximation, the theory for decelerated rising of bubbles of a radius of 0.4 mm is refined.
- The approach is valid for large Marangoni numbers and slow sorption kinetics.
- A method for determination of adsorption and desorption rate constants is proposed.
- The method can be applied to local velocity profiles of rising bubbles at surfactant concentrations above a critical value.

### GRAPHICAL ABSTRACT



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### ABSTRACT

In spite of the high level in the theory of steady rear stagnant caps (RSC) and its influence on steady rising, its practical application is mostly impossible because the coefficients for the adsorption and desorption rates are separately unknown. The determination of  $k_a$  and  $k_d$  separately is an actual task for the adsorption dynamics as whole. While steady RSC and steady rising retardation by surfactants are described in literature in details, only few papers are devoted to the modeling of the decelerated rising. Moreover, steady rising depends on the ratio  $k_a/k_d$  and its investigation is not helpful for the determination of  $k_d$ . In contrast a possibility to determine  $k_d$  (or  $k_a$  independently) from measurements of decelerated rising was shown by Zholkovskij et al. (2000).

However, experimental applications of this theory is difficult because of the condition  $Re < 1$ , that corresponds to small bubbles which surface is immobilized by impurities even in super clean water. This constraint may be eliminated due to the results presented by Cuenot et al. (1997), where the modeling of decelerated rising is accomplished numerically for  $Re = 100$ . However, direct application of this research is possible for a few surfactants, corresponding to the Marangoni number  $Ma = 61$ , as assumed in this simulation work. An equation is obtained for the determination of  $k_d$

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in a broad range of large  $Ma$  numbers from measurements of decelerated rising at  $Re = 200$  (bubble radius  $400 \mu\text{m}$ ) in this work. This equation is obtained on the basis of an equation for slow adsorption kinetics, a quasi-steady approximation and an equation for surfactant accumulation derived by Zholkovskij et al. (2000) as well as due to incorporation in this theory the vorticity distribution, as calculated by Fdhila and Duineveld (1996) for  $Re = 200$ . For the determination of  $k_d$  it is sufficient to measure the time required for the onset of maximal surface retardation for the concentrations above the critical concentration, i.e. the minimum concentration required for the onset of the minimum rising velocity.

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## 1. Introduction

Rising air bubbles in solutions of surface active molecules is of importance for many applications [1]. This is true, for example for all types of flotation processes as it is one of the main elementary steps in this technology [2]. The velocity of a rising bubble is remarkably influenced by the adsorption to and desorption from the bubble surface of the surface active molecules. After a certain period of time the adsorption and desorption fluxes are balanced and a steady state of the rising velocity is reached. This dynamic state includes not only the hydrodynamic field around the rising bubble but also the adsorption layer at the bubble's surface.

Following the picture discussed by Levich [3], the surface of a rising bubble consists of two parts—a leading part which is continuously expanded and a rear part which is compressed. The leading bubble part will be filled continuously by adsorbing molecules. On the contrary on the compressed rear part of the rising bubble adsorbed surface active molecules desorb. Therefore, a concentration gradient of adsorbed molecules exists at the bubble surface, i.e. there is an increase of the adsorbed amount  $\Gamma(\theta)$  from the leading pole towards the rear bubble cap. When we define the polar angle of the rising bubble by  $\theta$  (see Fig. 1) the adsorbed amount increases from the leading pole at  $\theta = 0$  to the rear pole at  $\theta = \pi$ . As an extreme case we have the leading bubble pole free of any surface active molecule, whereas the so-called rear stagnant cap (RSC) is covered by a compressed adsorption layer, hence the surface around the leading pole is mobile while the RSC is immobile and the transition between these two surface states is characterized by the cap angle  $\psi$  (Fig. 1a).

The bubble detaching from the capillary tip rises with acceleration due to the Archimedes force. Its velocity increases, and after some time the drag force becomes equal to the Archimedes force that corresponds to the onset of an invariant terminal velocity. When the bubble rises in a surfactant solution, the accumulation of adsorbing species at the bubble continues after the drag force achieves the value of the Archimedes force. This leads to a further growth of the RSC with a concomitant increase in rising retardation and the velocity decrease (decelerated rising), i.e. to the formation of a maximum on the dependence of the velocity vs. time and distance, called local velocity profile (LVP) [4]. The bubble acceleration during the acceleration stage is much higher (far from LVP maximum) than during the deceleration stage. As result, inertial forces (memory forces [5]) are stronger during the accelerated rising, while their quantification is problematic. The quasi-steady approximation is more suitable for decelerated rising. Consequently, it was proposed in [6] to focus on decelerated rising.

Accurately measured local velocity profile of a rising bubble can provide information about the dynamic adsorption layer (DAL). In particular, information about the kinetics of adsorption and about the dependence of surface tension on solute concentration can be obtained from the LVP because they control the DAL structure. For this aim, however, a quantitative DAL theory is necessary. A serious limitation in the current state is that most of the theories are addressed so far to the steady DAL only [7–13]. However, exper-

iments provide information not only about the steady (terminal) velocity but also the transition of the velocity from zero in the moment of bubble detachment from the capillary tip to the terminal velocity. This transient process covers very often the major part of the LVP of bubble rising. Thus, a quantitative interpretation of experimental data requires the development of the theory for non-steady DAL.

The role of a DAL in micro-flotation and flotation was considered in detail in Chapters 10 and 11 of the monograph [14]. The presence of a DAL on the rising bubble surface determines the efficiency of particle-bubble interaction. The development of a rising bubble theory accounting for the DAL formation on its surface was earlier summarized in [15] and [16], in particular, the influence of the adsorption layer rheology on particle-bubble interactions was analyzed in [16].

In most experimental and practical applications (e.g. in flotation) the bubbles are rather large, therefore the theory for non-steady DAL should be extended to the case of large Reynolds numbers  $Re$ . Such generalization of the DAL theory is simplified, if we apply a quasi-steady approximation. Such an approximation is based on the assumption that the purely hydrodynamic relaxation is much faster than the adsorption equilibrium is achieved. It means that the momentary unsteady velocity is identified as a steady velocity corresponding to a momentary distribution of the solute at the bubble surface. If the bubble rising is accompanied by the formation of a RSC the velocity is determined by the amount of the solute,  $M(t)$ , accumulated at the bubble surface at the time moment  $t$ ,

$$U = U_{st} [M(t)] \quad (1)$$

where the velocity  $U_{st}$  is described by the theory of steady bubble rising with a steady DAL. This approach was proposed in [17]. As the quasi-steady approximation does not consider any non-steady hydrodynamic contributions, it is sufficient to quantify  $M(t)$ .

Considering a quasi-steady rising it is important to make sure, that this approximation may be valid for quantifying a non-steady rising at large  $Re$ . To analyze this question we can compare the regularities for steady rising [7] and for the transient stage of rising [18]. An agreement of the results in [7] with those in [18] can be established with respect to the dependence of rising velocity on the cap angle  $\psi$  (or  $\psi(t)$ ). This corresponds to the condition of Eq. (1), because  $M(t)$  is a simple function of  $\psi(t)$ , as will be shown below. Such agreement justifies the assumption about the possibility to extend the quasi-steady approximation [17] over large Reynolds numbers [6].

The balance of Marangoni and viscous stresses is characterized semi-quantitatively by the Marangoni number:

$$Ma = \frac{RT\Gamma_{\infty}}{\eta U} \quad (2)$$

where  $R$  is the gas constant,  $T$  is the absolute temperature,  $\Gamma_{\infty}$  is the maximum surface concentration,  $\eta$  is the viscosity,  $U$  is the characteristic rising velocity of the bubble. The Marangoni number  $Ma$  characterizes the surface pressure gradient that surface active

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