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# Methane bubble rise velocities under deep-sea conditions—Influence of initial shape deformation

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GRAPHICAL ABSTRACT

#### HIGHLIGHTS

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- Bubbles under deep-sea conditions rising faster than expected.
- Range of rise velocities is covered by common equations.
- Effect of immobilized bubble interface by contaminants in seawater is negligible.
- Influence of initial bubble shape deformation on rise behavior is significant.

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#### ABSTRACT

Terminal bubble rise velocities play a major role in industrial and environmental applications and the investigation of the rise velocities in different material systems and under different process conditions is of increasing interest.

With respect to deep-sea oil spills and natural gas seeps, the investigation of methane bubble rise velocities under high-pressure and low-temperature conditions is of importance. In this context, nearand far-field models have been developed that use a group of correlations which is valid for contaminated systems. With these equations the velocities of bubbles and drops are calculated with the assumption of an immobilized particle interface, due to the existence of seawater and the possibility of hydrate formation. Experimental results under deep-sea conditions are very rare and often contradicting.

To identify the physical processes which influence the rise behavior of methane bubbles under deep-sea conditions, laboratory experiments in a high-pressure vessel and under ambient conditions are conducted. Methane bubble rise velocities at  $4 \,^{\circ}$ C and  $20 \,^{\circ}$ C as well as 0.1 and 15.1 MPa are investigated in artificial seawater and demineralized water.

Contrary to expectations, our experimental results under deep-sea conditions ( $4 \circ C$ , 15.1 MPa) show a brought distribution of the rise velocities for similar volume equivalent bubble diameters.

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#### 1. Introduction

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http://dx.doi.org/10.1016/j.colsurfa.2016.01.041 0927-7757/© 2016 Elsevier B.V. All rights reserved. The investigation of single bubble rise velocities in quiescent liquids has been an important topic for several decades. The knowledge about the rise behavior of bubbles plays a major role in chemical and biochemical engineering as well as environmental

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Nomenclati	ure

$ ho_{a}$	Density of the ambient fluid (kg m <sup>-3</sup> )
$ ho_{gas}$	Density of the gas phase (kg $m^{-3}$ )
$\eta_{W}$	Dynamic viscosity of tap water (kg m <sup>-3</sup> )
$\eta_{a}$	Dynamic viscosity of the ambient fluid (kg m $^{-3}$ )
σ	Interfacial surface tension between the ambient
	fluid and the gas phase (N $m^{-1}$ )
g	Gravity constant (m s <sup>-2</sup> )
$d_{\rm eq}$	Volume-equivalent diameter of a particle (m)
$d^*$	Dimensionless volume-equivalent diameter
Ε	Sphericity
h	Travel distance (rise height) (mm)
и	Bubble rise velocity (m s <sup>-1</sup> )
$u^*$	Dimensionless bubble rise velocity
Eo	Eötvös number
Fr	Froude number
K <sub>F</sub>	Dimensionless liquid number
Мо	Morton number
Re	Reynolds number



**Fig. 1.** Scheme of rising bubbles without (a) and with contaminations (b) and the resulting velocity profiles.

processes, like wastewater treatment or global warming. Further, it is indispensable for the investigation of deep-sea oil spills as well as natural gas seeps. Here, high amounts of gas are flowing into the ocean and affecting the environment.

After the explosion of the Deepwater Horizon (DWH) oil rig in the Gulf of Mexico in 2010, oil and gas flowed in nearly 1500 m depth into the deep sea. The various attempts to close the ruptured pipe demonstrate how essential the knowledge about the behavior of the different phases (oil, gas, saltwater, hydrate) in the deep sea is. Especially the localization of underwater oil plumes and specific cleaning measures after such oil spills require a thorough understanding of the distribution of oil and gas in deep oceans. Moreover, in common near- and far-field models, rise velocities for single bubbles and droplets as well as their size distributions are major input parameters to predict the distribution of oil and gas [3,19]. In this context, especially the rise behavior of methane plays an important role, as it is the main component of natural gas.

As a consequence of the oil spill in 2010 in the Gulf of Mexico, the research group at the Hamburg University of Technology investigates the rise velocities of pure methane bubbles in artificial seawater and demineralized water (DI-water). The group is part of the Center for Integrated Modeling and Analysis of Gulf Ecosystems (C-IMAGE), which in turn is financed by the Gulf of Mexico Research Initiative (GOMRI). The objective of the investigations is to explain the different influencing parameters on methane bubble rise behavior under deep-sea conditions. Laboratory experiments show that the rise velocity of CH<sub>4</sub> bubbles in artificial seawater at 4°C and 20°C and pressures up to 15 MPa varies strongly with diameter and shape of the bubble. Due to the controversial discussions in the literature about the influencing parameters on methane bubble rise velocities, further experiments under ambient conditions in DI-water have been conducted. With respect to the complexity of hydrate formation, this process is not addressed in this article, but will be clarified in a following publication.

#### 2. Theory for rising bubbles

The rise behavior of single bubbles in quiescent liquids has been investigated for many decades. One of the fundamental works has been published by Clift et al. [8], summarizing several correlations for the description of the rise behavior of particles. The comparison of the experimental data from different research groups indicates a wide distribution of the rise velocity as function of the volume equivalent (vol.-equivalent) diameter of the particles. Clift et al. conclude that this observed distribution is caused by experimental scatter or the influence of surfactants [8].

In the following, first the main difference of rising bubbles in pure and contaminated systems is shown. Second, the commonly used correlations for pure systems are explained and third, the correlations for contaminated systems are given with respect to the prediction of the rise behavior of bubbles in seawater.

### 2.1. Main difference of rising bubbles in pure and contaminated systems

The main difference in the rise behavior of gas bubbles in quiescent pure and contaminated liquids is the immobilization of the bubble interface.

In pure systems, no surface-active substances are present to be adsorbed at the bubble interface. The high mobility of the free interface enables an internal circulation inside the bubble that reduces the friction at the interface and leads to a smaller drag coefficient and higher rise velocity. With increasing bubble diameter, the bubble shape changes from spherical to ellipsoidal with the consequence of an increased pressure drop and lower bubble rise velocity (Fig. 1a).

In contaminated systems, surface-active agents accumulate at the bubble interface causing a more or less immobile surface. This immobilization inhibits the internal circulation of the bubble and causes a drag coefficient that is comparable to a rigid particle. Consequently, the rise velocity of such bubbles is much slower than that of bubbles with the same vol.-equivalent diameter in pure systems (Fig. 1b). This behavior has been taken into account by Hadamard and Rybczynski with a correction of the drag coefficient from  $\zeta = 24/\text{Re to } \zeta = 16/\text{Re } [8]$ .

For seawater a contaminated system is most often assumed due to suspended matter or other surface active substances.

#### 2.2. Correlations used for pure systems

A commonly used method to compare bubble rise velocities in process engineering applications is the plot of the dimensionless

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