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Mass transfer between bubbles and seawater

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HIGHLIGHTS

- Experiments monitoring evolution of size of bubbles in seawater has been conducted.
- Theory on mass transfer is compared to the experimental results.
- Experiments supports a specific correlation for the mass transfer coefficient.

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ABSTRACT

Mass transfer between bubbles and seawater is an important mechanism when determining how much gas reaches the atmosphere from gas sources at the seabed. The mass transfer coefficient is a governing parameter for the phenomenon. Experiments on small bubbles in seawater have been performed where the bubble size has been monitored. The observed evolution of the bubble size has been compared with theoretical predictions of the bubble size. Based on this comparison, it is shown that mass transfer correlations for contaminated conditions is more consistent with experiments than correlations for clean conditions. It is also learned that simultaneous desorption of gases dissolved in the liquid must be accounted for.

1. Introduction

Mass transfer between bubbles and seawater is an important mechanism in subsea blowouts and seepage from natural sources at the seabed. Being able to quantify mass transfer enables assessment of how much gas is dissolved in the ocean and how much gas reaches the atmosphere. Accumulation of methane in the atmosphere contributes to global warming. Although there are uncertainties in the significance of the marine contribution to atmospheric methane, studies exist which support the importance of natural gas seeps at the seabed (Judd et al., 1997). During accidental subsea blowouts from gas wells or pipelines, the amount and composition of gas reaching the atmosphere determines the potential for fire and explosions (Olsen and Skjetne, 2016).

Mass transfer between gas bubbles and surrounding liquid depends on solubility and the ability of the liquid to transport the gas species away from the bubble surface by convection and diffusion (Leifer and Patro, 2002). The contribution from convection and diffusion can be assigned to a mass transfer coefficient. The mass transfer rate is proportional this coefficient, which thus is essential in determining the fate of rising bubbles (Leifer and Patro, 2002; McGinnis et al., 2006). It is affected by several parameters including bubble size, diffusivity and level of contamination in the liquid. This requires thorough investigations of conditions where these correlations are valid before they can be applied. Numerous correlations exist for the mass transfer coefficient (e.g. Bird et al., 1960; Clift et al., 1978; Frössling, 1938; Higbie, 1935; Hughmark, 1967; Zheng and Yapa, 2002).

In the study presented here, an objective has been to assess if some of the previously published correlations are consistent with experimental data on bubbles in seawater. Although many experiments on mass transfer from bubbles have been conducted, very few of these have been performed with seawater. Some of the experiments with fresh water are still relevant for the topic. Takemura and Yabe (1999) conducted experiments on rising CO_2 bubbles in degassed water, both clean and contaminated. They designed a camera system where the camera moved alongside the rising bubbles. Alves et al. (2005) performed experiments in a downward flowing water column where they could maintain a fixed position of air bubbles for observations with various levels of contamination. More recently (Aoki et al., 2015) measured bubble size evolution on CO2 bubbles in a vertical pipe with controlled levels of surfactants. All of these investigations conclude that mass

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Nomenclature

A	surface area (m^2)
C	coefficient
c	concentration (kg/m^3)
D	diffusivity (m^2/s)
Eo	Eotvos number
d	diameter (m)
g	constant of gravity (m^2/s)
H	Henri's constant (kg/m^3 Pa)
J	flux (kg/m^2 s)
k	mass transfer coefficient (m/s)
$\dot{m}_i$	mass transfer rate (kg/s)
P	pressure (Pa)
Re	Reynolds number
Sc	Schmidt number
μ	viscosity (Pa s)
ρ	density (kg/m^3)
σ	surface tension (Nm)

Indexes

b	bubble
D	drag
i	species
l	liquid
t	terminal

transfer varies with contamination and that mass transfer decreases with increasing contamination. Mass transfer of bubbles in seawater was studied by Rehder et al. (2002) by measuring the evolution of bubble size on large bubbles (2–8 mm) released at deep waters (>400 m) outside Monterey. In order to match these experimental results Leifer and Patro (2002) and McGinnis et al. (2006) needed to apply correlations for mass transfer coefficients for clean or partly contaminated conditions. Correlations for fully contaminated conditions were inconsistent with the experiment. Concentration of surfactants were not measured.

The study presented here focusses on smaller bubbles (<1 mm) in shallow seawaters (<80 m). Experiments were conducted in a counter-current system, similar to the experiments of Alves et al. (2005), fed by a continuous supply of seawater. Temporal evolution of bubble size was monitored over durations of up to 20 min. The theory for mass transfer on bubbles is then compared with the experimental results and various correlations for the mass transfer coefficients results are assessed. Theory, experimental setup and results from the study are described with the goal of acquiring more information regarding the mass transfer between bubbles and seawater.

2. Theory

A challenge in modelling gas-liquid mass transfer for bubbles is to choose a proper mass transfer coefficient. The mass transfer coefficient quantifies how fast species are moving across an interphase. The mass transfer coefficient typically depends on how fast the species diffuse or are convected to and from the interphase. Typically for a gas bubble in a liquid, the limiting transport rate is located on the liquid side of the interphase. Therefore, species diffusion in the liquid and convective transport to and from the boundary layer on the liquid side will determine the mass transfer coefficient. The relevant convective velocity scale for transport to and from the liquid interface is the so called slip velocity between the bubble and the bulk liquid. Mass transfer also depends on whether surfactants and other contaminants are present at the interphase. Thus, correlations for the mass transfer coefficient exist for both clean and contaminated systems. A clean system is typically distilled water. Since seawater is naturally occuring and not treated in any way, it intuitively feels like it is a contaminated system. Note that an intermediate condition known as partly contaminated is normally also introduced (see below). The theory below describes mass transfer of a species to or from a gas bubble with a surrounding liquid in clean and contaminated systems.

The concentration of dissolved gaseous species in the ocean are normally very small and thus the principles of Fickian diffusion is assumed. Since diffusion and mass transfer is much faster in gas than in liquids, we can assume that all mass transfer resistance exist on the liquid side of the gas-liquid interphase. Mass transfer of species i from a bubble to the surrounding liquid is then given by Ranz and Marshall (1952)

$$\dot{m}_i = A_b J_i = \pi d_b^2 \cdot k_i^l \cdot (c_i^{sol} - c_i^l) \quad (1)$$

Here d_b is the bubble diameter (representing surface area), k_i^l is the mass transfer coefficient for species i in the surrounding liquid, c_i^{sol} is the solubility of species i in the surrounding liquid and c_i^l is the concentration of species i in the surrounding liquid. The solubility c_i^{sol} depends on partial pressure, temperature and salinity. For an ideal gas mixture we can apply Henry's law and use (Leifer and Patro, 2002)

$$\dot{m}_i = A_b J_i = \pi d_b^2 \cdot k_i^l \cdot (H_i P_i - c_i^l) \quad (2)$$

where P_i is the partial pressure of species i, H_i is Henry's constant[1] for solubility of species i. Use of this should be limited to moderate pressures. In the comparison between experiments and theory below Eq. (2) is applied, but for deep sea conditions Eq. (1) should be applied.

The mass transfer coefficient is a governing parameter when estimating the mass transfer rate. The coefficient strongly depends on whether contaminants are present or not. This is seen in Fig. 1. Based on penetration theory, Higbie (1935) derived a correlation for mobile interphases (clean conditions). This is considered as a theoretical upper limit on the mass transfer coefficient. Contaminated water usually has surface active components known as surfactants. In seawater this is mainly polysaccharides (Sakugawa and Handa, 1985). Surfactants will immobilize the interphase. Frössling derived an expression for rigid interphases (contaminated conditions) from laminar boundary layer theory that acts as a lower limit. Several other correlations also exist, either based on more advanced theory or experimental observations, e.g. (Bird et al., 1960; Hughmark, 1967). If contaminants are present in relatively small concentrations such that fast rising bubbles (typically large bubbles) are able to shed the surrounding layer of contaminants, the term partly contaminated is used. In partly contaminated conditions, small bubbles will behave as contaminated and large bubbles as clean. The transition can be explained by a stagnant cap model (Sadhal and Johnson, 1983) which was supported by the experiments of Takemura and Yabe (1999) and Alves et al. (2005). Based on these findings, correlations for partly contaminated systems have been derived, e.g. (Clift et al., 1978). Normally only distilled water qualifies as clean conditions. Thus partly contaminated or contaminated is expected to be the condition experienced by the bubbles in seawater. Even if we narrow the condition

[1] Note that numerous definitions of Henry's constants exist since both pressure and concentration can be specified in various units.