



# A new model for hydrodynamics and mass transfer of hydrated bubble rising in deep water



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## HIGHLIGHTS

- A new model was developed to predict hydrated bubble rising in deep water.
- Synergy between hydrate shell growth, bubble hydrodynamics, and mass transfer was considered.
- A correlation of mass transfer coefficient for hydrated bubble dissolution was proposed.
- The accuracy of the model was verified by literature experimental data.
- The rising process of methane bubble in deep water was investigated.

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## ABSTRACT

Hydrate shell growth characteristics have a significant influence on the migration process of a gas bubble rising in deep water, which has raised concerns across of broad range of environmental fields including marine chemistry, global warming, and ocean carbon dioxide disposal. In this work, a new coupled model of the behaviors of hydrated bubbles is developed. This model considers the bubble hydrodynamics, gas dissolution, hydrate nucleation time, and dynamic growth of the hydrate shell. In combination with published experimental data, the present model proposes new mass transfer correlations involving hydrated bubble dissolution and water permeation across the hydrate shell to describe the effects of the bubble interface and the hydrate shell structure on the mass transfer processes. Using the proposed model, the rising process of methane bubbles released at depths of 1000–2000 m in the California coast is simulated. The simulated results show that the hydrate nucleation time decreases rapidly as the water depth increases. Furthermore, the formed hydrate shell can reduce the gas bubble rising velocity and shrinkage rate, because it alters the mechanical force balance and gas dissolution rate. This work adds further insights into quantitatively characterizing the upward migration and mass transfer of a hydrated bubble in deep water.

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## 1. Introduction

Gas hydrates are crystalline compounds comprised of hydrogen-bonded water cages and trapped gas molecules (Sloan and Koh, 2008). Hydrates can play an important role in various applications, but they also pose some key issues in the environmental field. Recently, the hydrate formation characteristics and their associated governing effects have been widely studied in a complicated multiphase flow scenario (Yang et al., 2016; Zhao et al., 2017). For gas bubbles in water-dominated systems, the

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hydrate film formed on the bubble surface can prevent direct contact between the water and gas. In this regard, the disposal of greenhouse gases such as carbon dioxide into the ocean is expected to be an effective option to mitigate global warming. Indeed, the stable storage of greenhouse gases is promoted when such gases are injected into the deep ocean (Abe et al., 2016; Mori, 1998; Ogasawara et al., 2001; Teng et al., 1996) because gas dissolution into the liquid is significantly suppressed by coverage of a hydrate shell on the gas bubble. For the light methane gas, it can not only leak from a well blowout (Topham, 1984; Yapa and Chen, 2004; Yapa et al., 2001; Zheng and Yapa, 2002; Zheng et al., 2003) or pipeline burst (Wang et al., 2017), it can also be released from sea-floor sediments due to hydrate decomposition, crustal movement, shear stress from bottom currents, and climate change (Li and Huang, 2016; McGinnis et al., 2006). Since methane has more than

## Nomenclature

$A_0$	water permeation parameter, m	$n$	hydration number
$C$	gas concentration, kg/m <sup>3</sup>	$P$	pressure, Pa
$C_D$	drag coefficient, dimensionless	$Pe$	Peclet number, dimensionless
$C_{ins}$	gas concentration at the gas/water or hydrate/water interface, kg/m <sup>3</sup>	$r$	radial distance, m
$C_\infty$	gas concentration in the sea water, kg/m <sup>3</sup>	$R$	bubble radius, m
$D$	diffusion coefficient of gas in the water, m <sup>2</sup> /s	$Re$	Reynolds number, dimensionless
$d$	bubble diameter, m	$t$	time, s
$Eo$	Eötvös number, dimensionless	$t_{nuclea}$	hydrate nucleation time, s
$f_{eq}$	gas fugacity at three-phase equilibrium condition, Pa	$T$	temperature, K
$f_g$	gas fugacity at the initial thermodynamic condition, Pa	$T_g$	temperature gradient of sea water, K/m
$F_B$	Basset force, N	$v$	bubble rising velocity, m/s
$F_D$	drag force, N	$v_r$	radial velocity, m/s
$F_G$	gravity, N	$v_\theta$	circumferential velocity, m/s
$F_m$	additional mass force, N	$w$	fracture width, m
$F_V$	buoyancy, N		
$g$	gravitational acceleration, m/s <sup>2</sup>	<i>Greek letters</i>	
$J_{comp}$	gas compression rate, kg/s	$\sigma$	interfacial tension of water/guest-fluid, N/m
$k$	mass transfer coefficient, m/s	$\theta$	water-side contact angle on the fracture wall, rad
$L$	fracture length per unit area, 1/m	$\beta$	angle, rad
$L_c$	characteristic length, m	$\delta$	thickness of hydrate shell, m
$m$	mass of a hydrated bubble comprised of gas and hydrate shell, kg	$\mu$	water viscosity, Pa·s
$\dot{m}_{diss}$	gas dissolution rate, kg/s	$\varphi$	angle between the fracture extending direction and radial direction, rad
$\dot{m}_{perm}$	permeation rate of water, kg/s	$\rho_g$	gas density, kg/m <sup>3</sup>
$M_g$	molar mass of gas, kg/mol	$\rho_h$	hydrate density, kg/m <sup>3</sup>
$M_w$	molar mass of water, kg/mol	$\rho_w$	water density, kg/m <sup>3</sup>
		$\rho_b$	average density of bubble, kg/m <sup>3</sup>

20 times the global warming potential as the same mass of carbon dioxide, its transfer from seawater to the atmosphere is expected to be a potential problem (McGinnis et al., 2006). When gas undergoes vertical transport in the form of hydrated bubbles, the hydrate growth behaviors on the bubble surface will influence the buoyancy and mass transfer rates of the composite bubble (gas and hydrate), lengthening its migration process and lifetime.

In deep ocean scenarios, the stability of the hydrate phase persists because of the low temperature and high pressure. At present, several mathematical models have been developed to describe bubble migration in deep water with reference to the intrinsic kinetics mechanism of hydrate formation (Li and Huang, 2016; Topham, 1984; Yapa and Chen, 2004; Yapa et al., 2001, 2010; Zheng and Yapa, 2002; Zheng et al., 2003). Yapa et al. (2010) conducted one of the most comprehensive works in this regard. To simulate bubble migration in a plume of oil/gas blowouts, their model considers hydrate formation and decomposition, gas dissolution, gas diffusion within the hydrate, and hydrate shell reformation. Compared to the field data (Rehder et al., 2009), the model results show that the abrupt change of the measured shrinkage rate of a gas bubble occurs because of the complete conversion of the gas bubble into a hydrate particle. However, Li and Huang (2016) have a different explanation for this phenomenon. Introducing an empirical hydrate-coated area ratio, Li and Huang's model assumes that the gas can only dissolve into the water from an exposed region of the bubble surface. The "partly coverage" characteristic of the hydrate shell was concluded to account for the change in the shrinkage rate (Li and Huang, 2016).

In laboratory and in situ experiments, however, the hydrate shell was observed to rapidly cover the entire surface of an ascending gas bubble (Brewer et al., 1998; Maini and Bishnoi, 1981; Ogasawara et al., 2001). Furthermore, coverage of a hydrate shell never prevents but only retards the gas bubble shrinkage process (Mori, 1998; Ogasawara et al., 2001; Sun et al., 2010). Although

the gas dissolution is reduced significantly once a hydrate shell forms, its process continues at an appreciable rate and dominates the hydrate decomposition rate (Abe et al., 2016; Mori and Mochizuki, 1997). Therefore, mass transfer becomes the control step for the growth of a hydrated bubble (Sun et al., 2010). Abe et al. (2016) conducted a hydrate film growth experiment. The experimental conditions, in which a water flow was imposed and there was nearly no dissolved gas in the liquid medium, were similar to the scenario of a hydrated bubble rising in deep water. Notably, the hydrate film needed only 10 s to completely cover the gas/water interface. However, due to the gas dissolution and hydrate decomposition at the water/hydrate interface, the hydrate film growth could not increase continuously. Instead, the film remained almost constant at several micrometers thick, which is consistent with the estimated value of Ogasawara et al. (2001) via bubble rising experiments. Given these findings, the previous models may not completely agree with the experimental results, in which interphase mass transfer processes, including the phase migration across the hydrate shell and gas dissolution into water, were observed to be the dominant factors for the hydrated bubble behaviors.

In this work, we develop a fully coupled model for hydrate shell growth, bubble migration, and interphase mass transfer. In the model, new correlations of hydrated bubble dissolution and water permeation in the hydrate shell are experimentally determined. Moreover, the proposed model is applied to simulate the process of a methane bubble rising in deep water.

## 2. Model development

As shown in Fig. 1, gas bubbles rising in deep water can be a complicated process, relying on the multiple interactions such as bubble hydrodynamics, mass transfer, hydrate shell growth, and change of bubble geometry.

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