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Xiaohui Sun, Zhiyuan Wang, Baojiang Sun, Litao Chen, Jianbo Zhang

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## Modeling of dynamic hydrate shell growth on bubble surface considering multiple factor interactions

Xiaohui Sun, Zhiyuan Wang, Baojiang Sun<sup>\*</sup>, Litao Chen, Jianbo Zhang

*School of Petroleum Engineering, China University of Petroleum (East China), Qingdao 266580, China*

*\*Corresponding Author. School of Petroleum Engineering, China University of Petroleum (East China), 66 Changjiang West Road, Huangdao, Qingdao, 266580, China.*

*E-mail address: [sunbj1128@126.com](mailto:sunbj1128@126.com) (B. Sun)*

### Abstract

A unified mechanistic model of hydrate shell growth on a bubble surface is developed, considering the interactions of multiple thermodynamic, hydrodynamic, and mechanical factors. In the current study, the model is utilized in an interactive system consisting of a gas bubble, hydrate shell, and liquid medium; however, it can be easily simplified or modified for exploring other similar processes of hydrate film development. The main factors considered in the model are (i) mass transfer processes within the hydrate shell including gas diffusion and water permeation; (ii) updating of the hydrate pore-throat structure that affects the resistance of water imbibition; (iii) the dynamic collapse characteristics of the hydrate shell; and (iv) the gas dissolution process of the hydrated bubble related to the bubble motion and gas unsaturation. Good agreement is observed between the model and experimental results quoted in literature. Using this model, the dynamic growth rules of the hydrate shell on quiescent and moving bubbles are investigated. The hydrate shell collapse phenomenon is verified in this study. The simulated results show that variation of the hydrate shell geometry can be a continuous hydrate formation–collapse–rebuilding process, and the hydrated bubble shrinks gradually with time. Furthermore, more rapid bubble shrinkage, thinner shell thickness, and more frequent hydrate collapse can be generated at a high gas dissolution rate. This work adds further insights into quantitatively characterizing the hydrate shell behaviors and interphase mass transfer rules of a gas bubble.

**Keywords:** hydrate shell; mass transfer; water permeation; gas dissolution; collapse; hydrodynamic

### 1. Introduction

Gas hydrates are non-stoichiometric inclusion compounds, in which light gas molecules are trapped and captured in hydrogen-bonded cages formed by water molecules [1]. Hydrate utilization has gained popularity in the fields of energy and environment, such as in energy storage, solute separation, and ocean CO<sub>2</sub> disposal [2-5]. In these chemical processes, hydrate formation occurs between gas bubbles and water, substantially changing the interphase mass transfer rates and gas bubble lifetime. However, the phase transition of gas converting to solid hydrates can pose scientific and technological challenges in many scenarios. In submarine oil and gas pipelines, the formation of hydrates at the gas–liquid interface [6-8] may block the pipeline or cause it to burst [9], which can be a significant flow assurance problem. Similarly, during a deep-water oil/gas blowout, hydrate formation on gas bubbles/liquid droplets can be a concern for oil/gas containment and affects the environmental consequence of well blowouts [10]. Furthermore, as in the case of methane gas

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