

# Modeling the micromechanical interactions between clathrate hydrate particles and water droplets with reducing liquid volume



Chenwei Liu<sup>a,b</sup>, Yuxing Li<sup>b,\*</sup>, Weiyang Wang<sup>a</sup>, Sanbao Dong<sup>a</sup>, Mingzhong Li<sup>a,\*</sup>

<sup>a</sup> College of Petroleum Engineering, China University of Petroleum, Qingdao 266580, China

<sup>b</sup> Shandong Provincial Key Laboratory of Oil and Gas Storage and Transportation, China University of Petroleum, Qingdao 266580, China

## HIGHLIGHTS

- Hydrate film/shell grows along the water-hydrocarbon interface.
- Equations were developed for describing hydrate growth on liquid bridge surface.
- Model was proposed for simulating the interactions by considering hydrate formation.
- Effects of hydrate formation rate, water volume on adhesion force were discussed.

## ARTICLE INFO

### Article history:

Received 31 August 2016

Received in revised form 30 December 2016

Accepted 12 January 2017

Available online 14 January 2017

### Keywords:

Hydrate particle

Modified pendular liquid bridge model

Capillary force

Parabolic approximation

Reducing liquid volume

## ABSTRACT

The micromechanical interactions between hydrate particles and water droplets play an important role in determining hydrate agglomeration, which is a crucial cause of hydrate blockages/bedding in deepwater gas-oil flowlines. The amount of hydrate converted from water droplet during the interaction between hydrate particles and water droplets led to the reduction in liquid volume, which would significantly affect the interaction forces. The existing classic pendular liquid bridge model with fixed liquid volume is not adequate for this unique case. In this study, a modified pendular liquid bridge model has been developed by considering the conversion of hydrate from liquid droplet. Parabolic approximation and a rupture criterion are proposed to determine the liquid bridge profile and rupture distance, respectively. On the basis of experimental observations, the capillary force model between two plates is applied to predict the interaction forces. It is found that the evolution of the profile of bridge (including the hydrate coating section and the pure liquid section) can be predicted with a satisfactory accuracy. Subsequently, it ensures the good agreement between the measured and predicted interaction forces. The profile of the liquid bridge becomes unstable just before the rupture of the liquid bridge, resulting in a large deviation between the experimental results and the model predictions. The proposed model can be successfully used for investigating the effects of different factors, such as contact area and hydrate formation rate, on the interaction behavior/forces, which could help to provide new and critical insights into the hydrate agglomeration process. The developed models contribute a significant progress in existing models development on hydrate agglomeration, and thus could provide a more accurate evaluation of hydrate formation risk in gas-oil flowlines.

© 2017 Elsevier Ltd. All rights reserved.

## 1. Introduction

Gas hydrates are solid crystalline compounds with cage-like structures of water molecules. The cavities in these structures formed by the host water are usually occupied by suitably sized

gas molecules (e.g., methane, ethane, and carbon dioxide) at high pressures and low temperatures. The crude oil production pipelines with high pressures and low temperatures could provide favorable conditions for gas hydrate formation. The accumulation and aggregation of hydrates in pipelines can lead to pipeline blockage/bedding, thereby resulting in catastrophic operational failures. As gas and oil exploration and production move to ultradeep water (>8000 ft of sea water), the risk of hydrate formation increases significantly (Sloan and Koh, 2007).

\* Corresponding authors at: No. 66 Changjiang West Road, Qingdao 266580, China.

E-mail addresses: [liuchenwei\\_upc@hotmail.com](mailto:liuchenwei_upc@hotmail.com) (C. Liu), [liyix@upc.edu.cn](mailto:liyix@upc.edu.cn) (Y. Li), [wangweiyang@upc.edu.cn](mailto:wangweiyang@upc.edu.cn) (W. Wang), [dongsanbao2007@126.com](mailto:dongsanbao2007@126.com) (S. Dong), [limingzhong\\_upc@hotmail.com](mailto:limingzhong_upc@hotmail.com) (M. Li).

Turner (2005) in collaboration with Abrahamson proposed a conceptual model (see Fig. 1) to describe hydrate plug formation in oil-dominated systems where water droplets are entrained in the oil phase. The model suggests that hydrate agglomeration is the key step toward hydrate blockages. Specifically, after acquiring a thorough insight into hydrate agglomeration process, Fidel-Dufour (2004) suggested that the interactions between hydrate particle and unconverted water droplet in pipeline play an important role in the early stages of the agglomeration. Direct measurement of the interaction behaviors/forces between hydrate particle and hydrate particle or droplet is an effective method to investigate hydrate agglomeration mechanism. Yang et al. (2004) reported initial measurements of cohesive force between tetrahydrofuran hydrate particles using a micromechanical force (MMF) apparatus. However, in order to avoid suspected ice contamination in the measurements of tetrahydrofuran hydrates particles cohesive force, in the following work of Taylor et al. (2007a) and Dieker et al. (2009a,b), cyclopentane (CyC5) hydrate was investigated as a model hydrate. Dieker et al. (2009b) measured CyC5 hydrate-hydrate interparticle cohesive force as a function of sub-cooling and the effect of crude oil was also evaluated. Crude oils with high contents of acid and asphaltene were considered as to be more likely to exhibit nonplugging tendencies in flowlines. According to the temperature dependence of hydrate cohesion obtained by Yang et al. (2004), Taylor et al. (2007a), and Dieker et al. (2009a,b), the measured interparticle forces were attributed to a capillary bridge which may be formed from a liquid layer present on hydrate surfaces. Aman et al. (2011) discussed the possible mechanisms to account for the hydrate particle adhesion force. On the basis of fundamental interparticle force theories and the quasi-liquid layer observed on ice surface, they suggested that the interparticle interaction was dominated by capillary cohesion due to the presence of “quasi-liquid layer” on hydrate particle. Furthermore, they presented a hydrate interparticle force model (Fig. 2 and Eq. (1)) depending on the interaction regime, which predicted well the experimental hydrate cohesion force. Lee et al. (2014a) compared the cohesive forces of CyC5 hydrates with and without thermodynamic inhibitors, and suggested that the mechanism for the cohesive force between hydrate particles depended on many factors, such as the amount of unconverted water, temperature, and particle roughness. Recently, Lee et al. (2014b) developed a high-pressure MMF apparatus to directly measure the cohesive forces between gas hydrate particles. The experimental results suggested that the average cohesive force of gas hydrates was about 10 times higher than that of CyC5 hydrates in CyC5 liquid. The newly developed high pressure MMF apparatus will be central to better understand the agglomeration of hydrates in multiphase flowlines.

$$F = \begin{cases} \frac{2\pi \cos(\theta_p) R^*}{1+H/2d} + \pi\gamma \sin(\varphi_p) \sin(\varphi_p + \theta_p) R^* & t \leq 30s \\ \tau_t(\pi\chi^2) & t \geq 30s \end{cases} \quad (1)$$

where  $R^*$  is the harmonic mean radius of two particles (Aman et al., 2011).

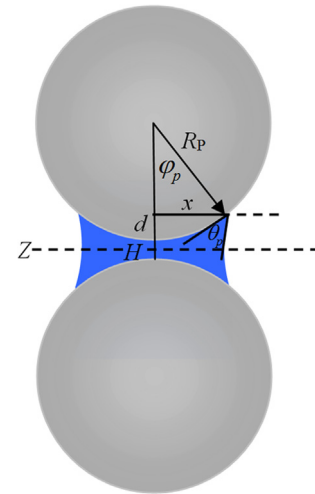


Fig. 2. Schematic of capillary cohesion for particle–particle geometries (redrawn from Aman et al. (2011)). Relevant parameters: embracing angle ( $\varphi_p$ ), contact angle ( $\theta_p$ ), liquid bridge immersion depth ( $d$ ), particle separation distance ( $H$ ), particle radius ( $R_p$ ), and the liquid bridge symmetry plane ( $Z$ ).

The aforementioned experimental and theoretical works were focused primarily on the interactions between hydrate particles. As another important interaction force, the interaction/adhesion force between hydrate particle and unconverted water droplet was also reported in recent few available literatures (Song et al., 2010a,b; Cha et al., 2013; Lee et al., 2015; Liu et al., 2015). Song et al. (2010a,b) utilized an apparatus fabricated with a microbalance and z-axis stage to study the interactions between CyC5 hydrate and water. In their work, the effects of substrate morphology and chemical additives on the initial contact force were investigated. They found that, for anionic and cationic surfactants, the increase in the concentration of the surfactants decrease in the initial contact force along with the overall adhesion energy. However, nonionic polymer inhibitors showed almost constant contact force as well as adhesion energy independent of the concentration. With the same microbalance system, Cha et al. (2013) investigated the effects of water droplet volume on the interfacial dynamic behavior between the CyC5 hydrates and water droplets. They suggested that the water volume effects on the liquid wetting of the probe and the size of capillary bridges could provide important insight into hydrate agglomeration in the presence of free water phase inside gas/oil pipelines. Lee et al. (2015) investigated the effects of salt on the crystal growth and adhesion force of clathrate hydrates and found that the presence of salt did not profoundly influence the change in the adhesion force of the hydrate probe with surfactant-free solutions, whereas it drastically reduced the adhesion force of the hydrate system with surfactant dissolving aqueous solutions. Recently, we also studied the CyC5 hydrate particle-droplet interactions using a micromechanical force (MMF) apparatus, and the effects of several fundamental factors,

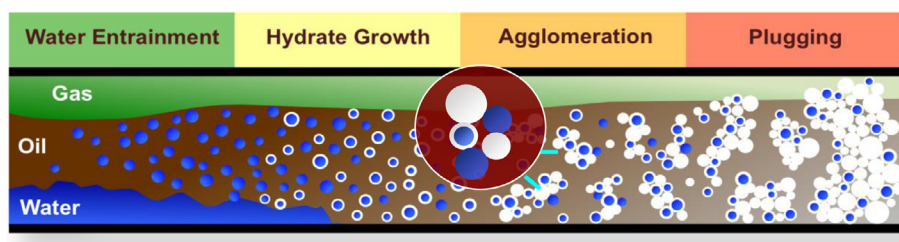


Fig. 1. Conceptual picture for hydrate formation in oil-dominated systems. (Modified from Turner (2005) & J. Abrahamson).

Download English Version:

<https://daneshyari.com/en/article/6467586>

Download Persian Version:

<https://daneshyari.com/article/6467586>

[Daneshyari.com](https://daneshyari.com)