



## Full Length Article

## Investigation on natural gas hydrate dissociation from a slurry to a water-in-oil emulsion in a high-pressure flow loop

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## ABSTRACT

Investigations of natural gas hydrate dissociation from a slurry to a water-in-oil emulsion are conducted in a high-pressure hydrate experimental flow loop equipped with on-line microcosmic instruments including a focused beam reflectance measurement (FBRM) probe and a particle video microscope (PVM) probe. Three stages of the dissociation processes are observed and sketched in a conceptual mechanistic diagram, including initial dissociation with agglomeration, dissociation with breakage and re-aggregation, and thorough dissociation. The influences of initial pressure, flow rate, and water cut on hydrate dissociation are discussed. The pressure drop is increased by hydrate particle agglomeration, which increases the risk of hydrate plugging; thus, hydrate systems require close monitoring during the hydrate slurry dissociation process. Based on the classical kinetics theory, an improved hydrate dissociation model is developed by introducing a coupled heat- and mass-transfer influence coefficient and by considering the changes in the hydrate dissociated area based on the chord length data recorded by the FBRM probe. The intrinsic rate constant for natural gas hydrate dissociation is within the range of  $1.41 \times 10^3$ – $7.18 \times 10^4 \text{ mol}\cdot\text{Pa}^{-1}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$ , and the activation energy of dissociation ranges from 68.2 to 72.3  $\text{kJ}\cdot\text{mol}^{-1}$ . The reported investigation is significant for the application of hydrate slurries in risk-management methods to address issues of hydrate flow assurance in deep-water systems.

## 1. Introduction

Gas hydrates are polycrystalline, non-stoichiometric, and clathrate-structured solids consisting of water molecules and small gas molecules such as methane, ethane, propane, and carbon dioxide, wherein water molecules form hydrogen-bonded cage structures containing guest molecules of the gas species [1–3]. As an important flow assurance issue, natural gas hydrate formation and blockages in petroleum transportation systems have attracted significant attentions from both academic and industrial communities [4,5]. Common remedies for hydrate flow assurance include traditional control methods of injecting thermodynamic inhibitors or applying thermal insulation [6,7], which guarantee operational conditions outside of the hydrate formation region. However, the cost and technical limitations of these traditional methods are increased with increasing depth for deep-water offshore petroleum exploitations [8]. A risk-management technique utilizing

cold flow and hydrate slurry has been proposed [9–11], wherein hydrates are allowed to form and made to flow with the help of anti-agglomerants (AAs) [12,13]. Significant efforts have been expended over the past decades to promote the application of this technology, including studies on the mechanisms of hydrate formation, deposition, blockage, dissociation, and remediation [14–23], the rheological properties of hydrate slurry [24–26], and the flow characteristics of hydrate slurry [27–29].

Studies on gas hydrate dissociation are important in harvesting gas from hydrate deposits [30–34] and dissociating hydrate blockages in pipeline systems [4,5,35]. However, little work has focused on the characteristics of hydrate dissociation from a slurry to a water-in-oil emulsion in an oil-dominated flow system [36]. The operation conditions of the hydrate slurry delivery system may enter the hydrate dissociation region because of the increasing environmental temperature and decreasing operational pressure in such systems [5]. Therefore, this

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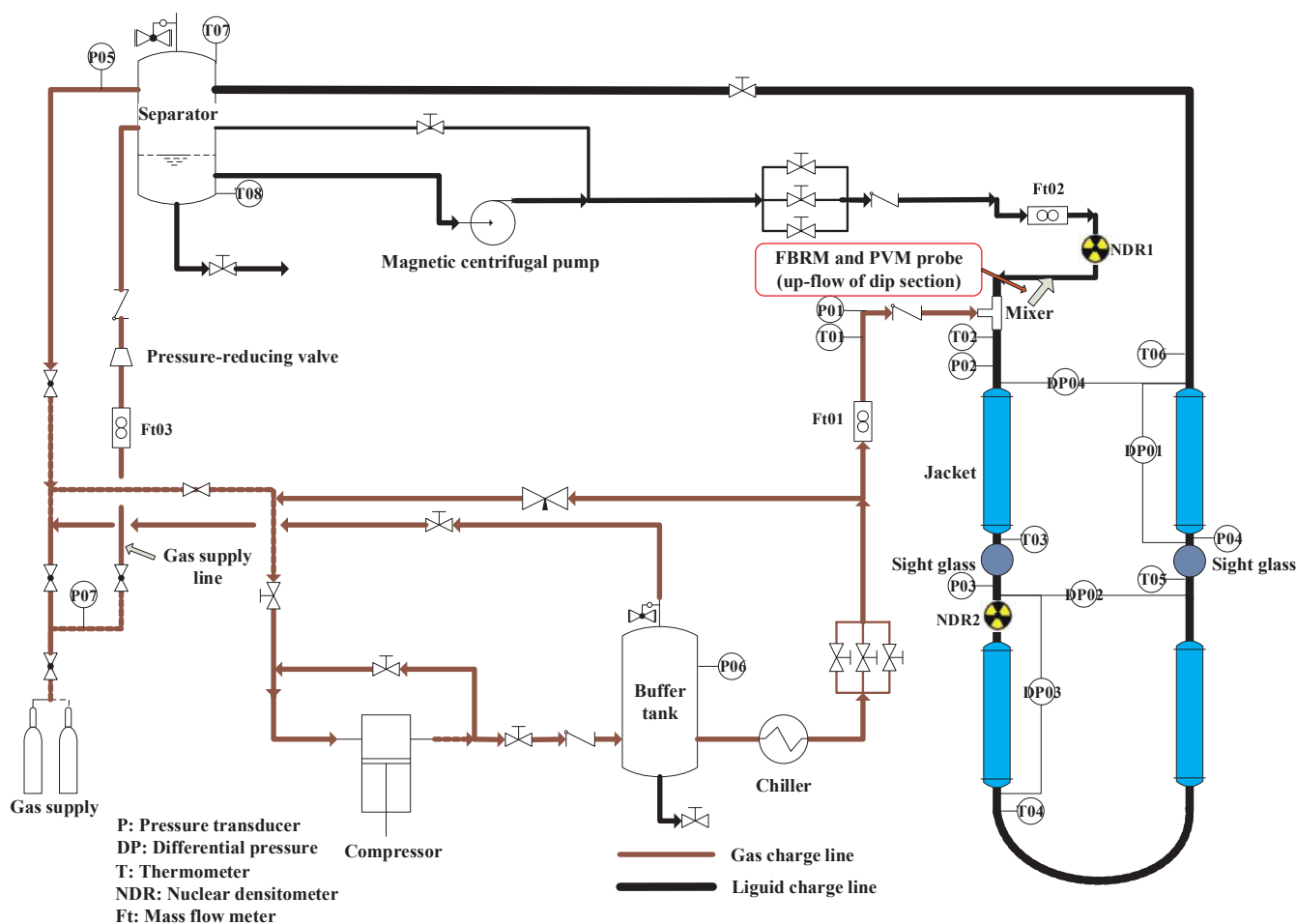


Fig. 1. Schematic of a high-pressure hydrate experimental flow loop.

study focuses on the hydrate dissociation mechanism in flow systems before applying hydrate slurry in risk-management methods to address hydrate flow assurance issues in deep-water systems. Though the results and experiments bear little resemblance to the scenarios in exploring hydrate deposits, this study is important for the application of hydrate risk management strategy in deep-water fields, which is also a valuable reference for some other production scenarios related to hydrate dissociation. As Song et al. [37] addressed that hydrate dissociation in flow system was a key study for the hydrate dissociation in the initial stage of hydrate formation and the production restart in the later stage of hydrate plug remediation. Meanwhile, it is an important reference for the exploitation of marine natural gas hydrates through solid fluidization [38], in which hydrates are transported and dissociated in a multiphase flow under different mining rates in the production system. And, the results of this study are valuable for the application of being phase change materials for CO<sub>2</sub> and THF hydrate slurry [39–41].

Recently, most studies on hydrate dissociation have focused on hydrate reservoir exploration and field production tests using different porous media or sediment systems [42–46], which provide important background information for the investigation of hydrate dissociation from a slurry to a water-in-oil emulsion in an oil-dominated flow systems [47–49]. Boxall et al. [47] and Chen et al. [48] reported on the mechanism of hydrate dissociation in stirring reactors. The hydrate dissociation rate can be enhanced by increasing the experimental temperature or decreasing the experimental pressure [50,51]. Meanwhile, the agglomeration of wetted hydrate particles in the initial hydrate dissociation stage was observed in detail by FBRM and PVM data. However, the influence of hydrate particle behaviors on hydrate dissociation has not been discussed widely. Therefore, the agglomeration

characteristics of hydrate particles during hydrate slurry dissociation are an important research subject.

After the classical hydrate dissociation kinetics model was developed by Kim et al. [52], Clark et al. [53–56] determined the intrinsic rate constants and activation energies for the dissociation of different hydrates. In these models, heat- and mass-transfer were not considered because of the high stirring rate. Sun et al. [57] modified the intrinsic kinetics model for hydrate dissociation in porous media. Liang et al. [58] developed a mathematical model to describe the kinetics of methane hydrate dissociation below the ice point based on an ice-shielding mechanism. Windmeier et al. [59–61] recognized that mass-transfer was the dominant factor near the dispersed hydrate-liquid interface during hydrate dissociation and developed the Consecutive Desorption and Melting (CDM) model. Therefore, it is necessary to improve existing models or establish a new hydrate dissociation model by considering heat-transfer [62,63], mass-transfer [64–66], intrinsic kinetics [30,67], and the changes in the dissociated surface induced by particle agglomeration [36,48].

In this work, experiments on natural gas hydrate dissociation from a slurry to a water-in-oil emulsion are conducted in a high-pressure hydrate experimental flow loop. Based on the data recorded from on-line particle analyzers (FBRM and PVM probes), three stages of the dissociation process are observed and sketched in a conceptual mechanistic diagram including initial dissociation with agglomeration, dissociation with breakage and re-aggregation, and thorough dissociation. In addition, the influences of initial pressure, flow rate, and water cut on hydrate dissociation are discussed. Based on the classical kinetics theory, an improved hydrate dissociation model is developed by introducing a coupled heat- and mass-transfer influence coefficient and

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