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The Effect of Dynamics and Mass Transfer Limitation on the Kinetic Growth Rate of Methane Gas Hydrates

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Abstract

Hydrate management aims to prevent impediments to fluid flow in pipes caused by hydrate deposition. To achieve this goal effectively, a comprehensive understanding of the thermodynamics and kinetics of hydrate formation is essential for predicting their equilibrium conditions, quantifying the amount of deposition, and estimating the associated risk. Hydrate growth studies are a relatively new field, and independent laboratory experiments have shown that hydrate growth is controlled by heat transfer, mass transfer, and its intrinsic kinetic property. At the commencement of growth, the intrinsic kinetics control the growth rate, and this mechanism gradually fades with the inclusion of mass transfer as growth progresses. This phase of transmission from intrinsic kinetic mechanism to mass transfer is not adequately explored in previous literature, and a common diffusivity constant that controls the mass transfer mechanism of gas hydrates is vital. Addressing these issues is crucial for estimating the growth rate of hydrants, which justifies this paper. Experimental data from the published literature were analyzed, and regression analysis was applied to model the behavior of methane gas consumption rates and stirring speeds during hydrate growth. Additionally, we examined how the diffusivity coefficient of methane hydrates depends on system temperature. R-squared values of 97.05% and 70.39% indicate good model fitness. The relationship between flow dynamics and intrinsic kinetics as a driving force is discussed, and threshold speeds in rpm for negligible mass transfer inclusion are presented for further methane hydrate growth studies. This paper seeks to advance the currently available knowledge in hydrate growth studies and, in a broader sense, flow assurance: risk analysis and management.

Keywords: Diffusivity, flow assurance, intrinsic kinetics, mass transfer, methane hydrates

INTRODUCTION

Gas hydrates are solid crystalline substances composed of water and gas. The water molecules exist in an ice-like cage with a variable structure, and gas molecules are trapped within this cage. They occur in extreme environments characterized by high pressure and low temperature, such as permafrost regions and deep-sea floors, and in oil and gas transportation flowlines. The crystallization of hydrates is considered stochastic and is not yet fully explained. In recent decades, there has been increased attention to gas hydrates, especially methane hydrates, largely because of the large quantities of these clathrates found on the deep-sea floor and in permafrost regions [1]. These natural gas hydrates have huge potential as an energy resource, accounting for at least twice that of all other fossil fuel deposits. Another reason for the growing interest in hydrates is that they have become a common problem in the production and transportation of oil and gas, existing as plugs in transportation pipelines. In pipelines, two sources of methane for hydrate formation exist: the free water and gas phases. Dissolved methane diffuses from the water phase, which is initially saturated with methane, to the hydrate film over time. Simultaneously, methane from the free gas phase and water from the aqueous phase diffuse through the hydrate film. All these

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processes cause pore filling and film growth. The flow assurance industry has been plagued with gas hydrate-related challenges for a long time, leading to safety issues, as well as high remedial and maintenance costs; and methods, such as heat treatment, chemical treatment, and depressurization operations, have been undertaken to control gas hydrate formation effectively. However, in addition to controlling the thermodynamic conditions of hydrate growth within the pipeline or in the subsurface, we are faced with a more challenging but important concept: managing their kinetic growth [2–5].

Existing theories describe kinetic hydrate formation as a two-step process: an initial stochastic nucleation phase, followed by a kinetic growth stage. During nucleation, transient molecular clusters and localized structuring are believed to precede the formation of stable hydrate nuclei. In contrast, the subsequent growth stage is generally governed by three principal mechanisms: intrinsic surface reaction kinetics, mass transfer limitations, and heat transfer constraints. Depending on the thermodynamic and hydrodynamic conditions of the system, one or more of these mechanisms may dominate hydrate growth behavior.

Numerous modeling approaches have been developed to represent the hydrate growth stage. Early formulations proposed multistep kinetic expressions to characterize hydrate formation rates, whereas later models integrated crystallization kinetics with gas–liquid mass transfer phenomena. Other frameworks have emphasized that hydrate growth may be primarily controlled by gas diffusion through the liquid film at the gas–liquid interface [6]. The diversity of these models reflects the complexity of hydrate formation and highlights that both nucleation and growth mechanisms require further systematic investigation (Figure 1).

This study focuses on the influence of hydrodynamic conditions—specifically, stirring or mixing rates—on methane hydrate formation, as evaluated through gas consumption measurements. Attention is given to the transition from intrinsic kinetic control to regimes increasingly influenced by mass transfer resistance. Examining the early formation stage, characterized by a constant slope in the gas consumption curve and, therefore, a constant formation rate, provides insight into the dominant growth mechanism [7]. Furthermore, evaluating the effective mass transfer diffusivity beyond this regime contributes to a clearer understanding of hydrate growth dynamics. Such an analysis is essential for addressing key challenges in hydrate-based carbon sequestration and methane recovery processes.

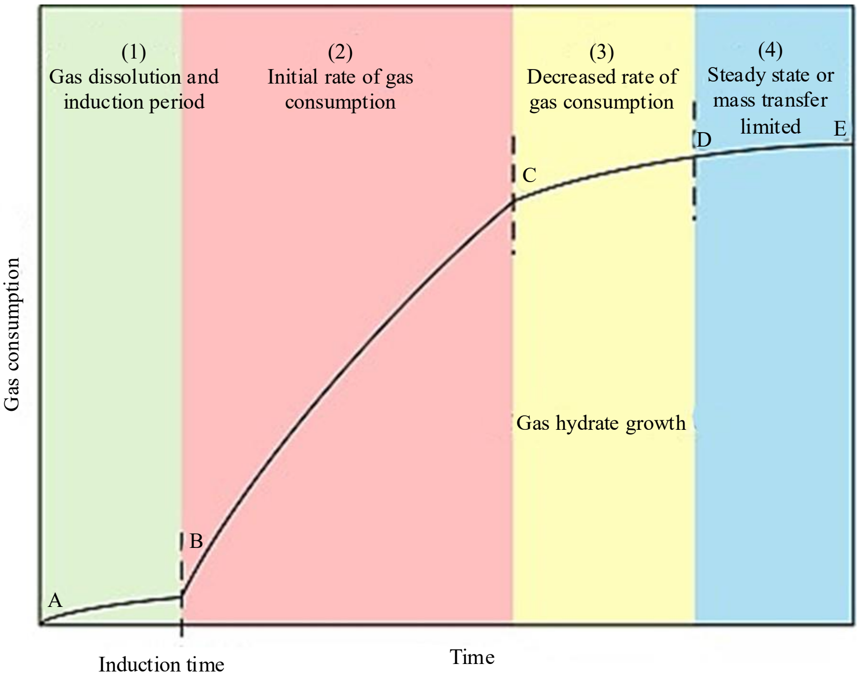

Figure 1. Typical gas uptake curve showing different hydrate growth stages [33].

Stirring Speed Versus Hydrate Growth

In stirred systems that are mostly designed to mimic pipeline turbulent flow and multiphase systems, heat and mass transfer play a significant role, whereas in systems with no heat or mass restriction, the hydrate growth rate is controlled by intrinsic kinetics.

It is common knowledge that stirring may increase reaction rates by increasing collision rates, increasing gas dispersion in water (increasing gas available for the reaction), and increasing the gas–liquid contact area (as the bubble-sized gas decreases with stirring), resulting in a more dispersed and finely mixed system. The constant initial slope stage in hydrate growth occurs over a longer period at higher stirring rates [8]. This process has been used both in industrial processes and in hydrate research studies for promoting reaction rates. It is also important to note that the system may alter the influence of stirring in place; for example, the amount of solid hydrate in a liquid/oil–hydrate type mix will differ with stirring in a gas system with hydrate presence. Stirring significantly reduces the induction time and increases the growth rate. These experiments are typically conducted at stirring speeds of 300 rpm or higher to ensure turbulent flow conditions in the liquid phase around the stirrer [9].

Experiments on stirring rates and kinetic growth of hydrates have also suggested a dependence on the hydrate growth-controlling mechanism. In a previous experimental study, the hydrate growth rate during stage I was observed to increase with increasing stirring rate. This behavior was attributed partly to the presence of pre-dissolved gas in the liquid phase, which creates a supersaturated condition prior to the onset of growth. Under such circumstances, mass transfer limitations were considered negligible during this initial stage [10–14].

Moreover, stage I is relatively short in duration, and although a rapid temperature increase occurs owing to the exothermic nature of hydrate formation, we assumed that this transient thermal effect does not significantly influence the growth kinetics. Collectively, these observations indicate that hydrate formation during growth in stage I is predominantly governed by intrinsic kinetic mechanisms rather than mass or heat transfer constraints.

During stage-II growth, mass- and heat-transfer restrictions begin to play a more significant role. A higher temperature build-up causes a drop in the temperature driving force (reducing the effect of intrinsic kinetics). In addition, the presence of hydrate crystals increases the viscosity of the mixture and impairs both mass-and heat transfer [15–17].

The experimental results of Meindinyo [18] for stirring rates between 350 rpm and 1200 rpm (at 6, 7, and 8°C) show a peak growth rate for methane gas hydrate between 500 rpm (for 6 and 8°C) and 575 rpm (for 7°C), in contrast to the experimental observations of previous investigations, in which the peak hydrate growth rate was observed at stirring speeds of approximately 400 rpm. Increasing the stirring rate beyond this value did not result in any further enhancement of the growth rate [19, 20]. This plateau behavior suggests that, above a certain rotational speed, external mass and heat transfer resistances are sufficiently minimized, and the system transitions toward intrinsic kinetic control. In other words, additional hydrodynamic intensification no longer contributes meaningfully to hydrate growth.

Similarly, kinetic studies examining methane hydrate formation across rotational speeds ranging from 300 rpm to 600 rpm demonstrated the influence of stirring on gas consumption rates. The results indicate that agitation enhances hydrate formation up to a threshold level, beyond which further increases in rotation speed provide negligible improvement in the observed growth rate [21, 22].

Their data suggest that more information on the relationship between stirring rates and methane conversion could be explored. They suggest that stirring at a rate of approximately 600 rpm leads to considerable rippling on the surface and the entrapment of bubbles. Their data indicates that the increase in methane consumption from 500 rpm to 600 rpm is significantly greater than the increase observed from 400 rpm to 500 rpm, implying a nonlinear relationship.

Diffusive Parameter in Methane Hydrate Growth

The diffusion coefficient is the proportionality factor in Fick's law. This law expresses the mass of a substance diffusing through a surface over time, which is proportional to the concentration gradient of this substance. Diffusivity is a key property that influences the stability and growth of methane hydrates. Methane hydrate diffusivity defines the rate at which methane molecules move through the hydrate structure. This property is crucial for understanding the kinetics of hydrate formation and dissociation. While factors that would normally influence the diffusion coefficient are temperature and pressure, hydrate structure, porosity, permeability, and concentration gradient, studies on the methane hydrate diffusion coefficient suggest that, unlike the intrinsic kinetic constant, the diffusion coefficient parameter of methane hydrate is variable with temperature and the type of vacancy (or fraction of unoccupied cages) [23]. This dependence has been validated through multiple experimental investigations. Stage II hydrate growth is predominantly governed by diffusion processes, characterized by the diffusion coefficient (D). During this stage, gas transport through the progressively thickening hydrate layer becomes the rate-limiting step.

Neutron diffraction studies examining methane hydrate formation in randomly packed ice have demonstrated a clear temperature dependence of hydrate growth, particularly during long-duration reactions that extend to completion. These findings indicate that, although diffusion through the hydrate layer dominates stage II, intrinsic kinetic reactions may still contribute to the overall growth behavior [24–27].

Computational approaches have also been employed to quantify methane diffusivity. Path sampling techniques that incorporate reactive flux analysis and kinetic Monte Carlo simulations have been used to derive expressions for the diffusion coefficient as a function of water-vacancy concentration at low temperatures (e.g., 225 K and 250 K). In addition, molecular dynamics simulations have been applied to investigate diffusion phenomena in methane and carbon dioxide hydrate systems, providing molecular-level insights into gas transport mechanisms within hydrate structures [28].

They estimated methane diffusion coefficients at 250 K and discussed the importance of water vacancies in creating channels for the movement of methane gas between cages. Ranieri et al. studied the fast diffusion of methane at the interface of two clathrate structures [24]. This translational diffusion was measured by quasielastic neutron scattering. 2D and 3D translational diffusion coefficient models were presented. Their fast model estimated translational diffusion coefficients between different hydrate structures in the order of 10^{-8} m^2s^{-1}, much faster than the methane diffusion coefficient for cage-to-cage hopping from other literature, which is in the order of 10^{-15} to 10^{-16} m^2s^{-1} at 250 K. In this study, we also suggest an expression for the diffusivity parameter in methane hydrates and explore the implications of these findings for flow assurance and energy production [29].

METHODS AND ANALYSIS

Stirring Rate Versus Methane Hydrate Growth

The graph of the experiment by Vysniauskas and Bishnoi [30] on the effect of stirring rate on kinetic growth, where methane hydrate consumption volumes are plotted against variable stirring speeds between 300 rpm and 600 rpm. The experiments were conducted in a semi-batch stirred reactor at temperatures between 274 K and 284 K and a pressure range of 3 MPa to 10 MPa. Data points from the 1067 stirring rates contained within the chart are taken for each consumption versus. speed plot and used to recreate a plot in Figure 2; however, a linear fitness function that expresses the relationship at different rotational speeds is introduced.

This data (in Figure 2) shows the relationship between system dynamics (speed, degree of agitation) and the formation of new methanogen hydrates (from the volumes of methanogen gas consumed).

R-squared accuracy metrics are provided for the fitness functions in Figure 2 to measure their quality. The use of R-squared statistics as a standard metric for evaluating regression analyses is widely accepted across scientific domains. It measures the squared correlation between actual outcome values and the

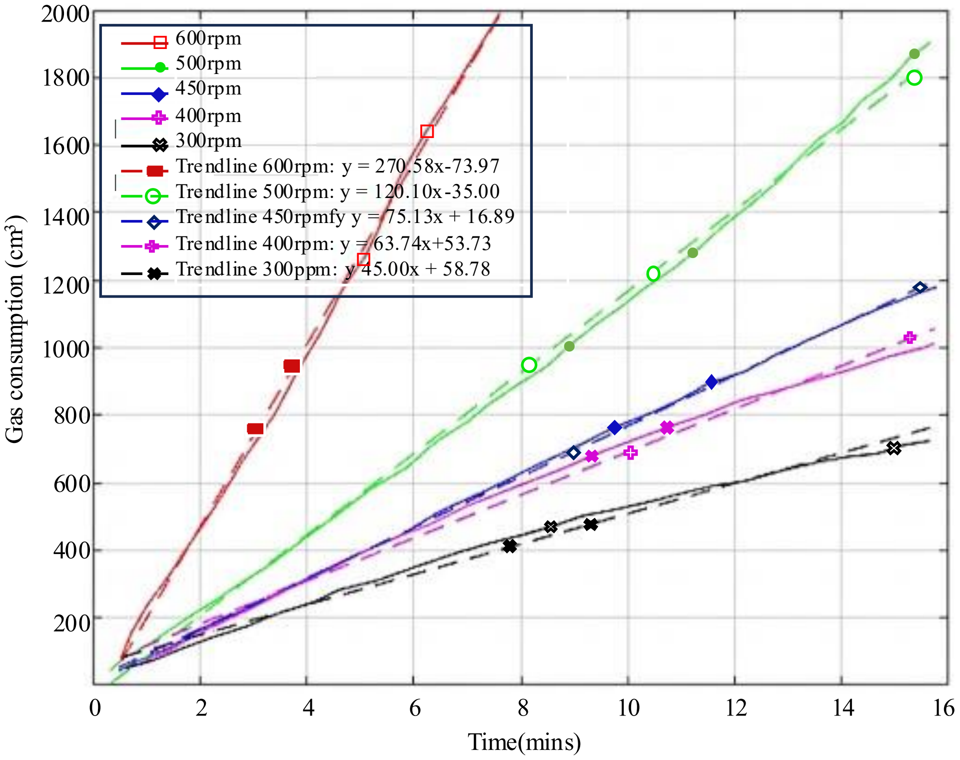

Figure 2. Plot of methane gas consumption against time for variable speeds. Adapted from Vysniauskas and Bishnoi [30].

Table 1. Equation and accuracy for the rate of growth at variable speed.

Rotation speed (rpm)	**Rate of consumption (cm^3/min)**	**R-squared value**
600	270.58x – 73.965	0.9983
500	120.19x – 34.999	0.9975
450	75.133x + 16.886	0.9993
400	63.745x + 53.731	0.9916
300	44.996x + 0.9884	0.9884

values predicted by the model. The higher the R-squared value, the better the model represents the data. R-squared can be estimated from Equation (1) as follows:

$$R-squared = \frac{Model\ sum\ of\ squares}{Total\ sum\ of\ squares} = \frac{\sum_{i=1}^{n}(\hat{y}_i-\bar{y})^2}{\sum_{i=1}^{n}(y_i-\bar{y})^2} \tag{1}$$

Where, $\hat{y}_i$ is the predicted outcome, y_i is the observed outcome and $\bar{y}$ is the average of the observed outcomes. Table 1 presents the R-squared values for the equations of each trendline in Figure 2. This R-squared value indicates a high-accuracy representation of each equation [31].

The direct relationship between each observed rotation speed and the amount of gas taken up and converted into hydrates is expressed in the second column of Table 1. This raises the question of how the consumption rates vary with changing rotation speeds. Figure 3 shows the exponential correlation between the stirring speed and gas consumption rate, as expressed in Equation (2), which also provides more information on the nonlinear behavior between the stirring speed and consumption rate.

$$r = 6.214e^{0.006v} \tag{2}$$

Where, r is the rate of gas consumption in cm^{3}min^{-1}, v is the stirring speed interpreted as the speed (or velocity), in rpm, of the fluid flowing within the pipeline in a dynamic/turbulent system.

An R2 value of 97.05% indicates good fitness of the model to the experimental data. As an example, for an intrinsic kinetic-controlled system with negligible mass and heat transfer interferences, the methane hydrate peak growth rate may be estimated from Equation (2). Elimination of mass transfer constraints and minimization of heat transfer are achieved at high stirring speeds, typically at rates with peak growth defined as the threshold rates. At rates lower than the threshold rate, there is a high probability of encroachment of the mass transfer constraint, whereas only stage-1 intrinsic kinetics and negligible heat constraints are expected to exist as the driving force at stirring speeds around threshold values. Further research on threshold speeds in hydrate growth kinetics is warranted [32].

We may assume a stirring speed threshold of 400 rpm, beyond which the methane kinetic parameter is constant, and the heat and mass resistance around the hydrate particle are eliminated. Using Equation (2), we estimate the maximum methane consumption rate.

$$r = 6.214e^{(0.006\times400)} = 68.5\ cm^3/min$$

Currently, the existing literature lacks sufficient experimental data to establish a relationship between threshold rates and the temperature of the system. However, Figure 4 shows a visual representation of the available threshold rates for methane gas hydrates in the literature [33].

Methane Hydrate Diffusivity

This study performs a regression analysis on reported values [15, 17] and models [23] for methane diffusion coefficients at selected temperatures. The models of Peters et al. [23] is given in Equation (3).

$$7\times10^{-15}X\ m^2/s \quad @250K \tag{3a}$$

$$1\times10^{-15}X\ m^2/s \quad @225K \tag{3b}$$

Where, X in Equation (3) represents the fraction of unoccupied cages or vacancies, which provide a geometric opening for the passage of methane from an occupied ‘donor’ cage to an adjacent ‘acceptor’ cage. The X range for hydrates is defined by Peters et al. [23] is $0.02 < X < 0.1$. However, methane hydrates generally have an occupancy between 95.8 and 100% [2, 17]. Therefore, we define X as 0.042 $(100 - 95.8\%)$ for a maximum vacancy in methane hydrates.

The regression analysis yielded a line of best fit. The fitness accuracy of this model was 70.39%, which is considered high for studies in the pure science field. This is because the behavior of molecules and/or particles, in this case, methane molecules diffusing through hydrate structures, can be reasonably predicted to some degree of accuracy in scientific research. A comparison of the results obtained in this study with those from previous experimental studies is shown in Figure 5, and the expression for the regression line is shown in Equation (4).

$$D = (3E - 8e^{0.0725T})\times10^{-16}\ m^2/s \tag{4}$$

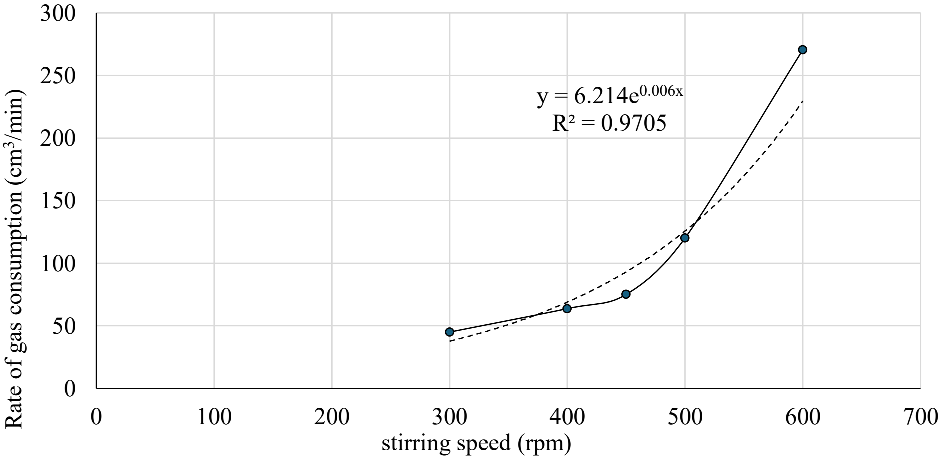

Figure 3. Effect of stirring speed on kinetic growth.

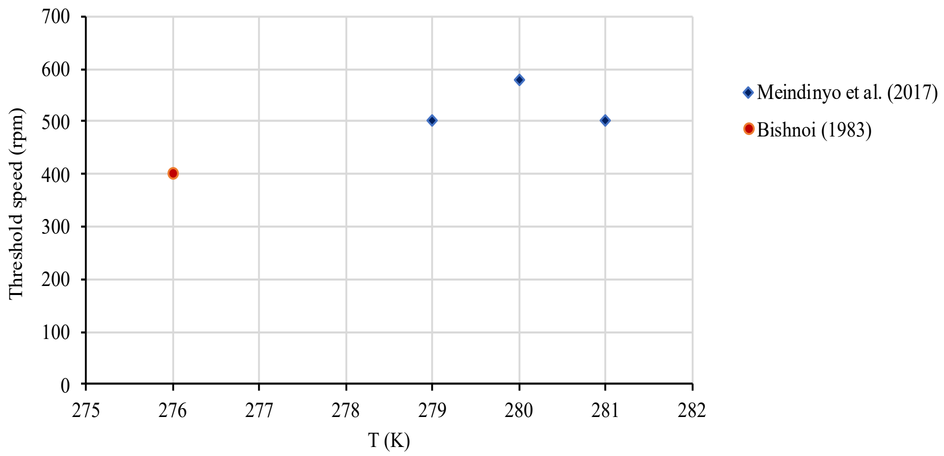

Figure 4. Threshold speeds for methane hydrate versus temperature.

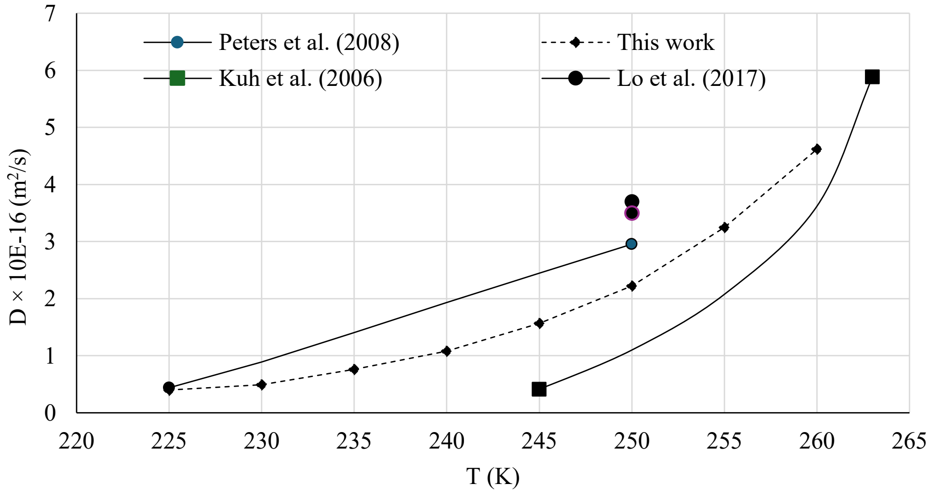

Figure 5. Comparative plot for diffusion coefficient in methane hydrates.

CONCLUSION AND RECOMMENDATION

Research on the kinetic growth of hydrates and the mechanisms that drive this growth is limited, indicating a need for further investigation. Experiments using stirrers have been conducted to simulate fluid dynamics in flowlines. Among the three widely accepted mechanisms that control growth, intrinsic kinetics can be sustained at high dynamics characterized by shear rates. This promotes the uptake of gas and the conversion of methane gas into hydrates. We developed a model to estimate the rate of methane gas uptake and its conversion into hydrates, measured in $cm^3/min/rpm$, within a stirred system. Additionally, we created a model that estimates the diffusivity coefficient as a function of temperature. While previous studies agree that the diffusion coefficient during hydrate growth is temperature-dependent, none, to the author's knowledge, have specifically defined this relationship. Furthermore, although existing studies on methane hydrates are limited, they generally support the notion that increasing system dynamics enhances growth through intrinsic kinetics. Our work represents the first attempt to define the relationship between the consumption rate and stirring speed. The data available for threshold stirring speeds, where the effects of diffusion and heat transfer are negligible, are few and conflicting, showing no trend. R-squared statistical metrics were implemented to define the fitness of the developed models, and both models yielded good correlations with the experimental data. This study could be improved by further investigating methane hydrates, especially when further experimental data become available. The application of the models developed in this work is limited to methane hydrates and reproducibility typically within the range of experimental conditions.

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