RESEARCH PAPER

Improving Gas Hydrate Formation Efficiency: Insights into Mass Transfer and Kinetics with Bio-Surfactants

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ABSTRACT

This study investigates the role of bio-surfactants in enhancing the efficiency of gas hydrate formation, with a focus on optimizing mass transfer and reaction kinetics. Gas hydrates, crucial for energy and environmental applications, often face challenges related to slow formation rates and inefficient mass transfer. By incorporating bio-surfactants, known for their ability to alter surface properties and improve solubility, we demonstrate significant improvements in these critical aspects. Our findings reveal that bio-surfactants not only accelerate gas hydrate formation but also enhance the overall reaction kinetics, leading to more efficient and rapid synthesis. This research provides new insights into leveraging bio-surfactants as a viable strategy for optimizing gas hydrate processes, offering promising implications for both industrial applications and fundamental scientific understanding. Gas hydrates offer significant potential for diverse applications such as gas mixture separation, carbon dioxide capture, transportation and sequestration, methane storage and transport, and seawater desalination. A major challenge in these applications is reducing the initiation time for hydrate formation and accelerating the growth rate of hydrates during the water-to-hydrate reaction. This paper explores how different surface types impact gas hydrate formation across various reactions, with a specific focus on the role of bio-surfactants. Factors such as temperature, pressure, and surface characteristics play a crucial role in the formation and growth of gas hydrates. While traditional methods like mechanical mixing and chemical additives have been used to enhance hydrate formation, they often face limitations in efficiency and may have adverse environmental effects.

Keywords: *Bio surfactants Hydrate reaction, Surface tension, Mass transfer coefficient.*

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1.1 INTRODUCTION:

Gas hydrates, also known as methane hydrates, form when methane molecules are encased within a lattice of water molecules. Gas hydrates are classified based on their molecular structure, forming crystalline lattice structures known as clathrates. A unique characteristic of clathrates is the absence of chemical bonding between the gas and water molecules, allowing for relatively easy separation. The water molecules stabilize the structure through van der Waals forces, creating a steady configuration [1]. Gas hydrate morphology plays a crucial role in determining the physical properties of gas hydrates [2]. The unique characteristics of methane hydrates are observed at temperatures below the melting point of ice, specifically between 242 and 271 K [3-4].

The formation and preservation of significant natural gas hydrate deposits depend on various geological structures, akin to conventional fuel reservoirs [5-7]. To characterize the effects of pore size on hydrate equilibrium conditions in a porous medium, it is essential to refine and expand upon the existing experimental work [8–18].

Methane in natural gas hydrates can originate from either bacterial processes or thermogenic sources [19]. The three common crystal structures of natural gas hydrates are structure I (sI), structure II (sII), and structure H (sH). Structures I and II each consist of one small and one large cavity, whereas structure H contains two small cavities and one large cavity. All three structures, sI, sII, and sH, share the basic pentagonal dodecahedron (12 pentagons, 5^{12}) cage configuration [20].

Gas hydrates form under specific temperature and pressure conditions. By analyzing the pressure and temperature gradients with depth, the thickness of the gas hydrate stability zone can be determined. Phase-boundary data, combined with subsurface temperature conditions, clearly indicate that the upper limit of the gas hydrate stability region is approximately 150 meters in permafrost (continental polar) regions where surface temperatures are below 0°C. In marine sediments, the upper limit is around 300 meters when the temperature nears 0°C. The lower limit of the gas hydrate stability zone is determined by geothermal gradients, with the maximum depth reaching up to 2000 meters below the surface. However, the actual lower limit is typically much less, depending on local

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**Author for Correspondence: kirti.zare@dypiu.ac.in* conditions. The pore pressure gradient exceeds the hydrostatic pressure gradient, and regions with high pore pressure and thicker gas hydrate layers are considered more stable. Sodium chloride salts lower the formation of gas hydrates. Therefore, when evaluating the gas hydrate stability zone, it is important to consider the potential impact of pore water salinity and gas formation chemistry [21]. Clathrate hydrates play a significant role in intermolecular compounds and contribute substantially to reserves [22, 23]. It has been observed that methane hydrate is stronger than water ice, as indicated by initial experiments conducted by researchers [24].

Surfactants are surface-active agents that reduce the surface tension of liquids. They can function as detergents, emulsifiers, or foaming agents. A surfactant molecule consists of a hydrophilic head, which is attracted to water, and a hydrophobic (lipophilic) tail, which repels water. Thus, a surfactant contains both water-soluble and water-insoluble components. The hydrophobic tail moves away from the bulk water phase toward the air, while the hydrophilic head remains in the water.

This alignment reduces the surface free energy, thereby lowering surface tension and increasing surface viscosity. The diffusion of surfactants to the surface continues until equilibrium is reached. In the bulk aqueous phase, surfactants form aggregates known as micelles, which can be spherical or cylindrical. The shape of these aggregates depends on the chemical structure of the surfactants [25].

A novel experimental methodology utilizing thin glass capillaries as both optical cells and micro reactors has been developed to observe and quantify the generation, growth kinetics, and porous structure of gas hydrates promoted by additives. By combining optical microscopy and micro-Raman spectroscopy, this method provides detailed insights into the hydrate formation process, including conversion rates, storage capacities, and porosity. The study applied this methodology to surfactants SDS and AOT, revealing that both promote an initial slow growth of hollow hydrate crystals followed by a rapid conversion phase.

It was observed that while SDS leads to a higher final conversion with lower hydrate porosity, AOT promotes faster conversion rates but results in higher porosity. This approach offers valuable data for optimizing gas-hydrate-based technologies, such as gas separation and storage [26].

This paper reviews the mechanisms by which various surfactants promote gas hydrate formation, discussing factors affecting the process and introducing mathematical models for kinetics and thermodynamics. It highlights a debate over whether surfactants like SDS form micelles during hydrate formation, with differing opinions on how carbon chain length influences hydrate formation. Future research directions include clarifying the mechanisms of surfactant promotion—capillary forces versus micelle formation—using advanced imaging techniques, studying the effects of ions and porous media similar to natural sediments on hydrate formation, and improving kinetic and thermodynamic models to better predict and optimize hydrate formation processes. Practical evaluation parameters such as induction time and gas storage density are also emphasized [27].

Our study explores the application of bio surfactants to reduce surface tension, thereby enhancing mass transfer and improving the structural formation of hydrates. We conducted a series of experiments using different surface types and bio surfactants to observe their effects on the initiation and growth rates of gas hydrates. The bio surfactants were selected based on their environmental compatibility and effectiveness in reducing surface tension.

1.2 Classification of surfactants:

Based on their separation in water, surfactants are classified into four types:

a. **Anionic Surfactants:** These surfactants have a higher hydrate formation kinetics and account for about 50% of global production. They contain an anionic functional group at their head.

b. **Cationic Surfactants:** Cationic surfactants are more expensive due to the high pressure required for the hydrogenation reaction during their synthesis.

c. **Non-Ionic Surfactants:** These surfactants, which do not ionize in aqueous solutions, make up about 45% of the overall market. They are considered secondary surfactants.

d. **Zwitterionic Surfactants:** These surfactants exhibit both anionic and cationic dissociation within a single molecule.

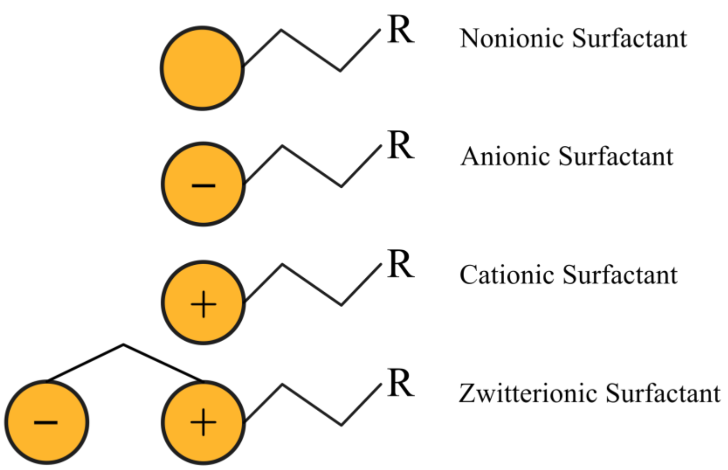


Figure No 1: Different classes of surfactant [25]

Bio surfactants are amphipathic molecules with both hydrophilic and hydrophobic properties. This allows the monomers to form structures such as micelles, micellar tubes, bilayers, and vesicles. Bio surfactants are also known for their ability to enhance the formation of emulsions between immiscible liquids, such as hydrocarbons and water. Gas hydrate is an ice-like crystalline structure where gas molecules are encased within cages formed by water molecules [28-29].

As hydrates can form at temperatures both below and above water's freezing point without chemical bonding between the water and gas molecules [30]. Research indicates that gas hydrates form in oil and gas systems under high pressure and low temperature. They are categorized into three structural types—Type I, Type II, and Type H—each with varying numbers and sizes of cages [31].

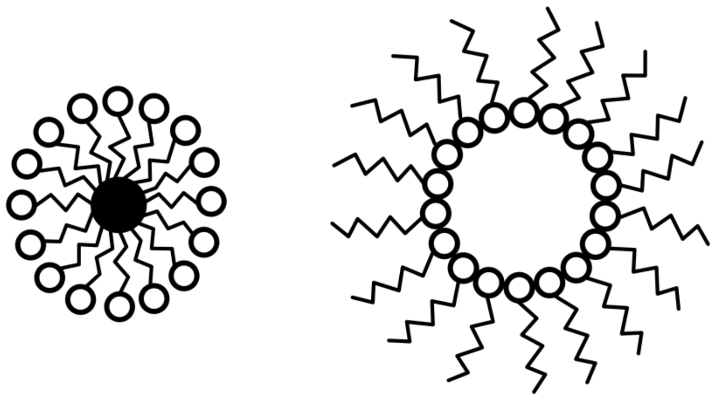


Figure No 2: Formation and emulsion with hydrocarbon & water[32]

Bio surfactants perfectly fit the criteria for green additives. They exhibit remarkable stability under various conditions, including temperature, salinity, and pH, and are effective in enhancing the kinetic properties of hydrate formation. While synthetic surfactants are typically used as kinetic promoters for hydrate formation, bio surfactants offer an environmentally friendly alternative. The use of these green additives has proven to be highly beneficial for gas hydrate formation, reducing environmental concerns related to disposal and toxicity.

This study evaluated nonionic surfactants as kinetic inhibitors for methane hydrate formation at 8.70 MPa and temperatures between 274.15 and 277.15 K. Results revealed that while Span and Tween series had minimal impact on methane hydrate phase boundaries, Tween-80 significantly delayed nucleation time but promoted the overall rate of hydrate formation. Tween-80, with an optimal concentration of 2.5% (v/v), showed superior induction time performance compared to the commercial inhibitor PVP, though PVP reduced the rate of formation and had higher gas consumption. The study suggests using induction time as a key performance indicator for hydrate inhibition, with Tween-80 outperforming PVP in this aspect. The classical nucleation theory and modified Englozes model effectively predicted induction time and formation rate with maximum errors of 5.70% and 4.93%, respectively [32].

Surfactants significantly enhance hydrate formation and accumulation, which could be leveraged to create an economical method for storing natural gas in gas hydrates.

This has potential applications in providing fuel for power plants during peak demand, among other uses. Anionic surfactant molecules help by bringing gas and structured water together for hydrate crystal nucleation and by adsorbing on solid surfaces at the gas/water interface to facilitate crystal agglomeration. In ocean sediments, bio surfactants behave similarly but with added complexity due to the varied porous media surfaces.

They selectively adsorb on specific surfaces, with sodium montmorillonite clay, commonly found in ocean sediments, favoring hydrate formation more than other minerals. In contrast, kaolinite surfaces tend to be less conducive and can even hinder hydrate formation. It's also notable that carbon dioxide reduces the catalytic effect of surfactants on hydrate formation due to its lower solubility with these surfactants. [33].

They examined the effects of temperature, initial water content, stirring rate, and reactor size on methane hydrate formation in a stirred semi-batch autoclave reactor at 90 bar pressure. Their findings indicated that sub cooling significantly influences methane hydrate formation, as increased sub cooling enhances the driving force for hydrate formation and growth. Additionally, they found that mass and heat transfer play a crucial role in hydrate formation within batch/semi-batch reactors [34].

They conducted experiments to assess the impact of impeller type and number on methane hydrate formation in a 5.71 L stirred tank reactor. Their results showed that the methane hydrate formation rate with Rushton turbine (RT) impellers was higher than with pitched blade turbines upward pumping (PBTU) impellers, although RT impellers consumed more energy. Tests with dual impellers yielded similar results to those with single impellers [35].

The induction time for methane, ethane, and carbon dioxide hydrates has been examined through both experimental and theoretical studies. In line with crystallization principles, induction time depends on the ratio of gas fugacity under experimental conditions to that under equilibrium conditions [36].

Several energy companies have reported significant flow assurance challenges, which require substantial efforts and costs to prevent hydrate blockages in oil and gas facilities. Gas hydrates can form quickly under favorable conditions, much faster than waxes, scales, or asphaltenes, and often without warning, particularly in offshore pipelines. While hydrate plugs form rapidly, the remediation process can take days or even months [37].

2. EXPERIMENTALLY PROCEDURE

The setup includes pressure gauges and temperature measurement equipment to facilitate hydrate separation. Before commencing each experiment, the system is meticulously inspected for leaks and blockages. The stirred tank reactor is then filled with the required amount of fluid, consisting of water and the bio-surfactant CAPB, and brought into contact with the hydrate-forming gas. The system pressure is raised to the desired experimental level by injecting the hydrate-forming gas, and the stirrer is operated at 400 rpm. As the water and gas interact, hydrates begin to form, indicated by a gradual decrease in system pressure.

The stirred tank reactor is subsequently filled with a precise amount of fluid, which is a mixture of water and the bio-surfactant CAPB (Cocamidopropyl Betaine). This specific composition of the fluid is chosen to facilitate the formation of hydrates. The bio-surfactant CAPB is known to enhance the stability and formation of hydrates by reducing surface tension and promoting better interaction between the water and the gas. Once the reactor is filled with the fluid, it is brought into contact with the hydrate-forming gas.

This step is crucial as the gas will dissolve in the fluid under high pressure and low temperature conditions, forming hydrates. The system is then pressurized to the desired experimental level by injecting the hydrate-forming gas into the reactor. The pressure level is carefully controlled and monitored using the pressure gauges installed in the system.

The stirrer within the reactor is set to rotate at a speed of 400 rpm. This agitation speed is selected to ensure that the fluid and gas are thoroughly mixed, which promotes the formation of hydrates. The rotational speed also helps in distributing the gas evenly throughout the fluid, increasing the contact surface area and enhancing the rate of hydrate formation.

As the water and gas come into contact under these controlled conditions, hydrates start to form within the reactor. The formation of hydrates is a gradual process, and one of the key indicators of this process is a decrease in system pressure. This decrease occurs because the gas is being consumed and incorporated into the solid hydrate structures. The pressure drop is continuously monitored, providing a real-time indication of the hydrate formation progress.

Overall, this detailed procedure ensures that the experimental conditions are optimized for hydrate formation, and the system is closely monitored to track the progress and efficiency of the process. A data acquisition system is used to obtain the relevant P-T data once the experiment begins.

When no notable pressure drop is observed inside the reactor, it indicates that the hydrate formation has reached saturation. This signifies that the maximum amount of hydrate has formed under the given experimental conditions, and no additional gas is being incorporated into the hydrate structure. This experiment is conducted using three different gases: methane, carbon dioxide, and natural gas. Each gas is tested separately to understand the specific characteristics and efficiency of hydrate formation with each type of gas.

Methane and carbon dioxide are pure gases, while natural gas is a mixture of several hydrocarbons, primarily methane, along with other gases such as ethane, propane, and butane.

By applying this equation, the amount of gas that has been incorporated into the hydrate structure can be accurately determined. This calculation is crucial for understanding the efficiency of the hydrate formation process and for comparing the performance of different gases.

Overall, the detailed analysis of the pressure drop, combined with the use of the Pitzer equation, provides a comprehensive understanding of the hydrate formation process for methane, carbon dioxide, and natural gas. This information is valuable for optimizing conditions for hydrate formation and for potential applications in gas storage and transportation.

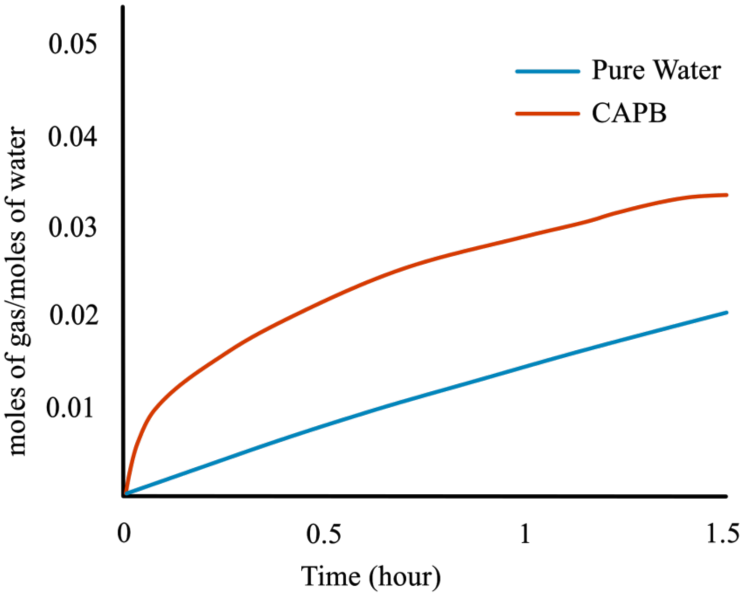
3. RESULT AND DISCUSSION

Pure methane gas was introduced into the stirred tank reactor at a pressure of 49.34 atmospheres and a temperature of 273.15 K (0°C). The experiment was conducted using a solution with 1% Cocamidopropyl Betaine (CAPB), a bio-surfactant that can influence hydrate formation. The kinetics of methane hydrate formation with varying concentrations of CAPB are illustrated in Graph No:1. This graph demonstrates how the presence and concentration of CAPB affect the rate at which methane hydrates form. To elaborate, the stirred tank reactor was set up under controlled conditions of high pressure and low temperature to facilitate the formation of methane hydrates.

The chosen pressure of 49.34 atmospheres and temperature of 273.15 K are specific conditions where methane is likely to form hydrates. CAPB, as a bio-surfactant, plays a significant role in hydrate formation by modifying the interface between the gas and the liquid phase. The concentration of CAPB used in this experiment was 1%, which is a critical factor in determining its impact on hydrate formation.

The graph shows how different levels of CAPB affect the rate at which methane hydrates are formed. In the experiment, the rate of hydrate formation was monitored over time, and the data was plotted to illustrate the kinetics.

The graph typically shows how quickly the pressure decreases as hydrates form, which corresponds to the gas uptake and the efficiency of hydrate formation. The shape of the curve in Graph No:1 reflects how the presence of CAPB influences the rate of methane hydrate formation, potentially highlighting optimal concentrations or revealing trends in hydrate formation behavior.

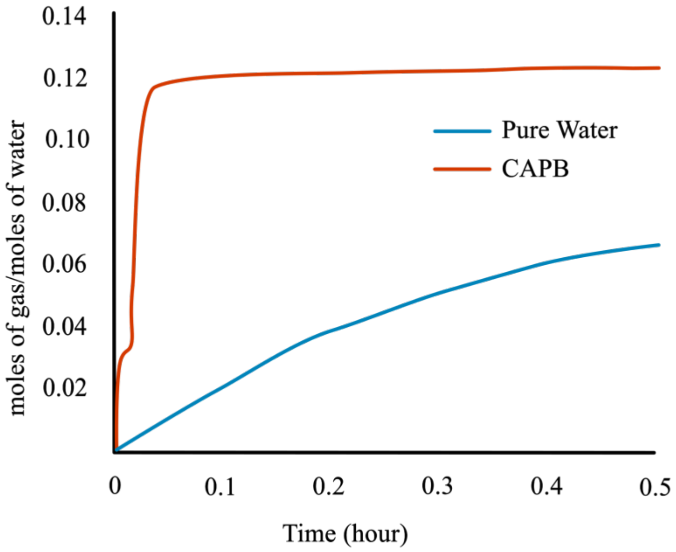


Graph No I: CAPB vs Methane Hydrate formation kinetics

The graph demonstrates that CAPB significantly enhances gas absorption in water compared to pure water. Over the 1.5-hour period, the gas absorption rate in CAPB is initially much higher and remains consistently greater than that in pure water, indicating that CAPB facilitates a more efficient gas uptake. This suggests that CAPB could be a valuable additive for processes requiring enhanced gas solubility in water, potentially leading to more efficient industrial or environmental applications where increased gas absorption is beneficial.

The graph illustrates that in the presence of 1% CAPB, the kinetics of carbon dioxide hydrate formation are significantly enhanced compared to pure water under conditions of 29.60 atmosphere and 273.15°C. Initially, the CAPB solution shows a much steeper rise in gas absorption, indicating a rapid formation of hydrates. This increased rate of absorption continues over time, maintaining a higher level than pure water and ultimately suggesting that CAPB acts as an effective promoter for hydrate formation.

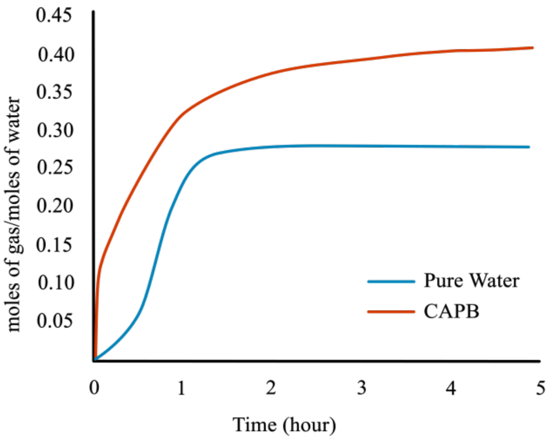
The leveling off towards the end indicates a higher overall capacity for gas absorption and hydrate formation in the CAPB solution, highlighting its potential utility in industrial applications where efficient gas hydrate formation is beneficial.



Graph No II: CAPB vs CO_2 hydrate formation kinetics

The graph demonstrates that in a stirred tank reactor containing natural gas at 9.60 atmosphere and 273.15°C with 1% CAPB, the kinetics of gas hydrate formation are significantly enhanced compared to pure water. The CAPB solution shows a rapid initial increase in gas absorption, indicating that CAPB acts as a nucleation promoter, enhancing the solubility and formation rate of hydrates. This higher absorption rate persists throughout the experiment, with the CAPB solution maintaining a greater overall gas uptake. As the system approaches saturation, the CAPB solution levels off at a higher absorption rate than pure water, highlighting CAPB's effectiveness in promoting efficient gas hydrate formation, which is advantageous for industrial applications.

The graph shows that in a stirred tank reactor with natural gas at 9.60 atmosphere and 273.15°C, the presence of 1% CAPB significantly enhances gas hydrate formation compared to pure water. The CAPB solution exhibits a rapid initial increase in gas absorption, quickly reaching a plateau at around 0.12 moles of gas per mole of water within a short time, indicating a swift and efficient hydrate formation. In contrast, pure water shows a slower, more gradual increase in gas absorption. This demonstrates that CAPB is an effective promoter of gas hydrate formation, significantly improving the rate and capacity of gas uptake, which is beneficial for industrial applications requiring efficient and rapid gas hydrate production.



Graph No III: CAPB vs natural gas formation kinetics

The graph III CAPB vs natural gas formation kinetics illustrates the gas hydrate formation kinetics in a stirred tank reactor with natural gas at 9.60 atmosphere and 273.15°C, comparing pure water to a 1% CAPB solution over five hours. The CAPB solution (orange line) shows a rapid initial increase in gas absorption, reaching about 0.38 moles of gas per mole of water within the first hour and then gradually approaching a plateau near 0.40 moles. In contrast, the pure water (blue line) also shows an initial increase but at a slower rate, leveling off around 0.27 moles of gas per mole of water. This indicates that the CAPB significantly enhances the gas hydrate formation process, resulting in a higher absorption capacity and quicker attainment of equilibrium. Thus, CAPB proves to be an effective promoter for gas hydrate formation, optimizing both the speed and efficiency of the process compared to pure water.

These findings have profound implications for industrial applications. In gas mixture separation, faster hydrate formation can lead to more efficient separation processes. For carbon dioxide capture, enhanced hydrate growth rates can improve the feasibility of sequestration techniques. In methane gas storage and transport, optimized hydrate formation can lead to safer and more economical storage solutions. Additionally, the use of bio surfactants in seawater desalination through hydrate formation offers a novel approach to improving the efficiency of desalination processes.

4. CONCLUSION

Surfactants are highly effective in enhancing the rate of hydrate formation reactions, making them valuable for improving hydrate nucleation rates. This study focuses on evaluating the impact of the bio-surfactant CAPB on the kinetics of hydrate formation for various gases, including methane, carbon dioxide, and natural gas, using a stirred tank reactor. The results demonstrated that the presence of CAPB significantly improves gas hydrate formation kinetics compared to water alone. The experimental results demonstrated that bio surfactants significantly reduce the induction time for hydrate formation and accelerate the growth rate of hydrates. By decreasing surface tension, bio surfactants enhance the mass transfer between gas and water phases, leading to a more efficient and rapid formation of gas hydrates. We observed that certain bio surfactants outperformed traditional chemical surfactants, offering a more sustainable and effective solution. By enhancing the kinetic properties of hydrate formation, CAPB increases methane storage capacity and aids in desalination processes. In conclusion, bio-surfactants like CAPB serve as safe, eco-friendly kinetic promoters in gas hydrate applications, offering potential benefits for methane separation, storage, and transportation. The use of bio surfactants presents a significant advancement in the formation and growth of gas hydrates. By enhancing mass transfer and structural formation, bio surfactants can improve the efficiency and effectiveness of various industrial applications, providing both economic and environmental benefits. Future research should focus on optimizing bio surfactant formulations and exploring their interactions with different gas and water mixtures to further enhance the practical applications of gas hydrates.

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