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Original Research Article

The Adverse Effect of SDS on the Growth Rate of Double Tetra *n*-butylammonium Chloride + Methane Semiclathrate Hydrate

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ABSTRACT

Accelerating gas hydrate formation is crucial to exploiting its positive applications, as these ice-like structures can serve as a vast and clean source of energy. In this research, we delve into the influence of sodium dodecyl sulfate (SDS) on the growth rate of double tetra n-butylammonium chloride (TBAC) +methane semiclathrate hydrate. To investigate this, we conducted experiments utilizing a stirred batch cell with a total volume of 169 cm^3. The cell's temperature was maintained at 278.15 K, while the initial pressures were set at 6 and 8 MPa. Comparing the results with pure water, SDS showed a significant positive effect on the growth rate of methane hydrate formation. However, when 400 ppm of SDS was introduced, it had a detrimental impact on the average growth rate of the double (TBAC + methane) semiclathrate hydrate within just 50 minutes of the process.

HIGHLIGHTS

- A thermodynamic and kinetic promoter mixture is used for hydrate formation
- The addition of 3 and 5 wt% TBAC increased the average growth rate of hydrate formation
- Using TBAC and SDS together decreased growth rate compared to SDS solution and TBAC solution.

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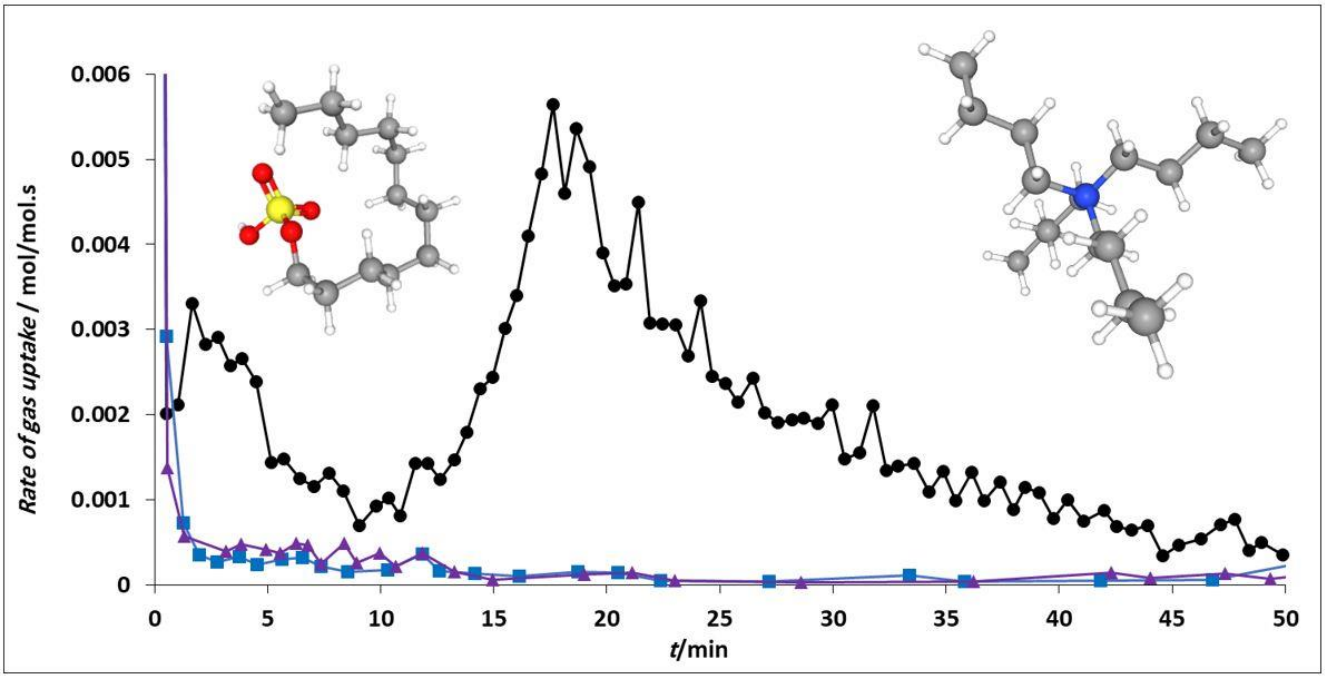
GRAPHICAL ABSTRACT



1. INTRODUCTION

Methane hydrates, also known as methane clathrates, represent a unique class of crystalline structures in which methane molecules are encaged within cavities formed by molecules of water. These hydrates occur in nature under specific conditions of high-pressure and low-temperature, commonly found in permafrost regions and marine sediments [1]. Gas hydrates have a wide range of positive utilization. They can be used for storage of gases, gas separation, wastewater treatment, juice concentration, energy recovery, gas transportation, water desalination, and CO_2 capturing [2-8]. Despite the immense promise methane hydrates hold as a clean energy resource, their commercial viability hinges on understanding and controlling the kinetics of their formation [5, 9-11]. The formation process involves the nucleation of hydrate crystals and their subsequent growth, which is influenced by various factors, including temperature, pressure, guest molecules and the presence of additives [12-14]. A thorough comprehension of the kinetics is essential to develop efficient and cost-effective methods for hydrate production. Moreover, an in-depth study of formation of gas hydrates in the presence of specific additives can shed light on their potential role in enhancing or inhibiting the process, leading to improved exploitation strategies.

Three typical structures can be identified when it comes to gas hydrates, including: Structure sI that is the most common and well-studied gas hydrate structure. It consists of water molecules forming hexagonal cages with one central guest molecule. The cages are arranged in a cubic lattice structure. Structure sII is similar to Structure sI, but has a larger cage size. The water molecules form pentagonal dodecahedral cages, accommodating multiple guest molecules. It is often found at higher pressures and structure sH that has a hexagonal prism structure and consists of two types of cages - large and small. Large cages contain guest molecules, while small cages can be empty. This structure is commonly found in certain clathrates, including gas hydrates [1, 15]. In 1940, Fowler *et al.* made a groundbreaking

discovery of a novel hydrate structure formed by tetra n-butyl ammonium salts. These structures, known as semiclathrates, involve the replacement of certain water lattice sites with guest molecules (anions) [16-21]. Semiclathrate hydrates (SCHs) are a promising phase change material (PCM) for storing and transporting the cold energy, as well as for improving the energy efficiency of cooling processes [21-24].

The phase equilibrium of semiclathrate hydrates has been investigated by numerous researchers in recent years [25-31]. However, despite the understanding of the thermodynamics governing semiclathrate hydrate formation, there has been relatively less emphasis on studying the kinetics of these hydrates [32-34]. The aim of this study is to explore how the growth rate of double (TBAC+ methane) semiclathrate hydrate is affected by SDS. This research provides insight into the differentiated behavior of gas hydrate formation in the SDS presence and its implications for energy-related and other applications. This highlights the need for further research on the role of additives in shaping the properties and behavior of gas hydrates to pave the way for innovative and sustainable energy solutions. The innovation of this study is the targeted study of the effects of SDS on the growth rate of specific gas hydrate structures, investigating both beneficial and detrimental effects, and providing useful insights in the fields of energy, environment, and materials science.

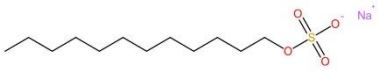
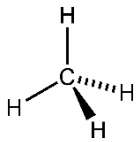
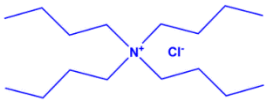
2. EXPERIMENTAL

2.1. Materials and Apparatus

The information regarding the chemicals used in this study, including their names, purities, molecular structures, and suppliers, is presented in Table 1. A visual representation of the experimental setup employed to study the kinetics of methane+TBAC semiclathrate hydrate formation is displayed in Fig 1.

The main component of the equipment is a high-pressure cell with an inner volume of 169 cm^3 and a maximum operating pressure of 20 MPa. To ensure proper mixing within the cell, a rocking cell stirrer is utilized, which operates at a speed of 10 rpm. The temperature of the cell is carefully regulated using a cooling system that circulates a mixture of 50% ethylene glycol and water as a coolant. Pressure measurements are taken using a 0.01 MPa scale pressure transducer, while temperatures are monitored using a 0.1 K scale thermocouple (PT100).

Table 1. The properties of the material used in the experiments

Material	CAS No.	Suppliers	Purity	Chemical structure
sodium dodecyl sulfate[a]	151-21-3	Merck	99% wt%	
methane	74-82-8	Technical Gas Service	99.95 mole%	
tetra-n-butylammonium chloride[b]	1112-67-0	Merck	95 mole%	

[a]*SDS;* [b]*TBAC.*

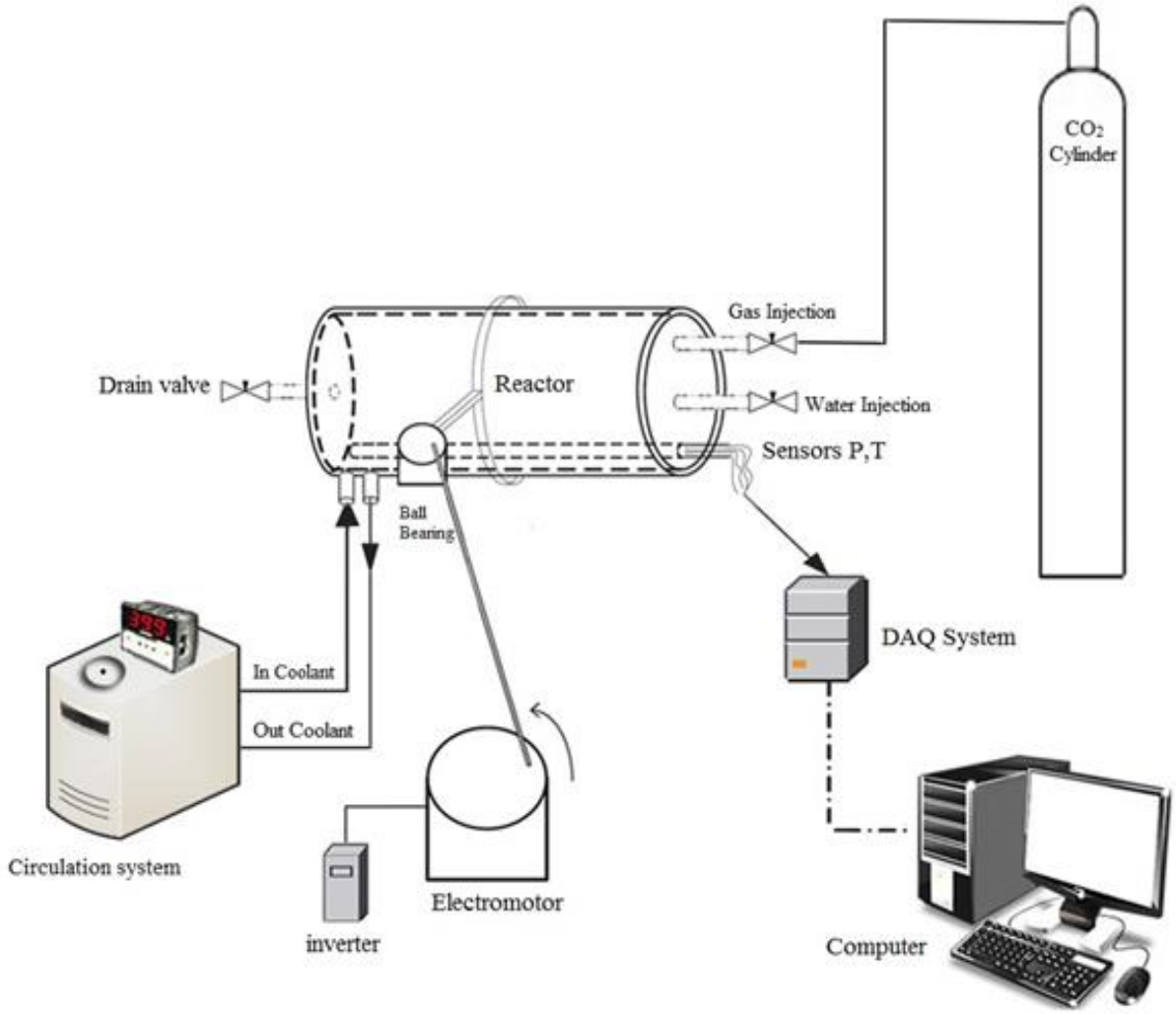


Fig 1. Visual representation of the experimental apparatus

2.2. Procedure

Various solutions of TBAC and SDS were prepared in an aqueous form. Afterwards, a precise amount of the prepared solution was added to the batch crystallizer. The temperature of the crystallizer was regulated to 278.15 K by means of a circulator. Gas was subsequently introduced into the crystallizer until the desired pressure levels were reached. To agitate the solution within the crystallizer, a rocking cell stirrer was activated at a speed of 10 rpm.

3. RESULTS AND DISCUSSION

To examine the influence of SDS on the growth rate of (TBAC+methane) semiclathrate hydrate various aqueous solutions were prepared, including: (a) Aqueous solutions containing 3 wt% and 3 wt% TBAC, (b) An aqueous solution with 400 ppm SDS, and (c) A TBAC + SDS mixture. Throughout the experiments, a constant temperature of 278.15 K and pressure of 6 MPa and 8 MPa were maintained within the cell to study the rate of TBAC+methane semiclathrate hydrate formation under these conditions.

The average growth rate (R_{av}) within the first 50 minutes of the process was evaluated using the

following equation which was used to evaluate the average rate of gas uptake (R_{50}) within 50 min of the process.

$$R_{50} = \frac{\Delta n_{Methane,50}}{n_{sol} \cdot \Delta t} \quad (1)$$

Where, $\Delta n_{Methane,50}$, n_{sol}, and Δt represent the number of moles of methane converted to gas hydrate, the number of moles of feed water used, and the time period, respectively.

Equation (2) was utilized to determine the quantity of methane uptake throughout the 50 min of the process.

$$\Delta n_{Methane,50} = n_{Methane,0} - n_{Methane,50} \quad (2)$$

Where, $n_{Methane,0}$ and $n_{Methane,50}$, respectively, stand for the methane mole number in the gas phase at time t = 0 and time 50 min, and can be calculated by the following equations.

$$n_{Methane,0} = \frac{P_0 V_0}{Z_0 RT} \quad (3)$$

$$n_{Methane,50} = \frac{P_{50} V_{50}}{Z_{50} RT} \quad (4)$$

Where, subscripts 0 and 50, respectively, are the reactor conditions at beginning and time 50 min, and Z is evaluated using PR EOS [35].

$$Z^3 - (B-1)Z^2 + (A-3B^2-2B)Z - (AB-B^2-B^3) = 0 \quad (5)$$

$$A = \frac{(a_c \alpha)P}{(RT)^2} \quad (6)$$

$$B = \frac{bP}{RT} \quad (7)$$

$$a_c = 0.45724 \frac{R^2 T_c^2}{P_c^2} \quad (8)$$

$$b = 0.07780 \frac{RT_c}{P_c} \quad (9)$$

Here, α and m are PR parameters that obtained as [36],

$$\alpha = (1 + m(1 - T_r^{0.5}))^2 \quad (10)$$

$$m = 0.3746 + 1.5423\omega - 0.2669\omega^2 \quad (11)$$

Where, ω is the acentric factor of methane.

Table 1 presents the growth rate of TBAC + methane semiclathrate hydrate during the first 50 minutes of the process. Fig 2 and 3 illustrate the influence of TBAC on the growth rate of methane hydrate per mole of feed solution at a constant T = 278.15 K and pressures of 6 MPa and 8 MPa, respectively.

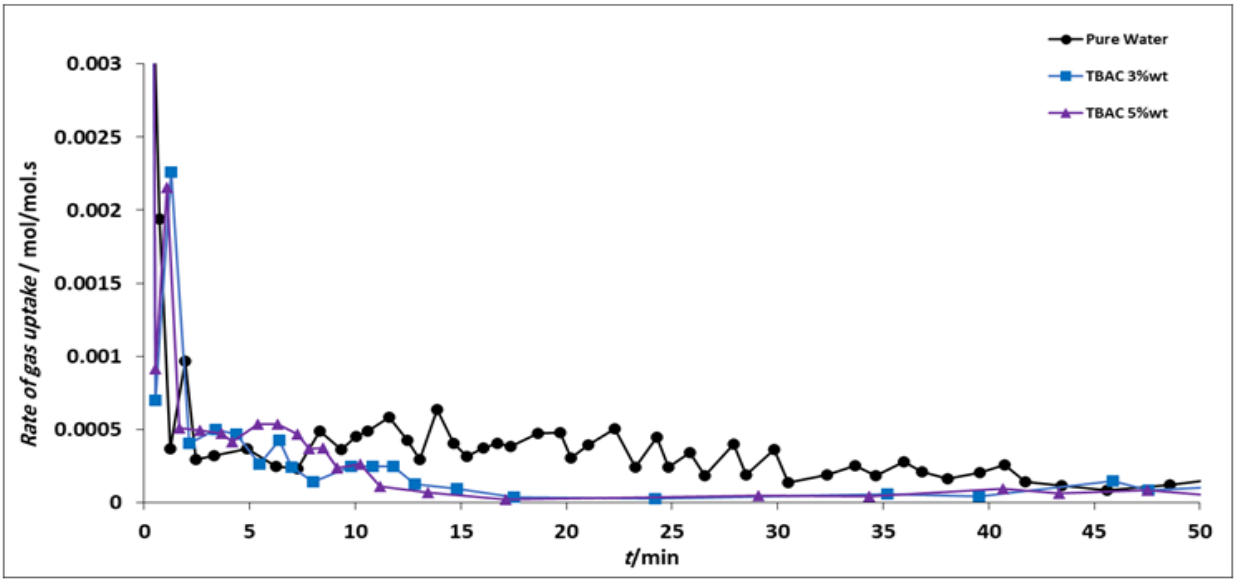


Fig 2. Growth rate of double (TBC+methane) semiclathrate hydrate per mole of feed solution at P_0 = 6 MPa and T = 278.15 K

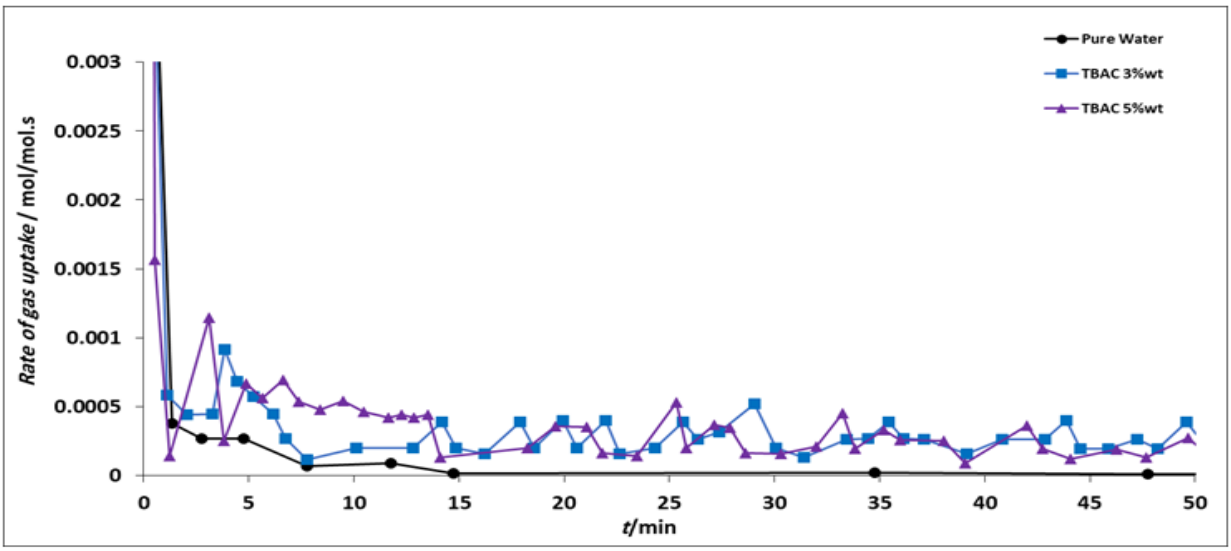


Fig 3. Growth rate of double (TBAC+methane) semiclathrate hydrate per mole of feed solution at P_0 = 8 MPa and T = 278.15 K

Based on the data presented in Fig 2 and 3, as well as Table 1, it is evident that the inclusion of 3 wt% and 5 wt% of TBAC has not a significant influence on the average growth rate of methane at initial pressure of 6 MPa, while a positive effect can be seen at P_0 = 8 MPa.

At P_0 = 8 MPa, the growth rate of formed hydrate in water is 0.40 mmol/mol.s, whereas in the TBAC solution (5 wt%), it is 0.70 mmol/mol.s. This indicates that the average growth rate of formed hydrate in the TBAC solution is 75% higher than in pure water under the mentioned thermodynamic conditions. By incorporating TBAC, the equilibrium pressure of methane hydrate formation is reduced, leading to a substantial increase in the driving force behind the process. This significant reduction in pressure moderates the thermodynamic conditions during methane hydrate formation [37-41]. By moderating Thermodynamics conditions, such as reducing the equilibrium pressure through the utilization of TBAC, the growth rate of the process is increased. This is because the reduction in pressure enhances the driving force of the process, leading to more efficient and faster methane uptake into the gas hydrate structure.

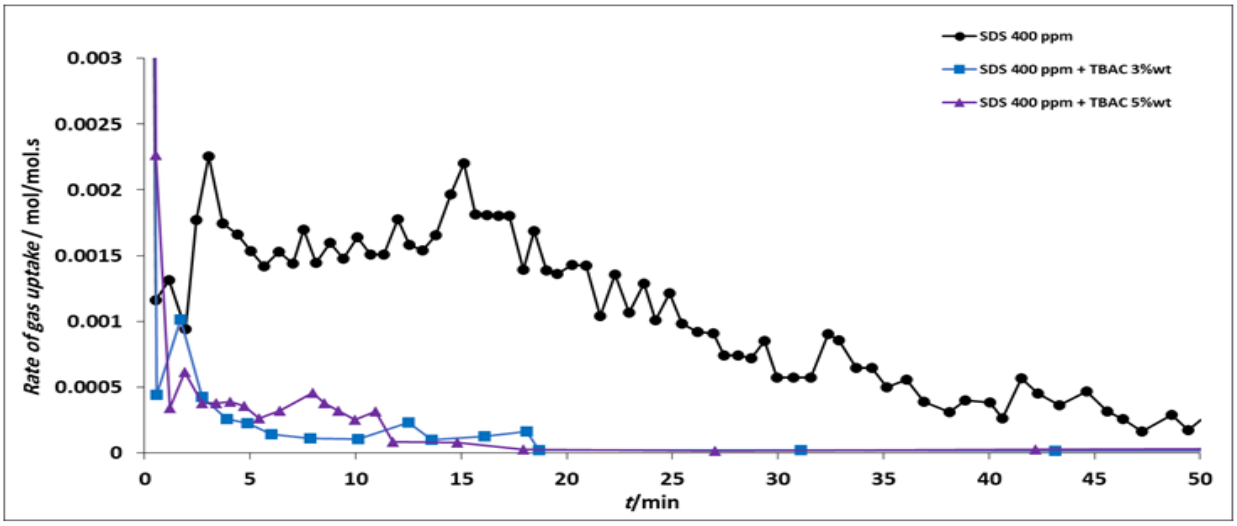


Fig 4. The effect of TBAC on the growth rate of double (TBAC+methane) semiclathrate hydrate per mole of feed solution in presence of 400 ppm SDS at P_0 = 6 MPa and T = 278.15 K

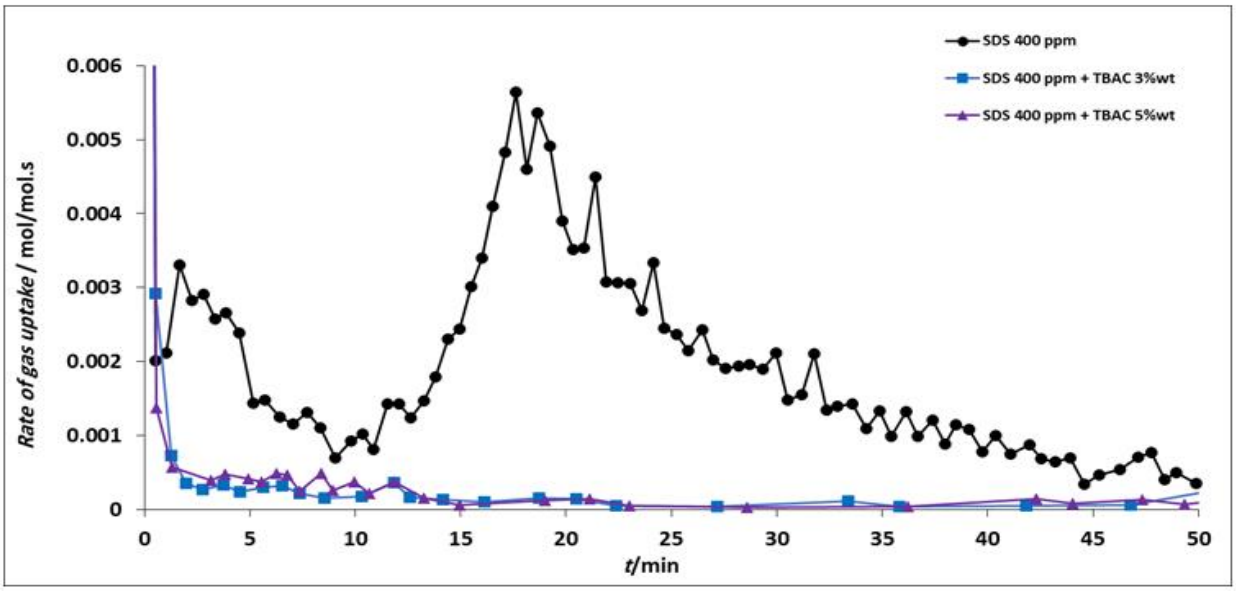


Fig 5.The effect of TBAC on the growth rate of double (TBAC + methane) semiclathrate hydrate per mole of feed solution in presence of 400 SDS at P_0 = 8 MPa and T = 278.15 K

In Fig 4 and 5, and as supported by the data in Table 1, a comparison was made between the influence of SDS alone and SDS + TBAC on the growth rate of TBAC+methane semiclathrate hydrate. The investigation revealed that the average growth rate of TBAC+methane semiclathrate hydrate for the solution of SDS+TBAC is lower than that of the SDS solution, consistently observed at 278.15 K and both tested pressures. This negative effect on the growth rate might be attributed to the interaction between the released ions of TBAC and SDS within the aqueous solution. Such interactions could lead to changes in the process, affecting the growth rate of the hydrate crystals.

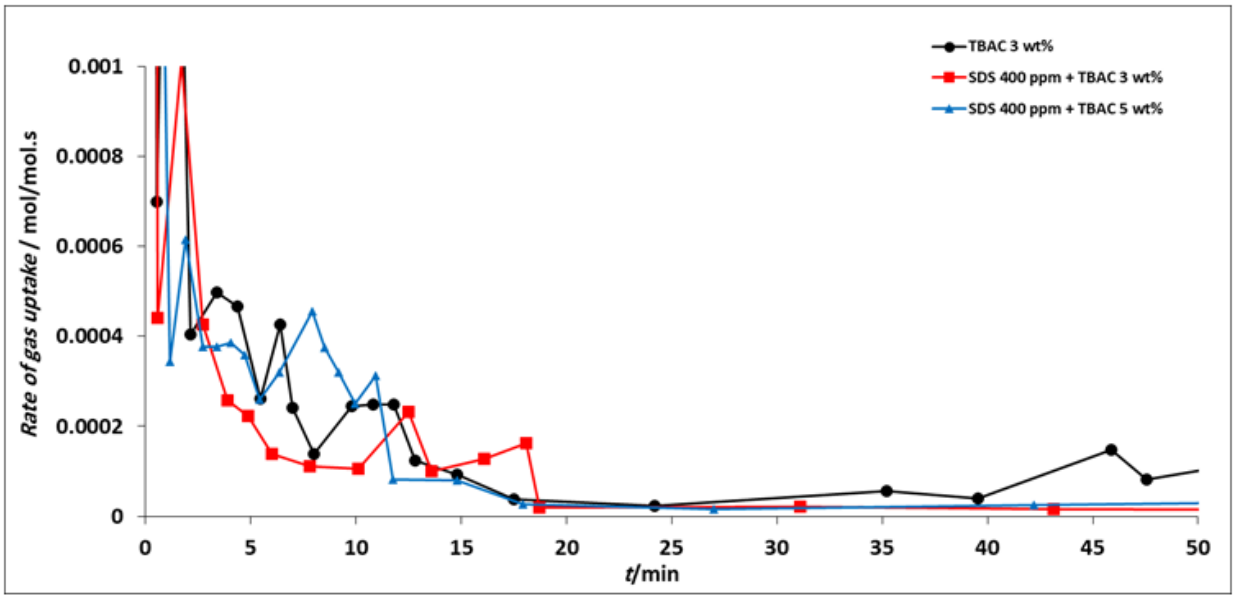


Fig 6. The effect of SDS on the growth rate of double (TBAC+methane) semiclathrate hydrate per mole of feed solution at P_0 = 6 MPa and T = 278.15 K

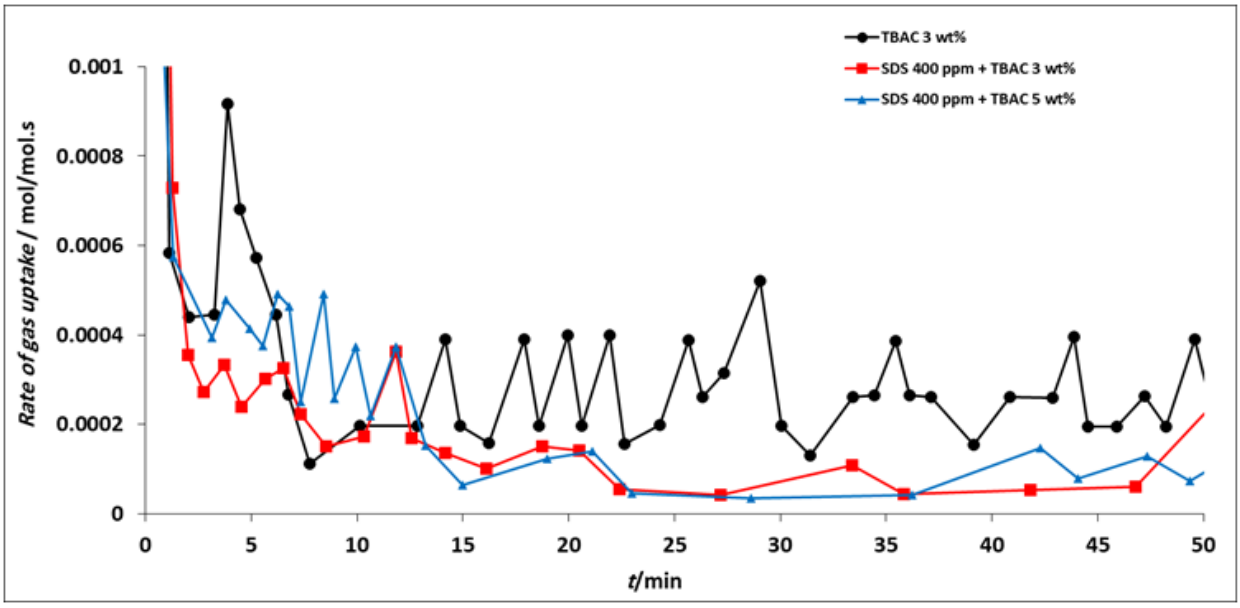


Fig 7. The effect of SDS on the growth rate of double (TBAC + methane) semiclathrate hydrate per mole of feed solution at P_0 = 8 MPa and T = 278.15 K

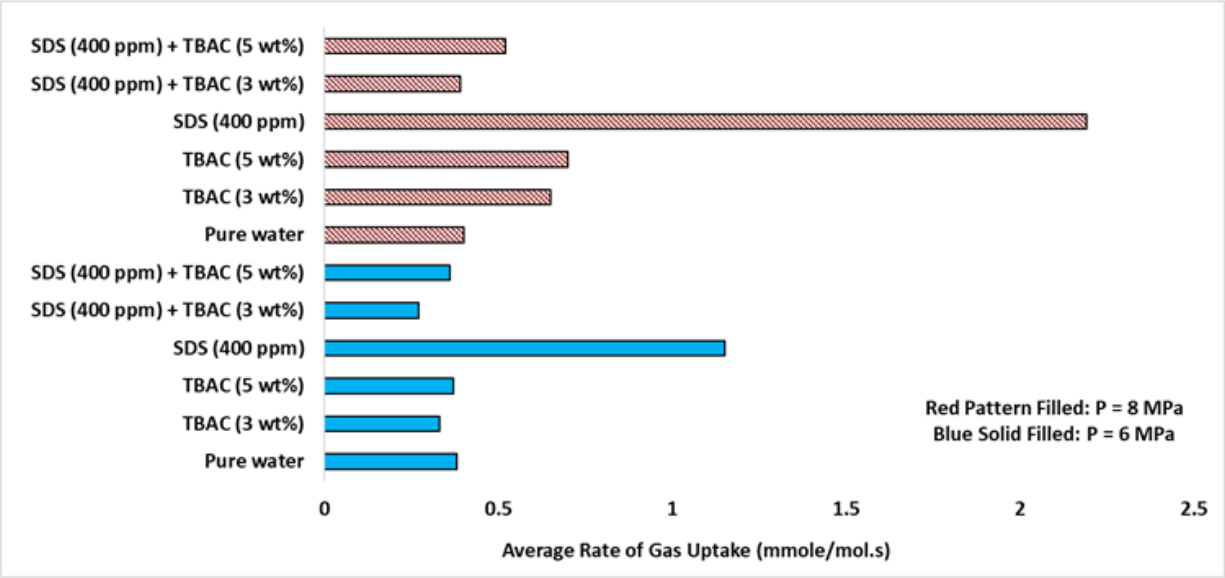


Fig 8. Average growth rate of double (TBAC+methane) semiclathrate hydrate during a 50-minute timeframe of the process at 278.15 K

Fig 6 and 7 compare the growth rates of (TBAC + methane) semiclathrate hydrate under the influence of TBAC and SDS+TBAC. The results revealed that in SDS+TBAC solutions, the growth rate of TBAC+methane semiclathrate hydrate is lower than when only TBAC is present.

Fig. 8 illustrates the impact of various concentrations of used additives (SDS and TBAC) on the average growth rate of formed hydrate at 278.15 K and P_0 = 6 and 8 MPa. From the figure, it is evident that using SDS (400 ppm) yields the most significant improvement in the rate of gas uptake. However, when SDS and TBAC are present together, the average rate of methane uptake is lower than that observed in the both SDS-only TBAC-only solutions.

4. CONCLUSION

The influence of SDS on the average growth rate of (TBAC+methane) semiclathrate hydrate within a 50-minute period was investigated. Experimental results obtained at 278.15 K and P_0 = 6 and 8 MPa demonstrated that the addition of 3 wt% and 5 wt% TBAC led to an increase in the average rate of methane uptake. However, when both TBAC and SDS were used simultaneously, their interaction resulted in a negative effect on the rate of methane uptake compared to the use of the aqueous solution of SDS alone.

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