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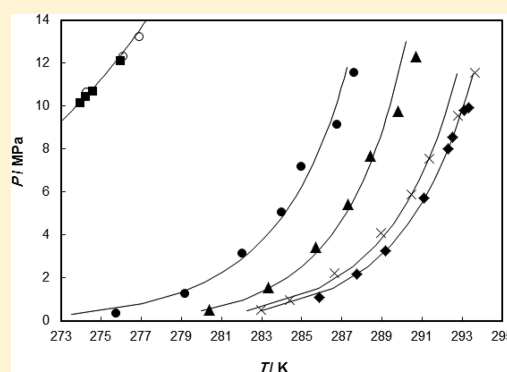
Experimental Measurement and Thermodynamic Modeling of Hydrate Dissociation Conditions for the Argon + TBAB + Water System

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ABSTRACT: Gas hydrate (clathrate hydrate) dissociation data at low to moderate pressures are necessary for practical applications, e.g., for gas separation. However, it is often known or recognized that the separation conditions encountered exist at high pressures and low temperatures. One solution to this problem is the application of the quaternary ammonium salts (QAS) semiclathrate hydrates such as tetra-*n*-butyl ammonium bromide (TBAB) which can moderate the hydrate dissociation conditions. The addition of TBAB to the solution enables a shift in the pressure-temperature diagrams to lower pressures and higher temperatures. In this study, the isochoric pressure search method^{1,4,13,28,34–37} was used to measure hydrate dissociation conditions for the argon + TBAB + water system. The measurement of hydrate dissociation conditions were performed at various TBAB concentrations, namely, (0, 0.05, 0.10, 0.20, and 0.30) mass fraction. The experiments were performed in the temperature range of (274 to 293.6) K and pressures ranging from (0.34 to 12.32) MPa. Results indicate that TBAB has a significant promotion effect on argon hydrate formation. In addition, increasing the TBAB concentration increases its promotion effect. A thermodynamic model was developed to predict the hydrate dissociation conditions for the system of argon + TBAB + water. The results show good agreement between the experimental measurements and model results.



1. INTRODUCTION

Gas hydrates, or clathrate hydrates, are crystalline, non-stoichiometric solids composed of water and some molecules of suitable size known as hydrate formers, which are formed at appropriate conditions of temperature and pressure (typically, high pressure and low temperature).^{1–40} The guest molecules are trapped in the cavities of hydrogen-bonded water molecules and do not participate in the hydrate lattice structure.¹ Gas hydrates are a subset of the compounds called clathrates which means barrier and indicates crystalline inclusion.^{1,22–25,30,31} Depending on the size of the hydrate former, and pressure and temperature conditions, gas hydrates can be classified into three typical types of structures: structure I (sI), structure II (sII) and structure H (sH).^{1–40}

This crystalline compound is usually found in gas transportation systems and pipelines for petroleum processing and creates serious economic and operational problems.¹ However, clathrate hydrates also have many positive applications in the industries such as gas storage and transportation, carbon dioxide capturing, gas separation, air-conditioning systems, water desalination technology, and concentration of dilute aqueous solutions.^{1,4–6,10–12,20,21,39,40}

The major problems in the implementation of gas hydrate technology for applications in industries are their slow formation rate and the high pressure dissociation conditions.^{1,5,6,16} In gas hydrate formation processes, one of the common ways to moderate the gas hydrate equilibrium conditions is using gas hydrate promoters. The usual hydrate promoters may be categorized into two groups: water-soluble and water-insoluble promoters.^{1,6}

Water-insoluble promoters may include some heavy hydrocarbons (alkanes, alkenes and alkynes) such as cyclohexane (CH), cyclopentane (CP), cyclobutane (CB), methylcyclohexane (MCH), methylcyclopentane (MCP), cycloheptane (CHP), 1,4-dimethylcyclohexane, 2,2-dimethylbutane, and cyclooctane (CO), which when placed in the large cavities of structure H and II hydrates, create a shift in the equilibrium conditions to the lower pressures.^{1–3,5,7,9,11,19} However, these promoters have some undesirable properties such as toxicity, volatility, and flammability in a hydrate-based process.^{1–3,5–7,9,11,19}

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Water-soluble promoters may be classified into two groups of kinetic and thermodynamic promoters.^{1–3,5,7,9,11,19} Sodium dodecyl sulfates (SDS) or kinetic promoters which were introduced by Zhong and Rogers¹⁶ have almost no effect on gas hydrate equilibrium phase data (T – P) and promote the rate of hydrate formation depending on their concentrations. Zhong and Rogers¹⁶ reported that a SDS solution with a concentration of 284 ppm would enhance the ethane hydrate formation rate about 700 times more than that of the pure water.¹⁶

Water-soluble thermodynamic promoters are additives that shift the equilibrium conditions of hydrate formation to higher temperatures or lower pressures. These types of promoters may be classified into two types of molecules: organic additives (e.g., tetrahydrofuran (THF), acetone, and 1,4-dioxane and 1,3-dioxalane)^{1–3,5–7,9,11,17–21} which do not participate in the hydrate cavities, but rather affect the activity of water and, the group of (mainly) quaternary ammonium salts (QAS) which produce semiclathrate hydrates and change the structure of water cages in the traditional clathrate hydrates.^{4,13,15,22–31,33,39,40}

Quaternary ammonium salts such as tetra-*n*-butyl ammonium bromide (TBAB), tetra-*n*-butyl ammonium chloride (TBAC), tetra-*n*-butyl ammonium fluoride (TBAF), and tetra-*n*-butyl phosphonium bromide (TBPB) were discovered by Fowler et al.²² These promoters can form semiclathrate hydrate crystals without any guest molecule at atmospheric pressure.^{23–25} Owing to the low equilibrium pressure conditions of the gas hydrates of QASs, these additives can be used in processes such as storage and transportation of natural gas and cold storage application in air conditioning systems. For instance, there are some studies on the application of pure semiclathrate hydrates of QAS (water + QAS hydrate system) in air conditioning systems as a Phase Change Material (PCM).^{15,26,27} Semiclathrate hydrates have the same properties as conventional clathrate hydrates; however, the difference lies in their structures where for semiclathrate hydrates the guest molecule not only occupies the hydrate cage but contributes to the water lattice structure.^{23–25,30,31} In TBAB semiclathrate hydrates, the positive/cation portion of the TBA salt can be found in the center of four cavities (two tetrakaidecahedra and two pentakaidecahedra) while the bromide anion, Br[–], forms hydrogen bonds with the water molecules and hence participates in the water lattice.³¹ When low molecular weight gases such as nitrogen, methane, and carbon dioxide are present in the system, the empty dodecahedral cavities are occupied by these small molecules. This kind of structure leads to stronger hydrogen bonding and as a result higher stability when compared with the conventional clathrate hydrate structure.³¹ The tetrabutyl ammonium bromide salt can form five different clathrate hydrate structures with 24, 26, 32, 36, and 38 hydration numbers.³⁰

In this study, the dissociation data of hydrates for the system of argon + TBAB aqueous solutions at concentrations of TBAB of (0, 0.05, 0.10, 0.20, and 0.30) mass fractions were measured. A thermodynamic model based on the models of Chen and Guo³² and Joshi et al.³³ was applied to predict the hydrate dissociation conditions for the system of argon + aqueous solutions of TBAB.

2. EXPERIMENTAL SECTION

2.1. Materials. The details of the purity and supplier of the chemicals used in this study are reported in Table 1. In all experiments, Ultrapure Millipore Q water with an electrical

Table 1. Details of the Chemicals Used in This Study

chemical	supplier	purity
argon	Air Liquide	0.999 mol fraction
TBAB	Sigma-Aldrich	more than 0.99 mass fraction
^a water		

^aUltrapure Millipore Q water with an electrical resistivity of 18 MΩ-cm was used in all experiments.

resistivity of 18 MΩ-cm was used to prepare the desired TBAB aqueous solutions which were (0.05, 0.10, 0.20, and 0.30) mass fraction TBAB. TBAB aqueous solutions were prepared using the gravimetric method with an analytical balance (Mettler Toledo Balance, model no. AB204-S) with an uncertainty of ± 0.0001 g.

2.2. Equipment. The schematic diagram of the experimental apparatus used in this study is shown in Figure 1. The main part of the experimental setup was a high-pressure equilibrium cell constructed from stainless steel with an internal volume of approximately 40 cm³. The cell can withstand pressures up to 16 MPa. A mechanical stirrer placed at the top of the equilibrium cell agitates the gas and aqueous solution inside the cell. A thermostatic temperature bath controller, model TFX200 supplied by PolyScience, was used to control the temperature of the bath fluid housing the equilibrium cell. This temperature controller includes an immersion circulator pump with an internal temperature probe to heat or cool the bath liquid. In addition, an immersion cooler supplied by PolyScience which consists of an evaporator, condenser, compressor, and throttling valve is used for cooling the liquid bath. A platinum resistance thermometer (Pt-100) was used to measure the equilibrium temperature. A standard temperature calibration unit, CTH 6500, supplied by WIKA with an uncertainty of ± 0.03 K for the temperature range of 73.15 K to 473.15 K was used to calibrate the temperature sensors. The combined uncertainty in the temperature measurement was 0.05 K. The pressure of the cell was measured using a calibrated WIKA pressure transducer with a pressure limit of 25 MPa and a combined measurement uncertainty of ± 0.01 MPa. The pressure and temperature of the equilibrium cell were monitored and logged using an Agilent data acquisition unit.

2.3. Experimental Procedure. In this study, the isochoric pressure search method which has been proven in a number of previous studies^{1,4,13,28,34–37} was used to measure the hydrate dissociation conditions for the system of argon + aqueous solutions of TBAB. At the start of the measurement, the cell was evacuated using a vacuum pump over a period of 30 min to remove the air and any contaminants inside the cell from previous experiments; then approximately 16 cm³ of the desired aqueous solutions of TBAB was filled into the cell. The cell was again evacuated quickly to remove the air. Then the equilibrium cell was immersed into the temperature controlled bath, and the temperature was set far from the hydrate zone. Later, by introducing argon from its cylinder into the cell, the pressure of the cell was increased until the desired pressure was achieved. The stirrer was turned on and set to a speed of 600 rpm to mix the gas and aqueous solution inside the cell. After the pressure had stabilized in the cell, the temperature was slowly decreased and hydrate formation was determined by the sharp drop in the pressure reading. The temperature of the equilibrium cell was then increased step-by-step to dissociate the hydrate. A step-change of 0.1 K per hour was used to increase the temperature of the cell until decomposition of the total semiclathrate

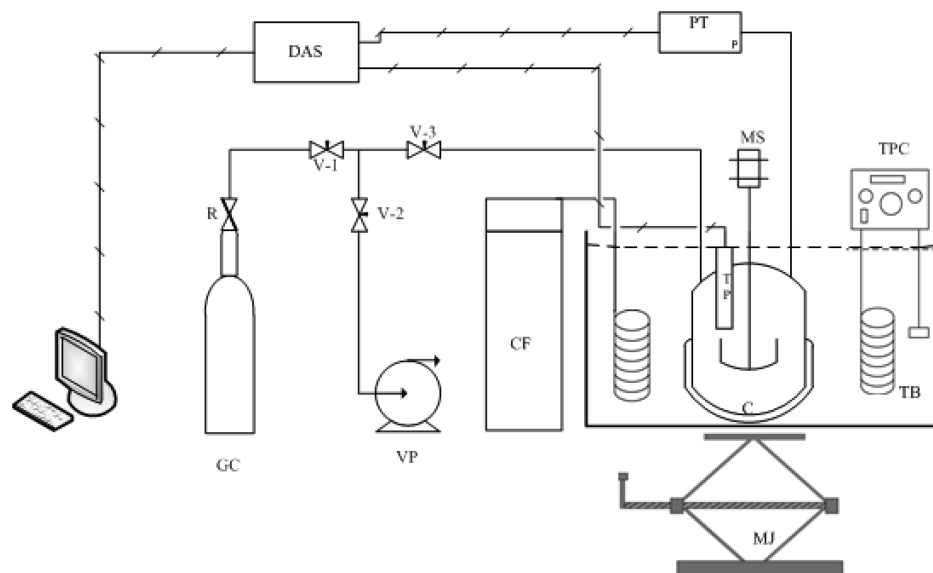


Figure 1. Schematic diagram of the apparatus used in this study: C, cell; CF, coldfinger; DAS, data acquisition system; GC, gas cylinder; MJ, mechanical jack; MS, mechanical stirrer; PT, pressure transmitter; R, regulator; TB, thermostated bath; TP, temperature probe; TPC, temperature programmable circulator; Vi, valve; VP, vacuum pump.

hydrate. For certain mixtures with higher concentrations of the salt, the interval time after the heating step was increased to 5 h to allow the system to reach an equilibrium pressure. During the experiment, both the temperature and pressure readings of the cell were recorded. Figure 2 shows an example of the

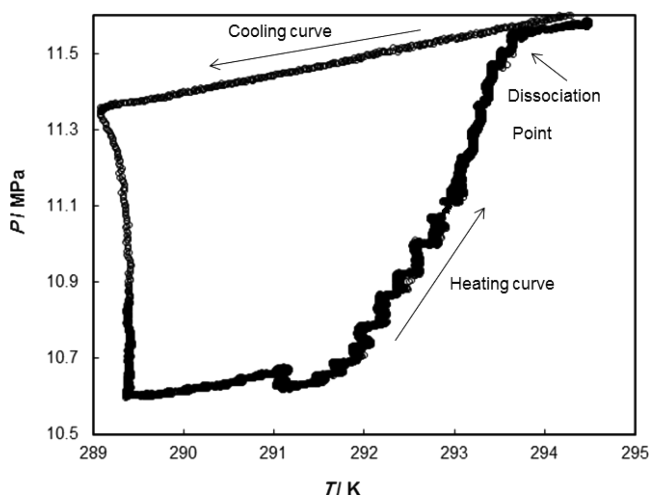


Figure 2. Primary cooling and heating curve for semiclathrate hydrate of argon + 0.20 mass fraction aqueous solutions of TBAB.

heating curve for the determination of the hydrate dissociation point. As observed in this figure, the point at which the slope of the pressure–temperature plot changes severely was determined as the hydrate dissociation point.

3. THERMODYNAMIC MODELING

In this study, phase equilibria of hydrates of argon in aqueous solutions of TBAB were modeled on the basis of the approach of Joshi et al.³³ The model of Joshi et al.³³ was an extension of the work of Chen and Guo³² for predicting phase equilibria of hydrates of CH₄, CO₂, and N₂ in aqueous solutions of TBAB. In the approach of Chen and Guo,³² hydrate formation consists

of two steps. First, a stoichiometric basic hydrate is formed through a quasi-chemical reaction where the dissolved gas molecules in water form unstable clusters with a number of water molecules surrounding the guest molecule. Second, the low molecular weight gas molecules such as Ar, N₂, O₂, CH₄ which are dissolved in water, are trapped into the linked cavities. According to Chen and Guo,³² the equation below is applied for the prediction of gas hydrate phase equilibria:

$$f = f^0 (1 - \theta)^{\alpha'} \quad (1)$$

where f and f^0 are described as the fugacity of gas species in the gas phase and in equilibrium with the unfilled basic hydrate, respectively. In eq 1, α' and θ are defined as

$$\alpha' = \frac{\lambda_1}{\lambda_2} \quad (2)$$

$$\theta = \frac{Cf}{1 + Cf} \quad (3)$$

where λ_1 and λ_2 are the number of linked cavities per water molecule and the number of gas molecules per water molecule in the basic hydrate, respectively. Parameter, θ , in eq 3 refers to the cage occupancy in which f and C represent the fugacity of the small guest molecule (argon in this study) in the gas phase and the Langmuir constant, respectively.

Equation 4 is used to evaluate the Langmuir constants for the guest–host interaction:^{1,36,38}

$$C = \frac{4\pi}{kT} \int_0^\infty \exp\left[-\frac{w(r)}{kT}\right] r^2 dr \quad (4)$$

In the above equation, T represents the temperature, r stands for the radius, k is the Boltzmann's constant and $w(r)$ is the overall cell potential which can be obtained from the Kihara potential function with a spherical core using the following equation:

$$w(r) = 2z\varepsilon \left[\frac{(\sigma^*)^{12}}{\bar{R}^{11}r} \left(\delta^{10} + \frac{\alpha}{\bar{R}} \delta^{11} \right) - \frac{(\sigma^*)^6}{\bar{R}r} \left(\delta^4 + \frac{\alpha}{\bar{R}} \delta^5 \right) \right] \quad (5)$$

where

$$\delta^{\bar{N}} = \frac{1}{\bar{N}} \left[\left(1 - \frac{r}{\bar{R}} - \frac{\alpha}{\bar{R}} \right)^{-\bar{N}} - \left(1 + \frac{r}{\bar{R}} - \frac{\alpha}{\bar{R}} \right)^{-\bar{N}} \right] \quad (6)$$

z is defined as the coordination number of the cavity, ε is the characteristic energy, and α is the radius of the spherical molecular core. \bar{R} symbolizes the cavity radius and \bar{N} is an integer, equal to 4, 5, 10, or 11. $\sigma^* = \sigma - 2\alpha$, where σ denotes the collision diameter. The Kihara potential parameters, α , ε , and σ , which are used in this study are presented in Table 2.

Table 2. Kihara Potential Parameters Used in This Study⁴⁵

compound	$(\alpha \cdot 10^{10})/\text{m}$	$(\varepsilon \cdot 10^{10})/\text{m}$	$(\sigma/k)\text{K}$
argon	0.184	2.9434	170.50

As previously mentioned, the semiclathrate hydrate of TBAB has a different structure compared with the traditional structures I and II in clathrate hydrates. There are two types of semiclathrate structures, type A and B, depending on the concentration of TBAB and the hydrate numbers.^{26,33,39,40} Shimada et al.³¹ and Oyama et al.²⁶ stated that the hydration numbers for type A and B are 26 and 38, respectively. In additions, for systems with a concentration of TBAB above 0.18 mass fraction, type A is more stable and has a higher melting point than type B and vice versa.^{8,26,33,39,40} At concentrations below 0.18 mass fraction of TBAB, type B is more stable compared to type A.^{8,26,33,39,40} The properties of type A and B include the hydrate number, number of linked cavities, and number of gas molecules, as well as λ_1 , λ_2 , and α' for a unit cell of TBAB semiclathrate hydrate. The information is summarized in Table 3.^{8,33}

Table 3. Properties of Type A and B for a Unit Cell of TBAB Semiclathrate Hydrate³³

	type B	type A
hydrate number	38	26
number of linked cavity ³³	1.75	1.65
number of gas molecule	3	3
λ_1^a	1.75/38	1.65/26
λ_2^b	3/38	3/26
α'^c	1.75/3	1.65/3

^aNumber of linked cavities per water molecule. ^bNumber of gas molecules per water molecule. ^cNumber of linked cavities per number of gas molecules.

The following equation is used to evaluate the fugacity of gas species in equilibrium with the unfilled basic hydrate (f^0):

$$f^0 = f^0(T) f^0(P) f^0(a_w) \quad (7)$$

where

$$f^0(T) = A' \exp\left(\frac{B'}{T - C'}\right) \quad (8)$$

$$f^0(P) = \exp\left(\frac{\beta P}{T}\right) \quad (9)$$

$$f^0(a_w) = a_w^{-1/\lambda_2} \quad (10)$$

In the above equations, P and a_w represent the pressure and water activity, respectively. The Antoine constants for argon, A' , B' , and C' presented by Chen and Guo³² were used to evaluate the $f^0(T)$ for argon in eq 8. The β structural constant in eq 9 depends on the weight fraction of TBAB.³³ Table 4

Table 4. Structural Parameter β for Semiclathrate of TBAB³³

TBAB mass fraction	structural parameter, $(\beta \cdot 10^5)/\text{K} \cdot \text{Pa}^{-1}$
0.05	0.697
0.10	0.450
0.20	0.350
0.30	0.300

presents β constants for all the ranges of TBAB concentrations applied in this study.³³ It should be noted that α' and β (structural parameters) depend only on the concentration of TBAB and are independent of the type of the guest molecule.³³

The following equation is applied for the calculation of the activity of water which depends on the concentration of TBAB in the aqueous phase as well as the type of guest molecule:³³

$$a_w = \frac{1}{1 - x_{\text{TBAB}}} \exp\left(\frac{k_1}{T} - k_2\right) \quad (11)$$

where x_{TBAB} is the mole fraction of TBAB and, k_1 and k_2 are guest (gas) dependent parameters.

The Valderrama modification of the Patel and Teja equation of state (VPT EoS)⁴¹ accompanied by the nondensity-dependent (NDD) mixing rule⁴² were used to predict the fugacity of argon in the gas phase (f parameter in eq 1). Details of the EoS and mixing rule used can be found elsewhere.^{2,36,37,41,42} The acentric factors (ω) and critical properties P_c , T_c , and Z_c for argon are reported in Table 5.

Table 5. Values for Acentric Factors (ω) as Well as Critical Temperatures (T_c), Pressures (P_c), and Compressibility Factors (Z_c) Used in This Study⁴⁶

component	T_c/K	P_c/MPa	ω	Z_c
argon	150.86	4.898	0.0	0.291

4. RESULTS AND DISCUSSION

To test the reliability of the experimental setup and the experimental procedure used in this study, four dissociation data points for argon simple hydrate were measured. As seen in Figure 3, there is a good agreement between the experimental data generated in this study and the data reported in the literature.⁴³

The results of the measurements of the hydrate dissociation conditions for the system of argon + aqueous solutions of TBAB ((0, 0.05, 0.10, 0.20 and 0.30) mass fraction of TBAB) are shown in Figure 3 and Table 6. As can be seen in Figure 3, TBAB has a drastic promotion effect on the argon hydrates. This promotion effect of TBAB on the argon hydrate formation is an advantage for the practical application of hydrate in gas separation technologies compared with processes using only the conventional clathrate hydrate method which is in the presence of pure water.

Also, evident in Figure 3 is that an increase in the concentration of TBAB from (0.05 to 0.20) mass fraction,

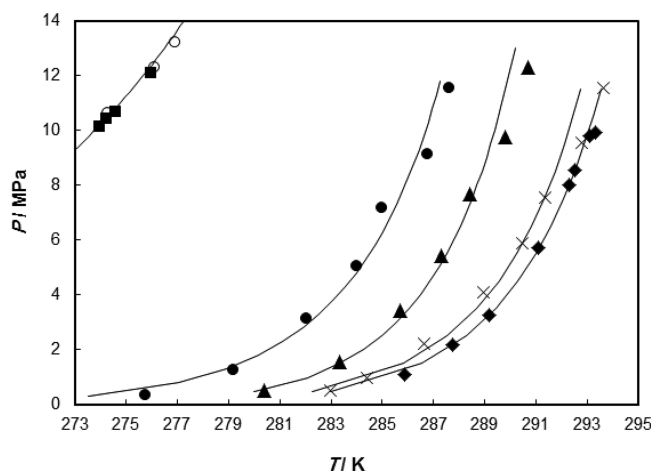


Figure 3. Experimental data and estimation of hydrate dissociation conditions for the system of argon + TBAB + water at various TBAB concentrations. Symbols represent experimental data: ●, this work, 0.05 mass fraction TBAB aqueous solution; ▲, this work, 0.10 mass fraction TBAB aqueous solution; ×, this work, 0.20 mass fraction TBAB aqueous solution; ◆, this work, 0.30 mass fraction TBAB aqueous solution; ■, this work, pure argon; ○, Marshal et al.,⁴³ pure argon; solid lines, this work, model results.

increases the promotion effects of TBAB on the argon hydrate formation significantly. Furthermore, as seen in Figure 3, the effects of TBAB on the promotion of the argon hydrate at the TBAB concentrations of (0.20 and 0.30) mass fractions at high pressures is almost identical. However, at pressures lower than 7.5 MPa, a solution of 0.30 mass fraction of TBAB has a greater promoting effect on the argon hydrate compared to 0.20 mass fraction of TBAB. To obtain the optimal concentration of TBAB for a separation process based on semiclathrate hydrates, some economic studies are essential.

In addition, the experimental results show that a small increase in the hydrate equilibrium temperature causes a large increase in the hydrate equilibrium pressure. Therefore, the measurement of the hydrate dissociation conditions should be done very slowly and carefully in order to prevent the generation of erroneous experimental data.⁴ For this reason, a step-change of 0.1 K per hour was used to increase the temperature of the cell and an adequate interval time (about 5 h) at each temperature step was taken during the hydrate heating procedure.

As clearly illustrated in Figure 3 and Table 6, there is a good agreement between the model predictions and the reported experimental measurements. The following objective function, F_{obj} , was minimized to obtain the optimum constants (k_1 and k_2) in eq 11 for the calculation of the water activity in the system of hydrates of argon + aqueous solutions of TBAB:

$$F_{obj} = \left(\frac{1}{NDP} \right) \sum_{k=1}^{NDP} \left(\frac{|T_{exp} - T_{cal}|}{T_{exp}} \right) \quad (12)$$

where NDP is the number of data points and, T_{exp} and T_{cal} refer to the experimental and calculated hydrate dissociation temperatures, respectively. Table 7 reports the constants (k_1 and k_2) in eq 11 for each concentration of TBAB. Since the available thermodynamic modeling in the literature for the prediction of the semiclathrate hydrate dissociation conditions are scarce,^{33,44} the thermodynamic modeling applied in this

Table 6. Experimental and Predicted Hydrate Dissociation Conditions for the System of Argon + Aqueous Solutions of TBAB

TBAB mass fraction ^a	T_{exp}/K	P_{exp}/MPa	T_{cal}/K	AD/K^b	$ARD/\%^c$
0	276.0	12.08	275.7	0.3	0.1
	274.6	10.66	274.4	0.2	0.1
	274.2	10.42	274.2	0.0	0.0
	274.0	10.12	273.9	0.1	0.0
	287.6	11.56	287.2	0.4	0.1
0.05	286.8	9.12	286.4	0.4	0.1
	285.0	7.17	285.5	0.5	0.2
	284.0	5.06	284.2	0.2	0.1
	282.0	3.16	282.4	0.4	0.1
	279.2	1.27	278.8	0.4	0.1
	275.8 ^d	0.34	273.9	1.9	0.7
	290.7	12.32	290.0	0.7	0.2
	289.8	9.76	289.3	0.5	0.2
	288.4	7.68	288.6	0.2	0.1
	287.3	5.43	287.5	0.2	0.1
0.10	285.7	3.44	286.0	0.3	0.1
	283.3	1.56	283.5	0.2	0.1
	280.4	0.51	280.0	0.4	0.1
	293.6	11.55	292.8	0.8	0.3
	292.8	9.57	292.1	0.7	0.2
	291.3	7.55	291.3	0.0	0.0
	290.5	5.90	290.5	0.0	0.0
	288.9	4.11	289.3	0.4	0.1
	286.6	2.22	287.1	0.5	0.2
	284.4	0.98	284.4	0.0	0.0
0.20	283.0	0.51	282.3	0.7	0.2
	293.3	9.91	293.0	0.3	0.1
	293.1	9.82	293.0	0.1	0.0
	292.5	8.54	292.5	0.0	0.0
	292.3	8.00	292.3	0.0	0.0
	291.1	5.74	291.1	0.0	0.0
	289.2	3.27	289.2	0.0	0.0
	287.8	2.19	287.8	0.0	0.0
	285.9	1.09	285.5	0.4	0.1
	285.9	1.09	285.5	0.4	0.1
0.30	293.3	9.91	293.0	0.3	0.1
	293.1	9.82	293.0	0.1	0.0
	292.5	8.54	292.5	0.0	0.0
	292.3	8.00	292.3	0.0	0.0
	291.1	5.74	291.1	0.0	0.0
	289.2	3.27	289.2	0.0	0.0
	287.8	2.19	287.8	0.0	0.0
	285.9	1.09	285.5	0.4	0.1
	285.9	1.09	285.5	0.4	0.1
	285.9	1.09	285.5	0.4	0.1

^aCombined standard uncertainty, u_c , in T , P , and mass fraction are $u_c(T_{exp}) = 0.05$ K, $u_c(P) = 0.01$ MPa, and $u_c(x_{TBAB}) = 0.0002$, respectively. ^bAbsolute deviation = $|T_{exp} - T_{cal}|$. ^cARD/% = $(|T_{exp} - T_{cal}|/T_{exp}) \cdot 100$, absolute relative deviation. ^dLikely represents hydrate of TBAB + water.

Table 7. Constants (k_1 and k_2) for the Water Activity Calculations in the System of Hydrates of Argon + Aqueous Solutions of TBAB

TBAB mass fraction	k_1	k_2
0.05	1130.704	3.591
0.10	1704.706	5.529
0.20	2334.051	7.402
0.30	2335.526	7.383

study can be useful for modeling other semiclathrate hydrate systems.

5. CONCLUSIONS

In this study, the isochoric pressure search method was used to measure hydrate dissociation data for the system of argon + aqueous solutions of TBAB at various concentrations of TBAB ((0, 0.05, 0.10, 0.20 and 0.30) mass fraction of TBAB). The results show that TBAB has a drastic promotion effect on the

formation of argon hydrates. In addition, with an increase in the concentration of TBAB, the promotion effect of TBAB on the hydrate dissociation conditions is increased (in the concentration ranges studied in the present work). A thermodynamic model based on the work of Chen and Guo³² and Joshi et al.³³ was applied to predict the hydrate dissociation conditions for the system of argon + aqueous solutions of TBAB. New constants for calculating the water activity in the system of hydrates of argon + aqueous solutions of TBAB were obtained.

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Notes

The authors declare no competing financial interest.

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