

Synthesis and Evaluation of Interfacial Properties and Carbon Capture Capacities of the Imidazolium-Based Ionic Liquid Surfactant

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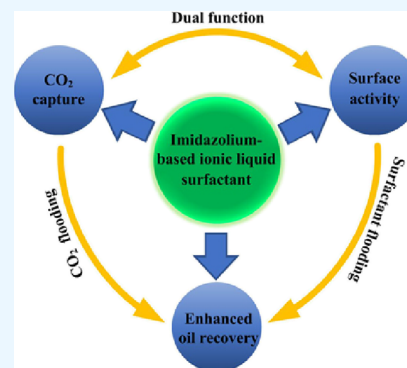
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ABSTRACT: Ionic liquid as a chemical flooding agent has broad application prospect in enhancing oil recovery. In this study, a bifunctional imidazolium-based ionic liquid surfactant was synthesized, and its surface-active, emulsification capacity, and CO₂ capture performance were investigated. The results show that the synthesized ionic liquid surfactant combines the characteristics of reducing interfacial tension, emulsification, and CO₂ capture. The IFT values for [C₁₂mim][Br], [C₁₄mim][Br], and [C₁₆mim][Br] could decrease from 32.74 mN/m to 3.17, 0.54, and 0.051 mN/m, respectively, with increasing concentration. In addition, the emulsification index values are 0.597 for [C₁₆mim][Br], 0.48 for [C₁₄mim][Br], and 0.259 for [C₁₂mim][Br]. The surface-active and emulsification capacity of ionic liquid surfactants improved with the increase in alkyl chain length. Furthermore, the absorption capacities reach 0.48 mol CO₂ per mol of ionic liquid surfactant at 0.1 MPa and 25 °C. This work provides theoretical support for further CCUS-EOR research and the application of ionic liquid surfactants.



1. INTRODUCTION

In the process of chemical flooding, the effect of spontaneous emulsification and oil–water interfacial tension reduction of oil-displacing agents with residual oil and residual oil can markedly improve oil recovery.^{1–3} As the key component of oil-displacing agents, the surfactant plays an irreplaceable role.^{4,5} Presently, the research of enhanced oil recovery by ionic liquid surfactants is gaining progressively more attention, and the understanding of its mechanism is also advancing steadily.^{6,7} One is that ionic liquid is a new type of green solvent, which is completely composed of ions.⁸ Second, the physicochemical properties of ionic liquid surfactants can be controlled by designing and selecting ionic structures. Ionic liquids have played an important role in organic synthesis, catalysis, crude oil dissolution, oil–water interfacial tension reduction, and so on.⁹ The imidazolium-based ionic liquid surfactant falls into a cross field of functional ionic liquids and surfactants, which covers an extensive range and has become one of the most widely studied surfactants at present.¹⁰

In recent years, various studies have been conducted around the application of ionic liquids to the oil and gas industry. It has been proved that ionic liquids have similar surface activity to surfactants and can form micelles in aqueous solutions.^{11,12} Ionic liquids show favorable interfacial behavior, thus lowering the oil–water interfacial tension to a significant value and promoting oil–water emulsification.¹³ Low oil–water interfacial tension, emulsification, and wettability alteration owing to adsorption are considered to be important factors for enhanced oil recovery.¹⁴ Evans et al.¹⁵ first investigated the critical micelle concentrations and micellar aggregation

numbers of ionic liquid surfactants in ethyl ammonium nitrate by classical and quasi-elastic light scattering. Merrigan et al.¹⁶ synthesized four kinds of imidazolium-based ionic liquid surfactants containing fluorine atoms, and the results showed that these imidazolium-based ionic liquid surfactants had the same molecular aggregation behavior as conventional surfactants. Baltazar et al.¹⁷ prepared 18 different monocationic and dicationic imidazolium-based ionic liquids and tested their critical micelle concentration (CMC) values and surface activities. The experimental results showed that the CMC values of imidazolium-based ionic liquids decreased moderately with the increase in alkyl chain length, and dicationic imidazolium-based ionic liquids exhibited much higher efficiencies at lowering the surface tension than monocationic imidazolium-based ionic liquids. Ao et al.¹⁸ explored the surface activity and thermodynamic properties of a series of imidazolium ionic liquid surfactants with four-methylene spacer groups. Results suggested that the surface activity of gemini imidazolium-based ionic liquid surfactants was higher than that of their corresponding monomers, and the hydrophilic groups were unfavorable for micellization, while the methylene units of the hydrophobic groups benefited

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micellization for imidazolium-based ionic liquid surfactants. Nandwani et al.¹⁹ found that when the carbon number of the alkyl side chain of imidazolium-based ionic liquid surfactants was consistent with that of traditional cationic surfactants, the critical micelle concentration of the former was significantly lower than that of the latter and showed higher adsorption efficiency, which could better reduce the surface tension. That is, the surface activity of imidazolium-based ionic liquid surfactants is superior to that of traditional surfactants. Manshad et al.²⁰ explored wettability alteration, IFT reduction, and oil displacement performance by screening four ionic liquids, namely, $[C_{12}mim][Cl]$, $[C_{18}mim][Cl]$, $[C_8Py][Cl]$, and $[C_{18}Py][Cl]$. The results showed that $[C_{18}mim][Cl]$ was the optimum ionic liquid, and its oil displacement efficiency was 13% higher than that of brine flooding. In conclusion, amphiphilic ionic liquid surfactants, as potential novel types of cationic surfactants, possess great application prospect in the field of chemical flooding.

Significantly, ionic liquids have emerged as potential absorbents for CO_2 capture and separation due to their prominent characteristics, such as negligible vapor pressure, non-corrosiveness, non-toxicity, designability, high thermal stability, higher CO_2 solubility, and selectivity.^{21–23} In particular, imidazolium-based ILs have a strong affinity toward CO_2 .²⁴ Since Blanchard et al.²⁵ first found that CO_2 can be effectively dissolved in the ionic liquid 1-butyl-3-methylimidazolium hexafluorophosphate ($[Bmim][PF_6]$) at 298 K and 40 MPa, a large number of scientific studies on the absorption of CO_2 by ionic liquids have been conducted successively, including conventional ionic liquids with physically adsorbed and functionalized ionic liquids with chemical adsorption. According to Henry's law, physical absorption refers to the attachment of CO_2 molecules to the adsorbent in the form of physical action.

The chemical absorption method refers to the effect of absorption through the chemical reaction between the chemical adsorbent and carbon dioxide. Anthony et al.²⁶ found that for ionic liquids with the same anion, the presence of imidazole cations with longer alkyl chains is conducive to improving the solubility of CO_2 . Sharma et al.²⁷ investigated the absorption capacities of a series of amino-functionalized imidazolium cation-based ionic liquids with different anions for CO_2 . The results showed that amine-functionalized ionic liquids exhibited higher CO_2 absorption capacity, which is positively correlated with pressure and negatively correlated with temperature. As a result, the capture of CO_2 by imidazolium-based ionic liquids has developed into a fascinating area of research.

However, a unified study on the CO_2 capture performance and surface activity of ionic liquids has not been reported so far. Combining CO_2 capture with surfactant flooding can further improve oil displacement efficiency by injecting ionic liquid surfactants after CO_2 capture into the reservoir to take advantage of the synergistic effect of CO_2 and ionic liquid surface activity (Figure 1). In this work, a series of amine-functionalized imidazolium cationic ionic liquids with different alkyl chain lengths were synthesized, and the surface activity, emulsification ability, and carbon dioxide capture performance of ionic liquid surfactants were investigated. The purpose is to lay a foundation for further research and development of high-efficiency and low-cost bifunctional ionic liquid surfactants with both surface activity and carbon dioxide capture capability

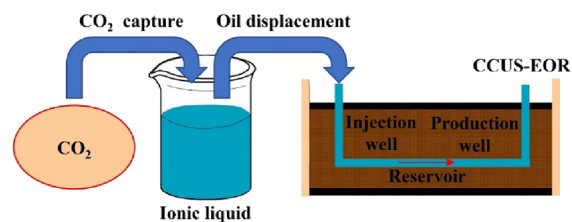


Figure 1. Oil displacement mechanism of the ionic liquid surfactant.

and to provide new ideas and strategies for carbon dioxide capture and oil displacement.

2. EXPERIMENTAL SECTION

2.1. Materials. The structures of imidazolium-based ionic liquid surfactants with different chain lengths $[C_nmim][Br]$, $n = 12, 14,$ and $16,$ which were synthesized in our laboratory, are shown in Figure 2. *N*-(3-Aminopropyl)-imidazole (purity,

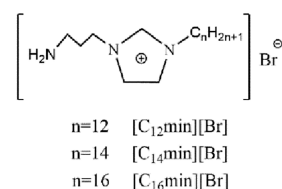


Figure 2. Chemical structures of imidazolium-based ionic liquid surfactants and their monomeric analogs.

>97), 1-bromododecane (purity, >98%), 1-bromotetradecane (purity, >98%), and 1-bromohexadecane (purity, >97%) were procured from Sigma-Aldrich Co. The reagents used such as isopropanol and ethyl acetate were purchased from Tianjin Kemiou Reagent Co. In all experiments, deionized water was used to configure the solution. Crude oil was obtained from one of CQ oil reservoirs. The viscosity of crude oil was 1.5 mPa·s at a reservoir temperature of 65 °C.

2.2. Methods. **2.2.1. Synthesis of Imidazolium-Based Ionic Liquid Surfactants.** According to the literature,^{28,29} this class of imidazolium-based ionic liquid surfactants was synthesized by mixing 0.1 mol of *N*-(3-aminopropyl)-imidazole and 0.15 mol of brominated *n*-alkanes (1-bromododecane, 1-bromotetradecane, or 1-bromohexadecane) in isopropanol (20 mL) and stirring for 24 h at 70 °C under nitrogen, and the reaction mechanism is shown in Figure 3.

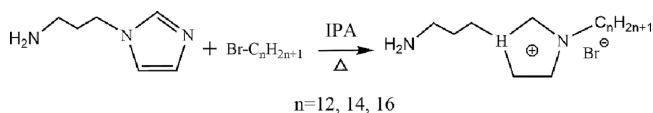


Figure 3. Reaction scheme for the synthesis of imidazolium-based ionic liquid surfactants.

After the reaction was finished, isopropanol and unreacted brominated *n*-alkanes were subsequently removed. The product was purified three times by recrystallization in ethyl acetate.

2.2.2. Critical Micelle Concentration. First, the aqueous solutions with different imidazolium-based ionic liquid surfactant concentrations were prepared. Then, the surface tension of prepared imidazolium-based ionic liquid surfactants was measured using an Easy Dyne K20 Tensiometer at 298 K. The surface tension of each solution was repeated three times,

and the average value was taken. The surface tension curve vs concentration of each imidazolium-based ionic liquid surfactant was plotted to determine the critical micelle concentration (CMC) of the individual imidazolium-based ionic liquid surfactants.

2.2.3. Interfacial Tension. The interfacial tension (IFT) between the imidazolium-based ionic liquid surfactant aqueous solution and crude oil was investigated using a JJ2000B tensiometer by a spinning drop method with a precision of 0.02 mN/m, and the temperature was maintained at 298 K.

2.2.4. Emulsification Capacity. The emulsification capacity of synthesized imidazolium-based ionic liquid surfactants was investigated using the high-temperature emulsification characterizer THE-II.³⁰ First, the dynamic emulsified oil ratio (E_0) between ionic liquid surfactants and crude oil was measured by the high-temperature emulsifying characterizer THE-II. The typical emulsification process is illustrated in Figure 4. The

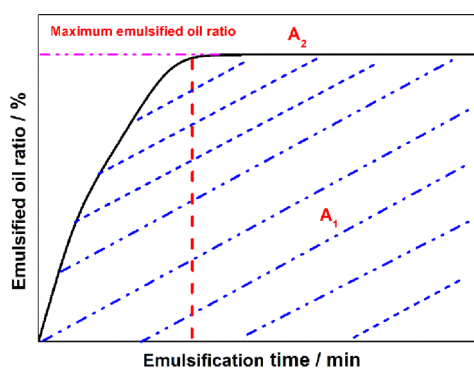


Figure 4. Typical dynamic emulsified oil rate curve.

dynamic emulsified oil ratio curve vs emulsification time was plotted for the determination of the emulsification index of the individual imidazolium-based ionic liquid surfactants, as shown in Figure 4. The emulsification index (EI) refers to the ratio of area A1 surrounded by the dynamic emulsified oil ratio curve and time axis to area A2 surrounded by the maximum emulsified oil ratio and time axis, as shown in eq 1. The emulsification index takes full account of the emulsification speed and emulsification amount, and the emulsification capacity has a positive correlation with the emulsification index.

$$EI = \frac{A_1}{A_2} = \frac{\int_0^{t_e} E_0(t) dt}{E_{\text{omax}} \times t_e} \quad (1)$$

2.2.5. CO₂ Absorption Performance. An ionic liquid surfactant was prepared as an aqueous solution for CO₂ absorption. The CO₂ absorption performance was investigated by the devices shown in Figure 5. In a specific experiment, about 10.0 g ionic liquid surfactant solution was loaded into the absorption chamber, the air in the system was removed by a vacuum pump, CO₂ was charged into the absorption chamber from a CO₂ reservoir, and the ionic liquid surfactant solution was stirred. The mass change of ionic liquid surfactant solution after absorbing CO₂ was recorded by weighing at fixed intervals. The sample solution was considered to have reached absorption equilibrium if the mass of the ionic liquid surfactant solution remained constant over 1 h. The molar fraction (η) of

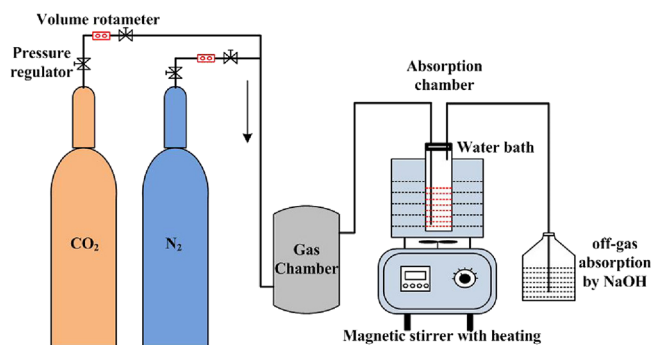


Figure 5. Schematic diagram of the CO₂ absorption apparatus.

CO₂ absorbed by ionic liquid surfactant solution was calculated by the following eq 2.

$$\eta = \frac{(m - m_0)/44}{m_1/M} \quad (2)$$

where m_0 is the initial mass of the absorption system (g), m is the mass of the absorption system during the adsorption process (g), M is the molar mass of the ionic liquid surfactant (g/mol), and m_1 represents the mass of the ionic liquid surfactant.

3. RESULTS AND DISCUSSION

3.1. Analysis of Critical Micelle Concentration. In the field of chemical enhanced oil recovery (EOR), the critical micelle concentration (CMC) is one of the crucial parameters for evaluating surface properties since it affects the oil displacement efficiency of surfactants in the reservoir. In general, the concentration of the injected surfactant solution is approximately two or three times the CMC value. Exploring the aggregation behavior of ionic liquid surfactants in solution through surface tension studies is highly vital to analyzing how ionic liquids will participate in complex systems as components. There are different methods to determine CMC, but the most common technique is to plot the surface tension versus surfactant concentration and to identify the turning point in the evolution curve as CMC.

The surface tension of ionic liquid surfactants was measured at different concentrations, and the results are shown in Figure 6. As shown in the plot, surface tension values initially decrease with the increase in ionic liquid surfactant concentration and do not decrease further after reaching a certain concentration. This specific concentration at which the surface tension remains steady is called CMC. Before reaching CMC, the dissolved ionic liquid surfactant molecules are uniformly dispersed as monomers, and the adsorption of ionic liquid surfactant molecules onto the air–water interface is in dynamic equilibrium. When the CMC of ionic liquid surfactants is attained, the surfactant molecules begin to aggregate to form micelles, minimizing free energy. Above CMC, the surface tension remains constant because the ionic liquid surfactant solution system is in a balanced state with an adsorbed monomolecular layer, free monomers, and micellized surfactant. From the surface tension plot, the minimum surface tension values of 31.02 mN/m for [C₁₆mim][Br], 32.04 mN/m for [C₁₄mim][Br], and 33.19 mN/m for [C₁₂mim][Br] were recorded, and the respective CMC values are 0.4, 2, and 9 mmol/L. The CMC value decreases in the order of [C₁₂mim][Br] > [C₁₄mim][Br] > [C₁₆mim][Br]. It is mainly

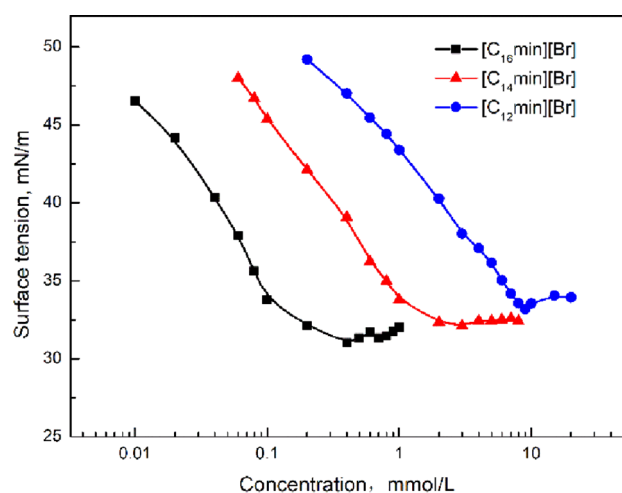


Figure 6. Effect of ionic liquid surfactant concentration on surface tension at 298 K and atmospheric pressure.

due to the increase in hydrocarbon chain length, and the hydrophobicity of ionic liquid surfactants is enhanced, which is conducive to the formation of micelles.³¹ Therefore, the longer the hydrophobic chain, the lower the CMC. It can be seen from the above studies that the micelle formation ability of $[C_{16}mim][Br]$ is superior to that of the other two ionic liquid surfactants.

3.2. Analysis of Interfacial Tension. Interfacial tension (IFT) reduction between crude oil and oil-displacing agents affects the oil recovery mechanism by causing a prominent increase in the capillary force that plays a crucial role in residual oil trapping in oil reservoirs.³² In chemical flooding, a larger capillary force is conducive to additional oil mobilization and lower residual oil saturation by reducing IFT. As an important parameter for the effective application of oil-displacing agents in EOR, many efforts have been made to further reduce the oil–water interfacial tension. Current technology can achieve very low IFTs (10^{-1} to 10^{-2} mN/m) or even ultra-low IFTs ($\leq 10^{-3}$ mN/m).

In this study, the IFT of the crude oil–water system was investigated, and the experimental results are shown in Figure 7. From Figure 7, it can be seen that the IFT values for $[C_{12}mim][Br]$, $[C_{14}mim][Br]$, and $[C_{16}mim][Br]$ decreased from 32.74 mN/m to 3.17, 0.54, and 0.051 mN/m, respectively, with increasing concentration. When the CMC of the ionic liquid was achieved, the IFT remained almost constant, and the IFT value rose slightly with the further increase in concentration. Therefore, the minimum IFT was obtained around the CMC of the ionic liquid surfactant. This is mainly because the IFT is related to the adsorption/desorption behavior of ionic liquid surfactant molecules at the oil–water interface. At CMC, the adsorption/desorption of the surfactant maintains a dynamic balance, resulting in a minimum value. It was also observed that the capacity of ionic liquid surfactants to reduce IFT became stronger with the increase in alkyl chain length. The reason is that the increase of methylene group in the cation structure leads to the increase in hydrophobicity, and the adsorption capacity of surfactant molecules at the oil–water interface is enhanced.³³ As a result, even a small amount of surfactant can significantly reduce the oil–water interfacial tension. Hezave et al.³⁴ also reported a similar conclusion in their study, that is, the capacity of the ionic liquid surfactants to reduce the oil–water interfacial

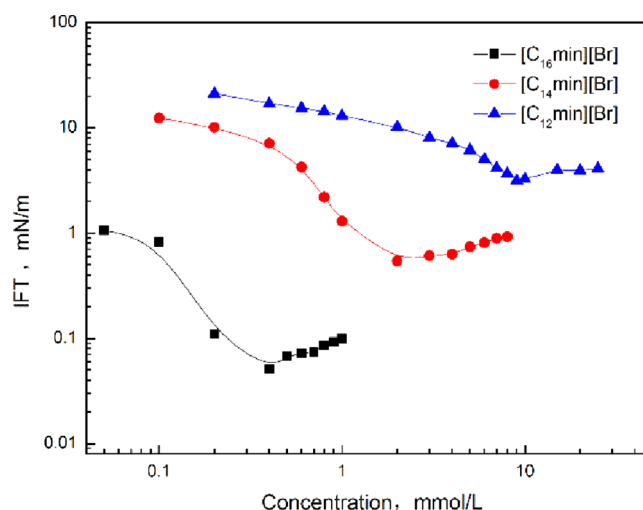


Figure 7. Effect of ionic liquid surfactant concentration on IFT between oil and water.

tension increases with increasing chain length, and the longer tails will result in a lower IFT at the same concentration.

3.3. Analysis of Emulsification Capacity. Emulsification of oil-displacing agents with crude oil constitutes was believed to be an important contribution to producing more oil.³⁵ One result of the emulsification is that the remaining oil and residual oil become more easily captured and displaced. Second, the in situ-formed emulsions enable the seal of the high-permeability areas efficiently, thus improving the oil displacement efficiency. In practical chemical flood, the emulsification capacity of crude oil–water mixture plays an essential role in remaining oil and residual oil displacing within porous media. Therefore, emulsification capacity is considered to be a vital parameter for screening surfactants for EOR application.

To explore the emulsification capacity of the synthesized ionic liquid surfactants, the dynamic emulsified oil ratio curves of each ionic liquid surfactant with crude oil were plotted respectively. The dosage of each ionic liquid surfactant was 0.5 mmol/L, and the corresponding emulsification experimental results are presented in Figure 8 and Table 1. As can be seen

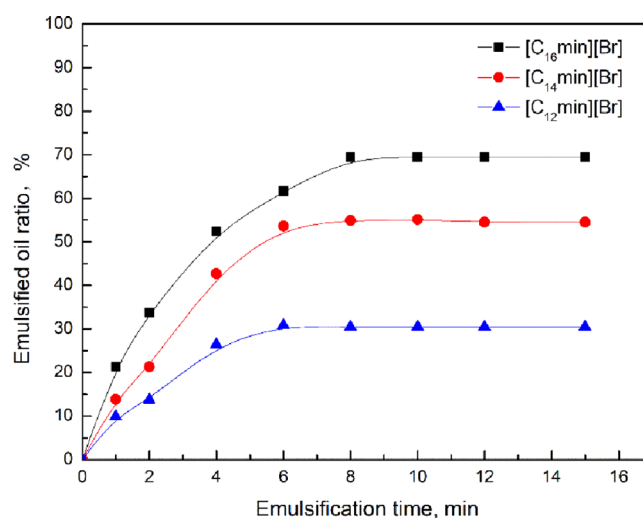


Figure 8. Dynamic emulsified oil ratio curves of ionic liquid surfactants.

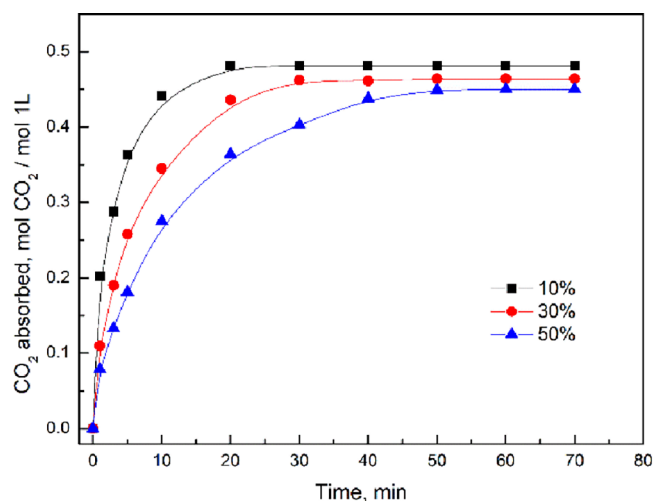
Table 1. Emulsification Results of Ionic Liquid Surfactants with Crude Oil

ionic liquid surfactant	emulsification time, min	emulsified oil ratio, %	EI
[C ₁₂ mim][Br]	6	30.5	0.259
[C ₁₄ mim][Br]	6	54.9	0.48
[C ₁₆ mim][Br]	8	69.4	0.597

from Figure 8 and Table 1, the emulsified oil ratio increases gradually with the extension of emulsification time until reaching the emulsification equilibrium. Furthermore, the maximum equilibrium emulsified oil ratio and the emulsification speed of three ionic liquid surfactant solutions were different. Among them, the emulsification speed of [C₁₆mim]-[Br] is slower than those of [C₁₂mim][Br] and [C₁₄mim][Br], but its maximum equilibrium emulsified oil rate is the largest. In addition, although [C₁₄mim][Br] and [C₁₂mim][Br] reach their maximum emulsified oil ratio at approximately the same time, [C₁₄mim][Br] with longer carbon chains has a greater maximum emulsified oil ratio. At the same emulsification speed, the surfactant with a higher equilibrium emulsified oil ratio has a stronger emulsification capacity. While the equilibrium emulsified oil ratio is the same, the surfactant with faster emulsification speed has stronger emulsification capacity. However, the emulsification speed and equilibrium emulsified oil ratio of ionic liquid surfactants are both different, and the emulsification capacity cannot be compared by using the emulsification speed or equilibrium emulsified oil ratio alone. Based on the emulsified oil ratio curves, the emulsification index values of 0.597 for [C₁₆mim][Br], 0.48 for [C₁₄mim][Br], and 0.259 for [C₁₂mim][Br] were calculated. The emulsification index can accurately quantify the emulsification capacity of the oil–water system regardless of the emulsification speed and equilibrium emulsified oil ratio. By comparing the emulsification index of the ionic liquid surfactants, it can be clearly seen that the emulsification capacity is enhanced in the order of [C₁₂mim][Br] < [C₁₄mim][Br] < [C₁₆mim][Br].

3.4. Analysis of CO₂ Capture Performance. Carbon dioxide capture, EOR utilization, and storage (CCUS-EOR) is the most practical and feasible large-scale carbon reduction technology.³⁶ When CO₂ is injected into the reservoir, the fluid properties under oil reservoir conditions are improved by IFT reduction, crude oil viscosity reduction, oil expansion, and extraction effect on lighter hydrocarbon components. Today, the CCUS-EOR technology can not only greatly improve the recovery of low-permeability oil fields but also balance the CO₂ concentration and achieve higher economic efficiency. Therefore, CCUS-EOR is an important part to achieve the “net” zero goal. Previous studies have shown that CO₂ can be captured by ionic liquids including physical adsorption, chemical absorption, etc. In this study, the amine-functionalized imidazolium cation-based ionic liquid surfactants were investigated as potential absorbents for CO₂ capture.^[37]

The capacities of CO₂ absorptions by [C₁₆mim][Br] solution of different concentrations (10, 30, and 50%) at 0.1 MPa and 25 °C were investigated, and the results are displayed in Figure 9. Here, as the solvent of the ionic liquid surfactant, water can reduce the viscosity of the absorption system and promote the CO₂ absorption process. It was found from Figure 9 that the lower the concentration of [C₁₆mim][Br], the faster the rate of CO₂ absorption. The main reason is that the

**Figure 9.** CO₂ absorption by the aqueous solution of ionic liquid surfactants.

absorption rate of CO₂ is closely related to the viscosity of the absorption system. For CO₂ absorption systems, the viscosity depends primarily on the concentration of [C₁₆mim][Br], and the higher concentration leads to higher viscosity and, consequently, slower absorption. At absorption equilibrium, the solution with [C₁₆mim][Br] concentrations of 10, 30, and 50% respectively presented molar CO₂ uptake capacity values of 0.481, 0.464, and 0.450. Although CO₂ capture capacity at all concentrations was close to 0.5 mol per mol [C₁₆mim][Br], there was a slight tendency for molar capacity to decrease as [C₁₆mim][Br] concentrations increased. This is due to the high viscosity of the absorption system and the subsequent absorption products inhibiting the absorption of CO₂.

According to the above research, the synthesized ionic liquid surfactant combines the characteristics of reducing interfacial tension, emulsification, and CO₂ capture. This study also provides theoretical support for further CCUS-EOR research and the application of ionic liquid surfactants. Therefore, the development of ionic liquid surfactants with stronger absorption capacity and more cost-effectiveness is an urgent follow-up work. In addition, the relationship between the molecular structure of the ionic liquid surfactant and its emulsifying capacity, the ability to reduce interfacial tension, and CO₂ capture capacity is the point of the research, especially the research on the oil-displacement mechanism of ionic liquid surfactants after absorbing CO₂ under reservoir conditions.

4. CONCLUSIONS

The present study focused on the development and performance analysis of ionic liquid surfactants for their application to CCUS-EOR. The synthesized imidazolium-based ionic liquid surfactants were evaluated for their surface-active, emulsification capacity, and CO₂ capture performance. It was found that the surface-active and emulsification capacity of ionic liquid surfactants improved with the increase in alkyl chain length. The IFT values for [C₁₂mim][Br], [C₁₄mim][Br], and [C₁₆mim][Br] could decrease from 32.74 mN/m to 3.17, 0.54, and 0.051 mN/m, respectively, with increasing concentration. Moreover, the emulsification index values are 0.597 for [C₁₆mim][Br], 0.48 for [C₁₄mim][Br], and 0.259 for [C₁₂mim][Br]. Meanwhile, the synthesized ionic liquid

surfactants exhibited good CO₂ capture performance. The absorption capacities reach 0.48 mol CO₂ per mol of ionic liquid surfactant at 0.1 MPa and 25 °C. Experimental results demonstrate the potential of the imidazolium-based ionic liquid surfactant as a bifunctional oil displacement agent for CO₂ capture and oil displacement.

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Notes

The authors declare no competing financial interest.

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