

Regeneration of CO₂-rich aqueous amine-ionic liquid blends in CO₂ capture process

Muthumari Perumal and Dhanalakshmi Jayaraman, Solvent Development for Clean Technology Lab, Department of Chemical Engineering, Sri Sivasubramaniya Nadar College of Engineering, Chennai, India

Abstract: In this present work, solvent regeneration of CO₂-rich aqueous solvent blends containing Monoethanolamine (MEA) and Ionic liquids was studied experimentally. The ionic liquids (ILs) used are tetrabutylammonium acetate [TBA][OAC] and tetrabutylammonium bromide [TBA][Br]. The solvent mixture composition of ILs are 2, 5, 10 wt% and their respective MEA as 28, 25, 20 wt%. CO₂ absorption and desorption process were performed using solvent blends. The recovery of solvent was performed for three different temperatures 358, 368 and 373 K. The findings reveal that solvent blends of MEA with [TBA][Br] exhibit higher regeneration efficiency and lower regeneration energy than MEA with [TBA][OAC] blends with a minimum composition of 2 wt% [TBA][Br]. Energy consumption for recovery of 2 wt% [TBA][Br] blend with 28 wt% MEA was found to be 65.66 KJ mole⁻¹ of CO₂ which is 41.97% less than 30 wt% MEA. Furthermore, physicochemical properties such as pH, density, viscosity, carbon loading and surface tension have been measured before and after CO₂ absorption and desorption process. © 2021 Society of Chemical Industry and John Wiley & Sons, Ltd.

Keywords: MEA; Ionic liquids; CO₂ desorption; regeneration energy

Introduction

Energy resources are facing higher demand due to globalization and the major contribution to this energy comes from the combustion of fossil fuels such as coal. A coal-fired power plant to generate electricity is the key contributor to carbon dioxide (CO₂) emissions.^{1–3} The schematic representation of CO₂ emissions from a coal-fired power plant is shown in Fig. 1. The rapid increase in CO₂ concentration in the atmosphere leads to global warming and other climatic changes in the environment.^{4,5} To eradicate these issues, Carbon Capture, Utilization and Storage

(CCUS) is a promising technique to provide a cleaner environment.^{6,7} CCUS technologies are post-combustion, pre-combustion and oxy-fuel combustion. Post-Combustion Carbon Capture (PCCC) is the most commonly used method to absorb CO₂ from the flue gas stream of coal-fired power plants through amine scrubbing.^{8,9} Amine scrubbing is a solvent-based CO₂ capture process to absorb CO₂ at ambient temperature (~40 °C) and strip the CO₂ by elevating the temperature of the CO₂-rich solution. The CO₂ desorption process is characterized by a high enthalpy of reaction and hence the regeneration energy cost for amine scrubbing is increased.¹⁰ Various

Correspondence to: J. Dhanalakshmi, Department of Chemical Engineering, Sri Sivasubramaniya Nadar College of Engineering, Kalavakkam, Chennai – 603110, Tamilnadu, India.

E-mail: dhanalakshmij@ssn.edu.in

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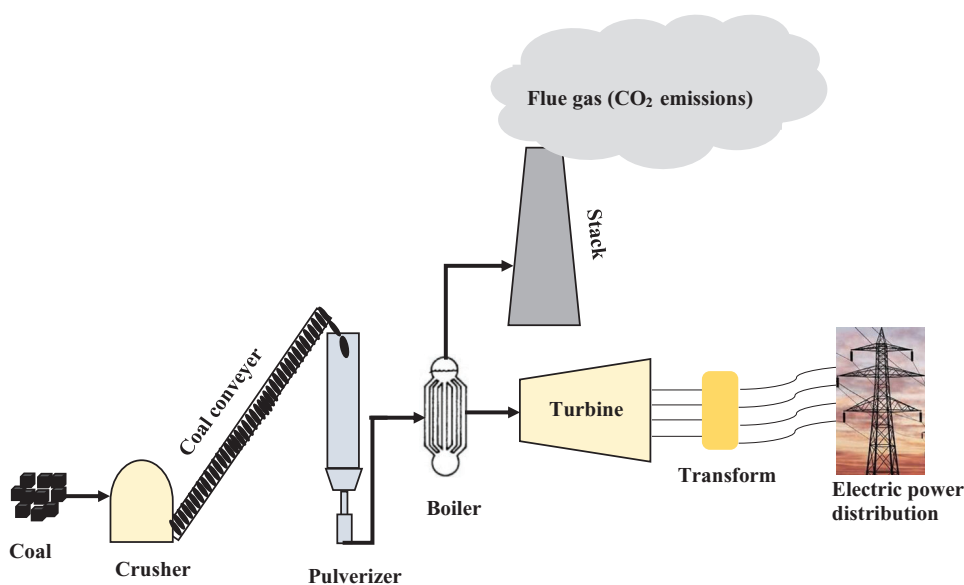


Figure 1. Emission of CO₂ from coal fired power plants.

alkanolamine solvents, namely monoethanolamine (MEA), diethanolamine (DEA), triethanolamine, (TEA) and methyldiethanolamine (MDEA) are used for the removal of CO₂ from flue gas in coal-fired power plants. Commercially, MEA is preferred as a benchmarking solvent because it has a higher absorption rate and is also relatively inexpensive. Nevertheless, the downside of MEA is higher solvent degradation and energy demand for solvent regeneration.^{11–14} Ionic liquids (ILs) act as potential solvents in both the CO₂ absorption and desorption processes due to their peculiar physico-chemical properties, including negligible vapour pressure, tunable cation/ anion combination and high thermal and chemical stability reported in the literature.^{15,16} Several studies have reported that various cations, such as imidazolium, pyridinium, pyrrolidinium and phosphonium-based ILs have more complex molecular structure and lower solubility was observed during CO₂ absorption. Therefore, choosing less complex cationic molecules such as ammonium-based ILs can promote higher CO₂ absorption and lower solvent regeneration.^{17–19} Babu *et al.*²⁰ investigated the simultaneous removal of CO₂ and SO₂ using aqueous 0.3–3.0 mol% tetra-*n*-butyl ammonium bromide [TBA][Br]. The highest gas uptake was reported at 0.3 mol% of aqueous [TBA][Br] itself because of its highest driving force. However, ILs have substantial drawbacks that limit their competitiveness when compared to traditional solvents, largely ascribed to their relatively

high viscosity, which limits their mass transfer capacity. Indeed, after absorbing CO₂, they become excessively viscous, causing solvent pumping problems as well as operating challenges throughout the CO₂ capture process.²¹ These limitations of individual solvents make them unsuitable for large-scale applications. Therefore, this experimental work is focused on the use of solvent blends, which can be easily obtained by mixing two solvents at different concentrations, resulting in low viscosity, relatively low cost and reduced energy consumption for solvent recovery.²²

Yang *et al.*²³ used a mixture of 1-butyl-3-methylimidazolium tetrafluoroborate [bmim][BF₄] and MEA for the CO₂ absorption and desorption process. It was found that the regeneration energy consumption was 37.2% less than the baseline MEA. Therefore, the results proved that the solvent blends exert less energy to recover the solvent. Taib and Murugesan,²⁴ investigated CO₂ solubility using ammonium-based ILs mixed with MEA as 20wt% bis (2hydroxyethyl) ammonium acetate with varying concentrations of aqueous MEA solution (5–20wt%). Similarly, 1-butyl-3-methylimidazolium tetrafluoroborate, an imidazolium-based IL was used as a blend of MEA and IL with the above concentration to study the solubility of CO₂. It was found that the blends of ammonium ILs and MEA relatively increased the solubility of CO₂ than the imidazolium-based IL blend mixture. Furthermore, physical properties such as density, viscosity and surface tension of solvents will

change during the absorption and desorption process. Changes in these properties will affect the mass transfer at the bottom of the absorber and the reaction kinetics in the stripper, thereby increasing the energy demand of CCS. Therefore, it is necessary to measure the physical properties during the absorption and desorption.^{25–30} From the extensive literature survey, it was noted that the regeneration study of solvents is less reported, especially for blended solvent composed of MEA and IL. Hence, the main objective of this study is to identify the appropriate solvent blends through experiments that will minimize the energy consumption for the solvent regeneration/CO₂ desorption process. In this study, ammonium-based ionic liquids, namely tetrabutylammonium bromide [TBA][Br] and tetrabutylammonium acetate [TBA][OAc] were used. The performance of the CO₂ desorption process is analysed using parameters such as CO₂ loading (carbon loading), regeneration temperature, desorption rate, regeneration efficiency and energy consumption during solvent regeneration. In addition, the physicochemical properties of a virgin (absence of CO₂), CO₂-rich and CO₂ lean solution were measured.

Materials and methods

Chemicals required

In this present investigation, aqueous MEA blended with ammonium-based ILs used as an absorbent for the CO₂ absorption and desorption process were: monoethanolamine (MEA, Merck, 98%), tetrabutylammonium acetate [TBA][OAc]. The purity of [TBA][OAc] (TCI Chemicals) was mentioned as >90% and the remaining <10% was found to be moisture content, which has been analyzed using the Karl Fisher titrator. The presence of moisture content does not make any significant changes in the experimental results obtained during the process. Tetrabutylammonium bromide ([TBA][Br], TCI chemicals, >98%). All the above solvents were used without further purification. Aqueous solvent blends containing MEA and ammonium-based ILs were prepared by weighing out the requisite amounts of corresponding solvents using a digital weighing balance with $\pm 0.01\%$ accuracy. The solvent blends are 28 wt% MEA+2 wt% [TBA][OAc], 25 wt% MEA+5 wt% [TBA][OAc], 20 wt% MEA+10 wt% [TBA][OAc], 28 wt% MEA+2 wt% [TBA][Br], 25 wt% MEA+5 wt% [TBA][Br] and 20 wt% MEA+10 wt%

[TBA][Br]. For the solvent blend preparation at the desired concentration, laboratory grade double distilled water was used.

Physicochemical properties of aqueous blends of MEA and ammonium-based IL solution

For the analysis of absorption and regeneration features, measurement of physicochemical properties such as pH, density, viscosity, CO₂ loading (carbon loading) and surface tension are very important and determined for all solvent blends of MEA and ammonium-based ILs. The pH of samples (virgin (absence of CO₂), CO₂-rich (CO₂ absorbed sample) and CO₂ lean solvent blends (regenerated sample)) were determined using a Thermo Scientific pH meter. The density of the samples was measured using a Sartorius weighing balance (Model No: BSA 224S, 0.1mg accuracy) through gravimetric analysis. The Cannon–Fenske opaque viscometer (Model No: 9721-F53) is a commercial instrument purchased from Cole-Parmer, USA with an accuracy of $\pm 0.3\%$. It was used to measure the viscosity of the solvent blends before and after the absorption/desorption process. The amount of CO₂ absorbed/desorbed in the solvent blends was measured using fabricated Chittick apparatus. The Chittick apparatus is custom made and fabricated, which was reported earlier in our previous study.³¹ The surface tension values of each and every sample were measured using a tensiometer. The heat capacity of solvent blends was measured using differential scanning calorimetry (NETZSCH DSC Model no: 204F1, Phoenix) with a temperature range of 303–393K at a constant heating rate of 5.0 (K min⁻¹).

Experimental procedure for CO₂ absorption

Before starting the experiment, the physicochemical analysis (pH, carbon loading, density, viscosity and surface tension) of the virgin sample (absence of CO₂) was performed. A solvent blend containing MEA and ammonium-based ILs was used for the CO₂ absorption process. In this study, a gas mixture of 15% by volume of CO₂ and 85% by volume of N₂ was purchased from Bharath Oxygen Company in Chennai, India. The cylinder consists of two pressure gauges, one for the cylinder tank pressure (0–3000 psi) and the other for the inlet gas pressure (0–15 psi). A known amount of

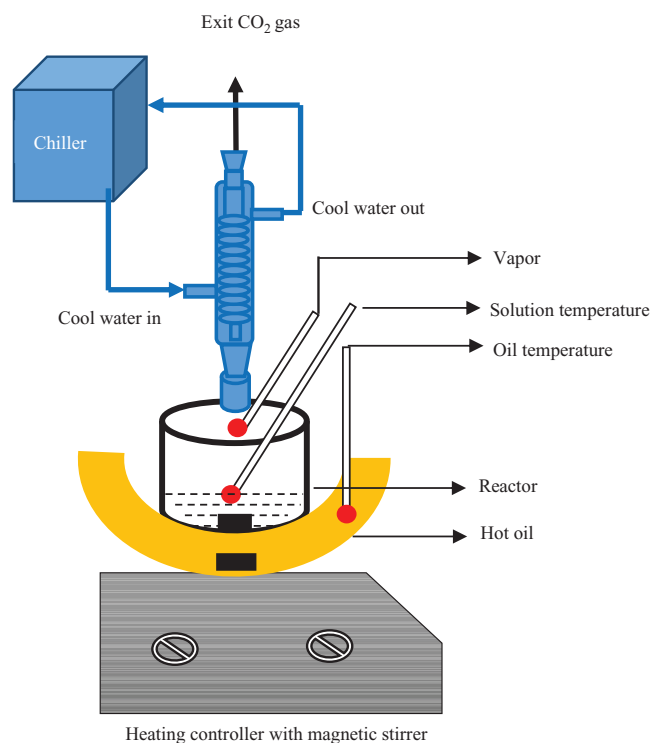


Figure 2. Solvent recovery experimental set up.

solvent blend was taken in a glass reactor and CO₂ inlet gas was fed into the reactor at a constant pressure of 0.5 psi. The absorption process is carried out continuously until the solution reaches saturation. The CO₂ saturation was qualitatively confirmed using a Horiba CO₂ analyser by monitoring both the inlet and exit concentrations of CO₂ until they became equal. The CO₂ loading (carbon loading) in the solvent blends was measured after absorption using the Chittick apparatus. The schematic representation of the absorption set up with detailed procedure was already discussed in our previous published work by Perumal *et al.*³¹

Experimental procedure for solvent regeneration

The experimental set up for solvent recovery is shown in Fig. 2. This setup is used to regenerate the solvent blends from CO₂-rich solutions for recovery and reuse. The solvent regeneration laboratory set-up comprises of thermocontroller heating system, temperature port for measuring oil, vapor and reaction mixture temperature. The condenser is mounted on the reactor, which is used to minimize the solvent vapor loss by circulating the cooling water. Hence, the non-condensable CO₂ gas is released during

regeneration process, then carbon lean solution was obtained. The ILs ([TBA][OAC], [TBA][Br]) mixed with MEA and its composition is kept constant at 30% by weight. On this basis, the following solvent blend compositions contain 2, 5, 10 wt% IL and their corresponding MEA are 28, 25 and 20 wt% respectively. The solvent blends are 28 wt% MEA+2 wt% [TBA][OAC], 25 wt% MEA+5 wt% [TBA][OAC], 20 wt% MEA+10 wt% [TBA][OAC], 28 wt% MEA+2 wt% [TBA][Br], 25 wt% MEA+5 wt% [TBA][Br], 20 wt% MEA+10 wt% [TBA][Br]. These solvent blends containing CO₂ after absorption are called CO₂-rich solution. In the reactor, 40 g of CO₂-rich solution was placed and the regeneration was performed experimentally at three different temperatures 373, 368 and 358 K respectively. The regeneration was performed for a time of 1 hr.³⁷ During the process, samples were collected at 5, 15, 30, 45 and 60 mins. Then, the CO₂ loading was measured using the Chittick apparatus.^{31,32} Following that, temperature was continuously recorded using a thermocouple (K-type) until 1 hr of regeneration. After 1 hr of regeneration, the sample (carbon lean solution) was collected and then the pH, viscosity, density, CO₂ loading and surface tension of the carbon lean solution were determined.

Results and discussion

The regeneration of the CO₂-rich aqueous blend containing MEA and ammonium-based ILs was performed at three different temperatures. The solvent mixture compositions are aqueous

28 wt% MEA+2 wt% [TBA][OAC], 25 wt% MEA+5 wt% [TBA][OAC], 20 wt% MEA+10 wt% [TBA][OAC], 28 wt% MEA+2 wt% [TBA][Br], 25 wt% MEA+5 wt% [TBA][Br], 20 wt% MEA+10 wt% [TBA][Br]. Important parameters such as regeneration temperature, desorption rate, CO₂ loading, regeneration efficiency, desorption kinetics and regeneration energy have been studied to understand the performance of the solvent regeneration process. Then, the physicochemical analysis of virgin, CO₂-rich and lean solvent blends was also determined and the results are discussed.

CO₂ absorption

The CO₂ absorption of aqueous blends containing MEA and ammonium-based ILs was performed. The CO₂-rich solution was achieved after the absorption

process. The mechanisms of CO₂ absorption in MEA follow the zwitterion concept. When CO₂ reacts with MEA, it will form a carbamate as an intermediate product. Then carbamate is hydrolyzed into bicarbonate, carbonate.³³ The interaction between the cation and anion of IL and CO₂ takes place through hydrogen bonding. The IL (tetrabutylammonium) cation reacts with MEA, CO₂, H₂O and forms carbamate, ammonium ions and ammonium hydroxide.³¹ The acetate anion reacts with CO₂, MEA, H₂O and may form compounds: acetic acid, carbonate, carbonic acid, bicarbonate. Whereas, bromide anion reacts with CO₂, MEA, H₂O, it may form bromic acid, hydrogen bromide (HBr). Based on the solvent absorption capacity, the CO₂-rich solution is obtained as shown in Table 4. From the table, it was observed that CO₂ loading of 2, 5 and 10 wt% of [TBA][OAC] in 28, 25 and 20 wt% MEA 0.459, 0.397 and 0.377 mole of CO₂/mole of solvent blend. The formation of acetic acid was rather than CO₂ absorption, which may reduce CO₂ loading while increasing [TBA][OAC] composition in MEA blends. In another case, 2, 5 and 10 wt% of [TBA][Br] in 28, 25 and 20 wt% MEA are 0.405, 0.392 and 0.345 mole of CO₂/mole of solvent blend, respectively. The absorption capacity of MEA-[TBA][OAC] blend was slightly higher than MEA-[TBA][Br] blend because acetate anion attracts more CO₂ via hydrogen bonding.

Solvent regeneration

Variation of temperature

The regeneration process of different CO₂-rich aqueous blends of MEA and ammonium-based IL was performed experimentally at three different temperatures 373, 368 and 358 K, respectively. This is very important to understand the solvent blend behaviour towards minimizing the energy consumption during regeneration. In order to maintain the temperature of the solution, the temperature was continuously recorded from the beginning to the end of 1 hr regeneration until it reaches the steady state operation. The rise in temperature of solution indicates that time required to reach the desired temperature. Using the heating controller, the desired temperature was set and maintained throughout the regeneration. Figure 3 depicts the variation of temperature during 1 hr of operation for all CO₂-rich solvent blends studied in this present work. During regeneration, the initial temperature ($T_{\text{CO}_2\text{ rich}}$) and final temperature ($T_{\text{CO}_2\text{ lean}}$)

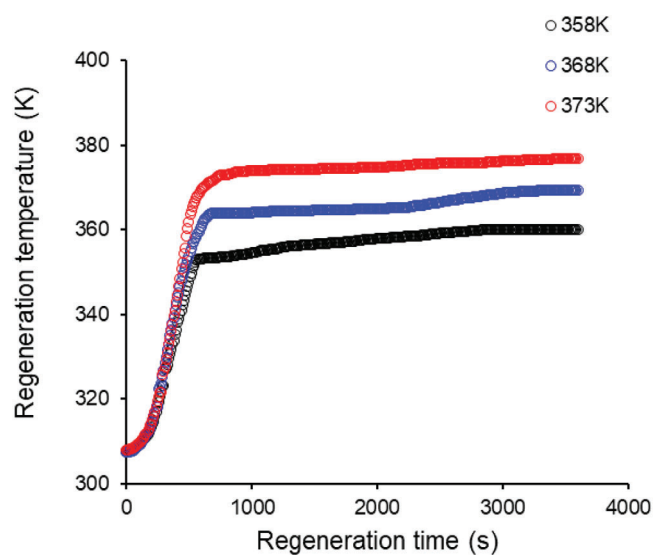


Figure 3. Temperature variation during regeneration.

of solvent blends were noted. The sensible heat calculation of solvent blends was measured using $T_{\text{CO}_2\text{ rich}}$, $T_{\text{CO}_2\text{ lean}}$, heat capacity, CO₂-rich and lean loading.

Variation of desorption rate

The CO₂ desorption rate was determined using the slope of the carbon loading-regeneration time curve reported by Zhang *et al.*³⁴ which is displayed in Fig. 4. According to Fig. 4, as the IL composition increases, the desorption rate of the CO₂-rich MEA-[TBA][OAC] blend increases. The synthesis of stable carbamate was minimized when [TBA][OAC] composition was raised in MEA blend. Because MEA reacts with CO₂ to form a stable carbamate, more energy is consumed to regenerate the solvent. The opposite trend is observed for the CO₂-rich MEA-[TBA][Br] blend, which indicates that the desorption rate decreases as the [TBA][Br] composition in MEA blend increases. On the other hand, increasing temperature, CO₂ desorption rate increases for all solvent blends. As the temperature rises, more collision occurs between the molecules, which results in a high rate of desorption of all solvent blends at 373 K. In addition, desorption rate decreases with an increase in desorption time as shown in Fig. 4. Especially 2%[TBA][OAC] indicates higher desorption rate because initially the solvent blend contains more amount of CO₂ and further gradually decreases as desorption time increases. For 10%[TBA][Br], desorption rate was suddenly increased due to stronger electrostatic interactions may occur in

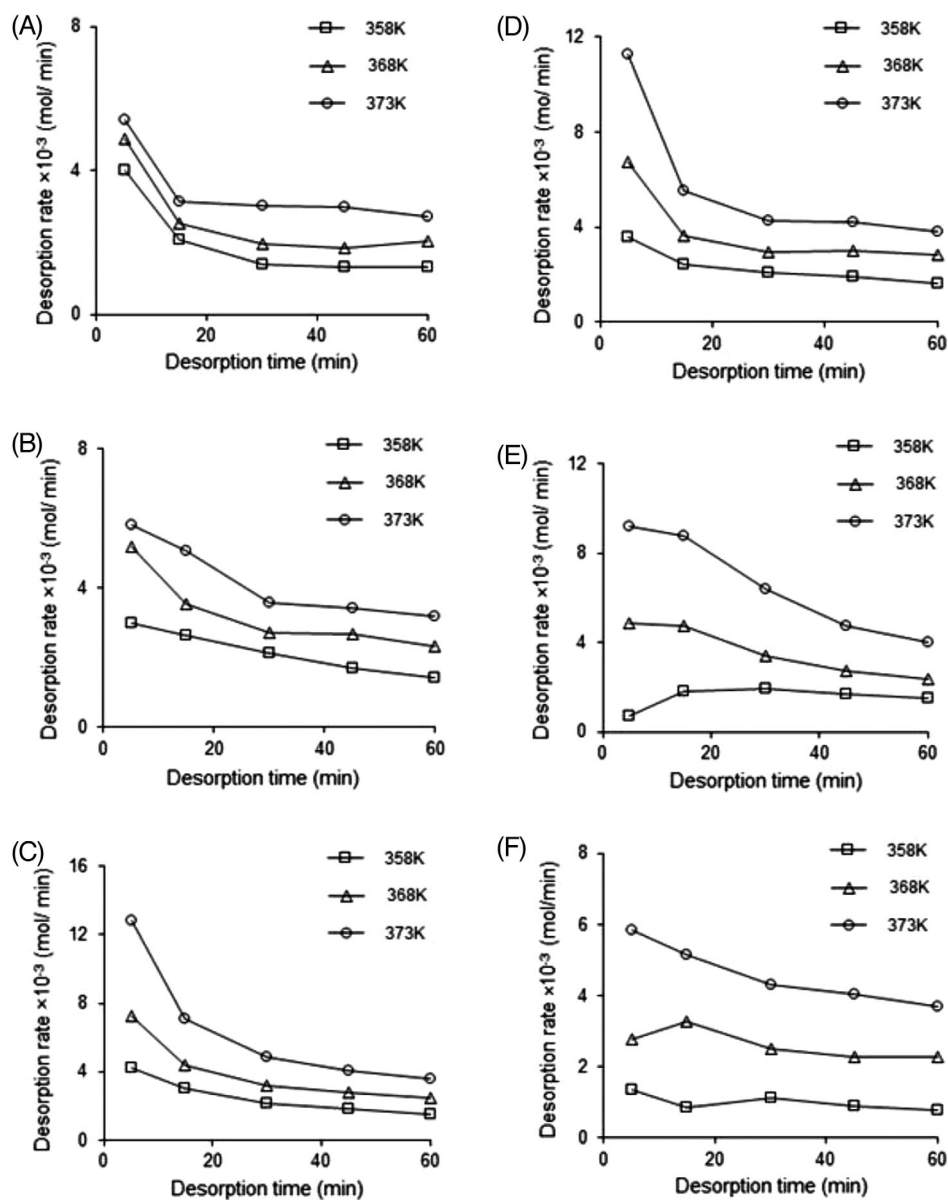


Figure 4. Influence of desorption time on desorption rate of solvent blends (A) 28 wt% MEA+2 wt% [TBA][OAC], (B) 25 wt% MEA+5 wt% [TBA][OAC], (C) 20 wt% MEA+10 wt% [TBA][OAC], (D) 28 wt% MEA+2 wt% [TBA][Br], (E) 25 wt% MEA+5 wt% [TBA][Br], (F) 20 wt% MEA+10 wt% [TBA][Br] at three different temperatures.

solution. It was also observed that the desorption rate of 2 wt% [TBA][Br] blend is almost similar to that of 10 wt% [TBA][OAC] blend. Therefore, it can be said that [TBA][Br] in MEA blend is a good choice for solvent blend which can achieve rapid desorption with minimal IL composition. The results obtained were compared with the 30 wt% MEA as earlier reported in Perumal *et al.*³¹ It has been observed that the CO₂ desorption rate of baseline MEA is lower than that of MEA with IL blends.

Influence of regeneration time and temperature

The regeneration performance is determined based on the regeneration efficiency and the residual CO₂ remaining in the regenerated solvent. The obtained results revealed that presence of residual CO₂ (lean carbon loading) in the regenerated solvent decreases with increasing the regeneration time and temperature as shown in Fig. 5. A similar trend was reported by

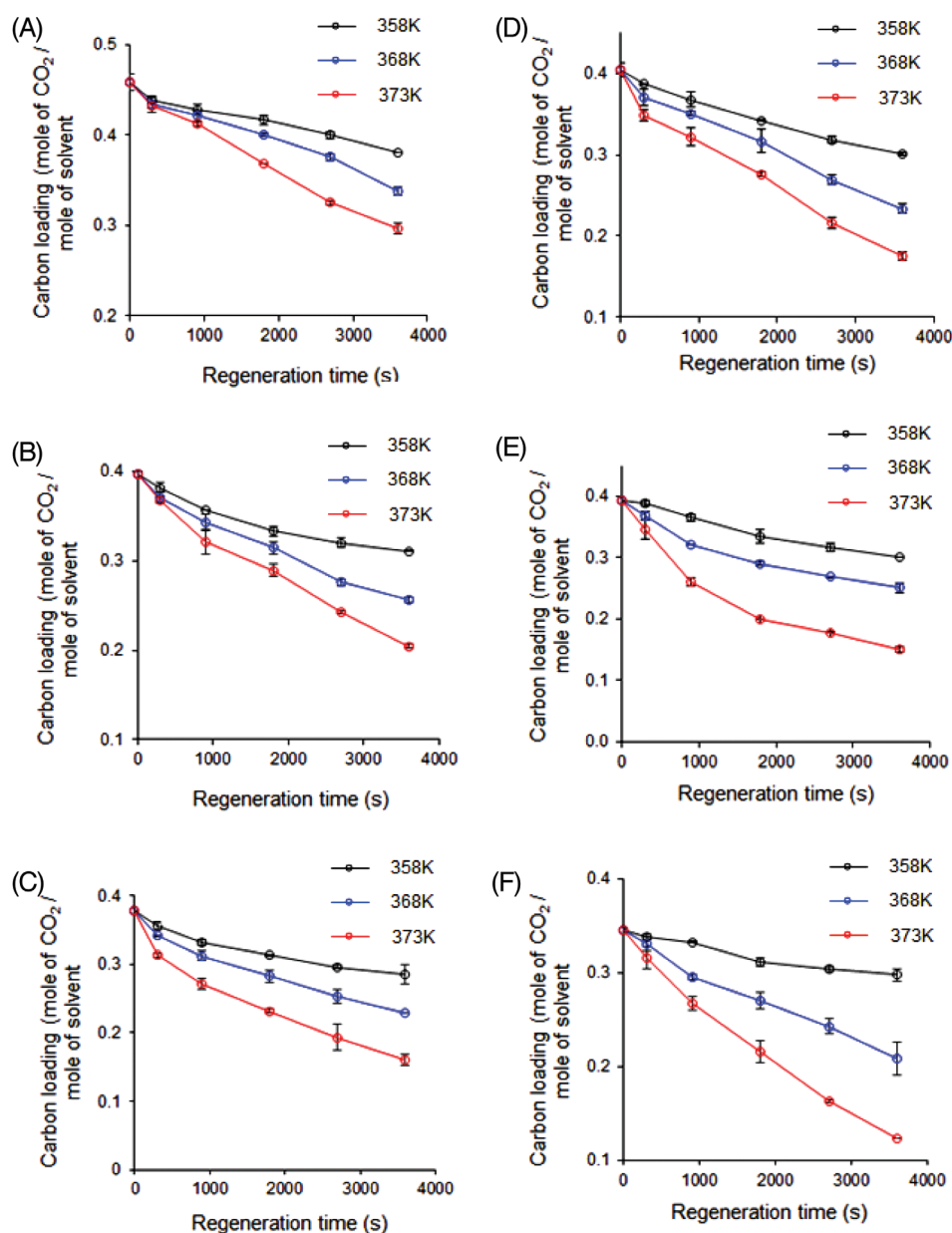


Figure 5. Effect of regeneration time on carbon loading of solvent blends (A) 28 wt% MEA+2 wt% [TBA][OAC], (B) 25 wt% MEA+5 wt% [TBA][OAC], (C) 20 wt% MEA+10 wt% [TBA][OAC], (D) 28 wt% MEA+2 wt% [TBA][Br], (E) 25 wt% MEA+5 wt% [TBA][Br], (F) 20 wt% MEA+10 wt% [TBA][Br] at three different temperatures.

Zhang *et al.*³⁴; Perumal *et al.*³¹ The results are compared with baseline MEA.³¹ The maximum CO₂ lean condition was obtained at 373 K for all solvent blends. Because the temperature provides the necessary heat for the thermal decomposition of all carbamate, carbonate, bicarbonate, acetic acid, bromic acid, hydrogen bromide during the reaction with CO₂-solvent blend, and convert into CO₂ free solvent blend. It was also found that [TBA][Br] IL mixtures

reaches faster carbon lean condition when compared to [TBA][OAC] blends because of its lower viscosity facilitates more release of CO₂.

Regeneration efficiency

The regeneration time and temperature are key parameters that directly affect the regeneration performance, which in turn impacts the regeneration

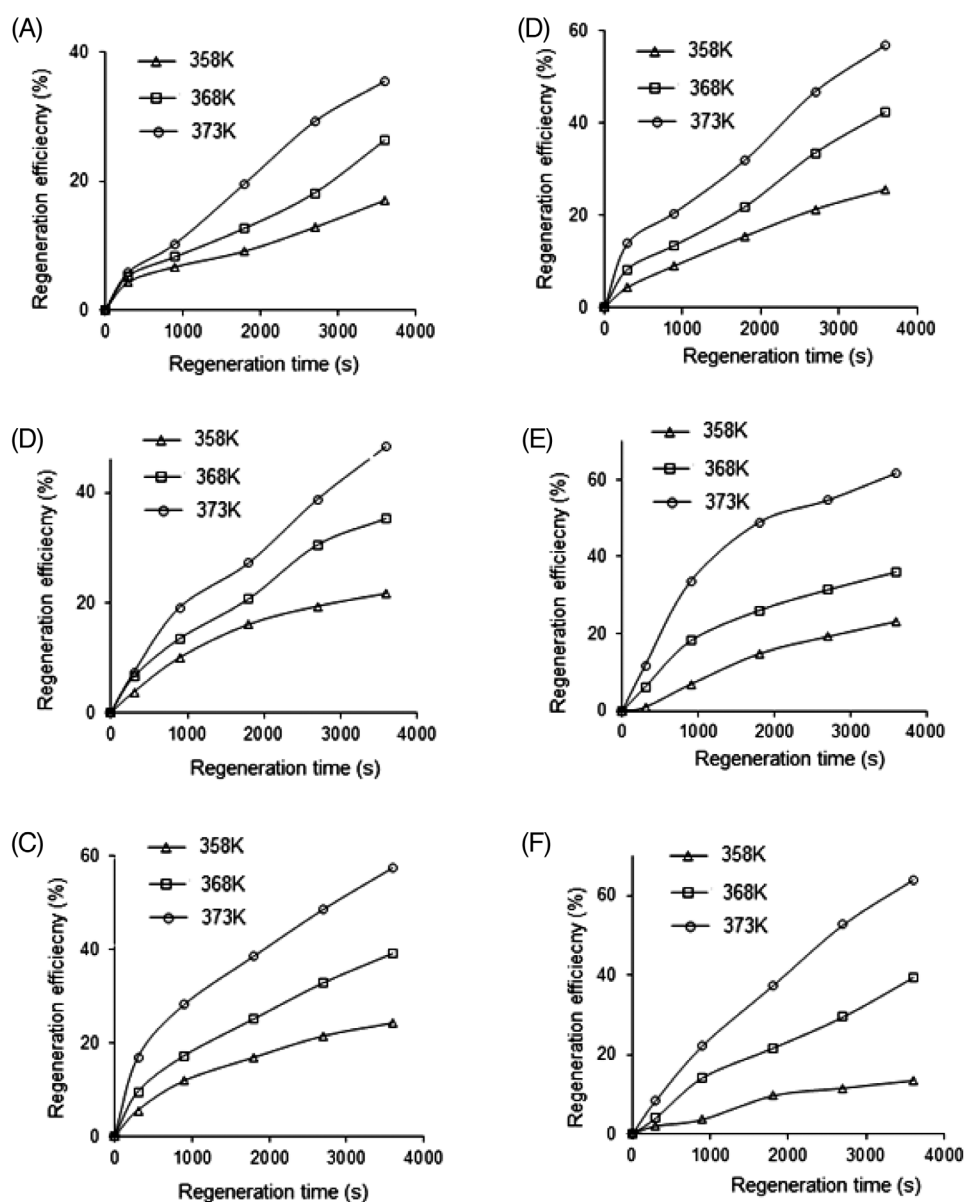


Figure 6. Influence of regeneration time on regeneration efficiency of solvent blends (A) 28 wt% MEA+2 wt% [TBA][OAC], (B) 25 wt% MEA+5 wt% [TBA][OAC], (C) 20 wt% MEA+10 wt% [TBA][OAC], (D) 28 wt% MEA+2 wt% [TBA][Br], (E) 25 wt% MEA+5 wt% [TBA][Br], (F) 20 wt% MEA+10 wt% [TBA][Br] at three different temperatures.

efficiency. The regeneration efficiency of various solvent blends was calculated as reported in Khan *et al.*⁸ and it was shown in Fig. 6. The regeneration efficiency increases with an increase in the temperature of both the solvent blends. This is due to higher kinetic energy occurring between the molecules in solution can dissociate the compounds that may results in the rapid release of CO₂. On the other hand, the regeneration efficiency gradually decreases with

increasing the IL composition in the aqueous MEA blend as shown in Fig. 6. The regeneration efficiency of MEA-[TBA][OAC] blend was significantly lower than MEA-[TBA][Br] blends because the dissociate of acetic acid consumes higher energy. The present results also revealed that [TBA][Br]-MEA blend has the superior regeneration efficiency because bicarbonate, carbonate and bromic acid formation with CO₂ which is easily separated into CO₂ free solvent blend at lower

temperature. Then the obtained results were compared with 30 wt% MEA results.³¹ The regeneration efficiency of 30 wt% MEA was lower than MEA-IL blend.

Regeneration efficiency

$$= 1 - \frac{\text{carbon loading of CO}_2 \text{ lean solution}}{\text{carbon loading of CO}_2 \text{ rich solution}} \times 100\% \quad (1)$$

Desorption kinetics and activation energy

The desorption kinetics for CO₂-rich solution containing blends of MEA and ammonium-based ILs is predicted using the following rate equation by varying temperature 358K, 368K and 373 K

$$\ln \left(\frac{C_A}{C_{A0}} \right) = -kt \quad (2)$$

where C_A is the CO₂ concentration at any given desorption time (mol/mol), C_{A0} is the initial concentration of CO₂-rich solution and k is the desorption rate constant.

This kinetics study is important to detect the suitable solvent by calculating its energy requirement for solvent recovery. Presence of higher activation energy indicates that CO₂-rich solution requires more energy demand to release the CO₂ for obtaining carbon lean solution. The kinetic parameter of temperature dependent reaction rate constant is used to calculate the activation energy with the help of Arrhenius equation and it is given below.³⁵

$$\ln k = \frac{-E_a}{RT} + \ln A \quad (3)$$

$$k = A e^{\left(\frac{-E_a}{RT} \right)} \quad (4)$$

where k is the reaction rate constant, A is the Arrhenius pre-exponential factor, E_a is the activation energy, R is the ideal gas constant and T is the temperature (K). The E_a was calculated by correlating $\ln k - 1/T \times 10^3$ plot for 358, 368 and 373 K which is shown in Fig. 7. From the plot, the slope is obtained and then it is substituted in Eqn 4 to calculate activation energy E_a . It was observed that E_a increased as increase of IL composition in solvent blends. For a minimum IL composition (2 wt% IL in MEA blend), the E_a was 58.62 and 60.17 KJ mol⁻¹ for [TBA][Br] and [TBA][OAC] respectively, which is lower than 88.22 KJ/mol for 30 wt% MEA as reported earlier in Perumal *et al.*³¹ (Eqns 5–10). From the obtained results, it was revealed that lower E_a promotes

higher rate of desorption which releases large amount of CO₂ from CO₂-rich solution.

$$K_{28 \text{ wt\% MEA} + 2 \text{ wt\% [TBA][OAC]}} = 3.08 \times 10^4 e^{\left(\frac{-7.238}{T} \right)}, \quad (5)$$

$$K_{25 \text{ wt\% MEA} + 5 \text{ wt\% [TBA][OAC]}} = 1.75 \times 10^5 e^{\left(\frac{-7.732}{T} \right)}, \quad (6)$$

$$K_{20 \text{ wt\% MEA} + 10 \text{ wt\% [TBA][OAC]}} = 5.98 \times 10^5 e^{\left(\frac{-8.106}{T} \right)}, \quad (7)$$

$$K_{28 \text{ wt\% MEA} + 2 \text{ wt\% [TBA][Br]}} = 3.20 \times 10^4 e^{\left(\frac{-7.051}{T} \right)}, \quad (8)$$

$$K_{25 \text{ wt\% MEA} + 5 \text{ wt\% [TBA][Br]}} = 8.34 \times 10^6 e^{\left(\frac{-9.133}{T} \right)}, \quad (9)$$

$$K_{20 \text{ wt\% MEA} + 10 \text{ wt\% [TBA][Br]}} = 3.97 \times 10^{15} e^{\left(\frac{-16.491}{T} \right)}. \quad (10)$$

Determination of heat capacity

The heat capacity of solvent blends was measured using differential scanning calorimetry (DSC) over a temperature range of 308–393 K. However, heat capacity data from 358 to 373 K were considered for present study to plot the DSC thermogram because solvent recovery was performed in this range. From the Fig. 8, increase in heat capacity for [TBA][OAC] in MEA blend was observed for increasing temperature as expected because of increase in kinetic energy of molecules possess rapid molecular collision which leads to rise in internal energy.³⁶ Whereas a slight decreasing trend was obtained for [TBA][Br] due to weaker molecular interactions between [TBA][Br] and MEA. It was also noted that heat capacity of aqueous solvents blends for both IL increases with increasing their IL concentration, comparatively lower heat capacity was observed [MEA-[TBA][Br]] than that of [MEA-[TBA][OAC]]. This is due to the size of anion limits the intermolecular electrostatic interactions results lower internal energy between the molecules. Thus, lower the heat capacity, lesser regeneration energy was obtained at a very low [TBA][Br] concentration. Hence, [MEA-[TBA][Br]] was comparatively exhibited a suitable solvent blend than that of [MEA-[TBA][OAC]].

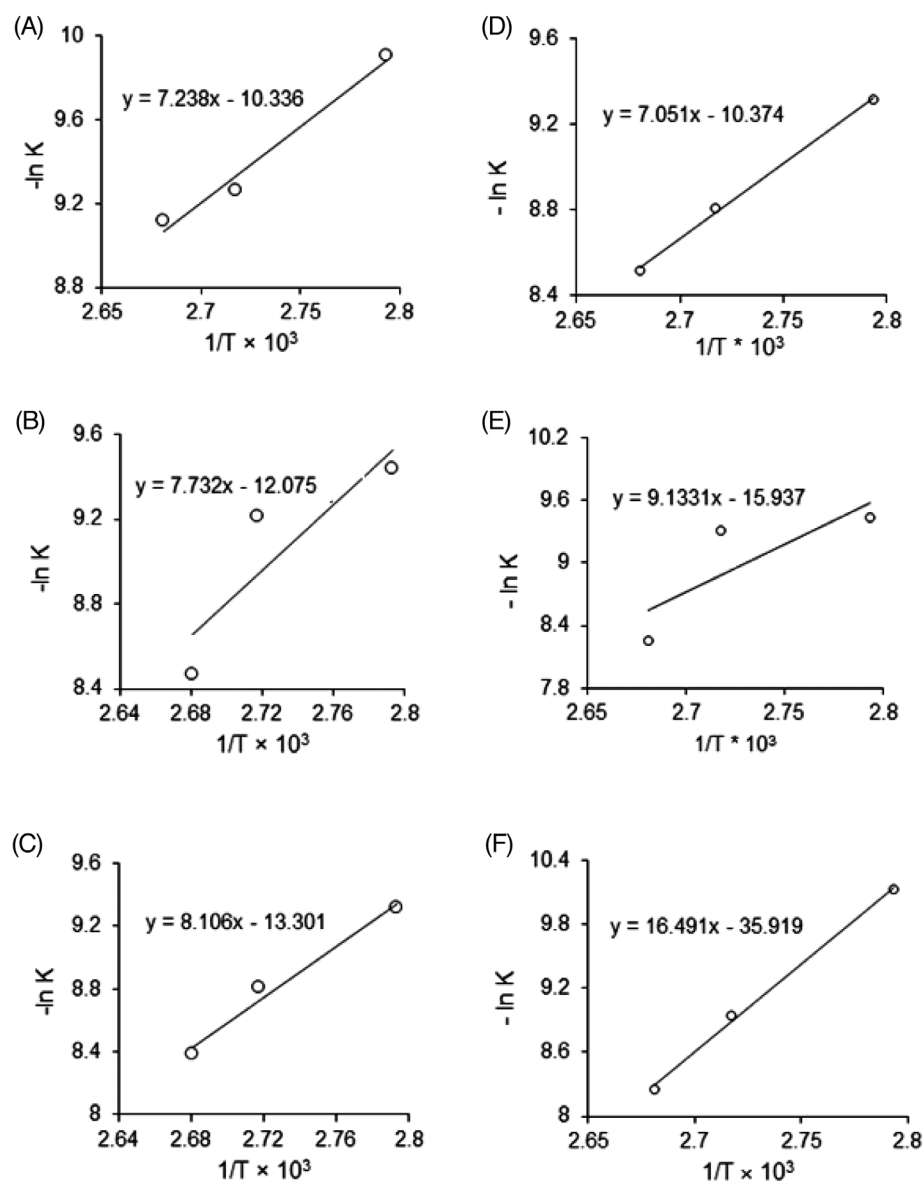


Figure 7. Arrhenius plots (activation energy) of solvent blends (A) 28 wt% MEA+2 wt% [TBA][OAC], (B) 25 wt% MEA+5 wt% [TBA][OAC], (C) 20 wt% MEA+10 wt% [TBA][OAC], (D) 28 wt% MEA+2 wt% [TBA][Br], (E) 25 wt% MEA+5 wt% [TBA][Br], (F) 20 wt% MEA+10 wt% [TBA][Br].

Regeneration energy

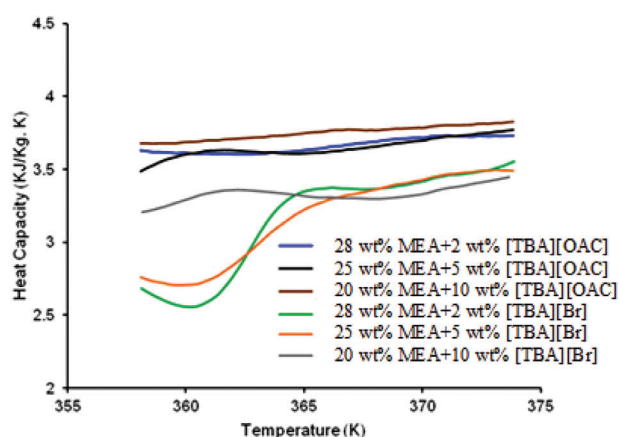
The regeneration energy penalty comprises of three sections (i) heat of absorption (ii) sensible heat and (iii) latent heat of vaporization.³⁷ The sensible heat is calculated by Eqn 11. The heat capacity of the solvent is predicted from the DSC analysis.

$$Q = \text{Solvent mass} \times \text{Heat capacity of solvent} \times \left(\frac{T_{\text{CO}_2\text{lean}} - T_{\text{CO}_2\text{rich}}}{\text{CO}_2\text{rich loading} - \text{CO}_2\text{lean loading}} \right) \quad (11)$$

The CO₂ loading of lean solution was calculated after regeneration. Table 1 shows sensible heat calculation for solvent blends regeneration. From the Table 1, it was observed that the regeneration energy for aqueous MEA-[TBA][OAC] blends was higher than various solvent blends with different regeneration temperatures. From an experimental data, it was noted that the regeneration energy of [TBA][OAC] in MEA blend mixtures decreases with increasing composition of ILs and vice versa for solvent blends of MEA with

Table 1. Regeneration energy of solvent blends.

Solvents	Sensible Heat (Regeneration energy) (KJ/mole CO ₂)		
	358 K	368 K	373 K
30wt%MEA ³¹	114.65	85.99	42.44
28 wt% MEA+2 wt% [TBA][OAC]	96.92	78.20	63.80
25 wt% MEA+5 wt% [TBA][OAC]	89.86	68.81	55.23
20 wt% MEA+10 wt% [TBA][OAC]	84.65	63.29	49.66
28 wt% MEA+2 wt% [TBA][Br]	66.53	51.65	41.05
25 wt% MEA+5 wt% [TBA][Br]	80.02	68.33	43.55
20 wt% MEA+10 wt% [TBA][Br]	143.69	61.31	43.13

**Figure 8. Heat capacity of solvent blends.**

[TBA][Br]. However, regeneration energy of MEA with [TBA][Br] solvent blends was significantly lesser than [TBA][OAC] blends. In addition, the regeneration energy of 30%MEA was reported earlier in Perumal *et al.*³¹ 114.65 KJ mol⁻¹ of CO₂, whereas 2 wt% [TBA][Br] in 28 wt% MEA consumes 66.53 KJ mol⁻¹ of CO₂ and 2 wt% [TBA][OAC] in 28 wt% MEA requires 96.92 KJ mol⁻¹ of CO₂ at 358 K. These results revealed that blends of [TBA][Br] require 41.97% lesser energy consumption for solvent regeneration than 30% MEA³¹ for recovery and reuse, as shown in Table 1.

Physicochemical property study

Variation of pH

Decrease in pH indicates the removal of acid gas CO₂ from CO₂-rich solution. From Table 2 and Fig. 9 it was observed that pH of virgin solvent was higher and it decreases after CO₂ loading. Again, pH gets increased with increasing temperature during solvent regeneration by releasing CO₂. At higher temperature,

pH was observed as high for all solvent blends. The pH of regenerated sample does not reach to virgin pH because there are few traces of CO₂ left in the solution.

Variation of density

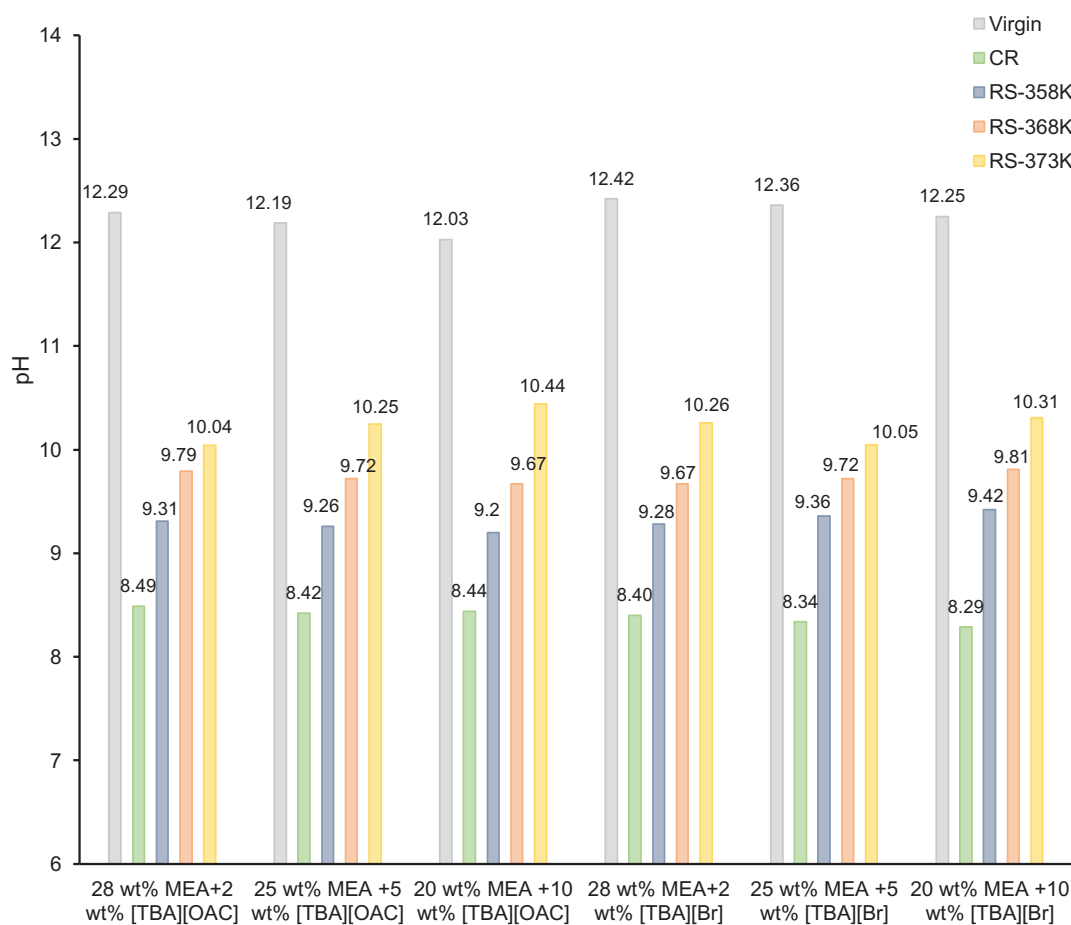
Density of MEA-[TBA][OAC] decreases with increasing their IL composition. This behaviour is due to the formation of acetic acid during the preparation of solvent blend. The opposite trend of density was observed for [TBA][Br] with MEA blend. The density of CO₂-rich solution containing solvent blends was higher than virgin solution due to presence of CO₂ as shown in Table 3 and Fig. 10. On the other hand, the density of the regenerated sample decreases with increasing temperature because CO₂ molecules are released from the CO₂-rich solution.

Variation of CO₂ loading

The variation of CO₂ loading for various solvent mixtures with MEA and ILs are given in Table 4 and Fig. 11. The CO₂ loading of CO₂-rich solvent blends of both ILs decreases while increasing the IL composition and regeneration temperature. The CO₂ loading of baseline 30 wt% MEA is higher than that of solvent blends. However, MEA requires more energy consumption for solvent regeneration. In the case of a blend of MEA and IL, the CO₂ loading is slightly lower than that of the baseline MEA. Nevertheless, it demands less energy consumption to regenerate the solvent. The performance of [TBA][Br] is significantly greater than [TBA][OAC] in MEA blend. From this study, it is evident that ILs with minimum composition blends with MEA exhibits less energy requirement for solvent regeneration and poses higher efficiency for CO₂ removal at the end of regeneration process.

Table 2. Variation of pH on virgin (0 min), CR (after 3 hr), and RS (after 1 hr).

Solvents	pH				
	Virgin	CR	RS-358K	RS-368K	RS-373K
28 wt% MEA+2 wt% [TBA][OAC]	12.29	8.49	9.31	9.79	10.04
25 wt% MEA +5 wt% [TBA][OAC]	12.19	8.42	9.26	9.72	10.25
20 wt% MEA +10 wt% [TBA][OAC]	12.03	8.44	9.2	9.67	10.44
28 wt% MEA+2 wt% [TBA][Br]	12.42	8.4	9.28	9.67	10.26
25 wt% MEA +5 wt% [TBA][Br]	12.36	8.34	9.36	9.72	10.05
20 wt% MEA +10 wt% [TBA][Br]	12.25	8.29	9.42	9.81	10.31

Abbreviations: CR, CO₂ rich; RS, regenerated solutions.**Figure 9.** Variation of pH on virgin; CR, CO₂-rich; RS, regenerated solutions.

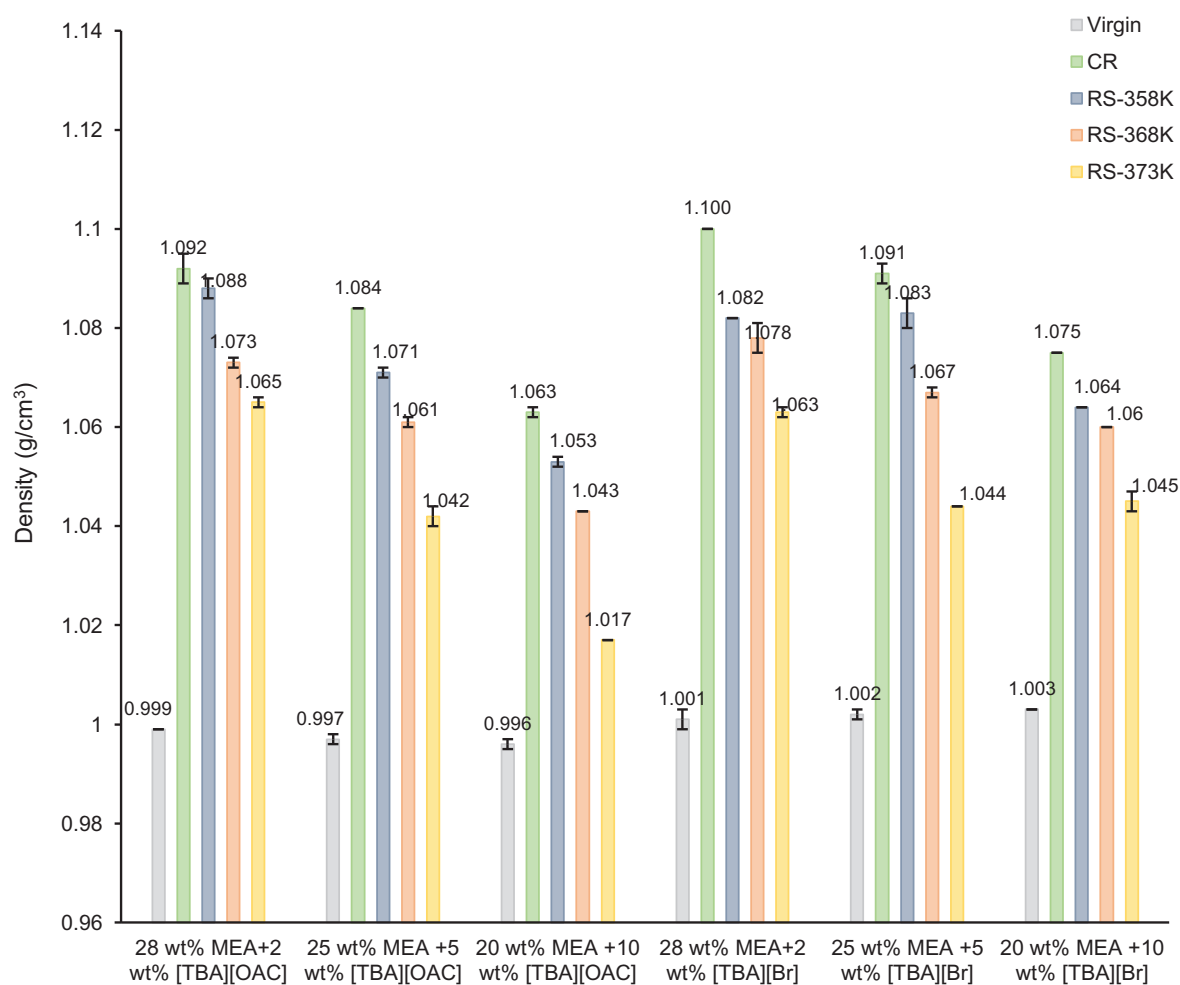
Variation of viscosity

The viscosity of the CO₂-rich solution is a key parameter in the solvent recovery process. Table 5 and Fig. 12 listed the viscosity of the virgin, CO₂-rich and regenerated solvent blends. It was observed that

viscosity of the solvent blends containing MEA and ILs increases as the composition of IL increases. On the other hand, the viscosity of solvent blends was increased dramatically because strong hydrogen bond occurs during CO₂ absorption. Subsequently, it was observed that the temperature increases the viscosity of

Table 3. Variation of density on virgin (0 min), CR (after 3 hr), and RS (after 1 hr).

Solvents	Density, $\times 10^3 \text{ Kg/m}^3$				
	Virgin	CR	RS-358K	RS-368K	RS-373K
28 wt% MEA+2 wt% [TBA][OAC]	0.999 \pm 0.001	1.092 \pm 0.003	1.088 \pm 0.002	1.073 \pm 0.002	1.065 \pm 0.000
25 wt% MEA +5 wt% [TBA][OAC]	0.997 \pm 0.000	1.084 \pm 0.000	1.071 \pm 0.000	1.061 \pm 0.001	1.042 \pm 0.002
20 wt% MEA +10 wt% [TBA][OAC]	0.996 \pm 0.001	1.063 \pm 0.001	1.053 \pm 0.001	1.043 \pm 0.000	1.017 \pm 0.000
28 wt% MEA+2 wt% [TBA][Br]	1.001 \pm 0.002	1.100 \pm 0.000	1.082 \pm 0.000	1.078 \pm 0.003	1.063 \pm 0.001
25 wt% MEA +5 wt% [TBA][Br]	1.002 \pm 0.001	1.091 \pm 0.002	1.083 \pm 0.003	1.067 \pm 0.001	1.044 \pm 0.000
20 wt% MEA +10 wt% [TBA][Br]	1.003 \pm 0.000	1.075 \pm 0.000	1.064 \pm 0.000	1.060 \pm 0.000	1.045 \pm 0.002

Abbreviations: CR, CO₂ rich; RS, regenerated solutions.**Figure 10. Variation of density on virgin; CR, CO₂-rich; RS, regenerated solutions.**

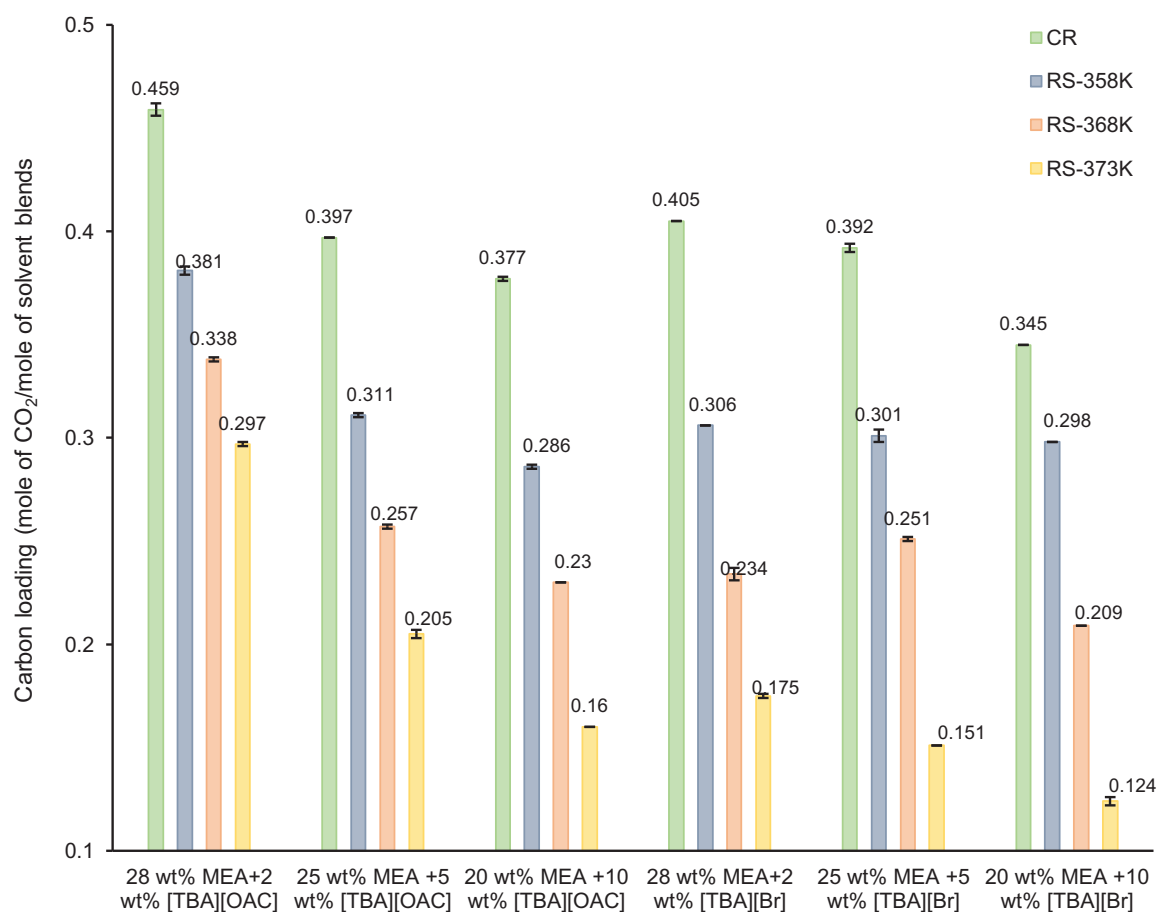
the regenerated solution (RS) decreases. This is because as the temperature increases, the intermolecular interaction between the molecules decreases, and then the viscosity decreases.

Effect of surface tension

The measured surface tension of virgin, CO₂-rich and lean solvent blends are listed in Table 6 and Fig. 13. From the table, it was noted that the surface tension of

Table 4. Variation of CO₂ loading on virgin (0 min), CR (after 3 hr), and RS (after 1 hr).

Solvents	CO ₂ loading (mole of CO ₂ / mole of solvent blend)			
	CR	RS-358K	RS-368K	RS-373K
28 wt% MEA+2 wt% [TBA][OAC]	0.459 ± 0.003	0.381 ± 0.006	0.338 ± 0.009	0.297 ± 0.013
25 wt% MEA +5 wt% [TBA][OAC]	0.397 ± 0.003	0.311 ± 0.010	0.257 ± 0.006	0.205 ± 0.002
20 wt% MEA +10 wt% [TBA][OAC]	0.377 ± 0.002	0.286 ± 0.017	0.230 ± 0.002	0.160 ± 0.003
28 wt% MEA+2 wt% [TBA][Br]	0.405 ± 0.008	0.306 ± 0.002	0.234 ± 0.006	0.175 ± 0.006
25 wt% MEA +5 wt% [TBA][Br]	0.392 ± 0.004	0.301 ± 0.000	0.251 ± 0.008	0.151 ± 0.005
20 wt% MEA +10 wt% [TBA][Br]	0.345 ± 0.002	0.298 ± 0.006	0.209 ± 0.018	0.124 ± 0.000

Abbreviations: CR, CO₂ rich; RS, regenerated solutions.**Figure 11. Variation of CO₂ loading on CR, CO₂-rich; RS, regenerated solutions.**

solvent blends decreases with rise in IL composition. This is because of more molecules tending to accumulate on the surface of the solvent at higher IL concentration. Generally, high surface tension results in more energy for regenerating the solvent. This behaviour was found in 30% MEA.³¹ For all solvent

blends, it was observed that the surface tension decreases as the regeneration temperature increases. Because the molecular attractive force between the molecules decreases as the temperature increases, which makes removal of CO₂ easier and reduces the surface tension.

Table 5. Effect of viscosity on virgin (0 min), CR (after 3 hr), and RS (after 1 hr).

Solvents	Viscosity (mPa.s)				
	Virgin	CR	RS-358K	RS-368K	RS-373K
28 wt% MEA+2 wt% [TBA][OAC]	2.11 ± 0.005	3.24 ± 0.007	3.20 ± 0.009	3.16 ± 0.006	3.01 ± 0.006
25 wt% MEA +5 wt% [TBA][OAC]	2.23 ± 0.006	3.34 ± 0.016	3.28 ± 0.013	3.19 ± 0.012	3.10 ± 0.015
20 wt% MEA +10 wt% [TBA][OAC]	2.30 ± 0.005	3.42 ± 0.012	3.38 ± 0.017	3.23 ± 0.024	3.17 ± 0.008
28 wt% MEA+2 wt% [TBA][Br]	1.98 ± 0.006	3.22 ± 0.007	3.17 ± 0.019	3.12 ± 0.016	2.93 ± 0.003
25 wt% MEA +5 wt% [TBA][Br]	2.07 ± 0.003	3.15 ± 0.003	3.13 ± 0.003	2.95 ± 0.009	2.79 ± 0.023
20 wt% MEA +10 wt% [TBA][Br]	2.27 ± 0.003	3.07 ± 0.003	3.02 ± 0.019	2.95 ± 0.003	2.75 ± 0.022

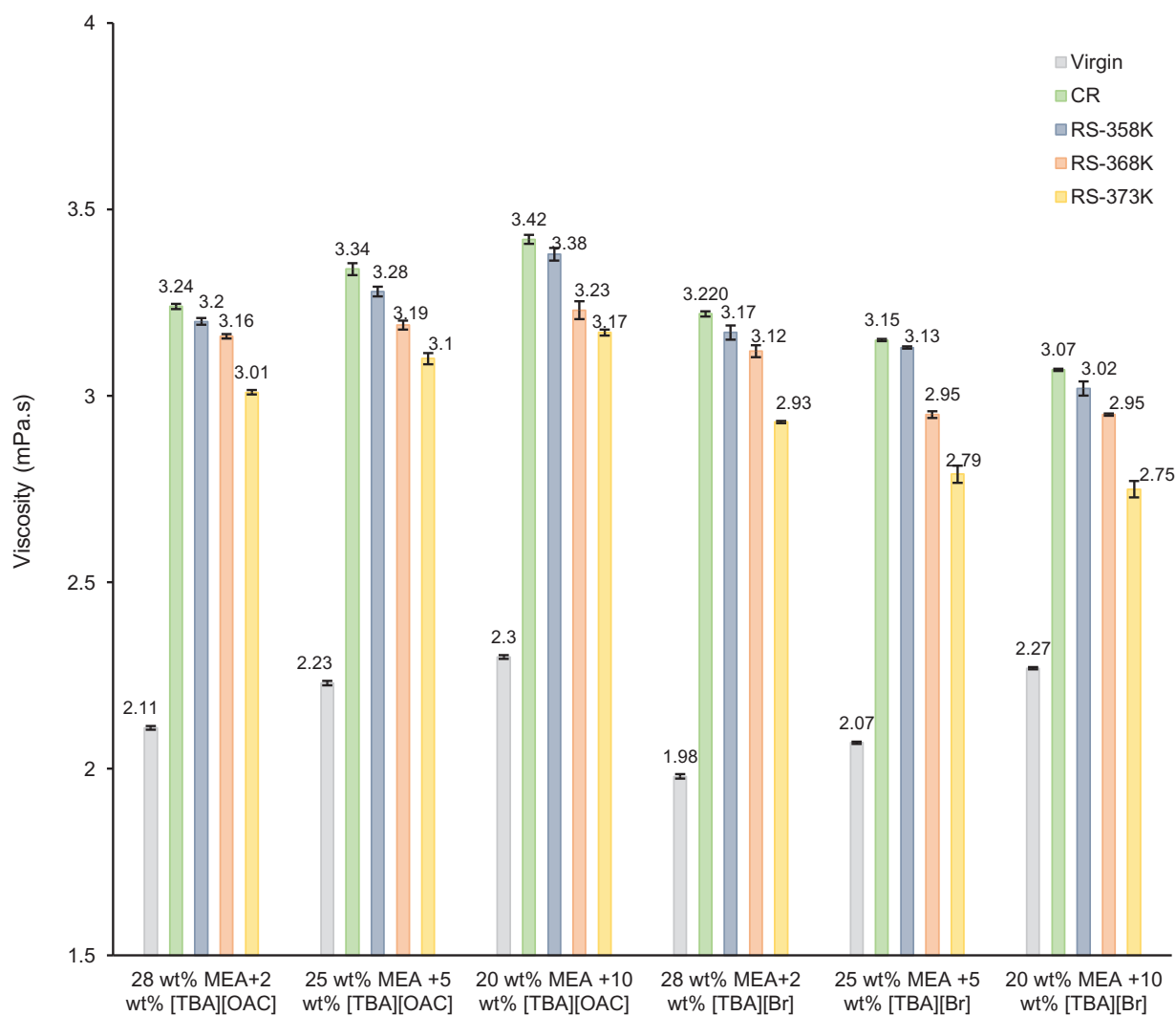
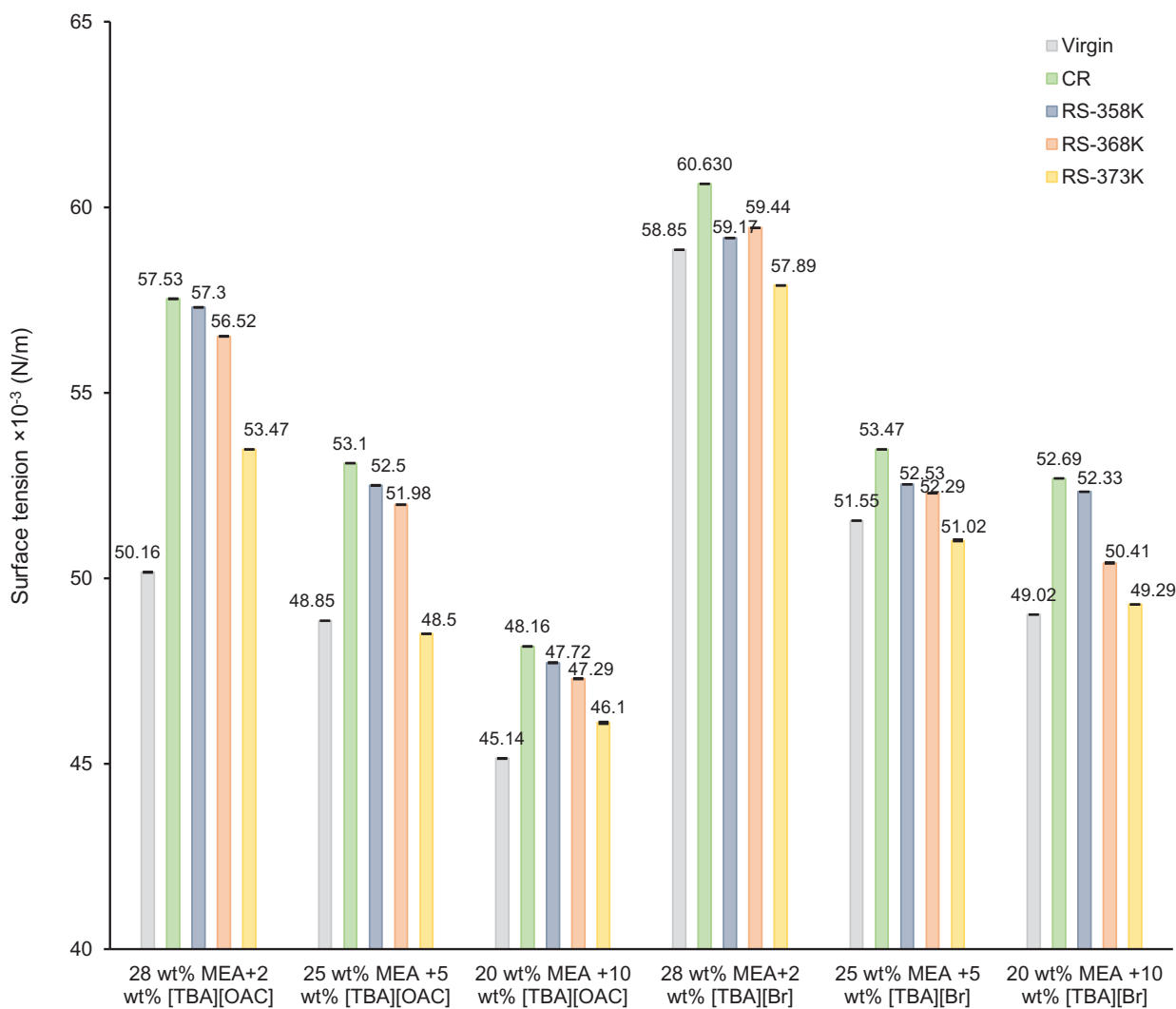
Abbreviations: CR, CO₂ rich; RS, regenerated solutions.**Figure 12. Effect of viscosity on virgin; CR, CO₂-rich; RS, regenerated solutions.**

Table 6. Effect of surface tension on (0 min), CR (after 3 hr), and RS (after 1 hr).

Solvents	Surface tension $\times 10^{-3}$ (N/m)				
	Virgin	CR	RS-358K	RS-368K	RS-373K
28 wt% MEA+2 wt% [TBA][OAC]	50.16 \pm 0.012	57.53 \pm 0.010	57.30 \pm 0.007	56.52 \pm 0.009	53.47 \pm 0.004
25 wt% MEA +5 wt% [TBA][OAC]	48.85 \pm 0.002	53.10 \pm 0.005	52.50 \pm 0.010	51.98 \pm 0.006	48.50 \pm 0.004
20 wt% MEA +10 wt% [TBA][OAC]	45.14 \pm 0.010	48.16 \pm 0.006	47.72 \pm 0.013	47.29 \pm 0.014	46.10 \pm 0.028
28 wt% MEA+2 wt% [TBA][Br]	58.85 \pm 0.001	60.63 \pm 0.003	59.17 \pm 0.005	59.44 \pm 0.001	57.89 \pm 0.005
25 wt% MEA +5 wt% [TBA][Br]	51.55 \pm 0.001	53.47 \pm 0.003	52.53 \pm 0.002	52.29 \pm 0.006	51.02 \pm 0.025
20 wt% MEA +10 wt% [TBA][Br]	49.0 \pm 0.001	52.6 \pm 0.001	52.3 \pm 0.003	50.4 \pm 0.017	49.2 \pm 0.004

Abbreviations: CR, CO₂ rich; RS, regenerated solutions.**Figure 13. Effect of surface tension on virgin; CR, CO₂-rich; RS, regenerated solutions.**

Conclusion

An experimental study of solvent regeneration was carried out on CO₂-rich aqueous solvent blends of MEA and ammonium-based ILs ([TBA][OAC], [TBA][Br]). The experimental results showed that the CO₂ content of all solvent blends decreases with the increase of regeneration time and temperature. The MEA blends with 2 wt% [TBA][Br] showed slight decrease in desorption rate when compared to 10% [TBA][OAC] with MEA blend used. Consequently, the regeneration efficiency of [TBA][Br] is relatively higher than that of [TBA][OAC] blends with MEA because of its lower viscosity. The extreme carbon lean condition was observed for MEA-[TBA][Br] blend using a minimum composition of 2 wt% [TBA][Br]. The regenerative energy requirement of the above mixture is 66.53 kJ mole⁻¹ of CO₂ and the energy consumed at 358 K is 41.97% less than the baseline MEA. Thus, the combination of 28 wt% MEA + 2 wt% [TBA][Br] can be a suitable solvent due to its lower energy consumption in solvent regeneration. This finding is essential for further research into solvent recycling and reuse, which can be expected to minimize solvent degradation and operating cost.

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Muthumari Perumal

Muthumari Perumal, currently pursuing doctorate study in the field of ionic liquids for solvent-based CO₂ capture process at Solvent Development for Clean Technology Lab, Department of Chemical Engineering at Sri Sivasubramaniya Nadar College of Engineering, Chennai. She received her

M.Tech in Biotechnology from Anna University Regional Campus, Coimbatore in 2017. She has published research articles in high reputed journals.



Dhanalakshmi Jayaraman

Dr Dhanalakshmi Jayaraman, working as Associate Professor in the Department of Chemical Engineering at Sri Sivasubramaniya Nadar College of Engineering, Chennai, since March 2015. She had received doctorate degree in chemical engineering from IIT Madras in the year 2014. Her research

work on the applications of ionic liquids for separation processes and prediction of phase equilibria in mixtures. As a Principal Investigator, she has received an External Research Grant of INR. 26.52 Lakhs from DST – SERB (Early Career Research Award), Delhi, India in the year 2016. She is the recipient of IIT Madras Travel Grant (2012) for AIChE conference, USA. She has published research papers in reputed refereed journals.