

Review

Ionic Liquids/Deep Eutectic Solvents-Based Hybrid Solvents for CO₂ Capture

Yanrong Liu ^{1,2}, Zhengxing Dai ³, Fei Dai ^{4,5,*} and Xiaoyan Ji ^{1,*}

- ¹ Energy Engineering, Division of Energy Science, Luleå University of Technology, 97187 Luleå, Sweden; yanrong.liu@ltu.se
- ² Swerim AB, Box 812, SE-97125 Luleå, Sweden
- ³ State Key Laboratory of Material-Oriented Chemical Engineering, Nanjing Tech University, Nanjing 211816, China; 201861104285@njtech.edu.cn
- ⁴ CAS Key Laboratory of Green Process and Engineering, Beijing Key Laboratory of Ionic Liquids Clean Process, State Key Laboratory of Multiphase Complex Systems, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China
- ⁵ School of Chemical Engineering, University of Chinese Academy of Sciences, Beijing 100049, China
- * Correspondence: fdai@ipe.ac.cn (F.D.); xiaoyan.ji@ltu.se (X.J.)

Received: 11 October 2020; Accepted: 26 October 2020; Published: 29 October 2020



Abstract: The CO_2 solubilities (including CO_2 Henry's constants) and viscosities in ionic liquids (ILs)/deep eutectic solvents (DESs)-based hybrid solvents were comprehensively collected and summarized. The literature survey results of CO₂ solubility illustrated that the addition of hybrid solvents to ILs/DESs can significantly enhance the CO₂ solubility, and some of the ILs-based hybrid solvents are super to DESs-based hybrid solvents. The best hybrid solvents of IL-H₂O, IL-organic, IL-amine, DES-H₂O, and DES-organic are [DMAPAH][Formate] (2.5:1) + H₂O (20 wt %) (4.61 mol/kg, 298 K, 0.1 MPa), [P₄₄₄₄][Pro] + PEG400 (70 wt %) (1.61 mol/kg, 333.15 K, 1.68 MPa), [DMAPAH][Formate] (2.0:1) + MEA (30 wt %) (6.24 mol/kg, 298 K, 0.1 MPa), [TEMA][CI]-GLY-H₂O 1:2:0.11 (0.66 mol/kg, 298 K, 1.74 MPa), and [Ch][Cl]-MEA 1:2 + DBN 1:1 (5.11 mol/kg, 298 K, 0.1 MPa), respectively. All of these best candidates show higher CO₂ solubility than their used pure ILs or DESs, evidencing that IL/DES-based hybrid solvents are remarkable for CO₂ capture. For the summarized viscosity results, the presence of hybrid solvents in ILs and DESs can decrease their viscosities. The lowest viscosities acquired in this work for IL-H₂O, IL-amine, DES-H₂O, and DES-organic hybrid solvents are [DEA][Bu] + H₂O (98.78 mol%) (0.59 mPa·s, 343.15 K), [BMIM][BF₄] + DETA (94.9 mol%) (2.68 mPa·s, 333.15 K), [L-Arg]-GLY 1:6 + H₂O (60 wt %) (2.7 mPa·s, 353.15 K), and [MTPP][Br]-LEV-Ac 1:3:0.03 (16.16 mPa·s, 333.15 K) at 0.1 MPa, respectively.

Keywords: ionic liquid; deep eutectic solvents; hybrid solvent; CO₂ solubility; Henry's constant; viscosity

1. Introduction

 CO_2 emission is an urgent issue due to its main contribution to global warming [1]. It has been evidenced that CO_2 capture is a promising route to mitigate CO_2 emissions, and in general, cost, energy demand, and environmental impact need to be considered when for selecting the potential CO_2 capture technologies [2]. At present, the absorption technology with 30 wt % MEA aqueous solution is the commercialized one. However, this technology with the corresponding solvent has the drawbacks of high energy demand (4.2 GJ/t CO_2), high cost ($0.19-1.31/t CO_2$), low thermal and chemical stability, and high volatility and corrosion [3–6], which underlines the necessity for developing greener and more efficient solvent for CO_2 capture.



2 of 20

Compared with the traditional amine-based solvent, the emerging new absorption solvents of ionic liquids (ILs) and deep eutectic solvents (DESs) have attracted more and more attention due to the merits of recyclability, good solvent stability, low energy demand, and environmentally friendly nature [7]. However, some of the ILs (e.g., $[P_{4442}][Cy-Suc]$, 2567 mPa·s at 303.15 K) [8] and DESs (e.g., [MTPP][Br]-GLY 1:4, 1658 mPa·s at 298.15 K) [9] have high viscosities that influence the rate of absorptions, inhibiting their industry applications. To cope with this disadvantage, hybrid ILs or DESs with water have been strongly recommended [9–14]. For example, Zhang et al. investigated the mass transfer feature in [BMIM][NO₃] + H₂O, evidencing that mass transfer increases with the increase of water content, e.g., the mass transfer of [BMIM][NO₃] (95 wt %) + H₂O (5 wt %) is $0.55 \times 10^5 \text{ m·s}^{-1}$, while it is $0.64 \times 10^5 \text{ m·s}^{-1}$ for [BMIM][NO₃] (90 wt %) + H₂O (10 wt %), which may due to the decrease of viscosity that from 35.74 ([BMIM][NO₃] (95 wt %) + H₂O (5 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([BMIM][NO₃] (90 wt %) + H₂O (10 wt %)) to 12.65 mPa·s ([B

Except for adding water to decrease the viscosity, organic solvents (e.g., PEG, TEG, and TG) can substitute water totally or partially to decrease the viscosity or to overcome high energy demand in IL + H_2O [15–17]. The experimental result indicates that TG can significantly decrease the viscosity of [P₆₆₆₁₄][4-Triz], especially at low temperatures, i.e., the viscosity of [P₆₆₆₁₄][4-Triz] is 4640 mPa·s at 278.15 K, while it is 163 mPa·s for [P₆₆₆₁₄][4-Triz] + TG (58.2 mol%) [18]. Liu et al. found that the addition of a certain amount of PEG200 to [Cho][Gly] + H_2O not only decreases the viscosity but also enhances the CO₂ solubility, as well as decreases the desorption enthalpy [17].

To further take the benefits of both neoteric and conventional solvents, IL–amine-based and superbase–amine-based DES hybrid absorbents have also been proposed and developed [19–25]. These hybrid solvents possess certain advantages of low energy demand, low water evaporation, and high CO₂ solubility compared to the commercialized MEA aqueous solution [26]. For example, Yang et al. reported that [BMIM][BF₄] (40 wt %) + MEA (30 wt %) + H₂O (30 wt %) has higher CO₂ solubility than MEA (30 wt %) + H₂O (70 wt %), and the energy demand reduced by 37.2% with respect to MEA (30 wt %) + H₂O (70 wt %) [21]. The CO₂ solubilities of four functionalized ILs in MDEA aqueous solution were investigated and compared with MEA + MDEA aqueous solution [27], showing that the CO₂ solubility of [N₂₂₂₂][Lys] (15 wt %) + MDEA (15 wt %) + H₂O (70 wt %) (0.64 mol/mol) > [N₁₁₁₁][Lys] (15 wt %) + MDEA (15 wt %) + H₂O (70 wt %) (0.64 mol/mol) > [N₁₁₁₁][Gly] (15 wt %) + MDEA (15 wt %) + H₂O (70 wt %) (0.36 mol/mol). For DES hybrid solvent of [Ch][Cl]-MEA 1:2 + DBN with volume ratio of 1:1, its CO₂ solubility improved from 3.29 to 5.11 mol/kg compared with [Ch][Cl]-MEA 1:2 at 298.15 K [25].

To develop the potential IL/DES-based hybrid solvents for CO_2 capture, CO_2 solubility (in accordance with Henry's constant for physical absorption) and viscosity are two key properties. Furthermore, the selectively for physical absorption and the CO_2 absorption enthalpy for chemical absorption are other concerns in development, while the research is still limited, especially when compared to those for CO_2 solubility and viscosity. Several reviews for IL/DES-based hybrid solvents from the aspect of CO_2 solubility, Henry's constant, and viscosity have been published. Babamohammadi et al. summarized the viscosities of IL + H₂O and IL + MEA/EG + H₂O until 2014 and the CO_2 solubilities of ILs (imidazolium- and ammonium-based ILs)-amine hybrid solvents since 2008 [28]. Gao et al. summarized the CO_2 solubility of 18 kinds of IL-amine based hybrid solvent until 2015. Huang et al. reviewed the advantages and disadvantages of five kinds of IL-hybrid solvents (i.e., IL-organic, normal IL-amine, normal IL aqueous-amine, functionalized IL-amine, and functionalized IL aqueous-amine) until 2016 for CO_2 capture [26], finding that IL-hybrid solvents can significantly reduce the viscosity. Lian et al. introduced the ILs-hybrid processes for CO_2 capture and compared the CO_2 solubilities for IL-DEA/DMEE/ethanol, indicating that IL-ethanol has the highest solubility of 2.3 mol/mol [29]. Recently, more IL-based hybrid solvents have been developed combined with property measurements, making it necessary to update the latest research progress. Meanwhile, to the best of our knowledge, there is no review article for the DES-based hybrid solvents.

To fulfill this gap and to promote the technology development on CO₂ capture in IL/DES-based hybrid solvents, this review summarizes the CO₂ solubilities (including Henry's constants) and viscosities of IL-based hybrid solvents since 2016 and DES-based hybrid solvents since 2014 to avoid the repetition of the published reviews. Finally, the best candidates for IL/DES-based hybrid solvents were obtained and compared with each other.

2. ILs-Based Hybrid Solvents

Regarding the CO₂ solubilities for 73 kinds of IL–H₂O and 37 kind of IL–organic-based hybrid solvents since 2016, 28 types of IL–amine hybrid solvents since 2018 together with 62 Henry's constants have been reported. The results were collected and summarized in Tables 1 and 2. The viscosities for 30 kinds of IL–H₂O and 121 IL–organic/organic aqueous solution hybrid solvents since 2016, and 15 kinds of IL–amine/amine aqueous solution hybrid solvents since 2018 have been determined, and these are listed in Table 3. The full names of ILs-based hybrid solvents are displayed in Table S1.

2.1. CO₂ Solubility and Henry's Constant

2.1.1. IL-H₂O

The effect of the addition of H₂O in [DMAPAH][Formate] (0.5:1, 1.0:1, 2.0:1, 2.5:1) was studied by Vijayaraghavan et al. [30]. Except for the molar ratio of amine to acid of 0.5:1, CO₂ solubility initially increased up to a certain water amount of 20 wt % (Figure 1); then, it steadily decreased as the H_2O concentration increased. As shown in Figure 1, the best candidate for CO_2 capture is [DMAPAH][Formate] (2.5:1) + H₂O (20 wt %) (5.69 mol/kg). The CO₂ solubility in [TMGH][Im] + H₂O (1–25 wt %) was studied by Li et al. [12]. The result indicates that CO₂ solubility first increased at a water content range of 1–7 wt %, and then, it decreased when the water content was larger than 7 wt % in [TMGH][Im], resulting in the best absorption capacity of 4.23 mol/kg for [TMGH][Im] + H₂O (7 wt %). Huang et al. reported that a $[P_{4442}][Suc]$ structure with basic anion can improve the reaction of CO_2 and [P₄₄₄₂][Suc] aqueous solution by forming bicarbonate and conjugate acid [31]. The result indicates that the CO₂ solubility of $[P_{4442}][Suc] + H_2O(3.3 \text{ wt }\%)$ (1.9 mol/mol) is slightly higher than $[P_{4442}][Suc]$ (1.87 mol/mol); however, the addition of 8.8 and 17.6 wt % of H_2O to $[P_{4442}][Suc]$ has a negative effect on their CO_2 solubilities compared with $[P_{4442}][Suc]$. As shown in Table 1, various compositions of [P₄₄₄₄][HCOO] in water were measured at 0.1 MPa in the temperature range of 248.75–324.65 K for CO₂ solubility and compared with [N₂₂₂₄][Ac] and [N₂₂₂₂][Ac] [32]. The results indicate that the CO₂ solubility of $[P_{4444}][HCOO] + H_2O$ first increased from 0.01 to 1 mol/mol at the water contents range of 29–66 mol% and then decreased with the increase of water contents of 70–91 mol%. Temperature also affects the absorption performance of $[P_{4444}][HCOO] + H_2O$. For example, at 273.15 K, the CO₂ solubility of $[P_{4444}][HCOO] + H_2O$ (<80 mol%) is higher than $[N_{2224}][Ac] + H_2O$ and $[N_{2222}][Ac] + H_2O$, while at 298.15 K, it is higher than $[N_{2224}][Ac] + H_2O$ at a water content less than 50 mol%, but it is lower at a water content high than 50 mol%. For 323.15 K, the CO₂ solubility in $[P_{4444}][HCOO] + H_2O (<80 \text{ mol}\%)$ is lower than $[N_{2222}][Ac] + H_2O$. Nathalia et al. [33] evidenced that there are three processes for CO₂ capture in [BMMIM][Im] and [BMMIM][Ac] aqueous solutions, i.e., physical, CO₂-imidazolium adduct generation, and bicarbonate formation, resulting in a maximum CO₂ solubility of 8.15 mol/mol in [BMMIM][Im] + H₂O (99.9 mol%). The CO₂ solubility in [P₄₄₄₃][Gly] + H₂O (59.9, 80.1, 90, 95 wt %) was measured at temperatures ranging from 278.14 to 348.05 K and pressures of 0.1–7.75 MPa [34]. A feature of physical absorption in these hybrid solvents was observed. The best CO₂ solubility of 2.44 mol/kg was acquired for $[P_{4443}][Gly] + H_2O$ (59.9 wt %) at 298.06 K and 4.6 MPa. Aghaie et al. tested the CO₂ solubilities of [HMIM][Tf₂N], [HMIM][FAP], and [BMIM][Ac] aqueous solutions [35]. The result indicates that the CO_2 solubility reduced by 45% in these three ILs aqueous solutions compared to the solubility of CO_2 in pure IL at 298 K and water content of 10 wt %.



Figure 1. Effects of water on CO₂ solubility in [DMAPAH][Formate]. The values in Figure 1 are cited from Vijayaraghavan et al. [30]. Copyright 2018 Elsevier.

2.1.2. IL-Organic/Organic Aqueous Solution

Huang et al. investigated the CO₂ capture capacity in [TETAH][Lys] + ethanol + H₂O [36], finding that the CO₂ solubility first increases with the increase of volume ratio of ethanol in water from 10:0 to 5:5 v/v, and then, it decreases from 5:5 to 2:8 v/v. Compared with the reported results, the maximum CO₂ solubility of 2.45 mol/mol for [TETAH][Lys] + ethanol + H₂O (5:5 v/v) is higher than those of [P₆₄₄₄][Lys] [37], [C₂NH₂MIM][Lys] [38], and [TETAH][Lys] + H₂O. Taheri et al. measured the CO₂ solubility of [AMIM][Tf₂N] + methanol at temperature and pressure ranges of 313.2–353.2 K and 0.98–6.19 MPa, respectively, indicating that the presence of methanol in [AMIM][Tf₂N] enhances the CO₂ solubility and results in a maximum of 3.89 mol/mol [39].

In order to overcome the drawbacks of high CO₂ capture enthalpy and water volatilization in ILs aqueous solution, PEG200 was introduced in [Cho][Gly] + H_2O [17]. CO₂ solubility was measured in such solvent at 308.15-338.15 K and pressure lower than 0.68 MPa, and the CO₂ desorption enthalpy was estimated. Owing to its physical-chemical properties, [Cho][Gly] (30 wt %) + PEG200 (30 wt %) + H_2O (40 wt %) has a higher CO₂ solubility (0.41–1.23 mol/kg) and regeneration efficiency (95%) compared with [Cho][Gly] (30 wt %) + H_2O (70 wt %). Li et al. found that the addition of PEG200 to [Cho][Pro] not only improves the absorption rate but also enhances the desorption efficient, resulting in a maximum CO₂ solubility of about 0.6 mol/mol for [Cho][Pro] + PEG200 with the mass ratios of 1:1, 1:2, 1:3, respectively [40]. PEG400 was introduced to [P₄₄₄₄][Gly], [P₄₄₄₄][Ala], and [P₄₄₄₄][Pro] [41], evidencing that $[P_{4444}][Pro] + PEG400$ (70 wt %) (1.61 mol/kg) > $[P_{4444}][Gly] + PEG400$ (70 wt %) $(1.58 \text{ mol/kg}) > [P_{4444}][Ala] + PEG400 (70 \text{ wt }\%) (1.57 \text{ mol/kg})$. The effect of three types of PEG (i.e., PEG200, PEG300, and PEG400) and water content on the CO₂ solubility in [DETAH][Br] and [DETAH][BF₄] at 293.15 K and 0.1 MPa was investigated by Chen et al. [42]. The result evidenced that the CO₂ solubility follows the order of [DETAH][Br] + PEG200 (1.18 mol/mol) > [DETAH][Br] + PEG300 (0.87 mol/mol) > [DETAH][BF₄] + PEG200 (0.65 mol/mol) > [DETAH][Br] + PEG300 (0.32 mol/mol) at a mass ratio of [DETAH][Br]:PEG = 1:4. Additionally, the CO₂ solubility in [DETAH][Br] + PEG200 + H_2O (4.7 wt %) (1.18 mol/mol) is higher than that in [DETAH][Br] + PEG200 + H_2O (1.3 wt %) (1.05 mol/mol), which may be because water weakens the interaction of the IL cation and anion and enhances the interaction with CO_2 . Due to the high CO_2 solubility of PEO1000 (0.35 mol/mol, 323 K, 4.98 MPa), it is introduced to [N₄₁₁₁][Tf₂N]. A maximum CO₂ solubility of 1.16 mol/mol was acquired at 323 K, 4.99 MPa for [N₄₁₁₁][Tf₂N] + PEO1000 (75 mol%), which is higher than

that of pure $[N_{4111}][Tf_2N]$ (0.14 mol/mol, 323 K, 5 MPa) [43]. Additionally, a higher amount of PEO1000 in $[N_{4111}][Tf_2N]$ corresponds to higher CO₂ solubility, which is attributed to the strong interaction between CO₂ and PEO. However, Jiang et al. [44] found that increasing the molar fraction of TEG in [BMIM][BF₄]/[BMIM][BF₄] + H₂O results in a decrease of CO₂ solubilities, which is on the contrary of the result from Lepre et al. [43]. Moreover, with increasing the [BMIM][BF₄] contents in [BMIM][BF₄] + TEG mixtures, the Henry's constant is increased (Table 2) [44]. The Henry's constant of [BMIM][BF₄] + TEG is higher than [BMIM][BF₄] but lower than TEG, indicating that CO₂ is more soluble in [BMIM][BF₄]. The CO₂ solubilities of [P₆₆₆₁₄][3-Triz] + TG (30 mol%) and [P₆₆₆₁₄][4-Triz] + TG (30 mol%) at 313.15–353.6 K and pressure less than 3 MPa were measured by Fillion et al. [18]. The CO₂ solubility of [P₆₆₆₁₄][4-Triz] + TG (30 mol%) is 2.23 mol/mol at 313.15 K and 3.03 MPa, which is higher than [P₆₆₆₁₄][3-Triz] + TG (30 mol%) (1.55 mol/mol, 313.15 K, and 2.68 MPa). [TEPAH][2-MI] combined with propan-1-ol (NPA) and EG was used for CO₂ capture [45]. The result indicates that the CO₂ solubility in [TEPAH][2-MI] + NPA + EG can reach up to 1.72 mol/mol, which was much higher than that of [C₃OHmim]Cl + MEA (0.3 mol/mol) [46], AMP + MEA + H₂O (0.5 mol/mol) [47], [P₆₆₆₁₄][Gly] (1.26 mol/mol) [48], and TETA + AMP + ethanol (1.03 mol/mol) [49].

2.1.3. IL-Amine/Amine Aqueous Solution

Three base-rich diamino ILs of [DMAPAH][Formate], [DMEDAH][Formate], and [DMAPAH] [Octanoate] were synthesized with different molar ratios of base to acid (0.5:1, 1.0:1, 2.0:1, and 2.5:1, respectively) and hybrid with MEA for CO₂ capture, respectively [30]. According to Table 1 and Figure 2, the hybrid solvents of the synthesized ILs with an additional MEA showed enhanced CO₂ solubility, which agrees with the studies from Zeng et al. [50] and Meng et al. [51] that applied MDEA and DMEE as the hybrid solvents to [DMAPAH][Ac] and [N₁₁₁₁][Lys], respectively. Among them, [DMAPAH][Formate] (2.0:1) + MEA (30 wt %) with 6.24 mol/kg was identified to be the best one for CO₂ capture. The CO₂ capture performance in [BMPyrr][DCA] (5 wt %) + DEA (35 wt %) + H₂O (60 wt %) and [BMPyrr][DCA] (10 wt %) + DEA (30 wt %) + H₂O (60 wt %) were studied by Salleh et al. [52] and compared with DEA (40 wt %) + H₂O (60 wt %). The result indicates that the CO₂ solubility increases with increasing the [BMPyrr][DCA] amount in the hybrid solvent. However, the CO₂ solubilities of these two hybrid solvents are lower than those in DEA (40 wt %) + H₂O (60 wt %).



Figure 2. Effects of MEA on CO₂ solubilities in [DMAPAH][Formate] and [DMAPAH][Octanoate] [30]. Copyright 2018 Elsevier.

In conclusion, (1) a certain amount of water in ILs (mainly for chemical-based ILs) can enhance the CO₂ solubility, due to the decrease in viscosity and the formation of new products (e.g., carbamate and bicarbonate). However, excess water in ILs corresponds to a low ILs concentration and results in the decrease of CO₂ solubility; (2) the IL–organic and IL–organic aqueous solution as absorbents exhibit remarkable CO₂ capture performances, including high absorption capacity and low desorption enthalpy. The organic molecular weight, type, and water content in ILs can affect their CO₂ capture performance. Based on the summarized result, the organic solvent with low molecular weight together with a certain amount of water is beneficial for capturing CO₂; (3) IL–MEA shows better CO₂ capture performance than that of IL–MDEA and IL–DMEE; additionally, the IL–amine based hybrid solvent has higher CO₂ solubility than that of IL–H₂O and IL–organic hybrid solvents. The best for each of them are [DMAPAH][Formate] (2.0:1) + MEA (30 wt %) (6.24 mol/kg, 298 K, 0.1 MPa), [DMAPAH][Formate] (2.5:1) + H₂O (20 wt %) (4.61 mol/kg, 298 K, 0.1 MPa), and [P₄₄₄₄][Pro] + PEG400 (70 wt %) (1.61 mol/kg, 333.15 K, 1.68 MPa). Sometimes, the presence of water in IL–organic/amine hybrid solvents improves the CO₂ solubility.

2.2. Viscosity

2.2.1. IL-H₂O

The viscosities of IL–H₂O hybrid solvents are given in Table 3. Figure 3 displays the viscosities of $[DEA][Bu] + H_2O [14]$; the result indicated that the [DEA][Bu] has a strong hygroscopic characteristic. The viscosity of $[DEA][Bu] + H_2O$ decreases with the increase of water content and temperature. Yasaka et al. found that the viscosities of $[P_{4444}][HCOO] + H_2O$ decreased with water contents from 25 (356 mPa·s) to 91 mol% (14.4 mPa·s), corresponding to an increase of the CO₂ solubility between the water content of 25 and 50 mol%, and then, they decrease from 50 to 91 mol%, which is regarded as the typical property of carboxylate ILs [32]. Aghaie et al. measured the viscosities of $[HMIM][Tf_2N]$, [HMIM][FAP], and [BMIM][Ac] aqueous solutions at 298–333 K, 2 MPa, and water mass percentages of 0.1, 1, 2, 5, and 10 wt %, respectively [35]. The result indicates that water has a significant effect on [BMIM][Ac] viscosity, e.g., the viscosity of [BMIM][Ac] decreased from 47.64 (pure IL) to 3.77 (10 wt % H₂O) mPa·s at 333 K. However, for $[HMIM][Tf_2N]$ and [HMIM][FAP], their viscosities only slightly decrease at 0.1–10 wt % water. For example, the viscosity of [HMIM][FAP] at 333 K is 20.72 mPa·s, while it is 20.47 mPa·s for $[HMIM][FAP] + H_2O (10 wt %)$. Additionally, increasing the water amount in these three ILs results in the decrease of CO₂ solubility.



Figure 3. Viscosity of [DEA][Bu] + H₂O, 283.15–343.15 K, 0.1 MPa [14]. Copyright 2018 Elsevier.

2.2.2. IL–Organic/Organic Aqueous Solution

For $[TETAH][Lys] + H_2O +$ ethanol, it was observed that its viscosity increased with the decrease amount of ethanol, corresponding to a decreased CO_2 solubility [36]. Liu et al. found that the viscosities of [Cho][Gly] (84.3-7.2 mPa·s), PEG200 (31.8-8.8 mPa·s), and [Cho][Gly] + PEG200 (70 wt %) $(101.3-28.6 \text{ mPa} \cdot \text{s})$ are much higher than that of [Cho][Gly]/H₂O + PEG200 (30 wt %) (7.96-3.43 mPa \cdot \text{s}) at 308.15–338.15 K [17]. To avoid the decrease of absorption rate by increasing PEG200, less than 30 wt % of PEG200 was recommended to [Cho][Gly] + PEG200 + H_2O . The viscosities of [P₄₄₄₄][Gly], [P₄₄₄₄][Ala], and [P₄₄₄₄][Pro] hybrid with PEG400 were measured at 298.15–393.15 K and 0.1 MPa [41]. The result indicates that the viscosities of these amino-acid ILs + PEG400 are about half with respect to the pure amino acid ILs at 298.15 K. Chen et al. reported that the viscosity of the [DETAH][Br] + PEG200 (80 wt %) is 71.7 mPa·s at 293.15 K and 0.1 MPa [42]. Despite the viscosities' increases with the increasing long-chain polymers, i.e., [N₄₁₁₁][Tf₂N] + PEO222 < [N₄₁₁₁][Tf₂N] + PEO500 < [N₄₁₁₁][Tf₂N] + PEO1000, $[N_{4111}]$ [Tf₂N] + PEO1000 was proposed for CO₂ capture due to the higher CO₂ solubility of the pure PEO1000 [43]. The viscosities for 18 kinds of ILs (imidazolium- and phosphonium-based) + TG at different of mole ratio of each of these ILs at 278.15–323.15 K and 0.1 MPa were studied [18], evidencing that the presence of TG can significantly decrease the viscosity, resulting in about 50 mPa·s for these hybrid solvents (Table 3).

2.2.3. IL-Amine

As shown in Table 3, the viscosities of the $[N_{1111}][Lys] + DMEE$ and $[BMIM][BF_4] + DETA$ hybrid solvents increased with increasing the content of ILs and decreasing temperature [51,53]. Based on the study of Meng et al. [51], the viscosity of $[N_{1111}][Lys] + DMEE$ significantly decreased when the IL is <60 wt % compared to the pure $[N_{1111}][Lys]$.

From Table 3, it can be found that the viscosities of IL-based hybrid solvents are very sensitive to H_2O , organic, and amine solvents. Their viscosities can significantly decrease with the increased amount of H_2O , PEG, TEG, TG, DMEE, and DETA; however, it increases with the increased amount of methanol. The lowest viscosity obtained from Table 3 for IL– H_2O , IL–organic, and IL–amine based hybrid solvents are [DEA][Bu] + H_2O (98.78 mol%) (0.59 mPa·s, 343.15 K), [N₁₁₁₄][Tf₂N] + PEO222 (75.06 mol%) (2.57 mPa·s, 353.15 K), and [BMIM][BF₄] + DETA (94.9 mol%) (2.68 mPa·s, 353.15 K) at 0.1 MPa, respectively.

IL-Based Hybrid Solvents	T/K	P/MPa	CO ₂ Solubility (mol CO ₂ /kg Absorbent)	CO ₂ Solubility (mol CO ₂ /mol IL)	Ref.				
IL-H ₂ O									
[DMAPAH][Formate] (0.5:1, 1.0:1, 2.0:1, 2.5:1) + H ₂ O (20, 33, 42, 49, 59, 75 wt %)	298.15	0.1	0.41–0.14, 2.73–0.77, 4.23–1.41, 4.61–1.41		[30]				
[P ₄₄₄₂][Suc] + H ₂ O (3.3, 8.8, 17.6 wt %)	293	0.1		1.9, 1.24, 0.81	[31]				
[TMGH][Im] + H ₂ O (1, 2, 3, 5, 7, 10, 15, 20, 25 wt %)	313.15	0.1	3.39-4.23		[12]				
[P ₄₄₄₄][HCOO] + H ₂ O (29, 32, 38, 50, 66, 70, 73, 79, 86, 91 mol%)	248.75-333.15	0.1		0.01-1	[32]				
[BMMIM][Im] + H ₂ O (67, 91, 99, 99.9 mol%)	/	2	1.29, 2.30, 0.83, 0.55	0, 0, 0.36, 8.15	[33]				
[BMMIM][Im] + H ₂ O (99 mol%)	/	1		0.45	[33]				
[BMMIM][Ac] + H ₂ O (67, 99.9 mol%)	/	2		0, 6.95	[33]				
[P ₄₄₄₃][Gly] + H ₂ O (59.9, 80.1, 90, 95 wt %)	278.09-348.10	0.103-7.53	0.091 - 2.44	0.23-12.13	[34]				
[HMIM][Tf ₂ N] + H ₂ O (0.1, 1, 2, 5, 10 wt %)	298-333	2		0.30-0.13	[35]				
[HMIM][FAP] + H ₂ O (0.1, 1, 2, 5, 10 wt %)	298-333	2		0.48-0.14	[35]				
[BMIM][Ac] + H ₂ O (0.1, 1, 2, 5, 10 wt %)	298-323	2		0.46-0.32	[35]				
[Cho][Gly] + H ₂ O (70 wt %)	308.15-338.15	0.0046-0.68	0.36-1.24	0.21 - 0.74	[17]				
IL-organic/organic aqueous solution									
[TETAH][Lys] + ethanol + H ₂ O	202	0.1		2.45 1.52	[26]				
(H ₂ O:ethanol = 8:2, 6:4, 5:5, 4:6, 3:7, 2:8 v:v)	505	0.1		2.40-1.00	[30]				
[AMIM][Tf ₂ N] + methanol (20, 50, 80 wt %)	313.2-353.2	0.98-6.19		2.15-3.89	[39]				
[Cho][Gly] (30 wt %) + PEG200 (10 wt%) + H ₂ O (60 wt %)	308.15-338.15	0.0054 - 0.68	0.37-1.22	0.22-0.72	[17]				
[Cho][Gly] (30 wt %) + PEG200 (20 wt%) + H ₂ O (50 wt %)	308.15-338.15	0.0039-0.68	0.41-1.21	0.24 - 0.72	[17]				
[Cho][Gly] (30 wt %) + PEG200 (30 wt%) + H ₂ O (40 wt %)	308.15-338.15	0.0065-0.68	0.41-1.23	0.24-0.73	[17]				
[Cho][Pro] + PEG200 (50, 67, 75 wt %)	308.15-353.15	0.0041 - 0.11		0.099-0.61	[40]				
[P ₄₄₄₄][Gly] + PEG400 (70 wt %)	333.15-413.15	0.088 - 1.7	0.19–1.58	0.19-1.23	[41]				
[P ₄₄₄₄][Ala] + PEG400 (70 wt %)	333.15-413.15	0.093-1.7	0.11-1.57	0.11-1.26	[41]				
[P ₄₄₄₄][Pro] + PEG400 (70 wt %)	333.15-413.15	0.096-1.71	0.15-1.61	0.17-1.41	[41]				
[DETAH][Br] + PEG200 (80 wt %)	293.15	0.1		1.18	[42]				
[DETAH][Br] + PEG300 (80 wt %)	293.15	0.1		0.87	[42]				
[DETAH][Br] + PEG400 (80 wt %)	293.15	0.1		0.32	[42]				
[DETAH][BF ₄] + PEG200 (80 wt %)	293.15	0.1		0.65	[42]				
[DETAH][Br] + PEG200 + H ₂ O (1.3, 4.7 wt %)	293.15	0.1		1.05, 1.18	[42]				
[N ₁₁₁₄][Tf ₂ N] + PEO1000 (10.44, 28.27, 50.22, 75.31 mol%)	323, 343	0.02-0.5		0.0057-1.16	[43]				
[BMIM][BF ₄] + TEG (20, 50, 80 mol%)	273.15-353.15	0.42-3.55		0.051-1.72	[44]				

Table 1. CO₂ solubility of ionic liquid (IL)-based hybrid solvents.

Table 1. Cont.

IL-Based Hybrid Solvents	T/K	P/MPa	CO ₂ Solubility (mol CO ₂ /kg Absorbent)	CO ₂ Solubility (mol CO ₂ /mol IL)	Ref.			
[BMIM][BF ₄] (56 mol%) + TEG (14 mol%) + H ₂ O (30 mol%)	293.15-333.15	0.38-4.17		0.051-0.96	[44]			
$[BMIM][BF_4] (35 \text{ mol}\%) + TEG (35 \text{ mol}\%) + H_2O(30 \text{ mol}\%)$	293.15-333.15	0.57-4.37		0.079-1.4	[44]			
[BMIM][BF ₄] (14 mol%) + TEG (56 mol%) + H ₂ O(30 mol%)	293.15-333.15	0.64 - 4.46		0.15-1.84	[44]			
[P ₆₆₆₁₄][3-Triz] + TG (30 mol%)	313.15-353.6	0.037-2.75		0.06-1.55	[18]			
[P ₆₆₆₁₄][4-Triz] + TG (30 mol%)	313.15-353.6	0.07-3.03		0.075-2.23	[18]			
[TEPAH][2-MI] + NPA + EG	303.15	0.1		1.72	[45]			
IL-amine/amine aqueous solution								
[BMPyrr][DCA] (5 wt %) + DEA (35 wt%) + H ₂ O (60 wt %)	333.15	0.5–0.7		0.49-0.71	[52]			
[BMPyrr][DCA] (10 wt %) + DEA (30wt%) + H ₂ O (60 wt %)	333.15	0.5-0.7		0.58-0.81	[52]			
[DMAPAH][Formate] (1.0:1, 2.5:1) + MEA	298.15	0.1	3.82, 4.52		[30]			
[DMEDAH][Formate] (1.0:1, 2.5:1) + MEA	298.15	0.1	3.52, 4.07		[30]			
[DMAPAH][Formate] (0.5:1, 1.0:1, 2.0:1, 2.5:1) + MEA (30 wt %)	298.15	0.1	2.85, 4.57, 6.24, 5.89		[30]			
[DMAPAH][Octanoate] (0.5:1, 1.0:1, 2.0:1, 2.5:1) + MEA (30 wt %)	298.15	0.1	2.29, 2.69, 3.30, 3.65		[30]			
[DMAPAH][Ac] + MDEA (20, 33, 43, 50, 60, 67, 80 mol%)	308.15	0.0–3.0		0–3.13	[50]			
[DMAPAH][Ac] + MDEA (50 mol%)	298.15-328.15	0.0-3.0	0-3.21		[50]			
[N ₁₁₁₁][Lys] + DMEE (95, 90, 80, 60, 40 wt %)	303	0.1	0.28-1.69	1.22-0.61	[51]			
[N ₁₁₁₁][Lys] + DMEE (80 wt %)	313, 323	0.1		0.76, 0.72	[51]			

The CO₂ solubility unit with mole scale for reference [45] is mol CO₂/mol absorbent, while in [52] is mol CO₂/mol amine.

Table 2. Henry's constant of IL-based hybrid solvents

IL-Based Hybrid Solvents	T (K)	Henry's Constant (MPa)	Ref.			
IL-H ₂ O						
[P ₄₄₄₃][Gly] + H ₂ O (59.9, 80.1, 90%, 95 wt %)	278.14-348.05	2.8–5.05, 1.53–3.3, 0.87–2.81, 0.35–1.03	[34]			
[Cho][Gly] + H ₂ O (70 wt %)	308.15-338.15	40.56–58	[17]			
IL–organic						
[Cho][Gly] (30 wt %) + PEG200 (10 wt %) + H ₂ O (60 wt %)	308.15-338.15	36.6–52.2	[17]			
[Cho][Gly] (30 wt %) + PEG200 (20 wt %) + H ₂ O (50 wt %)	308.15-338.15	33-47.03	[17]			
[Cho][Gly] (30 wt %) + PEG200 (30 wt %) + H ₂ O (40 wt %)	308.15-338.15	31.49-46.79	[17]			
[BMIM][BF ₄] + TEG (20, 50, 80 mol%)	273.15-353.15	4.84-20.02, 5.65-22, 6.48-27.63	[44]			

IL-Based Hybrid Solvents	T/K	P/MPa	Viscosity (mPa·s)	Ref.
Ш	-H ₂ O			
[DEA][Bu] + H ₂ O (98.78, 95.49, 90.01, 85.7, 79.49, 69.36, 51.21, 33.16, 2.6 mol%)	283.15–343.15	0.1	2.24–0.59, 6.91–1.10, 19.26–2.25, 32.03–3.24, 56.72–4.81, 106.48–7.09, 158.58–9.28, 158.29–9.64, 130.18–8.70	[14]
[P ₄₄₄₄][HCOO] + H ₂ O (25, 50, 60, 74, 80, 91 mol%)	298.15	0.1	356, 146, 97, 48, 35.3, 14.4	[32]
[HMIM][Tf ₂ N] + H ₂ O (0.1, 1, 2, 5, 10 wt %)	298–333	2	69.18–18.6, 66.54–18.2, 63.71–17.54, 55.95–15.7, 45.05–13.01	[35]
[HMIM][FAP] + H ₂ O (0.1, 1, 2, 5, 10 wt %)	298–333	2	88.09–20.71, 84.54–20.70, 80.76–20.66, 70.4–20.6, 56.01–20.47	[35]
[BMIM][Ac] + H ₂ O (0.1, 1, 2, 5, 10 wt %)	298–333	2	389–45.30, 225.86–30.04, 135.5–20.45, 44.08–8.78, 14.34–3.77	[35]
IL-organic/orga	nic aqueous solution			
[TETAH][Lys] + ethanol + H ₂ O (H ₂ O:ethanol = 8:2, 6:4, 5:5, 4:6, 3:7, 2:8 v:v)	303	0.1	2.57, 3.00, 3.50, 3.81, 3.74, 3.51	[36]
[Cho][Gly] + PEG200 (70 wt %)	308.15-338.15	0.1	101.3–28.6	[17]
[Cho][Gly]/H ₂ O + PEG200 (30 wt %)	308.15, 338.15	0.1	7.96, 3.43	[17]
[P ₄₄₄₄][Gly] + PEG400 (70 wt %)	298.15-393.15	0.1	180.47-8.96	[41]
[P ₄₄₄₄][Ala] + PEG400 (70 wt %)	298.15-393.15	0.1	216.64–9.13	[41]
[P ₄₄₄₄][Pro] + PEG400 (70 wt %)	298.15-393.15	0.1	481–14.1	[41]
[DETAH][Br] + PEG200 (80 mol%)	293.15	0.1	71.7	[42]
[N ₁₁₁₄][Tf ₂ N] + PEO222 (26.23, 50.07, 75.06 mol%)	293.15-353.15	0.1	135.22–2.57	[43]
[N ₁₁₁₄][Tf ₂ N] + PEO500 (20.90, 44.22, 70.40 mol%)	293.15-353.15	0.1	135.22–7.51	[43]
[N ₁₁₁₄][Tf ₂ N] + PEO1000 (25.04, 50.22, 75.31 mol%)	318.15-353.15	0.1	51.72-17.22	[43]
[P ₆₆₆₁₄][4-NO ₂ imid] + TG (18.9, 40.4 mol%)	278.15-323.15	0.1	1503–57	[18]
[P ₆₆₆₁₄][4,5-CNimid] + TG (20.1, 40.1 mol%)	278.15-323.15	0.1	1051–63	[18]
[P ₆₆₆₁₄][Tf ₂ N] + TG (19.1, 36.6, 55.9, 66.4 mol%)	278.15-323.15	0.1	589–51	[18]
[P ₆₆₆₁₄][2-CH ₃ ,5-NO ₂ imid] + TG (15.1, 39.9 mol%)	278.15-323.15	0.1	2535–57	[18]
[P ₆₆₆₁₄][DCA] + TG (9.9, 20.3, 29.9, 50.1, 65.2 mol%)	278.15-323.15	0.1	1130–54	[18]
[HMIM][Tf ₂ N] + TG (9.9, 19, 27.6, 41.3, 54.4 mol%)	278.15-293.15	0.1	171–50	[18]
[P ₆₆₆₁₄][BrBnIm] + TG (10.2, 13.8, 21.8, 34.6, 47.8, 63.8 mol%)	278.15-323.15	0.1	4400–51	[18]
[P ₆₆₆₁₄][Ac] + TG (10.3, 20.7, 29.9, 40, 49.9 mol%)	278.15-323.15	0.1	1130–57	[18]
[HMMIM][Tf ₂ N] + TG (10.2, 20.5, 30.6, 39.9, 49.9, 59.9 mol%)	278.15-323.15	0.1	370–48	[18]
[P ₆₆₆₁₄][4-Triz] + TG (4.6, 12.6, 19.6, 24.9, 30.6, 39, 48.4, 58.2 mol%)	278.15-323.15	0.1	3290–53	[18]
[P ₆₆₆₁₄][3-Triz] + TG (8.3, 12.2, 20.7, 31, 39.8, 50.1, 70 mol%)	278.15-323.15	0.1	1180–51	[18]
[P ₄₄₄₁₂][3-Triz] + TG (6.2, 12.3, 15.5, 21.4, 30.5, 39.5, 49.5, 59.6 mol%)	278.15-323.15	0.1	1980–57	[18]

 Table 3. Viscosity of IL-based hybrid solvents.

Table 3. Cont.

IL-Based Hybrid Solvents	T/K	P/MPa	Viscosity (mPa·s)	Ref.
[P ₂₂₂₈][4-NO ₂ pyra] + TG (5.1, 10.2, 19.9, 30, 40.4, 50.3 mol%)	278.15-323.15	0.1	1010–55	[18]
[P ₂₂₂₈][4-NO ₂ imid] + TG (10, 20, 30.1, 40.1, 50.1 mol%)	278.15-323.15	0.1	700–55	[18]
[P ₂₂₂₈][2-CH ₃ ,5-NO ₂ imid] + TG (3.6, 6.7, 11.5, 23.3, 30, 39.9, 50, 59.8 mol%)	278.15-323.15	0.1	2730–51	[18]
[mm(butene)im][4-NO ₂ pyra] + TG (4.8, 10, 19.9, 29.9, 39.9, 50, 60 mol%)	278.15-323.15	0.1	4300–51	[18]
[P ₂₂₂₄][2-CH ₃ ,5-NO ₂ imid] + TG (4.8, 10.2, 20.1, 29.9, 40.1, 50.1 mol%)	278.15-323.15	0.1	1540–57	[18]
[pmmim][4-NO ₂ pyra] + TG (5, 10, 20.1, 30.2, 39.9, 49.9, 60 mol%)	278.15-323.15	0.1	5420–59	[18]
[TEPAH][2-MI] + NPA + EG	303.15	0.1	3.66	[45]
IL-a	amine			
[N ₁₁₁₁][Lys] + DMEE (95, 90, 80, 60, 40 wt %)	303–333	0.1	12.09–4.88, 20.70–6.98, 30.00–9.30, 80.46–20, 101.86–25.35	[51]
[BMIM][BF ₄] + DETA (94.9, 80.14, 70.02, 60.05, 50.08, 40.95, 30.26, 19.88, 10.52, 5.04 mol%)	298.15–333.15	0.1	6.71–2.68, 12.1–4.89, 17.35–6.69, 23.66–8.72, 31.54–11.19, 40.07–13.88, 52.59–17.57, 67.10–21.30, 82.58–24.18, 91.94–24.54	[53]

3. DESs-Based Hybrid Solvents

The CO_2 solubility data for 33 kinds of DESs-based hybrid solvents, together with viscosities for six types of DESs-based hybrid solvents since 2016, and Henry's constants for 21 kinds of DES-based hybrid solvents since 2013 have been reported, as summarized in Tables 4 and 5. The full names of the studied components of DESs are given in Table S1.

3.1. CO₂ Solubility

3.1.1. DES-H₂O

The CO₂ solubility of DESs using water as a hybrid solvent was investigated by Sarmad et al. at 298.15 K and pressure up to 2 MPa (Table 4) [9]. From this study, the CO₂ solubility of DES-based hybrid solvents can be affected by four factors. (1) The first is pressure: the CO₂ solubility increased with the increasing pressure. For instance, the CO₂ solubility of [TEMA][Cl]-GLY-H₂O 1:2:0.11 increased from 0.025 to 0.66 mol/kg at a pressure range of 0.14 to 1.74 MPa. (2) The second factor is the mole ratio of water: the CO₂ solubility of [TEMA][Cl]-GLY-H₂O 1:2:0.05 first decreased at pressure range of 0.23–0.85 MPa, and then, it increased from 1.25 to 1.98 MPa compared with [TEMA][Cl]-GLY 1:2. Upon increasing the mole ratio of water, the CO₂ solubility of [TEMA][Cl]-GLY-H₂O 1:2:0.05. This result agrees with the DES-based hybrid solvent of [BTMA][Cl]-GLY-H₂O 1:2:0.011 and 1:2:0.05. (3) The third factor is the type of hydrogen bond acceptor (HBA)—for example, the CO₂ solubility of [TEMA][Cl]-GLY-H₂O 1:2:0.05 (1.98 MPa, 0.66 mol/kg) > [BTMA][Cl]-GLY-H₂O 1:2:0.05 (2.02 MPa, 0.29 mol/kg).

Harifi-Mood et al. investigated the Henry's constants for [Ch][Cl]-EG aqueous solution at temperatures of 303.15–323.15 K [54]. As shown in Table 5, the results indicate that the Henry's constant of CO_2 increases with increasing water amount in the absorbent, corresponding to a decrease of CO_2 solubility. This result agrees with the measured Henry's constants of [Ch][Cl]-EG, [Ch][Cl]-GLY, and [Ch][Cl]-MA aqueous solution from 303.15 to 313.15 K by Lin et al. [55].

3.1.2. DES-Organic

The CO₂ solubilities in DESs–organic hybrid solvents are given in Table 4. The result indicates that the addition of 0.03 mol of acetic acid in [MTPP][Br]-LEV 1:3 significantly enhanced the CO₂ solubility and decreased the viscosity compared with [MTPP][Br]-LEV 1:3 [9].

A superbase can participate in the reaction of DES and CO₂, thus increasing the CO₂ solubility. Bhawna et al. studied the CO₂ solubility by three hybrid superbases of TBD, DBN, and DBU with DESs of [Ch][Cl]-Urea 1:2 and [Ch][Cl]-EG 1:2, respectively [25]. The result indicates that all of these three superbases can enhance the CO₂ solubility, and among them, TBD has the highest capacity, followed by DBU and DBN. The further addition of glycerol in these hybrid solvents decreased the CO₂ solubility. For the effect of these three superbases on different male ratio of DESs, it is found that [Ch][Cl]-MEA 1:2 + DBN (5.11 mol/kg) > [Ch][Cl]-MEA + TBD 1:4 (3.91 mol/kg) > [Ch][Cl]-MEA 1:2 + DBU (3.54 mol/kg). Additionally, the same phenomenon was observed that the addition of glycerol in these [Ch][Cl]-MEA-based hybrid solvents can decrease the CO₂ solubility.

An imidazole (Im)-derived DESs of [BMIM][Cl]-Im was synthesized for CO_2 capture by hybrid with DBN [56]. These hybrid solvents show remarkable CO_2 capture capacity up to 1.00 mol/mol, following the order of DBN-[BMIM][Cl]-Im 1:1:2 > DBN-[BMIM][Cl]-Im 1:1:1 > DBN-[BMIM][Cl]-Im 1:2:1. The theoretical calculation indicates that DBN plays a key role in the absorption process by forming a strong hydrogen bond with the derived [BMIM⁺-COO⁻].

In conclusion, the obtained best DES–H₂O hybrid solvent is [TEMA][Cl]-GLY-H₂O 1:2:0.11 (0.66 mol/kg, 298 K, 1.74 MPa), while it is [Ch][Cl]-MEA 1:2 + DBN 1:1 (5.11 mol/kg, 298 K, 0.1 MPa) for DES–organic hybrid solvent. These values are lower than the best IL–H₂O and IL–organic hybrid solvents, respectively.

DES-Based Hybrid Solvents	Amount/ Ratio	Temperature (K)	Pressure (MPa)	CO ₂ Solubility (mol CO ₂ /kg DES)	CO ₂ Solubility (mol CO ₂ /mol DES)	Viscosity (mPa·s)	Ref.
			DES-H ₂	0			
[BHDE][Cl]-GLY-H ₂ O	1:3:0.11	298.15	0.23-2.02	0.037-0.21		32.76-17.11	[9]
[BTMA][Cl]-GLY-H ₂ O	1:2:0.05	298.15	0.21-2.02	0.044-0.29		70.76-26.81	[9]
[BTMA][Cl]-GLY-H ₂ O	1:2:0.11	298.15	0.26-2.03	0.016-0.33		22.19-14.41	[9]
[TEMA][Cl]-GLY-H ₂ O	1:2:0.05	298.15	0.23-1.98	0.009–0.66		90.98-23.56	[9]
[TEMA][Cl]-GLY-H ₂ O	1:2:0.11	298.15	0.14 - 1.74	0.025-0.66		48.63-19.31	[9]
[L-Arg]-GLY 1:6-H ₂ O	10, 20, 30, 40, 50, 60 wt %	303.15–353.15	0.1			434.2–26.3, 179.3–19.5, 45.2–7.2, 17.7–4.4, 8.0–3.6, 4.6–3.0	[57]
			DES-orga	nic			
[Ch][Cl]-GLY-AC	1:1:1	298.15	0.26-2.01	0.052-0.43		138.51-28.81	[9]
[MTPP][Br]-LEV-AC	1:3:0.03	298.15	0.29-2.06	0.18-1.32		40.12-16.16	[9]
[Ch][Cl]-Urea 1:2 + TBD	1:10	333.15	0.1	0.42	0.68		[25]
[Ch][Cl]-Urea 1:2 + DBU	1:10	333.15	0.1	0.56	1.21		[25]
[Ch][Cl]-Urea 1:2 + DBN	1:10	333.15	0.1	0.76	1.11		[25]
[Ch][Cl]-Urea 1:2-GLY + TBD	5:40:10	333.15	0.1	0.4	0.66		[25]
[Ch][Cl]-Urea 1:2-GLY + DBU	5:40:10	333.15	0.1	0.45	0.81		[25]
[Ch][Cl]-Urea 1:2-GLY + DBN	5:40:10	333.15	0.1	0.27	0.40		[25]
[Ch][Cl]-EG 1:2 + DBU	1:10	333.15	0.1	0.7	1.16		[25]
[Ch][Cl]-EG 1:2 + DBN	1:10	333.15	0.1	0.77	1.06		[25]
[Ch][Cl]-EG 1:2 + TBD	1:10	333.15	0.1	0.63	0.95		[25]
[Ch][Cl]-EG 1:2-GLY + DBU	5:40:10	333.15	0.1	0.68	1.16		[25]
[Ch][Cl]-EG 1:2 + DBU	1:10	298.15	0.1	0.83	1.41		[25]
[Ch][Cl]-EG 1:2 + DBN	1:10	298.15	0.1	0.86	1.19		[25]
[Ch][Cl]-EG 1:2 + TBD	1:10	298.15	0.1	0.76	1.17		[25]
[Ch][Cl]-EG 1:2-GLY + DBU	5:40:10	298.15	0.1	0.64	1.12		[25]
[Ch][Cl]-EG 1:2-GLY + DBN	5:40:10	298.15	0.1	0.74	1.05		[25]
[Ch][Cl]-EG 1:2-GLY + TBD	5:40:10	298.15	0.1	0.73	1.17		[25]
[Ch][Cl]-MEA 1:2 + DBU	1:10	298.15	0.1	3.54	2.86		[25]
[Ch][Cl]-MEA 1:2 + DBN	1:10	298.15	0.1	5.11	6.70		[25]
[Ch][Cl]-MEA 1:4 + TBD	1:10	298.15	0.1	3.91	5.77		[25]
[Ch][Cl]-MEA 1:2-GLY + DBU	5:40:10	298.15	0.1	3.26	5.46		[25]

Table 4. CO₂ solubilities and viscosities of deep eutectic solvent (DES)-based hybrid solvents.

_

Table 4.	Cont.
----------	-------

DES-Based Hybrid Solvents	Amount/ Ratio	Temperature (K)	Pressure (MPa)	CO ₂ Solubility (mol CO ₂ /kg DES)	CO ₂ Solubility (mol CO ₂ /mol DES)	Viscosity (mPa∙s)	Ref.
[Ch][Cl]-MEA 1:2-GLY + DBN	5:40:10	298.15	0.1	1.67	2.28		[25]
[Ch][Cl]-MEA 1:2-GLY + TBD	5:40:10	298.15	0.1	3.63	5.56		[25]
[BMIM][Cl]-Im + DBN	1:1:1	298.15-328.15	0.1		0.81-1.02		[56]
[BMIM][Cl]-Im + DBN	1:1:2	298.15-328.15	0.1		0.88-0.97		[56]
[BMIM][Cl]-Im + DBN	1:2:1	298.15-328.15	0.1		0.91-1.07		[56]

The ratio of DES–organic for reference [9] is mole ratio, while in [25] is volume ratio.

Table 5. Henry's constant of DES–H₂O hybrid solvents.

DES-H ₂ O	Т (К)	Henry's Constant	Ref.
[Ch][Cl]-EG + H ₂ O (10, 20, 30, 40, 50, 60, 70, 80, 90 mol%)	303.15-313.15	27–296	[54]
[Ch][Cl]-EG + H ₂ O (20, 40, 60, 80 wt %)	303.15-313.15	3965.5-2805.5	[55]
[Ch][Cl]-GLY + H ₂ O (20, 40, 60, 80 wt %)	303.15-313.15	3818.8–3185.2	[55]
[Ch][Cl]-MA + H ₂ O (20, 40, 60, 80 wt %)	303.15-313.15	4021.6-2946.2	[55]

The Henry's constant unit in reference of [54] is MPa, while it is $kPa \cdot m^3 \cdot kmol^{-1}$ for reference [55].

3.2. Viscosity

The DESs consisting of glycerol as the hydrogen bond donor (HBD) exhibited high viscosity. Meanwhile, their viscosities increased considerably with an increase in the amount of dissolved CO_2 . As shown in Table 4, using water as a hybrid solvent in glycerol-based DESs can significantly decrease the viscosity of the DES [9]. For example, the viscosity of [BTMA][Cl]-GLY 1:2 decreased from 1017.67 to 70.76 mPa·s when adding a 0.05 molar ratio of water in [BTMA][Cl]-GLY 1:2 (i.e., [BTMA][Cl]-GLY-H₂O 1:2:0.05), but limiting the contribution of H₂O to CO₂ solubility. Meanwhile, increasing the water content of 0.11 mol in [BTMA][Cl]-GLY 1:2 results in a considerably reduced viscosity, which agrees with the results in the [TEMA][Cl]-GLY 1:3 decreased the viscosity from 32.76 to 17.11 mPa·s at 298.15 K and 0.23–2.02 MPa, and it increased the CO₂ solubility from 0.037 to 0.21 mol/kg. The viscosity of the DES decreased sharply with the increase of water contents, giving an option to lower the viscosity of the DES decreased sharply with the increase of water contents, giving an option to lower the viscosity [57].

In a word, adding water and organic solvents in DES can significantly decrease the viscosity.

4. Comparison of CO₂ Solubility and Viscosity

The obtained best candidates of IL–H₂O, IL–organic, IL–amine, DES–H₂O, and DES–organic hybrid solvents were compared with each other and their pure ILs and DESs (Figure 4). As shown in Figure 4, for either the IL-based or DES-based hybrid solvents, their CO₂ solubilies are higher than their pure IL/DES under the same condition. For example, the CO₂ solubility of [DMAPAH][Formate] (2.5:1) + H₂O (20 wt %) is 4.61 mol/kg at 298 K and 0.1 MPa, while it is 2.32 mol/kg for [DMAPAH][Formate] (2.5:1). This result indicates that IL/DES-based hybrid solvents are remarkable ones for CO₂ capture. Additionally, the IL-based hybrid solvent shows better CO₂ capture performance compared with the DES-based hybrid solvent, as shown in Figure 4. Figure 5 gives the comparison of viscosities for these IL/DES-based hybrid solvents and pure IL and DES at 333.15 K and 0.1 MPa. As shown in Figure 5, the addition of hybrid solvents can significantly decrease the viscosity compared to pure ILs and DESs, which are beneficial to accelerate mass transfer during capturing CO₂.



Figure 4. Comparison of CO₂ solubility for IL and IL-based hybrid solvent, as well as DES and DES-based hybrid solvent.

400 350 300

Viscosity (mPa·s)



Figure 5. Comparison of viscosity for IL and IL-based hybrid solvent, as well as DES and DES-based hybrid solvent at 333.15 K and 0.1 MPa.

Viscosity is the key factor for impeding the mass transfer of gas in absorbent [58]. For example, Gómez-Coma et al. investigated the viscosity and mass transfer performance of $[EMIM][Ac] + H_2O$ for CO₂ capture [59], finding that the viscosity of [EMIM][Ac] + H₂O decreased from 150 to 20 mPa·s with the increase of water content from 0-40 wt %. For the mass transfer coefficient, it is first increased from 1.7×10^{-5} to 9.34×10^{-5} m·s⁻¹ with the increasing of water from 0 to 30 wt % in [EMIM][Ac] and then decreased to $6.81 \times 10^{-5} \text{ m} \cdot \text{s}^{-1}$ when water content up to 40 wt %. Huang et al. evidenced that the low viscosity of IL-MEA aqueous solution corresponds to a high mass transfer performance [60], i.e., [EMIM][Br] (20 wt %) + MEA (5 wt %) + H₂O (75 wt %) (11.57 × 10⁶ mol·m⁻³·s⁻¹·Pa⁻¹, $1.23 \text{ mPa} \cdot \text{s}) > [\text{BMIM}][\text{Br}] (20 \text{ wt }\%) + \text{MEA} (5 \text{ wt }\%) + \text{H}_2\text{O} (75 \text{ wt }\%) (11.04 \times 10^6 \text{ mol} \cdot \text{m}^{-3} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}, \text{mol} \cdot \text{m}^{-3} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}) = 1.23 \text{ mPa} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1} \cdot \text{Pa}^{-1} \cdot \text{s}^{-1} \cdot \text{s}^{-1} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1} \cdot \text{s}^{-1} \cdot \text{s}^{-1} \cdot \text{s}^{-1$ $1.3 \text{ mPa} \cdot \text{s} > [\text{EMIM}][\text{Br}] (30 \text{ wt }\%) + \text{MEA} (5 \text{ wt }\%) + \text{H}_2\text{O} (65 \text{ wt }\%) (9.86 \times 10^6 \text{ mol} \cdot \text{m}^{-3} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1})$ $1.42 \text{ mPa} \cdot \text{s} > [BMIM][Br] (30 \text{ wt }\%) + MEA (5 \text{ wt }\%) + H_2O (65 \text{ wt }\%) (9.67 \times 10^6 \text{ mol} \cdot \text{m}^{-3} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1},$ 1.6 mPa·s). A similar phenomenon can be found in DESs hybrid solvent. Ma et al. indicated that a small amount of water in [BTMA][Cl]-GLY 1:2 not only decreases the viscosity but also improves the CO₂ solubility due to the increase of the mass transfer [61], while excess water in [BTMA][Cl]-GLY 1:2 results in a decrease of CO₂ solubility, which is in agreement with Li et al. [12].

5. Conclusions

This review summarizes the research work on developing ILs/DESs-based hybrid solvents (i.e., IL-H₂O, IL-organic/organic aqueous solution, IL-amine, DES-H₂O, and DES-organic) for CO₂ capture, including CO₂ solubility, Henry's constant, and viscosity. The results illustrate that the addition of hybrid solvents to ILs and DESs can decrease the viscosity and enhance the CO₂ solubility. IL-amine based hybrid solvents are super to IL-H₂O and IL-organic/organic aqueous solution, and some of the IL-based hybrid solvents show better performance than that of DES-based hybrid solvents. Additionally, some of the IL/DES hybrid solvents have higher CO₂ solubility compared to their pure IL/DES, indicating that the addition of hybrid solvent to IL/DES is possible to develop greener and more efficient absorbents for CO_2 capture. To develop the efficient IL/DES hybrid solvents for CO_2 capture, the following aspects are suggested for consideration to decrease the viscosity and increase the CO₂ solubility: (1) hybrid of functional ILs/DESs that have high CO₂ solubilities with a certain amount of water; (2) the addition of organic solvent which has a small molecular weight to the ILs/DESs; and (3) applying amine solvent which has good CO₂ capture capacity to ILs and DESs.

Supplementary Materials: The following is available online at http://www.mdpi.com/2073-4352/10/11/978/s1, Table S1: Full names and abbreviations of ILs, components of DESs and hybrid solvents.

Author Contributions: Writing—original draft preparation, Y.L.; investigation, Z.D., writing—review and editing, F.D., conceptualization and supervision, X.J. All authors have read and agreed to the published version of the manuscript.

Funding: This work is financially supported by Carl Tryggers Stiffelse foundation (No. 18:175). X.J. thanks the financial support from the Swedish Energy Agency (No. P47500-1) and K. C. Wang Education Foundation (No. GJTD-2018-04). F.D. thanks the financial support from the National Nature Science Foundation of China (21808223).

Conflicts of Interest: The author declares no conflict of interest.

References

- Zhou, W.; Wang, J.; Chen, P.; Ji, C.; Kang, Q.; Lu, B.; Li, K.; Liu, J.; Ruan, R. Bio-mitigation of carbon dioxide using microalgal systems: Advances and perspectives. *Renew. Sustain. Energy Rev.* 2017, 76, 1163–1175. [CrossRef]
- 2. MacDowell, N.; Florin, N.; Buchard, A.; Hallett, J.; Galindo, A.; Jackson, G.; Adjiman, C.S.; Williams, C.K.; Shah, N.; Fennell, P. An overview of CO₂ capture technologies. *Energy Environ. Sci.* **2010**, *3*, 1645. [CrossRef]
- Oko, E.; Zacchello, B.; Wang, M.H.; Fethi, A. Process analysis and economic evaluation of mixed aqueous ionic liquid and monoethanolamine (MEA) solvent for CO₂ capture from a coke oven plant. *Greenh. Gases* 2018, *8*, 686–700. [CrossRef]
- 4. Kothandaraman, A.; Nord, L.; Bolland, O.; Herzog, H.J.; McRae, G.J. Comparison of solvents for post-combustion capture of CO₂ by chemical absorption. *Energy Procedia* **2009**, *1*, 1373–1380. [CrossRef]
- 5. Lawal, A.; Wang, M.; Stephenson, P.; Obi, O. Demonstrating full-scale post-combustion CO₂ capture for coal-fired power plants through dynamic modelling and simulation. *Fuel* **2012**, *101*, 115–128. [CrossRef]
- 6. Ramdin, M.; de Loos, T.W.; Vlugt, T.J.H. State-of-the-art of CO₂ capture with ionic liquids. *Ind. Eng. Chem. Res.* **2012**, *51*, 8149–8177. [CrossRef]
- Zeng, S.J.; Zhang, X.P.; Bai, L.; Zhang, X.C.; Wang, H.; Wang, J.J.; Bao, D.; Li, M.D.; Liu, X.Y.; Zhang, S.J. Ionic-liquid-based CO₂ capture systems: Structure, interaction and process. *Chem. Rev.* 2017, *117*, 9625–9673. [CrossRef] [PubMed]
- 8. Huang, Y.J.; Cui, G.K.; Wang, H.Y.; Li, Z.Y.; Wang, J.J. Tuning ionic liquids with imide-based anions for highly efficient CO₂ capture through enhanced cooperations. *J. CO2 Util.* **2018**, *28*, 299–305. [CrossRef]
- 9. Sarmad, S.; Xie, Y.J.; Mikkola, J.-P.; Ji, X.Y. Screening of deep eutectic solvents (DESs) as green CO₂ sorbents: From solubility to viscosity. *New J. Chem.* **2017**, *41*, 290–301. [CrossRef]
- 10. Liu, H.J.; Pan, Y.; Yao, H.; Zhang, Y. Enhancement of carbon dioxide mass transfer rate by (ionic liquid)-in-water emulsion. *Adv. Mater. Res.* **2014**, *881–883*, 113–117. [CrossRef]
- Chu, C.Y.; Zhang, F.B.; Zhu, C.Y.; Fu, T.T.; Ma, Y.G. Mass transfer characteristics of CO₂ absorption into 1-butyl-3-methylimidazolium tetrafluoroborate aqueous solution in microchannel. *Int. J. Heat Mass Trans.* 2019, 128, 1064–1071. [CrossRef]
- 12. Li, F.F.; Bai, Y.G.; Zeng, S.J.; Liang, X.D.; Wang, H.; Huo, F.; Zhang, X.P. Protic ionic liquids with low viscosity for efficient and reversible capture of carbon dioxide. *Int. J. Greenh. Gas Control* **2019**, *90*, 102801. [CrossRef]
- 13. Zhang, X.; Bao, D.; Huang, Y.; Dong, H.F.; Zhang, X.P.; Zhang, S.J. Gas-liquid mass-transfer properties in CO₂ absorption system with ionic liquids. *AIChE J.* **2014**, *60*, 2929–2939. [CrossRef]
- Alcantara, M.L.; Santos, J.P.; Loreno, M.; Ferreira, P.I.S.; Paredes, M.L.L.; Cardozo-Filho, L.; Silva, A.K.; Lião, L.M.; Pires, C.A.M.; Mattedi, S. Low viscosity protic ionic liquid for CO₂/CH₄ separation: Thermophysical and high-pressure phase equilibria for diethylammonium butanoate. *Fluid Phase Equilibr*. 2018, 459, 30–43. [CrossRef]
- Li, J.; You, C.J.; Chen, L.F.; Ye, Y.M.; Qi, Z.W.; Sundmacher, K. Dynamics of CO₂ Absorption and Desorption Processes in Alkanolamine with Cosolvent Polyethylene Glycol. *Ind. Eng. Chem. Res.* 2012, *51*, 12081–12088. [CrossRef]
- 16. Yang, Z.-Z.; He, L.-N.; Zhao, Y.-N.; Li, B.; Yu, B. CO₂ capture and activation by superbase/polyethylene glycol and its subsequent conversion. *Energy Environ. Sci.* **2011**, *4*, 3971. [CrossRef]

- Liu, S.D.; Li, H.; Chen, Y.F.; Yang, Z.H.; Wang, H.L.; Ji, X.Y.; Lu, X.H. Improved CO₂ separation performance of aqueous choline-glycine solution by partially replacing water with polyethylene glycol. *Fluid Phase Equilibr*. 2019, 495, 12–20. [CrossRef]
- 18. Fillion, J.J.; Bennett, J.E.; Brennecke, J.F. The viscosity and density of ionic liquid + tetraglyme mixtures and the effect of tetraglyme on CO₂ Solubility. *J. Chem. Eng. Data* **2017**, *62*, 608–622. [CrossRef]
- 19. Sairi, N.A.; Ghani, N.A.; Aroua, M.K.; Yusoff, R.; Alias, Y. Low pressure solubilities of CO₂ in guanidinium trifluoromethanesulfonate-MDEA systems. *Fluid Phase Equilibr.* **2015**, *385*, 79–91. [CrossRef]
- Sairi, N.A.; Yusoff, R.; Alias, Y.; Aroua, M.K. Solubilities of CO₂ in aqueous N-methyldiethanolamine and guanidinium trifluoromethanesulfonate ionic liquid systems at elevated pressures. *Fluid Phase Equilibr.* 2011, 300, 89–94. [CrossRef]
- 21. Yang, J.; Yu, X.H.; Yan, J.Y.; Tu, S.-T. CO₂ capture using amine solution mixed with ionic liquid. *Ind. Eng. Chem. Res.* **2014**, *53*, 2790–2799. [CrossRef]
- Xu, F.; Gao, H.S.; Dong, H.F.; Wang, Z.L.; Zhang, X.P.; Ren, B.Z.; Zhang, S.J. Solubility of CO₂ in aqueous mixtures of monoethanolamine and dicyanamide-based ionic liquids. *Fluid Phase Equilibr.* 2014, 365, 80–87. [CrossRef]
- 23. Gao, Y.; Zhang, F.; Huang, K.; Ma, J.-W.; Wu, Y.-T.; Zhang, Z.-B. Absorption of CO₂ in amino acid ionic liquid (AAIL) activated MDEA solutions. *Int. J. Greenh. Gas Control* **2013**, *19*, 379–386. [CrossRef]
- 24. Baj, S.; Siewniak, A.; Chrobok, A.; Krawczyk, T.; Sobolewski, A. Monoethanolamine and ionic liquid aqueous solutions as effective systems for CO₂ capture. *J. Chem. Technol. Biotechnol.* **2013**, *88*, 1220–1227. [CrossRef]
- 25. Bhawna; Pandey, A.; Pandey, S. Superbase-added choline chloride-based deep eutectic solvents for CO₂ capture and sequestration. *ChemistrySelect* **2017**, *2*, 11422–11430. [CrossRef]
- 26. Huang, K.; Chen, F.-F.; Tao, D.-J.; Dai, S. Ionic liquid–formulated hybrid solvents for CO₂ capture. *Curr. Opin. Green Sustain.* **2017**, *5*, 67–73. [CrossRef]
- 27. Zhang, F.; Fang, C.-G.; Wu, Y.-T.; Wang, Y.-T.; Li, A.-M.; Zhang, Z.-B. Absorption of CO₂ in the aqueous solutions of functionalized ionic liquids and MDEA. *Chem. Eng. J.* **2010**, *160*, 691–697.
- 28. Babamohammadi, S.; Shamiri, A.; Aroua, M.K. A review of CO₂ capture by absorption in ionic liquid-based solvents. *Rev. Chem. Eng.* **2015**, *31*, 383–412. [CrossRef]
- 29. Lian, S.H.; Song, C.F.; Liu, Q.L.; Duan, E.H.; Ren, H.W.; Kitamura, Y. Recent advances in ionic liquids-based hybrid processes for CO₂ capture and utilization. *J. Environ. Sci.* **2021**, *99*, 281–295. [CrossRef]
- 30. Vijayaraghavan, R.; Oncsik, T.; Mitschke, B.; MacFarlane, D.R. Base-rich diamino protic ionic liquid mixtures for enhanced CO₂ capture. *Sep. Purif. Technol.* **2018**, *196*, 27–31. [CrossRef]
- 31. Huang, Y.J.; Cui, G.K.; Zhao, Y.L.; Wang, H.Y.; Li, Z.Y.; Dai, S.; Wang, J.J. Reply to the correspondence on "Preorganization and cooperation for highly efficient and reversible capture of low-concentration CO₂ by ionic liquids". *Angew. Chem.* **2019**, *58*, 386–389. [CrossRef]
- 32. Yasaka, Y.; Kimura, Y. Effect of temperature and water concentration on CO₂ absorption by tetrabutylphosphonium formate ionic liquid. *J. Chem. Eng. Data* **2016**, *61*, 837–845. [CrossRef]
- 33. Simon, N.M.; Zanatta, M.; dos Santos, F.P.; Corvo, M.C.; Cabrita, E.J.; Dupont, J. Carbon dioxide capture by aqueous ionic liquid solutions. *ChemSusChem* **2017**, *10*, 4927–4933. [CrossRef]
- 34. Chen, Y.; Guo, K.H.; Huangpu, L. Experiments and modeling of absorption of CO₂ by amino-cation and amino-anion dual functionalized ionic liquid with the addition of aqueous medium. *J. Chem. Eng. Data* **2017**, *62*, 3732–3743. [CrossRef]
- Aghaie, M.; Rezaei, N.; Zendehboudi, S. Assessment of carbon dioxide solubility in ionic liquid/toluene/water systems by extended PR and PC-SAFT EOSs: Carbon capture implication. *J. Mol. Liq.* 2019, 275, 323–337. [CrossRef]
- 36. Huang, Q.S.; Jing, G.H.; Zhou, X.B.; Lv, B.H.; Zhou, Z.M. A novel biphasic solvent of amino-functionalized ionic liquid for CO₂ capture: High efficiency and regenerability. *J. CO*₂ *Util.* **2018**, *25*, 22–30. [CrossRef]
- Qian, Y.H.; Jing, G.H.; Lv, B.H.; Zhou, Z.M. Exploring the general characteristics of amino-acid-functionalized ionic liquids through experimental and quantum chemical calculations. *Energy Fuels* 2017, *31*, 4202–4210. [CrossRef]
- 38. Zhou, X.B.; Jing, G.H.; Liu, F.; Lv, B.H.; Zhou, Z.M. Mechanism and kinetics of CO₂ absorption into an aqueous solution of a triamino-functionalized ionic liquid. *Energy Fuels* **2017**, *31*, 1793–1802. [CrossRef]
- Taheri, M.; Dai, C.N.; Lei, Z.G. CO₂ capture by methanol, ionic liquid, and their binary mixtures: Experiments, modeling, and process simulation. *AIChE J.* 2018, *64*, 2168–2180. [CrossRef]

- 40. Li, X.Y.; Hou, M.Q.; Zhang, Z.F.; Han, B.X.; Yang, G.Y.; Wang, X.L.; Zou, L.Z. Absorption of CO₂ by ionic liquid/polyethylene glycol mixture and the thermodynamic parameters. *Green Chem.* **2008**, *10*, 879. [CrossRef]
- 41. Li, J.; Dai, Z.D.; Usman, M.; Qi, Z.W.; Deng, L.Y. CO₂ /H₂ separation by amino-acid ionic liquids with polyethylene glycol as co-solvent. *Int. J. Greenh. Gas Con.* **2016**, *45*, 207–215. [CrossRef]
- 42. Chen, Y.; Hu, H. Carbon dioxide capture by diethylenetriamine hydrobromide in nonaqueous systems and phase-change formation. *Energy Fuels* **2017**, *31*, 5363–5375. [CrossRef]
- 43. Lepre, L.F.; Pison, L.; Siqueira, L.J.A.; Ando, R.A.; Costa Gomes, M.F. Improvement of carbon dioxide absorption by mixing poly(ethylene glycol) dimethyl ether with ammonium-based ionic liquids. *Sep. Purif. Technol.* **2018**, *196*, 10–19. [CrossRef]
- 44. Jiang, Y.F.; Taheri, M.; Yu, G.Q.; Zhu, J.Q.; Lei, Z.G. Experiments, modeling, and simulation of CO₂ dehydration by ionic liquid, triethylene glycol, and their binary mixtures. *Ind. Eng. Chem. Res.* **2019**, *58*, 15588–15597. [CrossRef]
- Liu, F.; Shen, Y.; Shen, L.; Sun, C.; Chen, L.; Wang, Q.L.; Li, S.J.; Li, W. Novel amino-functionalized ionic liquid/organic solvent with low viscosity for CO₂ capture. *Environ. Sci. Technol.* 2020, *54*, 3520–3529. [CrossRef]
- Huang, Q.; Li, Y.; Jin, X.B.; Zhao, D.; Chen, G.Z. Chloride ion enhanced thermal stability of carbon dioxide captured by monoethanolamine in hydroxyl imidazolium based ionic liquids. *Energy Environ. Sci.* 2011, 4, 2125. [CrossRef]
- 47. Fu, D.; Hao, H.M.; Liu, F. Experiment and model for the viscosity of carbonated 2-amino-2-methyl-1-propanol-monoethanolamine and 2-amino-2-methyl-1-propanol-diethanolamine aqueous solution. *J. Mol. Liq.* **2013**, *188*, 37–41. [CrossRef]
- Goodrich, B.F.; de la Fuente, J.C.; Gurkan, B.E.; Zadigian, D.J.; Price, E.A.; Huang, Y.; Brennecke, J.F. Experimental measurements of amine-functionalized anion-tethered ionic liquids with carbon dioxide. *Ind. Eng. Chem. Res.* 2011, 50, 111–118. [CrossRef]
- 49. Liu, F.; Jing, G.H.; Zhou, X.B.; Lv, B.H.; Zhou, Z.M. Performance and mechanisms of triethylene tetramine (TETA) and 2-Amino-2-methyl-1-propanol (AMP) in aqueous and nonaqueous solutions for CO₂ capture. *ACS Sustain. Chem. Eng.* **2017**, *6*, 1352–1361. [CrossRef]
- 50. Zheng, W.-T.; Huang, K.; Wu, Y.-T.; Hu, X.-B. Protic ionic liquid as excellent shuttle of MDEA for fast capture of CO₂. *AIChE J.* **2018**, *64*, 209–219. [CrossRef]
- 51. Meng, Y.N.; Wang, X.D.; Zhang, F.; Zhang, Z.B.; Wu, Y.T. IL-DMEE Nonwater system for CO₂ capture: Absorption performance and mechanism investigations. *Energy Fuels* **2018**, *32*, 8587–8593. [CrossRef]
- 52. Salleh, R.M.; Jamaludin, S.N. Thermodynamic equilibrium solubility of diethanolamine–N-butyl-1-methylpyrrolidinium dicyanamide [DEABMPYRR DCA] mixtures for carbon dioxide capture. *IOP Conf. Ser. Mater. Sci. Eng.* **2018**, 358, 012010.53. [CrossRef]
- 53. Ahmad, W.; Al-Ajmi, A.; Vakili-Nezhaad, G.R. Investigation of physico-chemical properties for the 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF₄])–diethylenetriamine (DETA) system for CO₂ capture. *J. Solut. Chem.* **2019**, *48*, 578–610. [CrossRef]
- 54. Harifi-Mood, A.R.; Mohammadpour, F.; Boczkaj, G. Solvent dependency of carbon dioxide Henry's constant in aqueous solutions of choline chloride-ethylene glycol based deep eutectic solvent. *J. Mol. Liq.* **2020**, *319*, 114173. [CrossRef]
- Lin, C.-M.; Leron, R.B.; Caparanga, A.R.; Li, M.-H. Henry's constant of carbon dioxide-aqueous deep eutectic solvent (choline chloride/ethylene glycol, choline chloride/glycerol, choline chloride/malonic acid) systems. *J. Chem. Thermodyn.* 2014, *68*, 216–220. [CrossRef]
- 56. Zhang, N.; Huang, Z.H.; Zhang, H.M.; Ma, J.W.; Jiang, B.; Zhang, L.H. Highly efficient and reversible CO₂ capture by task-specific deep eutectic solvents. *Ind. Eng. Chem. Res.* **2019**, *58*, 13321–13329. [CrossRef]
- 57. Ren, H.W.; Lian, S.H.; Wang, X.; Zhang, Y.; Duan, E.H. Exploiting the hydrophilic role of natural deep eutectic solvents for greening CO₂ capture. *J. Clean. Prod.* **2018**, *193*, 802–810. [CrossRef]
- Siani, G.; Tiecco, M.; Di Profio, P.; Guernelli, S.; Fontana, A.; Ciulla, M.; Canale, V. Physical absorption of CO₂ in betaine/carboxylic acid-based natural deep eutectic solvents. *J. Mol. Liq.* 2020, *315*, 113708. [CrossRef]
- 59. Gómez-Coma, L.; Garea, A.; Irabien, Á. Hybrid solvent ([emim][Ac]+water) to improve the CO₂ capture efficiency in a PVDF hollow fiber contactor. *ACS Sustain. Chem. Eng.* **2017**, *5*, 734–743. [CrossRef]

- 60. Huang, Z.L.; Deng, Z.Y.; Ma, J.Y.; Qin, Y.H.; Zhang, Y.; Luo, Y.B.; Wu, Z.K. Comparison of mass transfer coefficients and desorption rates of CO₂ absorption into aqueous MEA + ionic liquids solution. *Chem. Eng. Res. Des.* **2017**, *117*, 66–72. [CrossRef]
- 61. Ma, C.Y.; Sarmad, S.; Mikkola, J.-P.; Ji, X.Y. Development of low-cost deep eutectic solvents for CO₂ capture. *Energy Procedia* **2017**, *142*, 3320–3325. [CrossRef]

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



© 2020 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).