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# Exergy-based sustainability analysis of biogas upgrading using a hybrid solvent (imidazolium-based ionic liquid and aqueous monodiethanolamine)

Bilal Kazmi[1,*], Syed Imran Ali[1], Zahoor Ul Hussain Awan[2]

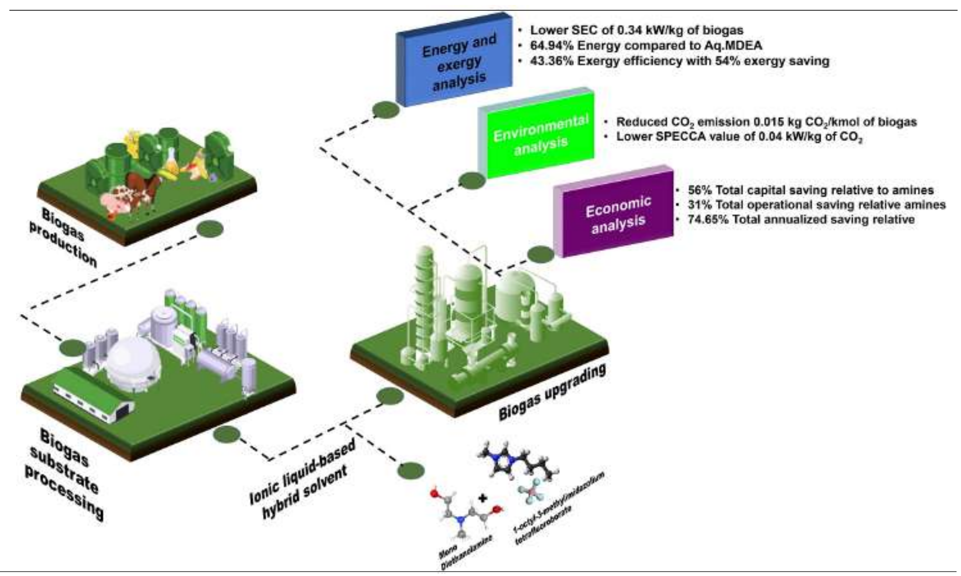
[1]*Department of Applied chemistry and chemical technology, University of Karachi, Pakistan.*

[2]*Department of Food Engineering, NED University of Engineering and Technology, Karachi, Pakistan.*

## HIGHLIGHTS

- Biogas upgrading by ionic liquid (IL)-based hybrid solvent combined with aqueous monodiethanolamine investigated.
- 5 wt% imidazolium-based IL with Aq.MDEA led to biomethane purity and recovery of ≥ 99 wt%.
- Hybrid solvent-based biogas upgrading led to an energy saving of 64.94% vs. the base system.
- Hybrid solvent-based biogas upgrading led to less exergy destruction of 54.25 MW with an overall exergy efficiency of 43.36%.
- The proposed solvent was eco-friendly, with a high $CO_2$ capturing rate (≥ 99 wt%) and less emission (0.015 kg $CO_2$/kmol).

## GRAPHICAL ABSTRACT

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## ABSTRACT

Energy may be generated in large quantities from fossil fuels, but this comes with environmental concerns. Thus, renewable resources like biogas, comprising carbon dioxide and methane, should be used alone or in combination with fossil fuels to mitigate the environmental footprints of energy generation systems. In this study, a new concept of hybrid solvent was presented, which combines 1-octyl-3-methylimidazolium tetrafluoroborate with aqueous mono diethanolamine for biogas upgrading process to provide high purity (≥ 99 wt%) and recovery (≥ 99 wt%) of biomethane. The process was simulated in ASPEN Plus® V.11. The thermodynamic framework was validated against experimental data, and rigorous regression was conducted to obtain binary parameters. To establish the efficacy of the suggested hybrid solvent, three scenarios were studied by altering the concentration of ionic liquid (5–20 wt%) linked with amine and compared to aqueous mono diethanolamine as the base case (50 wt%). The results showed that a hybrid solvent with 5 wt% 1-octyl-3-methylimidazolium tetrafluoroborate could increase $CH_4$ purity to 99% (mol%). The hybrid solvent led to an energy saving of 64.94% compared to the amine-based system. Thermodynamic irreversibilities showed that 5 wt% 1-octyl-3-methylimidazolium tetrafluoroborate improved exergy efficiency by 54% over the amine-based procedure. Environmentally, the hybrid solvent system also achieved a higher capture rate (99%) and lower emissions (0.017 kW/kmol). Comparing the economic prospects, 5 wt% 1-octyl-3-methylimidazolium tetrafluoroborate saved 56% on total capital cost, making it competitive from an investment perspective.



* Corresponding author at:
E-mail address: bilalkazmi91@gmail.com

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**Contents**



**Abbreviations**

| | |
|---|---|
| A/HS | Air to hybrid solvent ratio |
| Aq.MDEA | Aqueous mono diethanol amine |
| $CH_4$ | Methane |
| ChCl | Choline chloride |
| $CO_2$ | Carbon dioxide |
| DES | Deep eutectic solvent |
| EOS | Equation of state |
| FC | Flash Column |
| $H_2S$ | Hydrogen sulfide |
| HS | Hybrid solvent |
| HS/F | Hybrid solvent ratio to feed gas |
| IL | Ionic liquid |
| MEA | Monoethanolamine |
| $OmimBF_4$ | 1-octyl-3-methylimidaolium tetrafluoroborate |
| PC | Propylene carbonate |
| Peng Robin | Peng Robinson |
| PSE | Process system engineering |
| TAC | Total annualized cost |
| TCC | Total capital cost |
| TOC | Total operating cost |
| VLE | Vapor-liquid equilibrium |

## 1. Introduction

As a commodity and a resource, energy is an essential development means, mainly provided by fossil fuels like oil and coal. However, there is a growing consciousness about the harm burning fossil fuels poses to the planet and human health (EIA, 2019), advocating for a swift transition toward renewable energy resources. Negative effects on human health and the immediate environment could be peripheral to the greater damage to the global ecosystem (Cozma et al., 2013). Bio-based energy is a renewable resource that uses carbon-based organic material from plants and animals to generate electricity.

Biogas is a renewable and environmentally benign resource that can help countries achieve their goals of producing greener energy, easing the energy crisis, and reducing their carbon footprints (IEA Bioenergy Task 37, 2020). Biogas is produced through anaerobic digestion by microbial populations digesting organic materials. The obtained biogas is primarily composed of various components, including methane ($CH_4$), carbon dioxide ($CO_2$), hydrogen sulfide ($H_2S$), and water vapor (Taqvi and Kazmi, 2021). However, biogas must be improved by eliminating the $CO_2$ component before it can be utilized as a cleaner fuel in the form of biomethane. This upgrade aims to achieve a gas with the maximum feasible $CH_4$ content and consequently increase the calorific value of the product as a fuel.

Several methodologies of biogas upgrading have already been developed for enhancing the quality of biogas by the selective sequestration of $CO_2$ content from the biogas, including physical/chemical absorption (Menacho et al., 2022), pressure swing adsorption (Augelletti et al., 2016), membrane-based technology (HaiyanYang et al., 2022), cryogenic separation process (Yousef et al., 2018). The two most widely used industrial approaches are physical and chemical absorption. Chemical absorption is often seen as the most promising technology because it can manage a sizable flow of biogas while aiming to maintain a low partial pressure. Improvements in $CO_2$ loading, absorption rate, and capture efficiency are among the evolution of chemical absorption over time. However, chemical absorption is not eco-friendly, as the amines used in this process have a low vapor pressure leading to the loss of the solvent, causing corrosion in operational components and equipment, and generating thermally unstable byproducts. The fundamental drawback of the chemical absorption process, however, is the high temperature required for solvent regeneration, which significantly affects operational expenses. Hence, the primary goal of enhancing $CO_2$ removal from the biogas and producing pure biomethane using chemical absorption is to find solvents leading to a more sustainable and enhanced operation.

Ionic liquid (IL) research has recently intensified to create a $CO_2$ sequestration approach to overcome the drawbacks of biogas upgrading processes (Kazmi et al., 2022b). ILs are less hazardous and corrosive than amines, have high thermal stability, and have a virtually negligible solvent loss, making them a preferable solvent to amines (Kazmi et al., 2021a). However, IL's high viscosity reduces mass transfer and raises the expenses associated with pumping the solvent, making industrial use challenging. Nevertheless, blending an appropriate IL with a cosolvent like alkanolamine to generate a hybrid solvent might alleviate the IL's viscosity problem and serve as a solution to the biogas upgrading process. in this context, researchers have investigated the fast-growing idea of mixing IL with cosolvents in the aqueous phase to enhance the hybrid solvent's viscosity and capture potential. Fu et al. (2017) examined amino-functionalized IL and mono diethanol amine (MDEA) in an aqueous solution (Aq.MDEA). The hybrid solvent's $CO_2$ loading increases with cosolvent content, the aqueous phase effect in viscosity adjustment, and the system partial pressure for $CO_2$ absorption. Xiao et al. (2019) anticipated that imidazolium-based IL and alkanolamine might absorb $CO_2$. The study revealed that imidazolium-based IL combined with MDEA improved $CO_2$ separation and solved the IL viscosity issue, the major industrialization barrier.

Increasing the alkyl chain on the cation moiety of ILs could improve thermal stability, $CO_2$ sequestration, and hybrid solvent viscosity. Shojaeian and Haghtalab (2013) expanded the hybrid solvent concept based on imidazolium-based IL and believed that lowering IL concentration with amine would significantly alter $CO_2$ loading. Haider et al. (2021) also studied the prospects of using the hybrid solvent for upgrading biogas. The results showed that blending deep eutectic solvent (DES) with

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monoethanolamine (MEA) led to an energy saving of 72%, provided a higher capture rate of $CO_2$ with fewer emissions into the environment while providing the advantage of improving the process economy by 27.8% in terms of total annualized cost (TAC). Wang et al. (2022) also studied the hybrid solvent blend for biogas upgrading by mixing 1-butyl-3-methylimidazolium acetate [Bmim][Ac] with propylene carbonate (PC). The results suggested that the hybrid solvent seemed less energy intensive, providing 33% energy saving and 21% less capital investment than PC. Damanafshan et al. (2021) recently examined $CO_2$ solubility in the imidazolium-based IL/Aq.MDEA combination. The study revealed that a bigger alkyl group cation would improve $CO_2$ capture. Additionally, it was also predicted that as the concentration of IL in the solvent mixture rose above 10 wt%, it would hinder the hybrid solvent's removal ability since the higher concentrations of alkanol amine and IL would increase the hybrid solvent's viscosity, which would affect the mass transfer mechanism of $CO_2$ loading into the solution and also elevate the solution's viscosity. Most simulation studies concerning biogas upgrading use either IL or DES as the solvent, as shown in **Table 1**.

In light of the above, this study presents the process system engineering (PSE) perspective of a hybrid solvent showcasing 1-octyl-3-methylimidazolium tetrafluoroborate [Omim][$BF_4$] in combination with Aq.MDEA for preferential $CO_2$ sequestration from raw biogas feed at high pressures (>10 bar). This research aims to investigate a comprehensive framework by employing the hybrid solvent based on the selected solubility data to rigorously regress thermodynamic parameters, followed by

**Table 1.**
A summary of the latest works (simulated by Aspen Plus) on biogas upgrading using ionic liquid (IL) and deep eutectic solvent (DES) from the process system engineering perspective.

| Solvent system type | | Solvent class | $CH_4$ recovery (%) | $CH_4$ purity (%) | Solvent | Specific energy consumption | Economic aspect | | Reference |
|---|---|---|---|---|---|---|---|---|---|
| Single | Hybrid | | | | | | Capital cost | Operating cost | |
| Yes | No | DES* | 97 | 96.7 | Aqueous ChCl/Urea | 0.21 kWh/kg biogas | NA | NA | Ma et al. (2018b) |
| | | | | | | 0.21 kWh/kg biogas | NA | NA | |
| | | | | | | 0.22 kWh/kg biogas | NA | NA | |
| Yes | No | DES | NA | 97 | Deep eutectic solvent | 0.224 kW$_e$h/kg biogas | USD 0.35 × $10^6$ | USD 0.185 × $10^6$/yr | Xie et al. (2018) |
| Yes | No | DES | NA | 97 | Aqueous ChCl/Urea | 0.226 kW$_e$h/kg biogas | USD 0.48 × $10^3$ | USD 0.22 × $10^3$/yr | Ma et al. (2018a) |
| | | | | | | 0.229 kW$_e$h/kg biogas | USD 0.69 × $10^3$ | USD 0.248 × $10^3$/yr | |
| Yes | No | IL | >96 | >99 | [Bmim][$PF_6$] | 1.1048 kWh/kg biogas | USD 5.7 × $10^6$ | USD 1.38 × $10^6$/yr | Haider et al. (2019) |
| Yes | No | IL | NA | >96 | Aqueous [Bmmorp][OAc] | 147.5 kW/kg biogas | NA | NA | Ma et al. (2019) |
| Yes | No | IL | NA | 97 | [Bmmorp][OAc] + 20 wt% water | NA | USD 0.041× $10^6$ | USD 0.09 × $10^6$/yr | Wang et al. (2020) |
| | | | | | [Bmmorp][OAc] + 30 wt% water | | USD 0.048 × $10^6$ | USD 0.105 × $10^6$/yr | |
| Yes | No | DES | NA | 97 | ChCl/Urea | NA | USD 3.8 × $10^6$ | USD 0.77 × $10^6$/yr | Słupek et al. (2020) |
| | | | | | ChCl/Oxalic acid | | USD 4.1 × $10^6$ | USD 0.91 × $10^6$/yr | |
| Yes | No | IL | NA | NA | [$P_{2228}$][CNPyr] | 2.1 GJ/t$CO_2$ | NA | NA | Hospital-Benito et al. (2020) |
| | | | | | [$P_{66614}$][CNPyr] | 3.0 GJ/t$CO_2$ | | | |
| | | | | | [Bmim][acetate] | 3.4 GJ/t$CO_2$ | | | |
| Yes | No | IL & DES | >97 | >99 | [Bmim][$PF_6$] | 0.235 kWh/kg biogas | USD 33.5 × $10^6$ | USD 10.93 × $10^6$/yr | Haider et al. (2020) |
| | | | | | Aq.ChCl/urea | 0.296 kWh/kg biogas | USD 44.87 × $10^6$ | USD 9.53 × $10^6$/yr | |
| | | | | | | 0.286 kWh/kg biogas | USD 40.54 × $10^6$ | USD 16.57 × $10^6$/yr | |
| | | | | | | 0.244 kWh/kg biogas | USD 34.8 × $10^6$ | USD 10.6 × $10^6$/yr | |
| | | | | | | 0.244 kWh/kg biogas | USD 33.3 × $10^6$ | USD 9.3 × $10^6$ /yr | |
| | | | | | | 0.246 kWh/kg biogas | USD 32.7 × $10^6$ | USD 8.52 × $10^6$ /yr | |
| | | | | | | 0.245 kWh/kg biogas | USD 32.06 × $10^6$ | USD 7.85 × $10^6$/yr | |
| Yes | No | IL | >96 | >99 | [Bmim][$PF_6$] | 0.42 kWh/kg biogas | NA | NA | Long et al. (2022) |
| Yes | No | IL | NA | NA | [$P_{2228}$][CNPyr] | 1.8 GJ/t$CO_2$ | USD 10.71 × $10^6$ | USD 1.82 × $10^6$/yr | Hospital-Benito et al. (2021) |
| | | | | | [$P_{66614}$][CNPyr] | 2.8 GJ/t$CO_2$ | USD 11 × $10^6$ | USD 1.91 × $10^6$/yr | |
| | | | | | [Bmim][acetate] | 3.1 GJ/t$CO_2$ | USD 11.01 × $10^6$ | USD 1.93 × $10^6$/yr | |
| Yes | Yes | DES & MEA | 97 | 99 | ChCl/urea + $H_2O$ + MEA | 0.143 kWh/kg biogas | USD 31.15 × $10^6$ | USD 4.27 × $10^6$/yr | Haider et al. (2021) |
| | | | | | | 0.145 kWh/kg biogas | USD 30.34 × $10^6$ | USD 4.31 × $10^6$/yr | |
| | | | | | | 0.15 kWh/kg biogas | USD 30.61 × $10^6$ | USD 4.46 × $10^6$/yr | |
| Yes | Yes | IL+PC | 99 | 97 | [Bmim][Ac]+PC | 23.4 kW/kg biogas | NA | NA | Wang et al. (2022) |
| Yes | No | IL | NA | 97 | [$P_{2228}$][CNPyr] | 0.211 kWh/kg biogas | USD 4.25 × $10^6$ | NA | Moya et al. (2022) |

* *Abbreviations:* IL = Ionic liquid; DES = Deep eutectic solvent; MEA = Monoethanolamine; MDEA = Mono diethanolamine; PC = Propylene carbonate; ChCl = choline chloride; [Bmim][$PF_6$] = 1-butyl-3-methylimidazolium hexafluorophosphate; [Bmim][Ac] = 1-butyl-3-methylimidazolium acetate; [Omim][$BF_4$] = 1-octyl-3-methylimidazolium tetrafluoroborate; [$P_{2228}$][CNPyr] = triethyloctylphosphonium 2-cyanopyrrole; [$P_{66614}$][CNPyr] = trihexyltetradecylphosphonium 2-cyanopyrrole; NA = not available

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designing the process using Aspen Plus® to optimize and examine the process based on energy, exergy, environmental, and economic analysis. This article is structured as follows: *Section 2* presents the methodology used in the process. In *Section 3*, we outline the framework and underlying assumptions of the process simulation. *Section 4* describes the detailed process analysis based on the process system engineering perspective (energy, exergy, environment, economic) for assessing and enhancing the process variables that provide a robust model with minimal energy requirement. Finally, *Section 5* details the overall conclusion drawn from the data obtained and provides a future outlook.

## 2. Research methodology: proposed process

### *2.1. Process description*

A schematic presentation of the process and the main parameters (temperature, pressure, and flow rate) are shown in **Figure 1**. A raw biogas stream containing 62.6 wt% $CH_4$ and 37.4 wt% $CO_2$ (Haider et al., 2019) at a flow rate of 60,000 kg/h at 30 °C and 1 bar pressure was compressed to 40 bar using a multistage compressor before being fed into a 20-stage absorption column. Simultaneously, another stream based on a hybrid solvent was fed into the absorber to begin the absorption process. The hybrid solvent selectively separated $CO_2$ from the raw biogas stream, resulting in a biomethane stream from the top of the absorber with a high purity of ≥ 97 wt% for $CH_4$ and a recovery of ≥ 99 wt%. The bottom stream of the absorption column, which contained high amounts of hybrid solvent, $CO_2$, and traces of $CH_4$, was sent to the series of flash columns (FC-1 and FC-2) operating at a pressure of 13.25 bar and 4.1 bar, respectively, to maximize biomethane recovery and purity even further. The bottom stream from the flash column (FC-2) was then sent toward the stripping column *via* a valve that reduced its pressure to 1.2 bar. The rich stream, containing $CO_2$ and a hybrid solvent mixture, was injected into an 18-stage regeneration column to regenerate the solvent using air, which was then delivered into the column counter-currently *via* a compressed air blower. The bottom regenerated solvent stream was recycled back into the absorber column through the pump and reused in the process.

### *2.2. Process simulation and design*

This study proposes a process simulation-based design of an IL-based hybrid solvent for the selective removal of $CO_2$ from the raw biogas stream. The process design approach is illustrated in **Figure 2**. The main goal of the proposed approach is to investigate the hybrid solvent's potential to produce biomethane with high purity (≥ 97 wt%) and recovery (≥99 wt%) while using the minimum energy possible. The process design approach was designed specifically for absorption solvents, and a base case employing alkanolamine was created for performance comparison with the hybrid solvent. The choice of solvent was made based on the experimental results of the vapor-liquid equilibrium of $CO_2$ solubility, which implied that introducing imidazolium-based IL and MDEA as a co-solvent to the aqueous medium would increase $CO_2$ loading. Therefore, a thorough thermodynamic evaluation was conducted to identify the binary interaction parameters needed in the simulation domain to characterize the necessary interaction between the acid gas molecules and the hybrid solvent. The thermodynamic analysis and regression were used to characterize the hybrid solvent, while the Aspen database library has a clear definition of the solvent. Then, the process was modelled, and a comprehensive sensitivity analysis was performed on the two significant pieces of equipment, the regenerator and the absorber. The goal was to mark the proposed case's performance against the base, considering the design constraints. Furthermore, the PSE paradigm was used to assess the sustainability and acceptability of the process from the energy, exergy, environment, and economic perspectives. Finally, efforts were put to attain a best-case scenario, indicating a highly energy-effective and efficient process that would be both ecologically friendly and feasible from an economic standpoint.

## 3. Process assumption

Aspen Plus® V.11 was utilized for simulating and assessing various aspects of the biogas upgrading process using the hybrid solvent.

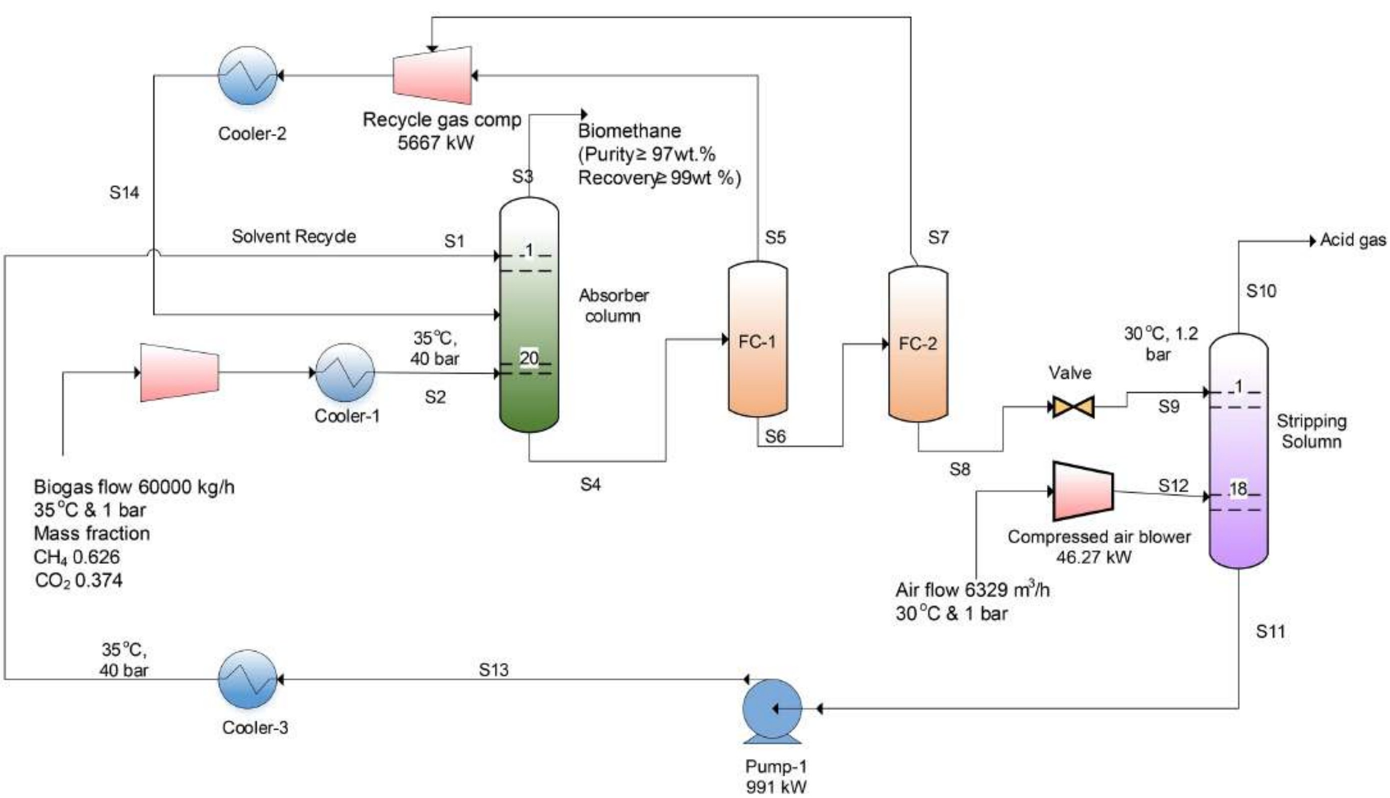


**Fig. 1.** Process flow diagram of the proposed hybrid solvent (5 wt% 1-octyl-3-methylimidazolium tetrafluoroborate+ 50 wt% MDEA + 45 wt% $H_2O$) for biogas upgrading.

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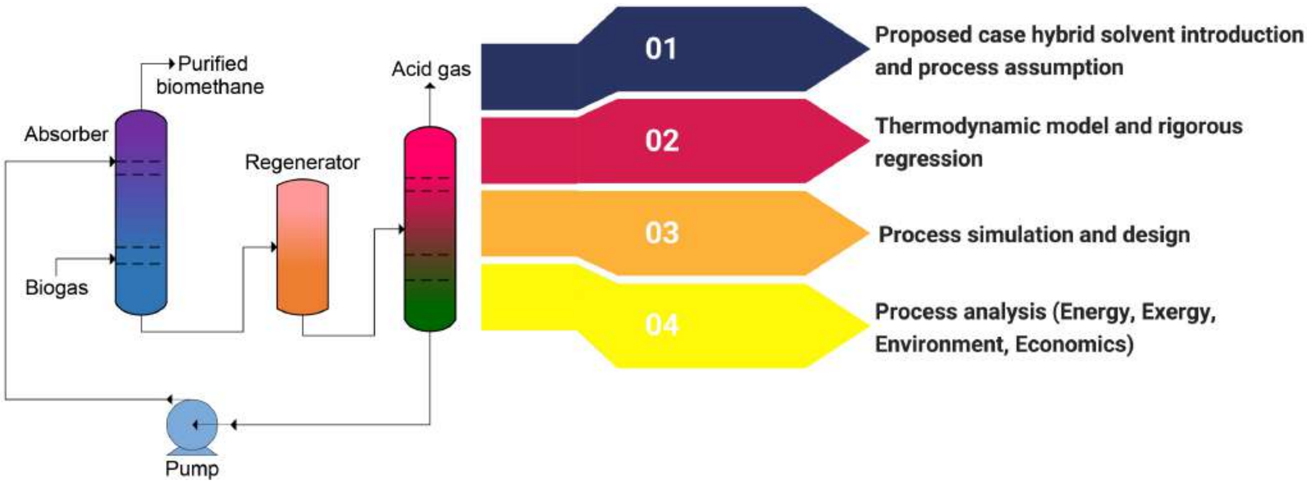


**Fig. 2.** Schematic presentation of the process design approach used in this study.

According to this theory, the basic assumptions used to represent both the proposed hybrid solvent-based and the alkanolamine-based processes were:

- Steady-state process.
- Negligible heat loss from the system to the surrounding.
- Purity and recovery of $CH_4$ content in biomethane to be ≥97 wt% and ≥99 wt%, respectively.
- For the pumps and compressor used in the system, isentropic efficiency was assumed to be 80%.

## 4. Thermodynamic model analysis and data regression

Process design is a vital aspect that determines the feasibility and proficiency of a process based on thermodynamic principles. In this context, thermodynamic model selection is critical for understanding the separation mechanism and estimating scalar and temperature-dependent properties. Therefore, Peng-Robinson (PengRobin), used as a primary thermodynamic model for hydrocarbon-based systems, was chosen because of its ability to forecast and inter-inspect these systems accurately. The binary interaction parameters were studied by performing rigorous regression and validation using Aspen Plus® based on experimental and predicted results. **Table 2** shows the vapor-liquid equilibrium (VLE-based) binary interaction parameters for the investigated hybrid solvent. Additional details can be found in the **Supplementary Information**.

**Table 2.**
Binary interaction parameter between the [Omim][$BF_4$], $CO_2$, $CH_4$, and MDEA based on the Peng Robinson equation-of-state model using rigorous regression evaluated in this work.

| *i* | *J* | $K_{Aij}$ | $K_{Bij}$ | Deviation |
|---|---|---|---|---|
| [Omim][$BF_4$] | $CO_2$ | 0.056 | -0.0028 | 7.39e-5 |
| [Omim][$BF_4$] | $H_2O$ | 5.24 | -0.021 | 0.016 |
| [MDEA] | [Omim][$BF_4$] | 0.86 | -0.0025 | 0.00026 |
| [MDEA] | $CO_2$ | 0.64 | -0.0028 | 0.0035 |
| **Average deviation** | | | | **0.0049** |

By connecting the VLE data for the hybrid solvent based on the Peng Robin equation, the minimal deviation was obtained between the estimated and experimental data, indicative of good agreement with the experimental data (**Tables S1-S3**). The estimated solubilities based on the VLE experimental points for the concerned components are shown in **Figures 3a**, **b**, **c**, and **d** for [Omim][$BF_4$], MDEA, $CO_2$, and $H_2O$, respectively.

### 4.1. Exergy analysis

Energy optimization is an important tool in process-based studies. However, energy analysis does not specify the system's thermodynamic irreversibilities (Ghorbani et al., 2021). Exergy analysis is crucial in this approach based on the second rule of thermodynamics. It identifies the irreversibilities in the process scheme's primary equipment (Hosseinipour and Mehrpooya, 2019). Exergy is mostly of two types: physical and chemical (**Eq. 1**). Physical exergy is solely addressed as a technique for analyzing the process in the present study because there are no chemical interactions, and all the process is considered as one control volume. As a result, the exergy destruction for each component involved in the process was estimated as shown in **Equation 2**:

$$Ex = Ex_{physical} + Ex_{chemical} \quad \text{Eq. 1}$$

$$\dot{E}x_{physical,K,i} = W \times \left[h_{k,i}(T,P) - h_o(T_o,P_o) - T_o\left(s_i(T,P) - s_o(T_o,P_o)\right)\right] \quad \text{Eq. 2}$$

where Ėk, I, h, s, W, and T represent the physical exergy rate, enthalpy, entropy, mass flow, and temperature for each k$^{th}$ process equipment involved, respectively.

Exergy efficiency defines the performance of each process equipment, and it explains the valuable work obtained from a specific system and can mathematically be written as shown in **Equation 3**.

$$\eta = \frac{ex_{gained}}{ex_{expanded}} = 1 - \frac{ex_{destroyed}}{ex_{expanded}} \quad \text{Eq. 3}$$

Hence, the exergy destruction and exergy efficiency equations for each of the equipment used in this study are listed in **Table 3**.

Systematically combining exergy analysis with life cycle assessment (LCA) could also be used to assess a process, as these two are correlated, allowing for accurate assessment of the quality and thermodynamic inefficiencies of a product or system, as well as the associated environmental implications or pathways the system can avoid through optimization and a greener approach, providing a process that is both efficient and eco-friendly. **Table 4** lists the correlation factor and its equation for assessing the proposed cases based on the systematic combination of exergy and environmental aspects (Blumberg et al., 2019).

### 4.2. Environmental analysis

Due to strict environmental rules being imposed globally, significant effort is being directed toward optimizing the environmental process. In light of that, the carbon capture rate, specific $CO_2$ emission, and specific energy needed for $CO_2$ capture were the major factors employed in this study to analyze the proposed cases (Haider et al., 2021). The carbon

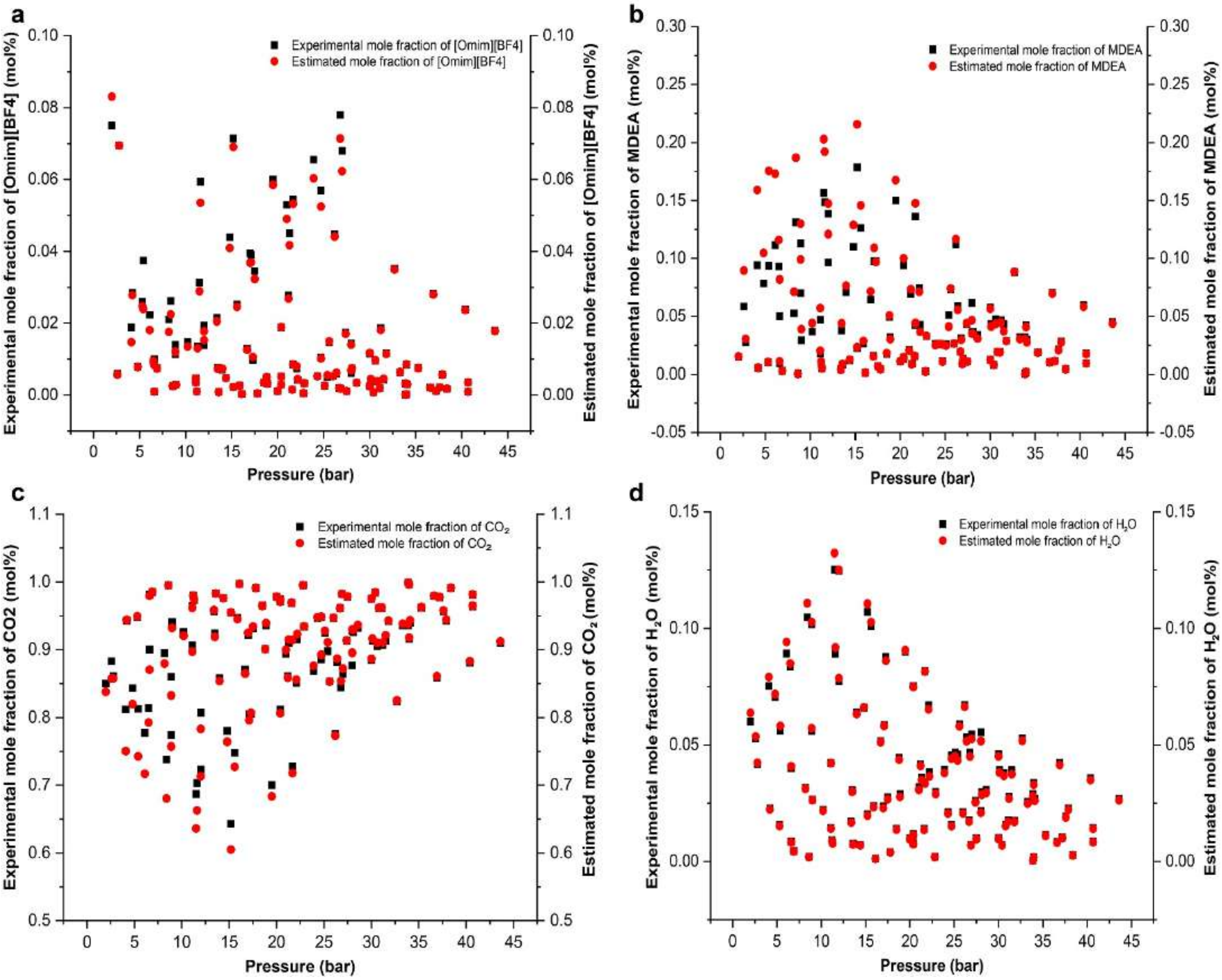


**Fig. 3.** Schematic Experimental and estimated results of the rigorous regression based on vapor-liquid equilibrium (VLE) data for the evaluation of binary interaction parameters for **(a)** [Omim][BF$_4$], **(b)** [MDEA], **(c)** $CO_2$, and **(d)** $H_2O$ systems.

**Table 3.**
Exergy destruction equations of various equipment analyzed for the proposed and base case scenarios.*

| Equipment | Exergy destruction |
|---|---|
| Absorber and regenerator | $\dot{I}_{abs\,\&\,regen} = \sum(\dot{m}.\dot{e})_{in} - \sum(\dot{m}.\dot{e})_{out}$ |
| Compressor and pump | $\dot{I}_{comp\,\&\,pump} = \sum(\dot{m}.\dot{e})_{in} - \sum(\dot{m}.\dot{e})_{out} + \dot{W}$ |
| Heat exchanger | $\dot{I}_{HEX} = \sum(\dot{m}.\dot{e})_{in} - \sum(\dot{m}.\dot{e})_{out}$ |
| Flash separator | $\dot{I}_{sep} = (\dot{m}.\dot{e})_{in} - \left((\dot{m}.\dot{e})_{out,liquid} + (\dot{m}.\dot{e})_{out,vapor}\right)$ |
| Coolers | $\dot{I}_{cooler} = (\dot{m}.\dot{e})_{in} - (\dot{m}.\dot{e})_{out}$ |

* Source: Kazmi et al. (2021b).

**Table 4.**
Equations associated with the exergy and life cycle systematic analysis for the proposed and base case scenarios.*

| Equation | Parameter Description |
|---|---|
| $f_{ei} = \frac{\sum Ex_{total\ destruction}}{\sum Ex_{total\ in}}$<br>$\Theta_i = f_{ei} \times C_{ei}$ | $f_{ei}$ = exergy destruction factor<br>$Ex_{total\ destruction}$= total exergy losses<br>$Ex_{total\ in}$= total exergy losses at inlet<br>$C_{ei}$ = coefficient related to environmental effect |
| $C_{ei} = \frac{1}{\sum Ex_{eff}}$ | $Ex_{eff}$ = exergy efficiency of the process |
| $\Theta_{ii} = \frac{1}{\Theta_i}$ | $\Theta_i$ = index related to environmental losses |
| $f_{es} = \frac{\sum Ex_{total\ des}}{\sum Ex_{output} + \sum Ex_{total\ des} + 1}$ | $f_{es}$ = exergy stability<br>$Ex_{total\ des}$ = total exergy losses<br>$Ex_{output}$ = total exergy losses at the outlet |

* Source: Kazmi et al. (2022a).

capture rate describes the ability of the solvent to remove $CO_2$ from the feed biogas. The following equation was used to determine carbon capture (Eq. 4).

$$Carbon\ capture\ rate\ (\%) = \frac{Sequestered\ CO_2}{Total\ CO_2\ in\ feed} \quad \text{Eq. 4}$$

The total $CO_2$ emission in the purified biomethane concerning the feed rate was characterized as specific $CO_2$ emissions. The specific primary energy consumption for $CO_2$ avoided (SPECCA) index indicates the energy required to reduce $CO_2$ emissions, computed using Equation 5.

$$SPECCA = \frac{Base\ case\ energy\ consumption - Proposed\ case\ energy\ consumption}{Base\ case\ CO_2\ emission - Proposed\ case\ CO_2\ emissions} \quad \text{Eq. 5}$$

### 4.3. Economic evaluation

Economic analysis is a method of understanding the process feasibility and prospects from an economic standpoint. Capital investment, which includes equipment expenditures, installation expenses, and infrastructure development, is the foundation of the analysis. The second is the cost associated with the process's operations, which is dependent on electricity costs, labor costs, and other variable expenses required for the process's operation—combining both of these costs to achieve a third important cost variable known as the TAC (Eq. 6), which forecasts the overall annual expenditure expected to occur in the process's proper functioning.

$$TAC = \left(\frac{Capital\ cost}{Payback\ period}\right) + Operatingcost \quad \text{Eq. 6}$$

The estimation of capital investment is mostly based on the equipment cost connected with sizing and other criteria. For this purpose, Dimian (2003) presented a methodology for calculating compressor and pump costs as a function of power consumption and certain operational parameters. Pumps and compressors with greater power ratings were analyzed independently, and the equation employed is shown in Table 5. The bare modulus-based method proposed by Richard Turton, which depends on various criteria, including the equipment's capacity, size, operational requirements, and material costs connected with its production, was used for calculating the cost of the absorber and regeneration column (Turton et al., 2012). For flash columns, the cost was evaluated using Guthrie's method (Scholz et al., 2013). It uses the vessel's height and diameter as a function to calculate the cost of the equipment. All the cost relationships used to calculate the equipment cost are described in Table 5. The cost of electricity was assumed to be USD 0.22 $kWh^{-1}$

**Table 5.**
Equipment cost evaluation equations for determining the Capital cost.*

| Equipment | Cost Function |
|---|---|
| Compressors and pumps | $Ce = \alpha + \beta S^n$ for compressor power < 3000 kW<br>S= equipment Power (kW), α &β = specific equipment constant<br>$C_e = 517.5 \times \left(\frac{Marshal\ and\ swift\ Cost\ index}{280}\right) \times P^{0.82} \times F_c$ for compressor power > 3000 kW<br>P = equipment power (kW), $F_c$ = correction factor |
| Flash vessel | $BC = Co\left(\frac{l}{lo}\right)^{\alpha}\left(\frac{d}{do}\right)^{\beta}$<br>$IC = UF.BC(MBF + MF - 1)$<br>l, d; length and diameter dimensions |
| Absorber, regenerator, coolers | $\log_{10} Fp = C1 + C2 \log_{10} P + C3\ (\log_{10} P)^2$ ;<br>$Fbm = B1 + B2(Fp)(Fm)$<br>$\log_{10} Cp = K1 + K2\ \log_{10} X + K3\ (\log_{10} X)^2$;<br>$Cbm = Fbm\ Cp$<br>$F_p$; factor associated with pressure (bar), X=factor on which equipment cost depends, (k, C, B) = constant associated with specific equipment, $F_m$ = factor associated with material of equipment |

* Source: Turton et al. (2012); García-Gutiérrez et al. (2016).

## 5. Results and Discussion

### 5.1. Energy analysis: optimized variables

The proposed hybrid solvent based on [Omim][$BF_4$] in combination with Aq.MDEA for the biogas upgrading process was optimized and analyzed for the best combination of IL with amine, leading to reduced energy requirement with enhanced biomethane purity. So, in this context, for the studied hybrid mixture consisting of IL([Omim][$BF_4$]) +Aq.MDEA, the concentration of IL varied from 5 to 20 wt%, as shown in Figure 4. Further, to analyze the proposed process prospects properly, Aq.MDEA (50 wt%) was chosen as the base case. Based on the literature, it is essential to note that pure IL viscosity is the primary block in its path toward industrialization. Hence, using a hybrid solvent combining the advantages of IL and blending it with an aqueous mixture of amine could be a promising strategy that can be applied, especially in the biogas upgrading process as a cutting-edge novel solvent possessing a higher capability than the conventional solvents. Therefore, process analysis was carried out based on the process system engineering prospects to assess the new avenues these hybrid solvents could offer in biogas upgrading technology. Table 6 lists the design parameters and constraints utilized in evaluating the proposed process for minimal energy requirement.

An initial sensitivity analysis was undertaken to alter the parameters of the proposed process in each analyzed scenario to identify the combination of IL+ Aq.MDEA with the lowest energy requirement. First, the hybrid solvent flow rate concerning the feed biogas was evaluated because it is directly related to energy usage (Fig. 5). The higher the solvent flow, the more energy is required in the solvent regeneration section, and the more solvent recirculation pumping is required. The hybrid solvent-to-feed (HS/F) ratio in the proposed process was tuned to produce a ≥ 99 wt% recovery with ≥ 97 wt% purity for $CH_4$ in the produced biomethane. The optimal flow rate of the selected hybrid solvent would also improve the absorber's trajectories, resulting in lower capital spending required for the equipment. Figure 5 illustrates the sensitivity analysis performed to optimize the HS/F ratio and theoretical absorber stages required to meet the purity and recovery goal. As stated earlier, enhancing the concentration of IL, i.e., [Omim][$BF_4$] above 10 wt%, would undoubtedly affect the $CO_2$ loading into the solvent. The results further show that Case-III, based on 5 wt% [Omim][$BF_4$] + 50 wt% MDEA + 45 wt% $H_2O$, provided the most effective $CO_2$ sequestration from the feed biogas as compared to Case-I and Case-II. Case-III, based on 5 wt% [Omim][$BF_4$], provided an effective separation with a reduced solvent ratio of 0.45; increasing the IL concentration above 10 wt% with 50 wt% MDEA would hinder the viscosity of the solvent and would result in less dilution of $CO_2$ into the hybrid solvent. Figure 5 shows that in the proposed Cases I-III, the HS/F was 0.55, 0.50, and 0.45, and the corresponding stages required in the absorber column for studied Cases I-III were 20, 20, and 18, respectively.

Based on design constraints, the flash column was placed next to the absorber column, which was fed with the bottom stream of the absorber column, and the pressure of the flash column was adjusted to obtain the target purity and recovery content of $CH_4$ in the biomethane stream. While managing the flash column pressure to enhance the biomethane at the top of the absorber, the recycle gas column power must also be taken into account, as an abruption of flash column pressure would significantly affect the power consumption by the recycle compressor. So, the flash column *in situ* pressure with the optimal HS/F ratio would also be essential for reducing the gas compression load, as shown in Figure 5. The results for the proposed cases are tabulated in Table 6. It can be concluded that the compression requirement reduced as we moved from Case-I to Case-III, respectively.

Moving on from the absorber column, it was critical to regenerate the solvent so it could be recirculated back into the system. Hence, air stripping was utilized as a model based on compressed air for regenerating the solvent in the proposed scenarios since air stripping would give the advantage of cutting off the thermal load in the form of a reboiler, which is the major concern of the conventional process. As with the absorber column, it was critical to optimize the HS/F ratio, as well as the airflow rate, in proportion to the rich stream flow containing a mixture of IL and Aq. MDEA, to provide a reduced energy consumption path with higher hybrid solvent recovery and $CO_2$ content reduction. Based on the results

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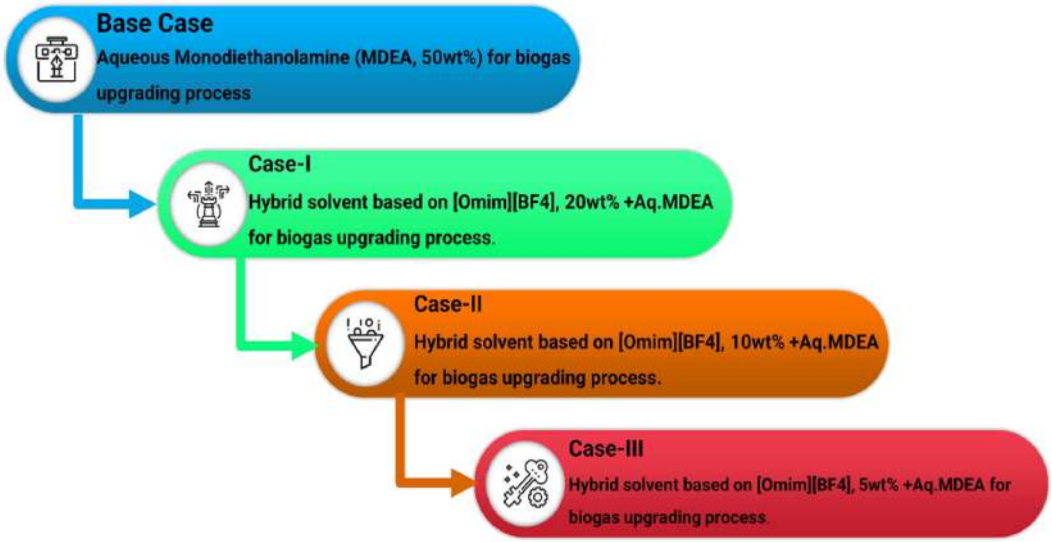


**Fig. 4.** Process analysis cases investigated for IL-based hybrid solvent used in biogas upgrading.

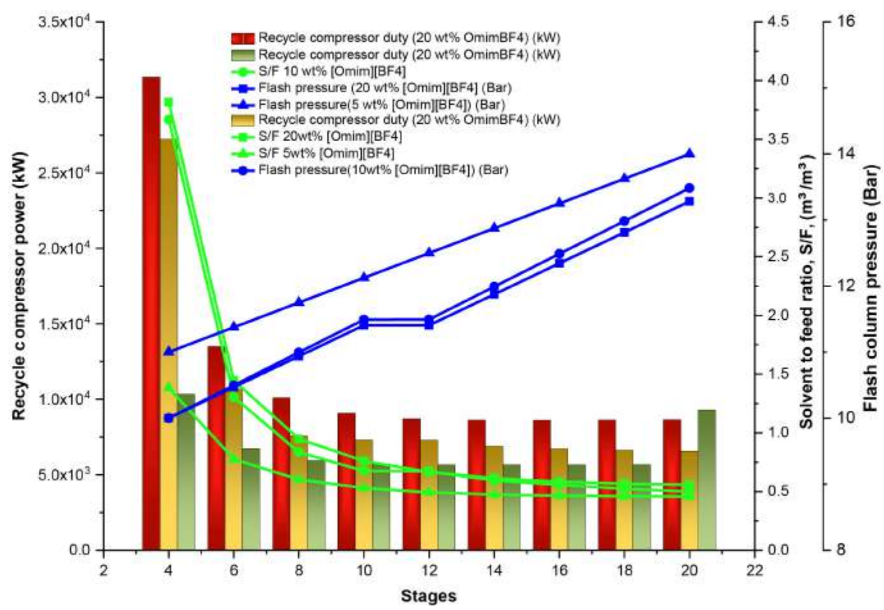


**Fig. 5.** The sensitivity analysis results for the absorber based on various stages for selecting the optimal solvent flow, corresponding flash column pressure, and recycle compressor duty.

shown in **Figure 6**, airflow to hybrid solvent (A/HS) became constant after certain stages of the column. For Case-I, 18 stages of the regenerator column were selected, corresponding to the A/HS of 0.53; for Case-II, 20 stages corresponding to the A/HS of 1.71; and for Case-III, 18 stages corresponding to the A/HS of 0.59. Nevertheless, the air compressor's airflow requirement in Case-I was relatively high, with high energy consumption. On the other hand, the concentration of IL reduced significantly due to the ease of breaking the bond between the $CO_2$ moieties and IL+Aq.MDEA mixture.

### *5.2. Environmental analysis*

**Figure 7** depicts the results, revealing that all cases had a $CO_2$ capture rate of 90%. Compared to the proposed hybrid solvent scenarios, the amine-based procedure (base case) had a lower $CO_2$ sequestration rate. Moreover, the specific $CO_2$ emission demonstrated that the hybrid solvents had lower $CO_2$ emissions, like in Case-III (0.015), which was 92.5% more favorable than the amine-based base.

SPECCA, on the other contrary, compares the methods based on the energy required to capture $CO_2$ and the corresponding emissions. The results presented in **Figure 7** demonstrate that Cases II and III had a lower SPECCA of 0.036 and 0.037, respectively. A lower SPECCA indicates that the process is very efficient and maximizes carbon capture. Furthermore, compared to the Aq.MDEA-based base case, the proposed cases based on a hybrid solvent were more ecologically sound in terms of energy requirement because the proposed cases were based on air stripping technology for regeneration, reducing their emission rate and overall thermal energy requirement.

### *5.3. Exergy analysis*

The total exergy destruction and efficiency for the processing equipment involved in the base case and Cases I-III are shown in **Figure 8**. It can be observed that Case-III, based on 5 wt% [Omim][$BF_4$]+50 wt% MDEA + 45 wt% $H_2O$, was the most efficient case providing an exergy saving of

**Table 6.** Design constraints and optimized variables employed based on the simulation results for the base case and proposed cases for biogas upgrading.

| Variables/Parameters | Base Case | Case-I | Case-II | Case-III |
|---|---|---|---|---|
| **Biogas upgrading** | | | | |
| Hybrid Solvent composition (wt%) | MDEA (50), $H_2O$ (50) | MDEA (50), $H_2O$ (30), [Omim][$BF_4$] (20) | MDEA (50), $H_2O$ (40), [Omim][$BF_4$] (10) | MDEA (50), $H_2O$ (45), [Omim][$BF_4$] (5) |
| **Design Constraints** | | | | |
| Purity of biomethane (wt%) | 0.99 | 0.9751 | 0.9751 | 0.9751 |
| Recovery of biomethane (wt%) | 0.99 | 0.988 | 0.98 | 0.99 |
| $CO_2$ removal (%) | 0.95 | 0.95 | 0.99 | 0.99 |
| **Absorber Column** | | | | |
| Biogas flow (kg/h) | 60000.0 | 60000.0 | 60000.0 | 60000.0 |
| Biogas flow ($m^3$/h) | 71686.05 | 71686.05 | 71686.05 | 71686.05 |
| Solvent flow ($m^3$/h) | 343.4 | 908.4 | 814.7 | 749.29 |
| Hybrid Solvent/Feed gas (HS/F) ratio ($m^3/m^3$) | 0.0047 | 0.55 | 0.5 | 0.45 |
| Absorber column stages | 20 | 20 | 20 | 18 |
| Absorber column pressure (bar) | 40 | 40 | 40 | 40 |
| Absorber column temperature (°C) | 35 | 35 | 35 | 35 |
| Flash column-I pressure (bar) | 2.8 | 13.28 | 13.48 | 13.25 |
| Flash column-II pressure (bar) | - | 4.5 | 4.3 | 4.1 |
| Recycle gas compressor power (kW) | - | 8640.0 | 6543.3 | 5667.5 |
| **Regenerator Column** | | | | |
| Regenerator column stages | 20 | 18 | 20 | 18 |
| Regenerator column temperature (°C) | 90 | 30 | 30 | 30 |
| Regenerator column pressure (bar) | 1.51 | 1.2 | 1.2 | 1.2 |
| Regenerator column reflux ratio | 1 | - | - | - |
| Thermal requirement for regenerator column (kW) | 44400 | - | - | - |
| Air flow rate ($m^3$/h) | - | 19666.3 | 22254.8 | 6329.83 |
| Air / Hybrid solvent (A/HS) rich ratio ($m^3/m^3$) | - | 0.53 | 1.71 | 0.59 |
| Power of pump for recycling (kW) | 508.3 | 8345.8 | 1096.1 | 991.4 |
| Power of air compressor (kW) | - | 166.7 | 162.6 | 46.27 |
| SEC (kWh /kg of biogas) | 0.97 | 0.89 | 0.36 | 0.34 |

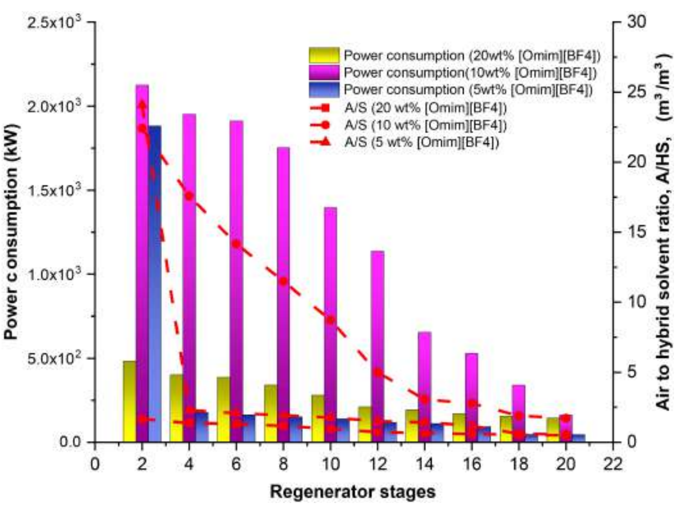


**Fig. 6.** Regenerator analysis to assess the optimal airflow rate concerning the solvent-rich stream at various regenerator stages and corresponding power consumption.

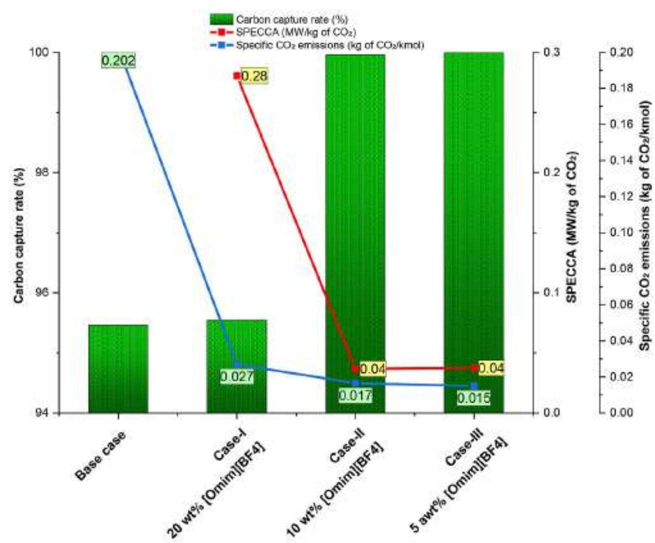


**Fig. 7.** Carbon capture rate (%), specific primary energy consumption for $CO_2$ avoided (SPECCA), and specific $CO_2$ emission (kg $CO_2$/kmol) for the ionic liquid-based hybrid solvent cases in comparison with the amine-based base case.

54.52% against the base case. This marked difference would be due to the fewer thermodynamic irreversibilities occurring in the process, providing a synergistic effect on the overall process. Additionally, from Figure 8, it can be seen that Case-III provided the lowest exergy destruction of 54.25 MW with the highest exergy efficiency of 43.36% compared to the base case (36.9%), Cases-I (29.48%), and Case-II (42.45%). The reason contributing to the lower exergy destruction of 54.25 MW in Case-III compared to the base

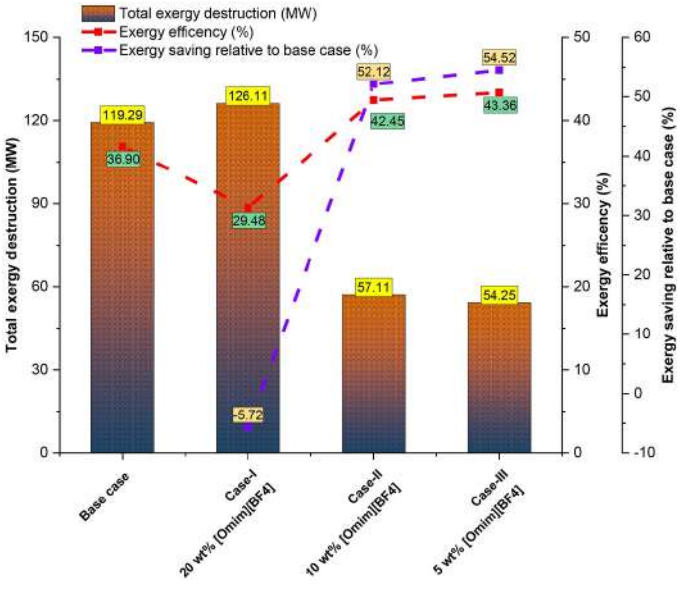


**Fig. 8.** Total equipment exergy destruction and exergy efficiency for each of the three studied ionic liquid-based hybrid solvent cases compared to the amine-based base case and corresponding exergy savings.

case (119.29 MW), Case-I (126.11 MW), and Case-II (57.11 MW) could be attributed to the lower energy requirement in the regenerator, which is the major contributor to exergy destruction.

To further mark the performance of the proposed process, exergy analysis was combined with various LCA variables to analyze the avenue of process enhancement further. **Figure 9** shows that the exergy destruction factor ($f_{ei}$) for Case-III had the lowest exergy destruction factor, i.e., 1.16, when compared to the other proposed cases. The lower $f_{ei}$ value accounts for the lower exergy losses in the hybrid system, which was nearly 79.56% lower than the Aq.MDEA-based base case. The environmental destruction index focuses on the assessment by combining entropy generation in a system as a result of irreversibilities occurring in the process and its negative impact on the ecological system.

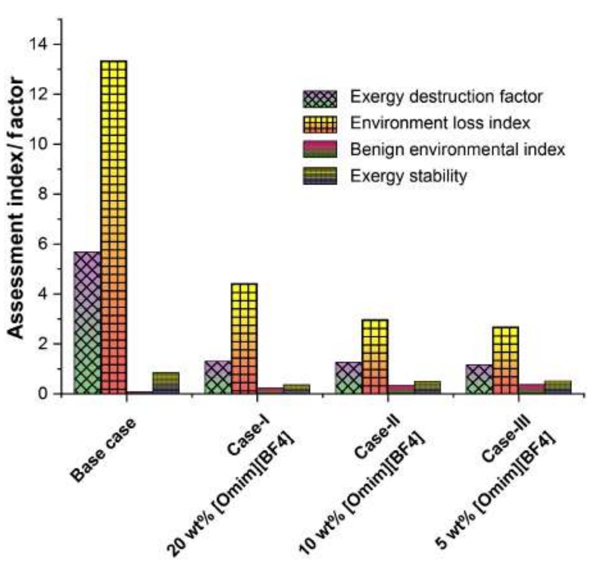


**Fig. 9.** Assessment of the proposed processes compared to the base case by combining the exergy and life cycle assessment analysis.

**Figure 9** also reveals that Case-III and Case-I were on the lower side, i.e., 2.67, which could be attributable to the process having lower exergetic losses and becoming less damaging to the environment than the other proposed cases. It is critical to emphasize that the hybrid solvent made the process more environmentally friendly, as it was 79.94% less harmful to the environment than the base case. Furthermore, to highlight the process positivity, the benign environmental index (Θii) was calculated, which was higher for Case-II and Case-III, i.e., 0.37, in comparison with the base case (0.07), indicating that they had a more positive impact on the environment due to their lower exergy losses, high exergy efficiency, and lower specific energy requirement. Furthermore, the analysis also quantified the process's operational capabilities, demonstrating that the hybrid solvent processes provided a more stable process with lower environmental losses than the base case, i.e., 56.5% for Case-I, 42% for Case-II, and 40.14% for Case-III.

### *5.4. Economic evaluation*

**Figure 10** depicts the economic analysis results, showing that Case-III, 5 wt% [Omim][$BF_4$] + 50 wt% MDEA +45 wt% $H_2O$, was the most economically viable option, with a total capital cost (TCC) of USD 2.11 × $10^7$, which was nearly 56% less than the base case and approximately 70% and 4% less than Case-I and Case-II, respectively. Similarly, the total operational cost (TOC) for Case-III was the lowest (USD 4.84 × $10^6$). Compared to the 50 wt% Aq. MDEA-based process (base case), Case-III delivered a TOC savings of 31% since the base case required more thermal energy for the regeneration of amines and a larger variable cost in process maintenance. Similarly, the TAC for Case-III was lower, i.e., USD 4.23 × $10^6$/yr among the proposed cases as well as in comparison with the base case. More specifically, Case-III saved 74.65% on TAC compared to the base case. At the same time, Case-I and Case-II led to TAC savings of 73.57% and 14.56 %, respectively. Hence, based on the economic projection and estimation, it can be concluded that hybrid solvent seemed to be a viable choice for the biogas upgrading process compared to the amine-based conventional process, and all the factors and indicators favored the use of hybrid solvent constituting IL blended with Aq.MDEA for the separation of $CO_2$.

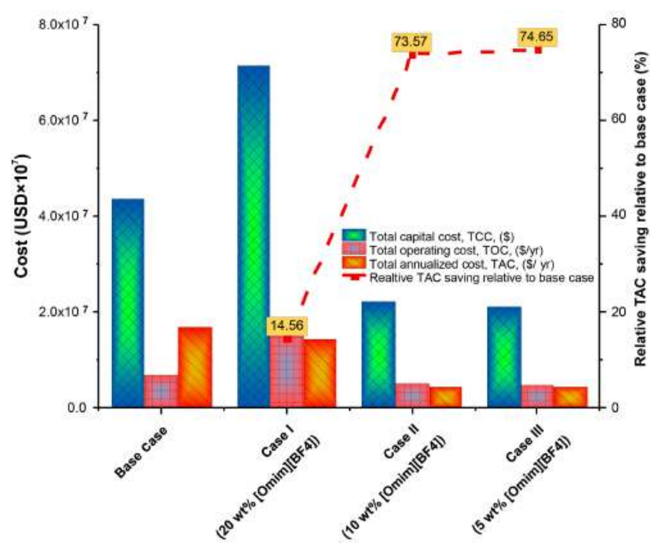


**Fig. 10.** Economic analysis of the base case and proposed hybrid solvent-based cases.

## 6. Limitations of the present study

ILs, regarded as "green solvents," can facilitate a better capture rate for eliminating $CO_2$-based impurities. However, some factors limit the use of ILs and may be regarded as a major impediment to their commercial implications. These factors include the IL's operation and process economy, its high operating costs, and perhaps most pertinently, its viscosity, which hinders the mass transfer operation of the process and also raises the power consumption requiring a large volume of the solvent. Even though ILs and their associated hybrid solvents offer significant process efficiency, as discussed earlier in the present work, the high cost and operational and technical challenges associated with ILs still prevent their practical adoption in biogas upgrading.

**7. Practical implication of the present study**

To practically implement the IL-based hybrid solvent for biogas upgrading, a strategic approach must be developed, beginning with rigorous experimental data at various temperatures and pressures, which can then be implemented using commercial process design software that provides a robust initial design. The proposed process design can be backed up by process system engineering-based analysis techniques that link the initial experimental evaluation of ILs with a wide range of analysis results obtained based on process design, serving as a stepping stone for their development to pilot-scale implementation.

**8. Conclusions and future perspectives**

It is vital to develop an energy-efficient, environmentally friendly, and economically viable process when using biogas as a renewable and clean fuel for energy generation while meeting environmental laws. The traditional biogas upgrading technology based on alkanol amine requires high amounts of energy for $CO_2$ removal, and amines are detrimental to the environment. This study provided a comprehensive understanding of using a hybrid solvent, including IL [Omim][$BF_4$], in combination with Aq.MDEA for the selective sequestration of $CO_2$ from raw biogas streams.

The hybrid solvent was designed to provide a synergistic effect with minimal energy, ease of operation with minimal losses, low environmental impact, and financial feasibility. The results showed 5 wt% [Omim][$BF_4$] in combination with Aq.MDEA delivered a 99 wt% recovery and 97 wt% purity of $CH_4$ with a lower specific energy consumption of 0.34 kW/kg biogas, resulting in an energy savings of around 64.94% when compared to the amine-based base case. Furthermore, the thermodynamic behavior was analyzed using exergy analysis, which provided a more in-depth understanding of the hotspots introducing thermodynamic irreversibilities into the process. In comparison with the Aq.MDEA, the result based on 5wt% [Omim][$BF_4$] showed less overall exergy destruction of 54.25 MW and exergy efficiency of 43.36%, providing a relative exergy saving of 54%.

The proposed hybrid solvent was also considered environmentally acceptable. According to the findings, 5wt% [Omim][$BF_4$] captured 99% of $CO_2$ with a reduced specific $CO_2$ emission of 0.015 kg $CO_2$/kmol of feed and a lower SPECCA of 0.04 kW/kg $CO_2$. A systematic study was also performed by combining the exergy and the LCA variables, which revealed that the 5wt% [Omim][$BF_4$] case was 79.94% less negative for the environment than the base case. Furthermore, the ϴii for this approach (Case-III) was higher, at 0.37, compared to 0.07 for the base case, indicating that this IL-based hybrid solvent system had a more positive impact on the environment due to lower exergy losses, high exergy efficiency, and lower specific energy requirement. The process was also studied by executing an economic evaluation, which showed that 5 wt% [Omim][$BF_4$] led to a relative saving of 56, 31, and 74.65% of TCC, TOC, and TAC when compared to the Aq.MDEA-based base case for biogas upgrading.

Future research should concentrate on analyzing the LCA of IL-based hybrid solvents. This analysis should be able to quantitatively evaluate the technical and environmental repercussions of the process across a broad domain. This could further assist in intensifying the application of the ILs and the hybrid solvents for carbon dioxide sequestration. In addition, for initial forecasting, process design, and simulation, based on rigorous solubility data with a variety of cations and anions combination of ILs with other components of gas should be explored. Furthermore, the obtained biomethane can be integrated with a liquefaction process based on mixed refrigerant to assess the process, providing a higher liquefaction rate and reduced overall energy requirement.

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**Bilal Kazmi** is currently a Research Scholar at the Department of Applied Chemistry and Chemical Technology at the University of Karachi. His research is focused on using ionic liquids for acid gas removal from natural gas and biogas based on a process system engineering perspective, biomass for biohydrogen production, natural gas, and biogas liquefaction process intensification. He has published more than 10 SCI papers in highly referred journals.

**Dr. Syed Imran Ali** completed his PhD in 2010 from the Eindhoven University of Technology, the Netherlands. His research was focused on exploring emulsion polymerization routes for the design and synthesis of anisotropic and hollow latex particles. Then, as a postdoctoral fellow, he extended the work towards responsive nanocapsules for drug release applications. In 2012, he joined Polymer & Petrochemical Engineering Department at NED University of Engineering and Technology, Pakistan, as Assistant Professor. Since August 2015, he has been working as an Associate Professor at the Department of Applied Chemistry & Chemical Technology, University of Karachi, Pakistan. His current research interests focus on polymeric systems for drug encapsulation and controlled release, polymer encapsulation of nanoparticles and polymeric nanocomposites for advanced applications, and Acid gas removal using ionic liquids.

**Prof. Dr. Zahoor Ul Hussain Awan** completed his PhD in 2014 at Chonbuk National University, South Korea, where his research was focused on developing an effective Lithium air-based energy storage device. He is currently working as a full-time Professor and serving as the Head of the Department of Food Engineering at the NED University of Engineering and Technology, Karachi, Pakistan. Prof. Dr. Zahoor has published more than 50 research papers in reputable journals and has been cited around 900 times (h-index: 16, i10-index:21, Source: Google Scholar). His current research interests focus on the ionic liquid assessment as an emergent solvent for acid gas removal from a process system engineering perspective, and synthesis and characterization of the nanocomposites for enhancing energy storage devices and advancing the hydrogel network.

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**Supplementary Information**

**Thermodynamic model and rigorous regression**

The experimentally determined vapor-liquid equilibrium (VLE) data of the systems $CO_2$ + [Omim][$BF_4$]+MDEA were implemented in Aspen Plus using the Peng Robinson (PengRobin) model thermodynamic model.

Since hydrocarbon processing requires high temperatures and pressures, PengRobin's standard cubical equation of state is a good fit for this type of application. The model is also consistent in the critical zone. So unlike other thermodynamic models using activity coefficients, this one does not show much variation. The PengRobin equation can be applied to polar, non-ideal chemical mixtures (**Eq. S1**):

$$P = \frac{RT}{V_m} - \frac{a}{V_m(V_m+b)+b(V_m-b)} \quad \text{Eq. S1}$$

$V_m$ = Molar volume

$$a = \sum_i \sum_j x_i x_j (a_i a_j)^{0.5}(1-k_{ij})$$

$$b = \sum_i x_i b_i$$

$$k_{ij} = k_{ij}^{(1)} + k_{ij}^{(2)} T + \frac{k_{ij}^{(3)}}{T}$$

**Table S1.**
The experimental and estimated $CO_2$ solubility ($X_{CO2}$) in $CO_2$ + [Omim][$BF_4$]+[MDEA] as a function of pressure.

| $X_{CO2(exp)}$ | $X_{CO2(est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 298.15 K | | | | | | | |
| 8.8300E-01 | 8.6950E-01 | 8.8300E-04 | -1.3400E-02 | 2.6000E+00 | 3.5143E+00 | 2.6000E-03 | 9.1430E-01 |
| 9.8100E-01 | 9.7890E-01 | 9.8100E-04 | -2.0900E-03 | 6.6000E+00 | 1.6231E+01 | 6.6000E-03 | 9.6308E+00 |
| 9.8000E-01 | 9.7930E-01 | 9.8000E-04 | -6.6000E-04 | 1.1200E+01 | 1.8637E+01 | 1.1200E-02 | 7.4368E+00 |
| 9.5500E-01 | 9.5520E-01 | 9.5500E-04 | 2.3600E-04 | 1.5200E+01 | 1.7619E+01 | 1.5200E-02 | 2.4187E+00 |
| 9.3600E-01 | 9.3890E-01 | 9.3600E-04 | 2.9590E-03 | 1.8900E+01 | 1.8162E+01 | 1.8900E-02 | -7.3840E-01 |
| 9.1500E-01 | 9.2070E-01 | 9.1500E-04 | 5.7180E-03 | 2.2200E+01 | 1.8918E+01 | 2.2200E-02 | -3.2821E+00 |
| 8.9800E-01 | 9.0520E-01 | 8.9800E-04 | 7.2520E-03 | 2.5400E+01 | 1.9901E+01 | 2.5400E-02 | -5.4988E+00 |
| 8.7700E-01 | 8.8490E-01 | 8.7700E-04 | 7.9460E-03 | 2.8000E+01 | 2.0820E+01 | 2.8000E-02 | -7.1802E+00 |
| 8.5000E-01 | 8.3960E-01 | 8.5000E-04 | -1.0390E-02 | 2.0000E+00 | 1.7077E+01 | 2.0000E-03 | 1.5077E+01 |
| 9.4300E-01 | 9.4430E-01 | 9.4300E-04 | 1.3390E-03 | 4.2000E+00 | 1.9802E+01 | 4.2000E-03 | 1.5602E+01 |
| 9.9500E-01 | 9.9490E-01 | 9.9500E-04 | -1.1125E-05 | 8.6000E+00 | 2.1055E+01 | 8.6000E-03 | 1.2455E+01 |
| 9.5700E-01 | 9.5840E-01 | 9.5700E-04 | 1.4000E-03 | 1.3400E+01 | 2.2681E+01 | 1.3400E-02 | 9.2805E+00 |
| 9.3100E-01 | 9.3430E-01 | 9.3100E-04 | 3.3950E-03 | 1.7500E+01 | 2.3229E+01 | 1.7500E-02 | 5.7291E+00 |
| 9.1000E-01 | 9.1520E-01 | 9.1000E-04 | 5.2440E-03 | 2.1300E+01 | 2.3576E+01 | 2.1300E-02 | 2.2759E+00 |
| 8.8600E-01 | 8.9340E-01 | 8.8600E-04 | 7.4000E-03 | 2.4700E+01 | 2.3753E+01 | 2.4700E-02 | -9.4660E-01 |
| 8.6400E-01 | 8.7330E-01 | 8.6400E-04 | 9.3560E-03 | 2.7000E+01 | 2.3780E+01 | 2.7000E-02 | -3.2199E+00 |
| 8.6100E-01 | 8.5900E-01 | 8.6100E-04 | -1.9600E-03 | 2.8000E+00 | 1.7183E+01 | 2.8000E-03 | 1.4383E+01 |
| 9.4800E-01 | 9.4910E-01 | 9.4800E-04 | 1.1650E-03 | 5.3000E+00 | 1.9676E+01 | 5.3000E-03 | 1.4376E+01 |
| 9.8500E-01 | 9.8490E-01 | 9.8500E-04 | -6.1412E-05 | 6.9000E+00 | 2.0453E+01 | 6.9000E-03 | 1.3553E+01 |
| 9.7300E-01 | 9.7330E-01 | 9.7300E-04 | 3.7000E-04 | 1.1300E+01 | 2.1691E+01 | 1.1300E-02 | 1.0391E+01 |
| 9.2100E-01 | 9.2490E-01 | 9.2100E-04 | 3.9850E-03 | 1.7000E+01 | 2.2253E+01 | 1.7000E-02 | 5.2527E+00 |
| 8.9400E-01 | 9.0030E-01 | 8.9400E-04 | 6.3860E-03 | 2.1000E+01 | 2.2500E+01 | 2.1000E-02 | 1.5003E+00 |
| 8.6900E-01 | 8.7760E-01 | 8.6900E-04 | 8.6690E-03 | 2.3900E+01 | 2.2560E+01 | 2.3900E-02 | -1.3395E+00 |
| 8.4400E-01 | 8.5510E-01 | 8.4400E-04 | 1.1142E-02 | 2.6800E+01 | 2.2582E+01 | 2.6800E-02 | -4.2179E+00 |
| Average deviation = 0.0023<br>Root mean square error = 0.0061<br>Average absolute = 0.0046 | | | | | | | |
| 313.15 K | | | | | | | |
| 8.4300E-01 | 8.2750E-01 | 8.4300E-04 | -1.5400E-02 | 4.8000E+00 | 6.7377E+00 | 4.8000E-03 | 1.9377E+00 |
| 9.4100E-01 | 9.3190E-01 | 9.4100E-04 | -9.0870E-03 | 9.0000E+00 | 1.4977E+01 | 9.0000E-03 | 5.9767E+00 |
| 9.8300E-01 | 9.8270E-01 | 9.8300E-04 | -2.9430E-04 | 1.3600E+01 | 2.4097E+01 | 1.3600E-02 | 1.0497E+01 |
| 9.9100E-01 | 9.9100E-01 | 9.9100E-04 | 4.0801E-05 | 1.7800E+01 | 2.6011E+01 | 1.7800E-02 | 8.2114E+00 |
| 9.6900E-01 | 9.6920E-01 | 9.6900E-04 | 2.8030E-04 | 2.1600E+01 | 2.4668E+01 | 2.1600E-02 | 3.0677E+00 |
| 9.2460E-01 | 9.2450E-01 | 9.2400E-04 | -6.0759E-05 | 2.5100E+01 | 2.7681E+01 | 2.5100E-02 | 2.5813E+00 |
| 9.3200E-01 | 9.3470E-01 | 9.3200E-04 | 2.7000E-03 | 2.8600E+01 | 2.4779E+01 | 2.8600E-02 | -3.8207E+00 |
| 9.1300E-01 | 9.1660E-01 | 9.1300E-04 | 3.6110E-03 | 3.1500E+01 | 2.5416E+01 | 3.1500E-02 | -6.0841E+00 |
| 8.1200E-01 | 7.5920E-01 | 8.1200E-04 | -5.2790E-02 | 4.1000E+00 | 6.0814E+00 | 4.1000E-03 | 1.9814E+00 |
| 9.0000E-01 | 8.7350E-01 | 9.0000E-04 | -2.6410E-02 | 6.6000E+00 | 1.1042E+01 | 6.6000E-03 | 4.4417E+00 |
| 9.6400E-01 | 9.6140E-01 | 9.6400E-04 | -2.5730E-03 | 1.1100E+01 | 2.1164E+01 | 1.1100E-02 | 1.0064E+01 |
| 9.9700E-01 | 9.9700E-01 | 9.9700E-04 | 1.7664E-05 | 1.6100E+01 | 2.6316E+01 | 1.6100E-02 | 1.0216E+01 |
| 9.7100E-01 | 9.7090E-01 | 9.7100E-04 | -9.1603E-07 | 2.0400E+01 | 2.4742E+01 | 2.0400E-02 | 4.3424E+00 |
| 9.4700E-01 | 9.4780E-01 | 9.4700E-04 | 8.1900E-04 | 2.4300E+01 | 2.4297E+01 | 2.4300E-02 | -2.8350E-03 |
| 9.2600E-01 | 9.2810E-01 | 9.2600E-04 | 2.1020E-03 | 2.8100E+01 | 2.4506E+01 | 2.8100E-02 | -3.5942E+00 |
| 9.0500E-01 | 9.0830E-01 | 9.0500E-04 | 3.3060E-03 | 3.0600E+01 | 2.4676E+01 | 3.0600E-02 | -5.9240E+00 |
| 8.1300E-01 | 7.4810E-01 | 8.1300E-04 | -6.4800E-02 | 5.4000E+00 | 9.6802E+00 | 5.4000E-03 | 4.2802E+00 |
| 8.9500E-01 | 8.7630E-01 | 8.9500E-04 | -1.8630E-02 | 8.2000E+00 | 1.6483E+01 | 8.2000E-03 | 8.2832E+00 |

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**Table S1.**
Continued.

| $X_{CO2(exp)}$ | $X_{CO2(est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 9.2600E-01 | 9.1870E-01 | 9.2600E-04 | -7.2740E-03 | 1.0200E+01 | 1.9643E+01 | 1.0200E-02 | 9.4431E+00 |
| 9.7600E-01 | 9.7530E-01 | 9.7600E-04 | -6.2610E-04 | 1.4400E+01 | 2.4057E+01 | 1.4400E-02 | 9.6572E+00 |
| 9.7400E-01 | 9.7380E-01 | 9.7400E-04 | -1.4360E-04 | 2.0400E+01 | 2.5114E+01 | 2.0400E-02 | 4.7143E+00 |
| Average deviation = -0.0075 | | | | | | | |
| Root mean square error = 0.0187 | | | | | | | |
| Average absolute = 0.0089 | | | | | | | |
| 323.15 K | | | | | | | |
| 8.1400E-01 | 7.9549E-01 | 8.1400E-04 | -1.8514E-02 | 6.5000E+00 | 9.2578E+00 | 6.5000E-03 | 2.7578E+00 |
| 9.0600E-01 | 8.9729E-01 | 9.0600E-04 | -8.7094E-03 | 1.1100E+01 | 1.6639E+01 | 1.1100E-02 | 5.5387E+00 |
| 9.4700E-01 | 9.4521E-01 | 9.4700E-04 | -1.7920E-03 | 1.5900E+01 | 2.3525E+01 | 1.5900E-02 | 7.6252E+00 |
| 9.7800E-01 | 9.7796E-01 | 9.7800E-04 | -3.6706E-05 | 2.0000E+01 | 2.7933E+01 | 2.0000E-02 | 7.9330E+00 |
| 9.7800E-01 | 9.7827E-01 | 9.7800E-04 | 2.7369E-04 | 2.7500E+01 | 2.9200E+01 | 2.7500E-02 | 1.7001E+00 |
| 9.6100E-01 | 9.6187E-01 | 9.6100E-04 | 8.6777E-04 | 3.1100E+01 | 2.8836E+01 | 3.1100E-02 | -2.2639E+00 |
| 9.4000E-01 | 9.4176E-01 | 9.4000E-04 | 1.7598E-03 | 3.4100E+01 | 2.8855E+01 | 3.4100E-02 | -5.2446E+00 |
| 7.7700E-01 | 7.2096E-01 | 7.7700E-04 | -5.6039E-02 | 6.1000E+00 | 9.0982E+00 | 6.1000E-03 | 2.9982E+00 |
| 8.6000E-01 | 8.3313E-01 | 8.6000E-04 | -2.6868E-02 | 8.9000E+00 | 1.4248E+01 | 8.9000E-03 | 5.3483E+00 |
| 9.2400E-01 | 9.1822E-01 | 9.2400E-04 | -5.7820E-03 | 1.3500E+01 | 2.1732E+01 | 1.3500E-02 | 8.2315E+00 |
| 9.6500E-01 | 9.6444E-01 | 9.6500E-04 | -5.5797E-04 | 1.8500E+01 | 2.6593E+01 | 1.8500E-02 | 8.0926E+00 |
| 9.9500E-01 | 9.9504E-01 | 9.9500E-04 | 3.7667E-05 | 2.2800E+01 | 2.9810E+01 | 2.2800E-02 | 7.0099E+00 |
| 9.8200E-01 | 9.8215E-01 | 9.8200E-04 | 1.4787E-04 | 2.6900E+01 | 2.9299E+01 | 2.6900E-02 | 2.3988E+00 |
| 9.6100E-01 | 9.6164E-01 | 9.6100E-04 | 6.3806E-04 | 3.0800E+01 | 2.8673E+01 | 3.0800E-02 | -2.1272E+00 |
| 9.3600E-01 | 9.3748E-01 | 9.3600E-04 | 1.4826E-03 | 3.3300E+01 | 2.8307E+01 | 3.3300E-02 | -4.9935E+00 |
| Average deviation = -0.0075 | | | | | | | |
| Root mean square error = 0.0169 | | | | | | | |
| Average absolute = 0.0082 | | | | | | | |
| 333.15 K | | | | | | | |
| 7.7400E-01 | 7.5157E-01 | 7.7400E-04 | -2.2434E-02 | 8.9000E+00 | 1.2618E+01 | 8.9000E-03 | 3.7184E+00 |
| 8.5800E-01 | 8.5024E-01 | 8.5800E-04 | -7.7612E-03 | 1.4000E+01 | 1.9603E+01 | 1.4000E-02 | 5.6026E+00 |
| 9.0100E-01 | 8.9920E-01 | 9.0100E-04 | -1.8028E-03 | 1.8800E+01 | 2.4887E+01 | 1.8800E-02 | 6.0865E+00 |
| 9.3400E-01 | 9.3398E-01 | 9.3400E-04 | -1.7714E-05 | 2.2900E+01 | 2.8368E+01 | 2.2900E-02 | 5.4681E+00 |
| 9.6100E-01 | 9.6130E-01 | 9.6100E-04 | 3.0154E-04 | 2.6700E+01 | 3.0833E+01 | 2.6700E-02 | 4.1333E+00 |
| 9.8400E-01 | 9.8417E-01 | 9.8400E-04 | 1.7050E-04 | 3.0400E+01 | 3.2684E+01 | 3.0400E-02 | 2.2838E+00 |
| 9.9600E-01 | 9.9605E-01 | 9.9600E-04 | 5.0255E-05 | 3.4000E+01 | 3.3593E+01 | 3.4000E-02 | -4.0705E-01 |
| 9.7700E-01 | 9.7737E-01 | 9.7700E-04 | 3.7192E-04 | 3.7200E+01 | 3.3046E+01 | 3.7200E-02 | -4.1536E+00 |
| 7.3800E-01 | 6.7915E-01 | 7.3800E-04 | -5.8849E-02 | 8.4000E+00 | 1.2475E+01 | 8.4000E-03 | 4.0752E+00 |
| 8.0700E-01 | 7.7969E-01 | 8.0700E-04 | -2.7312E-02 | 1.2000E+01 | 1.8018E+01 | 1.2000E-02 | 6.0183E+00 |
| 8.7100E-01 | 8.6305E-01 | 8.7100E-04 | -7.9488E-03 | 1.6700E+01 | 2.3782E+01 | 1.6700E-02 | 7.0820E+00 |
| 9.1500E-01 | 9.1343E-01 | 9.1500E-04 | -1.5692E-03 | 2.1700E+01 | 2.7618E+01 | 2.1700E-02 | 5.9176E+00 |
| 9.4700E-01 | 9.4697E-01 | 9.4700E-04 | -2.5367E-05 | 2.6000E+01 | 2.9989E+01 | 2.6000E-02 | 3.9887E+00 |
| 9.7500E-01 | 9.7519E-01 | 9.7500E-04 | 1.8755E-04 | 3.0000E+01 | 3.1958E+01 | 3.0000E-02 | 1.9578E+00 |
| 9.9900E-01 | 9.9901E-01 | 9.9900E-04 | 1.1422E-05 | 3.3900E+01 | 3.3685E+01 | 3.3900E-02 | -2.1540E-01 |
| 9.7900E-01 | 9.7928E-01 | 9.7900E-04 | 2.7786E-04 | 3.6600E+01 | 3.2845E+01 | 3.6600E-02 | -3.7548E+00 |
| 7.0300E-01 | 6.5448E-01 | 7.0300E-04 | -4.8523E-02 | 1.1600E+01 | 1.8451E+01 | 1.1600E-02 | 6.8512E+00 |
| 7.8000E-01 | 7.5938E-01 | 7.8000E-04 | -2.0625E-02 | 1.4800E+01 | 2.2387E+01 | 1.4800E-02 | 7.5867E+00 |
| 8.0500E-01 | 7.9274E-01 | 8.0500E-04 | -1.2260E-02 | 1.7100E+01 | 2.4104E+01 | 1.7100E-02 | 7.0037E+00 |
| 8.6100E-01 | 8.5719E-01 | 8.6100E-04 | -3.8130E-03 | 2.1200E+01 | 2.6743E+01 | 2.1200E-02 | 5.5429E+00 |
| 9.1300E-01 | 9.1278E-01 | 9.1300E-04 | -2.2044E-04 | 2.7400E+01 | 2.9242E+01 | 2.7400E-02 | 1.8423E+00 |
| 9.4200E-01 | 9.4231E-01 | 9.4200E-04 | 3.1127E-04 | 3.1800E+01 | 3.0611E+01 | 3.1800E-02 | -1.1893E+00 |
| 9.6200E-01 | 9.6232E-01 | 9.6200E-04 | 3.2040E-04 | 3.5300E+01 | 3.1629E+01 | 3.5300E-02 | -3.6706E+00 |
| 9.9100E-01 | 9.9107E-01 | 9.9100E-04 | 7.4638E-05 | 3.8400E+01 | 3.3365E+01 | 3.8400E-02 | -5.0349E+00 |
| Average deviation = -0.0087 | | | | | | | |
| Root mean square error = 0.0180 | | | | | | | |
| Average absolute = 0.0089 | | | | | | | |
| 343.15 K | | | | | | | |
| 7.2300E-01 | 6.9233E-01 | 7.2300E-04 | -3.0669E-02 | 1.2000E+01 | 1.6664E+01 | 1.2000E-02 | 4.6636E+00 |
| 8.0500E-01 | 7.9466E-01 | 8.0500E-04 | -1.0335E-02 | 1.7300E+01 | 2.3262E+01 | 1.7300E-02 | 5.9618E+00 |
| 8.5100E-01 | 8.4799E-01 | 8.5100E-04 | -3.0128E-03 | 2.2100E+01 | 2.7889E+01 | 2.2100E-02 | 5.7889E+00 |
| 8.8200E-01 | 8.8161E-01 | 8.8200E-04 | -3.8913E-04 | 2.6400E+01 | 3.0885E+01 | 2.6400E-02 | 4.4850E+00 |
| 9.1300E-01 | 9.1353E-01 | 9.1300E-04 | 5.2552E-04 | 3.0100E+01 | 3.2882E+01 | 3.0100E-02 | 2.7821E+00 |
| 9.3600E-01 | 9.3672E-01 | 9.3600E-04 | 7.1779E-04 | 3.3900E+01 | 3.4274E+01 | 3.3900E-02 | 3.7447E-01 |
| 9.5700E-01 | 9.5759E-01 | 9.5700E-04 | 5.9118E-04 | 3.7600E+01 | 3.5280E+01 | 3.7600E-02 | -2.3199E+00 |
| 9.8100E-01 | 9.8124E-01 | 9.8100E-04 | 2.4270E-04 | 4.0700E+01 | 3.6163E+01 | 4.0700E-02 | -4.5366E+00 |
| 6.8700E-01 | 6.2691E-01 | 6.8700E-04 | -6.0087E-02 | 1.1500E+01 | 1.6651E+01 | 1.1500E-02 | 5.1507E+00 |
| 7.4800E-01 | 7.1869E-01 | 7.4800E-04 | -2.9310E-02 | 1.5600E+01 | 2.1956E+01 | 1.5600E-02 | 6.3560E+00 |
| 8.1200E-01 | 8.0136E-01 | 8.1200E-04 | -1.0641E-02 | 2.0400E+01 | 2.6730E+01 | 2.0400E-02 | 6.3299E+00 |
| 8.5300E-01 | 8.4973E-01 | 8.5300E-04 | -3.2710E-03 | 2.5600E+01 | 2.9962E+01 | 2.5600E-02 | 4.3625E+00 |

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**Table S1.**
Continued.

| $X_{CO2(exp)}$ | $X_{CO2(est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 8.8500E-01 | 8.8435E-01 | 8.8500E-04 | -6.4753E-04 | 3.0000E+01 | 3.1928E+01 | 3.0000E-02 | 1.9279E+00 |
| 9.1600E-01 | 9.1635E-01 | 9.1600E-04 | 3.5164E-04 | 3.4000E+01 | 3.3379E+01 | 3.4000E-02 | -6.2085E-01 |
| 9.4300E-01 | 9.4358E-01 | 9.4300E-04 | 5.7816E-04 | 3.7900E+01 | 3.4524E+01 | 3.7900E-02 | -3.3758E+00 |
| 9.6400E-01 | 9.6445E-01 | 9.6400E-04 | 4.4742E-04 | 4.0700E+01 | 3.5344E+01 | 4.0700E-02 | -5.3559E+00 |
| 6.4300E-01 | 5.9756E-01 | 6.4300E-04 | -4.5436E-02 | 1.5200E+01 | 2.1783E+01 | 1.5200E-02 | 6.5829E+00 |
| 7.0000E-01 | 6.7918E-01 | 7.0000E-04 | -2.0820E-02 | 1.9500E+01 | 2.5312E+01 | 1.9500E-02 | 5.8122E+00 |
| 7.2800E-01 | 7.1448E-01 | 7.2800E-04 | -1.3518E-02 | 2.1700E+01 | 2.6736E+01 | 2.1700E-02 | 5.0365E+00 |
| 7.7600E-01 | 7.7100E-01 | 7.7600E-04 | -4.9995E-03 | 2.6200E+01 | 2.8981E+01 | 2.6200E-02 | 2.7809E+00 |
| 8.2400E-01 | 8.2396E-01 | 8.2400E-04 | -3.9507E-05 | 3.2700E+01 | 3.1140E+01 | 3.2700E-02 | -1.5601E+00 |
| 8.5900E-01 | 8.6013E-01 | 8.5900E-04 | 1.1272E-03 | 3.6900E+01 | 3.2308E+01 | 3.6900E-02 | -4.5924E+00 |
| 8.8100E-01 | 8.8251E-01 | 8.8100E-04 | 1.5061E-03 | 4.0400E+01 | 3.3071E+01 | 4.0400E-02 | -7.3290E+00 |
| 9.1000E-01 | 9.1134E-01 | 9.1000E-04 | 1.3418E-03 | 4.3600E+01 | 3.3840E+01 | 4.3600E-02 | -9.7602E+00 |
| Average deviation = -0.0094 | | | | | | | |
| Root mean square error = 0.0186 | | | | | | | |
| Average absolute = 0.0100 | | | | | | | |

**Table S2.**
The experimental and estimated [Omim][BF$_4$] solubility (X[Omim][BF$_4$]) in $CO_2$ + [Omim][BF4]+[MDEA] as a function of pressure.

| $X_{[Omim][BF4](exp)}$ | $X_{[Omim][BF4](est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 298.15 K | | | | | | | |
| 5.8500E-03 | 5.8028E-03 | 5.8500E-06 | -4.7231E-05 | 2.6000E+00 | 3.5144E+00 | 2.6000E-03 | 9.1440E-01 |
| 9.5000E-04 | 9.7060E-04 | 9.5000E-07 | 2.0596E-05 | 6.6000E+00 | 1.6231E+01 | 6.6000E-03 | 9.6309E+00 |
| 1.0000E-03 | 1.0195E-03 | 1.0000E-06 | 1.9464E-05 | 1.1200E+01 | 1.8637E+01 | 1.1200E-02 | 7.4368E+00 |
| 2.2500E-03 | 2.3116E-03 | 2.2500E-06 | 6.1576E-05 | 1.5200E+01 | 1.7619E+01 | 1.5200E-02 | 2.4187E+00 |
| 3.2000E-03 | 3.3738E-03 | 3.2000E-06 | 1.7378E-04 | 1.8900E+01 | 1.8162E+01 | 1.8900E-02 | -7.3848E-01 |
| 4.2500E-03 | 4.6007E-03 | 4.2500E-06 | 3.5071E-04 | 2.2200E+01 | 1.8918E+01 | 2.2200E-02 | -3.2821E+00 |
| 5.1000E-03 | 5.5906E-03 | 5.1000E-06 | 4.9062E-04 | 2.5400E+01 | 1.9901E+01 | 2.5400E-02 | -5.4988E+00 |
| 6.1500E-03 | 6.7716E-03 | 6.1500E-06 | 6.2160E-04 | 2.8000E+01 | 2.0820E+01 | 2.8000E-02 | -7.1802E+00 |
| 7.5000E-02 | 8.6439E-02 | 7.5000E-05 | 1.1439E-02 | 2.0000E+00 | 1.7077E+01 | 2.0000E-03 | 1.5077E+01 |
| 2.8500E-02 | 2.8568E-02 | 2.8500E-05 | 6.8478E-05 | 4.2000E+00 | 1.9802E+01 | 4.2000E-03 | 1.5602E+01 |
| 2.5000E-03 | 2.6022E-03 | 2.5000E-06 | 1.0215E-04 | 8.6000E+00 | 2.1055E+01 | 8.6000E-03 | 1.2455E+01 |
| 2.1500E-02 | 2.0838E-02 | 2.1500E-05 | -6.6246E-04 | 1.3400E+01 | 2.2681E+01 | 1.3400E-02 | 9.2805E+00 |
| 3.4500E-02 | 3.2068E-02 | 3.4500E-05 | -2.4323E-03 | 1.7500E+01 | 2.3229E+01 | 1.7500E-02 | 5.7291E+00 |
| 4.5000E-02 | 4.0772E-02 | 4.5000E-05 | -4.2285E-03 | 2.1300E+01 | 2.3576E+01 | 2.1300E-02 | 2.2759E+00 |
| 5.7000E-02 | 5.0645E-02 | 5.7000E-05 | -6.3545E-03 | 2.4700E+01 | 2.3753E+01 | 2.4700E-02 | -9.4669E-01 |
| 6.8000E-02 | 5.9723E-02 | 6.8000E-05 | -8.2775E-03 | 2.7000E+01 | 2.3780E+01 | 2.7000E-02 | -3.2199E+00 |
| 6.9500E-02 | 6.8521E-02 | 6.9500E-05 | -9.7863E-04 | 2.8000E+00 | 1.7183E+01 | 2.8000E-03 | 1.4383E+01 |
| 2.6000E-02 | 2.5066E-02 | 2.6000E-05 | -9.3364E-04 | 5.3000E+00 | 1.9676E+01 | 5.3000E-03 | 1.4376E+01 |
| 7.5000E-03 | 7.7039E-03 | 7.5000E-06 | 2.0394E-04 | 6.9000E+00 | 2.0453E+01 | 6.9000E-03 | 1.3553E+01 |
| 1.3500E-02 | 1.3420E-02 | 1.3500E-05 | -8.0296E-05 | 1.1300E+01 | 2.1691E+01 | 1.1300E-02 | 1.0391E+01 |
| 3.9500E-02 | 3.6288E-02 | 3.9500E-05 | -3.2121E-03 | 1.7000E+01 | 2.2253E+01 | 1.7000E-02 | 5.2527E+00 |
| 5.3000E-02 | 4.7635E-02 | 5.3000E-05 | -5.3648E-03 | 2.1000E+01 | 2.2500E+01 | 2.1000E-02 | 1.5004E+00 |
| 6.5500E-02 | 5.8088E-02 | 6.5500E-05 | -7.4125E-03 | 2.3900E+01 | 2.2560E+01 | 2.3900E-02 | -1.3396E+00 |
| 7.8000E-02 | 6.8389E-02 | 7.8000E-05 | -9.6106E-03 | 2.6800E+01 | 2.2582E+01 | 2.6800E-02 | -4.2179E+00 |
| Average deviation = -0.0015 | | | | | | | |
| Root mean square error = 0.0043 | | | | | | | |
| Average absolute = 0.0026 | | | | | | | |
| 313.15 K | | | | | | | |
| 7.8500E-03 | 8.2310E-03 | 7.8500E-06 | 3.8102E-04 | 4.8000E+00 | 6.7377E+00 | 4.8000E-03 | 1.9377E+00 |
| 2.9500E-03 | 2.9272E-03 | 2.9500E-06 | -2.2781E-05 | 9.0000E+00 | 1.4977E+01 | 9.0000E-03 | 5.9768E+00 |
| 8.5000E-04 | 8.7003E-04 | 8.5000E-07 | 2.0026E-05 | 1.3600E+01 | 2.4097E+01 | 1.3600E-02 | 1.0497E+01 |
| 4.5000E-04 | 4.5590E-04 | 4.5000E-07 | 5.9031E-06 | 1.7800E+01 | 2.6011E+01 | 1.7800E-02 | 8.2114E+00 |
| 1.5500E-03 | 1.5815E-03 | 1.5500E-06 | 3.1475E-05 | 2.1600E+01 | 2.4668E+01 | 2.1600E-02 | 3.0677E+00 |
| 2.6000E-03 | 2.6658E-03 | 2.6000E-06 | 6.5803E-05 | 2.5100E+01 | 2.7681E+01 | 2.5100E-02 | 2.5814E+00 |
| 3.4000E-03 | 3.5677E-03 | 3.4000E-06 | 1.6767E-04 | 2.8600E+01 | 2.4779E+01 | 2.8600E-02 | -3.8207E+00 |
| 4.3500E-03 | 4.6135E-03 | 4.3500E-06 | 2.6351E-04 | 3.1500E+01 | 2.5416E+01 | 3.1500E-02 | -6.0841E+00 |
| 1.8800E-02 | 1.5513E-02 | 1.8800E-05 | -3.2871E-03 | 4.1000E+00 | 6.0814E+00 | 4.1000E-03 | 1.9814E+00 |
| 1.0000E-02 | 8.7209E-03 | 1.0000E-05 | -1.2791E-03 | 6.6000E+00 | 1.1042E+01 | 6.6000E-03 | 4.4417E+00 |
| 3.6000E-03 | 3.6631E-03 | 3.6000E-06 | 6.3073E-05 | 1.1100E+01 | 2.1164E+01 | 1.1100E-02 | 1.0064E+01 |
| 3.0000E-04 | 3.0448E-04 | 3.0000E-07 | 4.4778E-06 | 1.6100E+01 | 2.6316E+01 | 1.6100E-02 | 1.0216E+01 |
| 2.9000E-03 | 2.9850E-03 | 2.9000E-06 | 8.4969E-05 | 2.0400E+01 | 2.4742E+01 | 2.0400E-02 | 4.3424E+00 |
| 5.3000E-03 | 5.5224E-03 | 5.3000E-06 | 2.2238E-04 | 2.4300E+01 | 2.4297E+01 | 2.4300E-02 | -2.8355E-03 |
| 7.4000E-03 | 7.7713E-03 | 7.4000E-06 | 3.7130E-04 | 2.8100E+01 | 2.4506E+01 | 2.8100E-02 | -3.5942E+00 |
| 9.5000E-03 | 9.9947E-03 | 9.5000E-06 | 4.9468E-04 | 3.0600E+01 | 2.4676E+01 | 3.0600E-02 | -5.9240E+00 |
| 3.7400E-02 | 2.5952E-02 | 3.7400E-05 | -1.1448E-02 | 5.4000E+00 | 9.6802E+00 | 5.4000E-03 | 4.2802E+00 |
| 2.1000E-02 | 1.7959E-02 | 2.1000E-05 | -3.0410E-03 | 8.2000E+00 | 1.6483E+01 | 8.2000E-03 | 8.2832E+00 |

**Table S2.**
Continued.

| $X_{[Omim][BF4](exp)}$ | $X_{[Omim][BF4](est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 1.4800E-02 | 1.3898E-02 | 1.4800E-05 | -9.0181E-04 | 1.0200E+01 | 1.9643E+01 | 1.0200E-02 | 9.4431E+00 |
| 4.8000E-03 | 4.9444E-03 | 4.8000E-06 | 1.4444E-04 | 1.4400E+01 | 2.4057E+01 | 1.4400E-02 | 9.6572E+00 |
| 5.2000E-03 | 5.3549E-03 | 5.2000E-06 | 1.5488E-04 | 2.0400E+01 | 2.5114E+01 | 2.0400E-02 | 4.7143E+00 |
| 1.0400E-02 | 1.0611E-02 | 1.0400E-05 | 2.1113E-04 | 4.8000E+00 | 6.7377E+00 | 4.8000E-03 | 1.9377E+00 |
| 1.4400E-02 | 1.4558E-02 | 1.4400E-05 | 1.5800E-04 | 9.0000E+00 | 1.4977E+01 | 9.0000E-03 | 5.9768E+00 |
| 1.8600E-02 | 1.8611E-02 | 1.8600E-05 | 1.0595E-05 | 1.3600E+01 | 2.4097E+01 | 1.3600E-02 | 1.0497E+01 |
| Average deviation = -0.00071<br>Root mean square error = 0.0025<br>Average absolute = 0.00095 | | | | | | | |
| 323.15 K | | | | | | | |
| 9.3000E-03 | 9.9133E-03 | 9.3000E-06 | 6.1325E-04 | 6.5000E+00 | 9.2578E+00 | 6.5000E-03 | 2.7578E+00 |
| 4.7000E-03 | 4.8174E-03 | 4.7000E-06 | 1.1744E-04 | 1.1100E+01 | 1.6639E+01 | 1.1100E-02 | 5.5387E+00 |
| 2.6500E-03 | 2.7370E-03 | 2.6500E-06 | 8.7026E-05 | 1.5900E+01 | 2.3525E+01 | 1.5900E-02 | 7.6252E+00 |
| 1.1000E-03 | 1.1211E-03 | 1.1000E-06 | 2.1113E-05 | 2.0000E+01 | 2.7933E+01 | 2.0000E-02 | 7.9330E+00 |
| 1.1000E-03 | 1.1122E-03 | 1.1000E-06 | 1.2190E-05 | 2.7500E+01 | 2.9200E+01 | 2.7500E-02 | 1.7001E+00 |
| 1.9500E-03 | 1.9915E-03 | 1.9500E-06 | 4.1530E-05 | 3.1100E+01 | 2.8836E+01 | 3.1100E-02 | -2.2639E+00 |
| 3.0000E-03 | 3.1086E-03 | 3.0000E-06 | 1.0864E-04 | 3.4100E+01 | 2.8855E+01 | 3.4100E-02 | -5.2446E+00 |
| 2.2300E-02 | 1.9011E-02 | 2.2300E-05 | -3.2892E-03 | 6.1000E+00 | 9.0982E+00 | 6.1000E-03 | 2.9982E+00 |
| 1.4000E-02 | 1.2767E-02 | 1.4000E-05 | -1.2328E-03 | 8.9000E+00 | 1.4248E+01 | 8.9000E-03 | 5.3483E+00 |
| 7.6000E-03 | 7.6687E-03 | 7.6000E-06 | 6.8714E-05 | 1.3500E+01 | 2.1732E+01 | 1.3500E-02 | 8.2315E+00 |
| 3.5000E-03 | 3.6237E-03 | 3.5000E-06 | 1.2366E-04 | 1.8500E+01 | 2.6593E+01 | 1.8500E-02 | 8.0926E+00 |
| 5.0000E-04 | 5.0444E-04 | 5.0000E-07 | 4.4394E-06 | 2.2800E+01 | 2.9810E+01 | 2.2800E-02 | 7.0099E+00 |
| 1.8000E-03 | 1.8337E-03 | 1.8000E-06 | 3.3698E-05 | 2.6900E+01 | 2.9299E+01 | 2.6900E-02 | 2.3988E+00 |
| 3.9000E-03 | 4.0320E-03 | 3.9000E-06 | 1.3198E-04 | 3.0800E+01 | 2.8673E+01 | 3.0800E-02 | -2.1272E+00 |
| 6.4000E-03 | 6.6876E-03 | 6.4000E-06 | 2.8761E-04 | 3.3300E+01 | 2.8307E+01 | 3.3300E-02 | -4.9935E+00 |
| Average deviation = -0.0001<br>Root mean square error = 0.0009<br>Average absolute = 0.0004 | | | | | | | |
| 333.15 K | | | | | | | |
| 1.1300E-02 | 1.2136E-02 | 1.1300E-05 | 8.3604E-04 | 8.9000E+00 | 1.2618E+01 | 8.9000E-03 | 3.7184E+00 |
| 7.1000E-03 | 7.5407E-03 | 7.1000E-06 | 4.4067E-04 | 1.4000E+01 | 1.9603E+01 | 1.4000E-02 | 5.6026E+00 |
| 4.9500E-03 | 5.2446E-03 | 4.9500E-06 | 2.9464E-04 | 1.8800E+01 | 2.4887E+01 | 1.8800E-02 | 6.0865E+00 |
| 3.3000E-03 | 3.4499E-03 | 3.3000E-06 | 1.4989E-04 | 2.2900E+01 | 2.8368E+01 | 2.2900E-02 | 5.4681E+00 |
| 1.9500E-03 | 2.0002E-03 | 1.9500E-06 | 5.0165E-05 | 2.6700E+01 | 3.0833E+01 | 2.6700E-02 | 4.1333E+00 |
| 8.0000E-04 | 8.0485E-04 | 8.0000E-07 | 4.8543E-06 | 3.0400E+01 | 3.2684E+01 | 3.0400E-02 | 2.2838E+00 |
| 2.0000E-04 | 1.9908E-04 | 2.0000E-07 | -9.1545E-07 | 3.4000E+01 | 3.3593E+01 | 3.4000E-02 | -4.0705E-01 |
| 1.1500E-03 | 1.1563E-03 | 1.1500E-06 | 6.2667E-06 | 3.7200E+01 | 3.3046E+01 | 3.7200E-02 | -4.1536E+00 |
| 2.6200E-02 | 2.2816E-02 | 2.6200E-05 | -3.3844E-03 | 8.4000E+00 | 1.2475E+01 | 8.4000E-03 | 4.0752E+00 |
| 1.9300E-02 | 1.8127E-02 | 1.9300E-05 | -1.1727E-03 | 1.2000E+01 | 1.8018E+01 | 1.2000E-02 | 6.0183E+00 |
| 1.2900E-02 | 1.3013E-02 | 1.2900E-05 | 1.1324E-04 | 1.6700E+01 | 2.3782E+01 | 1.6700E-02 | 7.0820E+00 |
| 8.5000E-03 | 8.8480E-03 | 8.5000E-06 | 3.4795E-04 | 2.1700E+01 | 2.7618E+01 | 2.1700E-02 | 5.9176E+00 |
| 5.3000E-03 | 5.5284E-03 | 5.3000E-06 | 2.2836E-04 | 2.6000E+01 | 2.9989E+01 | 2.6000E-02 | 3.9887E+00 |
| 2.5000E-03 | 2.5618E-03 | 2.5000E-06 | 6.1780E-05 | 3.0000E+01 | 3.1958E+01 | 3.0000E-02 | 1.9578E+00 |
| 1.0000E-04 | 9.9450E-05 | 1.0000E-07 | -5.4998E-07 | 3.3900E+01 | 3.3685E+01 | 3.3900E-02 | -2.1540E-01 |
| 2.1000E-03 | 2.1329E-03 | 2.1000E-06 | 3.2859E-05 | 3.6600E+01 | 3.2845E+01 | 3.6600E-02 | -3.7548E+00 |
| 5.9400E-02 | 5.0081E-02 | 5.9400E-05 | -9.3188E-03 | 1.1600E+01 | 1.8451E+01 | 1.1600E-02 | 6.8512E+00 |
| 4.4000E-02 | 3.9358E-02 | 4.4000E-05 | -4.6418E-03 | 1.4800E+01 | 2.2387E+01 | 1.4800E-02 | 7.5867E+00 |
| 3.9000E-02 | 3.5836E-02 | 3.9000E-05 | -3.1637E-03 | 1.7100E+01 | 2.4104E+01 | 1.7100E-02 | 7.0037E+00 |
| 2.7800E-02 | 2.6699E-02 | 2.7800E-05 | -1.1013E-03 | 2.1200E+01 | 2.6743E+01 | 2.1200E-02 | 5.5429E+00 |
| 1.7400E-02 | 1.7440E-02 | 1.7400E-05 | 3.9529E-05 | 2.7400E+01 | 2.9242E+01 | 2.7400E-02 | 1.8423E+00 |
| 1.1600E-02 | 1.1866E-02 | 1.1600E-05 | 2.6612E-04 | 3.1800E+01 | 3.0611E+01 | 3.1800E-02 | -1.1893E+00 |
| 7.6000E-03 | 7.8321E-03 | 7.6000E-06 | 2.3211E-04 | 3.5300E+01 | 3.1629E+01 | 3.5300E-02 | -3.6706E+00 |
| 1.8000E-03 | 1.8179E-03 | 1.8000E-06 | 1.7892E-05 | 3.8400E+01 | 3.3365E+01 | 3.8400E-02 | -5.0349E+00 |
| Average deviation = -0.00081<br>Root mean square error = 0.0023<br>Average absolute = 0.0010 | | | | | | | |
| 343.15 K | | | | | | | |
| 1.3850E-02 | 1.4668E-02 | 1.3850E-05 | 8.1795E-04 | 1.2000E+01 | 1.6664E+01 | 1.2000E-02 | 4.6636E+00 |
| 9.7500E-03 | 1.0425E-02 | 9.7500E-06 | 6.7488E-04 | 1.7300E+01 | 2.3262E+01 | 1.7300E-02 | 5.9618E+00 |
| 7.4500E-03 | 7.9804E-03 | 7.4500E-06 | 5.3042E-04 | 2.2100E+01 | 2.7889E+01 | 2.2100E-02 | 5.7889E+00 |
| 5.9000E-03 | 6.2837E-03 | 5.9000E-06 | 3.8367E-04 | 2.6400E+01 | 3.0885E+01 | 2.6400E-02 | 4.4850E+00 |
| 4.3500E-03 | 4.5777E-03 | 4.3500E-06 | 2.2766E-04 | 3.0100E+01 | 3.2882E+01 | 3.0100E-02 | 2.7821E+00 |
| 3.2000E-03 | 3.3204E-03 | 3.2000E-06 | 1.2042E-04 | 3.3900E+01 | 3.4274E+01 | 3.3900E-02 | 3.7447E-01 |
| 2.1500E-03 | 2.1953E-03 | 2.1500E-06 | 4.5261E-05 | 3.7600E+01 | 3.5280E+01 | 3.7600E-02 | -2.3199E+00 |
| 9.5000E-04 | 9.5151E-04 | 9.5000E-07 | 1.5134E-06 | 4.0700E+01 | 3.6163E+01 | 4.0700E-02 | -4.5366E+00 |
| 3.1300E-02 | 2.7760E-02 | 3.1300E-05 | -3.5403E-03 | 1.1500E+01 | 1.6651E+01 | 1.1500E-02 | 5.1507E+00 |
| 2.5200E-02 | 2.3835E-02 | 2.5200E-05 | -1.3647E-03 | 1.5600E+01 | 2.1956E+01 | 1.5600E-02 | 6.3560E+00 |
| 1.8800E-02 | 1.8744E-02 | 1.8800E-05 | -5.5797E-05 | 2.0400E+01 | 2.6730E+01 | 2.0400E-02 | 6.3299E+00 |

**Table S2.**
Continued.

| $X_{[Omim][BF4](exp)}$ | $X_{[Omim][BF4](est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 1.4700E-02 | 1.5062E-02 | 1.4700E-05 | 3.6231E-04 | 2.5600E+01 | 2.9962E+01 | 2.5600E-02 | 4.3625E+00 |
| 1.1500E-02 | 1.1935E-02 | 1.1500E-05 | 4.3547E-04 | 3.0000E+01 | 3.1928E+01 | 3.0000E-02 | 1.9279E+00 |
| 8.4000E-03 | 8.7655E-03 | 8.4000E-06 | 3.6552E-04 | 3.4000E+01 | 3.3379E+01 | 3.4000E-02 | -6.2085E-01 |
| 5.7000E-03 | 5.9248E-03 | 5.7000E-06 | 2.2477E-04 | 3.7900E+01 | 3.4524E+01 | 3.7900E-02 | -3.3758E+00 |
| 3.6000E-03 | 3.6982E-03 | 3.6000E-06 | 9.8232E-05 | 4.0700E+01 | 3.5344E+01 | 4.0700E-02 | -5.3559E+00 |
| 7.1400E-02 | 6.2468E-02 | 7.1400E-05 | -8.9324E-03 | 1.5200E+01 | 2.1783E+01 | 1.5200E-02 | 6.5829E+00 |
| 6.0000E-02 | 5.3911E-02 | 6.0000E-05 | -6.0887E-03 | 1.9500E+01 | 2.5312E+01 | 1.9500E-02 | 5.8122E+00 |
| 5.4400E-02 | 4.9463E-02 | 5.4400E-05 | -4.9375E-03 | 2.1700E+01 | 2.6736E+01 | 2.1700E-02 | 5.0365E+00 |
| 4.4800E-02 | 4.1611E-02 | 4.4800E-05 | -3.1888E-03 | 2.6200E+01 | 2.8981E+01 | 2.6200E-02 | 2.7809E+00 |
| 3.5200E-02 | 3.3473E-02 | 3.5200E-05 | -1.7267E-03 | 3.2700E+01 | 3.1140E+01 | 3.2700E-02 | -1.5601E+00 |
| 2.8200E-02 | 2.7332E-02 | 2.8200E-05 | -8.6836E-04 | 3.6900E+01 | 3.2308E+01 | 3.6900E-02 | -4.5924E+00 |
| 2.3800E-02 | 2.3372E-02 | 2.3800E-05 | -4.2849E-04 | 4.0400E+01 | 3.3071E+01 | 4.0400E-02 | -7.3290E+00 |
| 1.8000E-02 | 1.8009E-02 | 1.8000E-05 | 9.2864E-06 | 4.3600E+01 | 3.3840E+01 | 4.3600E-02 | -9.7602E+00 |
| Average deviation = -0.0011 | | | | | | | |
| Root mean square error = 0.0026 | | | | | | | |
| Average absolute = 0.0014 | | | | | | | |

**Table S3.**
The experimental and estimated MDEA solubility ($X_{MDEA}$) in $CO_2$ + [Omim][BF4]+[MDEA] as a function of pressure.

| $X_{MDEA(exp)}$ | $X_{MDEA(est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 298.15 K | | | | | | | |
| 5.8500E-02 | 7.0077E-02 | 5.8500E-05 | 1.1577E-02 | 2.6000E+00 | 3.5144E+00 | 2.6000E-03 | 9.1440E-01 |
| 9.5000E-03 | 1.2103E-02 | 9.5000E-06 | 2.6030E-03 | 6.6000E+00 | 1.6231E+01 | 6.6000E-03 | 9.6309E+00 |
| 1.0000E-02 | 1.1133E-02 | 1.0000E-05 | 1.1333E-03 | 1.1200E+01 | 1.8637E+01 | 1.1200E-02 | 7.4368E+00 |
| 2.2500E-02 | 2.2833E-02 | 2.2500E-05 | 3.3291E-04 | 1.5200E+01 | 1.7619E+01 | 1.5200E-02 | 2.4187E+00 |
| 3.2000E-02 | 2.8818E-02 | 3.2000E-05 | -3.1817E-03 | 1.8900E+01 | 1.8162E+01 | 1.8900E-02 | -7.3848E-01 |
| 4.2500E-02 | 3.4663E-02 | 4.2500E-05 | -7.8369E-03 | 2.2200E+01 | 1.8918E+01 | 2.2200E-02 | -3.2821E+00 |
| 5.1000E-02 | 3.8838E-02 | 5.1000E-05 | -1.2162E-02 | 2.5400E+01 | 1.9901E+01 | 2.5400E-02 | -5.4988E+00 |
| 6.1500E-02 | 4.4371E-02 | 6.1500E-05 | -1.7129E-02 | 2.8000E+01 | 2.0820E+01 | 2.8000E-02 | -7.1802E+00 |
| 1.5000E-02 | 1.5654E-02 | 1.5000E-05 | 6.5355E-04 | 2.0000E+00 | 1.7077E+01 | 2.0000E-03 | 1.5077E+01 |
| 5.7000E-03 | 5.7651E-03 | 5.7000E-06 | 6.5136E-05 | 4.2000E+00 | 1.9802E+01 | 4.2000E-03 | 1.5602E+01 |
| 5.0000E-04 | 5.0485E-04 | 5.0000E-07 | 4.8517E-06 | 8.6000E+00 | 2.1055E+01 | 8.6000E-03 | 1.2455E+01 |
| 4.3000E-03 | 4.2206E-03 | 4.3000E-06 | -7.9437E-05 | 1.3400E+01 | 2.2681E+01 | 1.3400E-02 | 9.2805E+00 |
| 6.9000E-03 | 6.6906E-03 | 6.9000E-06 | -2.0943E-04 | 1.7500E+01 | 2.3229E+01 | 1.7500E-02 | 5.7291E+00 |
| 9.0000E-03 | 8.6550E-03 | 9.0000E-06 | -3.4500E-04 | 2.1300E+01 | 2.3576E+01 | 2.1300E-02 | 2.2759E+00 |
| 1.1400E-02 | 1.0889E-02 | 1.1400E-05 | -5.1140E-04 | 2.4700E+01 | 2.3753E+01 | 2.4700E-02 | -9.4669E-01 |
| 1.3600E-02 | 1.2930E-02 | 1.3600E-05 | -6.7019E-04 | 2.7000E+01 | 2.3780E+01 | 2.7000E-02 | -3.2199E+00 |
| 2.7800E-02 | 3.2399E-02 | 2.7800E-05 | 4.5992E-03 | 2.8000E+00 | 1.7183E+01 | 2.8000E-03 | 1.4383E+01 |
| 1.0400E-02 | 1.0965E-02 | 1.0400E-05 | 5.6518E-04 | 5.3000E+00 | 1.9676E+01 | 5.3000E-03 | 1.4376E+01 |
| 3.0000E-03 | 3.0739E-03 | 3.0000E-06 | 7.3932E-05 | 6.9000E+00 | 2.0453E+01 | 6.9000E-03 | 1.3553E+01 |
| 5.4000E-03 | 5.4337E-03 | 5.4000E-06 | 3.3728E-05 | 1.1300E+01 | 2.1691E+01 | 1.1300E-02 | 1.0391E+01 |
| 1.5800E-02 | 1.5568E-02 | 1.5800E-05 | -2.3199E-04 | 1.7000E+01 | 2.2253E+01 | 1.7000E-02 | 5.2527E+00 |
| 2.1200E-02 | 2.0624E-02 | 2.1200E-05 | -5.7611E-04 | 2.1000E+01 | 2.2500E+01 | 2.1000E-02 | 1.5004E+00 |
| 2.6200E-02 | 2.5262E-02 | 2.6200E-05 | -9.3789E-04 | 2.3900E+01 | 2.2560E+01 | 2.3900E-02 | -1.3396E+00 |
| 3.1200E-02 | 2.9815E-02 | 3.1200E-05 | -1.3851E-03 | 2.6800E+01 | 2.2582E+01 | 2.6800E-02 | -4.2179E+00 |
| Average deviation = -0.00098 | | | | | | | |
| Root mean square error = 0.0053 | | | | | | | |
| Average absolute = 0.0027 | | | | | | | |
| 313.15 K | | | | | | | |
| 7.8500E-02 | 9.3590E-02 | 7.8500E-05 | 1.5090E-02 | 4.8000E+00 | 6.7377E+00 | 4.8000E-03 | 1.9377E+00 |
| 2.9500E-02 | 3.9933E-02 | 2.9500E-05 | 1.0433E-02 | 9.0000E+00 | 1.4977E+01 | 9.0000E-03 | 5.9768E+00 |
| 8.5000E-03 | 9.1926E-03 | 8.5000E-06 | 6.9258E-04 | 1.3600E+01 | 2.4097E+01 | 1.3600E-02 | 1.0497E+01 |
| 4.5000E-03 | 4.6365E-03 | 4.5000E-06 | 1.3649E-04 | 1.7800E+01 | 2.6011E+01 | 1.7800E-02 | 8.2114E+00 |
| 1.5500E-02 | 1.5571E-02 | 1.5500E-05 | 7.1467E-05 | 2.1600E+01 | 2.4668E+01 | 2.1600E-02 | 3.0677E+00 |
| 2.6000E-02 | 2.5600E-02 | 2.6000E-05 | -4.0014E-04 | 2.5100E+01 | 2.7681E+01 | 2.5100E-02 | 2.5814E+00 |
| 3.4000E-02 | 2.9563E-02 | 3.4000E-05 | -4.4368E-03 | 2.8600E+01 | 2.4779E+01 | 2.8600E-02 | -3.8207E+00 |
| 4.3500E-02 | 3.5621E-02 | 4.3500E-05 | -7.8789E-03 | 3.1500E+01 | 2.5416E+01 | 3.1500E-02 | -6.0841E+00 |
| 9.4000E-02 | 1.4769E-01 | 9.4000E-05 | 5.3692E-02 | 4.1000E+00 | 6.0814E+00 | 4.1000E-03 | 1.9814E+00 |
| 5.0000E-02 | 7.8793E-02 | 5.0000E-05 | 2.8793E-02 | 6.6000E+00 | 1.1042E+01 | 6.6000E-03 | 4.4417E+00 |
| 1.8000E-02 | 2.1378E-02 | 1.8000E-05 | 3.3777E-03 | 1.1100E+01 | 2.1164E+01 | 1.1100E-02 | 1.0064E+01 |
| 1.5000E-03 | 1.5311E-03 | 1.5000E-06 | 3.1148E-05 | 1.6100E+01 | 2.6316E+01 | 1.6100E-02 | 1.0216E+01 |
| 1.4500E-02 | 1.4797E-02 | 1.4500E-05 | 2.9669E-04 | 2.0400E+01 | 2.4742E+01 | 2.0400E-02 | 4.3424E+00 |
| 2.6500E-02 | 2.5588E-02 | 2.6500E-05 | -9.1167E-04 | 2.4300E+01 | 2.4297E+01 | 2.4300E-02 | -2.8355E-03 |
| 3.7000E-02 | 3.3830E-02 | 3.7000E-05 | -3.1699E-03 | 2.8100E+01 | 2.4506E+01 | 2.8100E-02 | -3.5942E+00 |
| 4.7500E-02 | 4.1835E-02 | 4.7500E-05 | -5.6655E-03 | 3.0600E+01 | 2.4676E+01 | 3.0600E-02 | -5.9240E+00 |

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**Table S3.**
Continued.

| $X_{MDEA(exp)}$ | $X_{MDEA(est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 9.3500E-02 | 1.7147E-01 | 9.3500E-05 | 7.7968E-02 | 5.4000E+00 | 9.6802E+00 | 5.4000E-03 | 4.2802E+00 |
| 5.2500E-02 | 7.5863E-02 | 5.2500E-05 | 2.3363E-02 | 8.2000E+00 | 1.6483E+01 | 8.2000E-03 | 8.2832E+00 |
| 3.7000E-02 | 4.6292E-02 | 3.7000E-05 | 9.2923E-03 | 1.0200E+01 | 1.9643E+01 | 1.0200E-02 | 9.4431E+00 |
| 1.2000E-02 | 1.2786E-02 | 1.2000E-05 | 7.8608E-04 | 1.4400E+01 | 2.4057E+01 | 1.4400E-02 | 9.6572E+00 |
| 1.3000E-02 | 1.3237E-02 | 1.3000E-05 | 2.3743E-04 | 2.0400E+01 | 2.5114E+01 | 2.0400E-02 | 4.7143E+00 |
| 2.6000E-02 | 2.5554E-02 | 2.6000E-05 | -4.4605E-04 | 4.8000E+00 | 6.7377E+00 | 4.8000E-03 | 1.9377E+00 |
| 3.6000E-02 | 3.4364E-02 | 3.6000E-05 | -1.6365E-03 | 9.0000E+00 | 1.4977E+01 | 9.0000E-03 | 5.9768E+00 |
| 4.6500E-02 | 4.3205E-02 | 4.6500E-05 | -3.2953E-03 | 1.3600E+01 | 2.4097E+01 | 1.3600E-02 | 1.0497E+01 |
| Average deviation = 0.0081<br>Root mean square error = 0.021<br>Average absolute = 0.010 | | | | | | | |
| 323.15 K | | | | | | | |
| 9.3000E-02 | 1.1167E-01 | 9.3000E-05 | 1.8673E-02 | 6.5000E+00 | 9.2578E+00 | 6.5000E-03 | 2.7578E+00 |
| 4.7000E-02 | 5.7426E-02 | 4.7000E-05 | 1.0426E-02 | 1.1100E+01 | 1.6639E+01 | 1.1100E-02 | 5.5387E+00 |
| 2.6500E-02 | 2.9319E-02 | 2.6500E-05 | 2.8186E-03 | 1.5900E+01 | 2.3525E+01 | 1.5900E-02 | 7.6252E+00 |
| 1.1000E-02 | 1.1421E-02 | 1.1000E-05 | 4.2083E-04 | 2.0000E+01 | 2.7933E+01 | 2.0000E-02 | 7.9330E+00 |
| 1.1000E-02 | 1.0943E-02 | 1.1000E-05 | -5.7023E-05 | 2.7500E+01 | 2.9200E+01 | 2.7500E-02 | 1.7001E+00 |
| 1.9500E-02 | 1.8426E-02 | 1.9500E-05 | -1.0738E-03 | 3.1100E+01 | 2.8836E+01 | 3.1100E-02 | -2.2639E+00 |
| 3.0000E-02 | 2.6570E-02 | 3.0000E-05 | -3.4298E-03 | 3.4100E+01 | 2.8855E+01 | 3.4100E-02 | -5.2446E+00 |
| 1.1150E-01 | 1.7209E-01 | 1.1150E-04 | 6.0590E-02 | 6.1000E+00 | 9.0982E+00 | 6.1000E-03 | 2.9982E+00 |
| 7.0000E-02 | 1.0079E-01 | 7.0000E-05 | 3.0791E-02 | 8.9000E+00 | 1.4248E+01 | 8.9000E-03 | 5.3483E+00 |
| 3.8000E-02 | 4.5341E-02 | 3.8000E-05 | 7.3405E-03 | 1.3500E+01 | 2.1732E+01 | 1.3500E-02 | 8.2315E+00 |
| 1.7500E-02 | 1.8489E-02 | 1.7500E-05 | 9.8882E-04 | 1.8500E+01 | 2.6593E+01 | 1.8500E-02 | 8.0926E+00 |
| 2.5000E-03 | 2.5389E-03 | 2.5000E-06 | 3.8860E-05 | 2.2800E+01 | 2.9810E+01 | 2.2800E-02 | 7.0099E+00 |
| 9.0000E-03 | 9.0234E-03 | 9.0000E-06 | 2.3446E-05 | 2.6900E+01 | 2.9299E+01 | 2.6900E-02 | 2.3988E+00 |
| 1.9500E-02 | 1.8748E-02 | 1.9500E-05 | -7.5207E-04 | 3.0800E+01 | 2.8673E+01 | 3.0800E-02 | -2.1272E+00 |
| 3.2000E-02 | 2.9465E-02 | 3.2000E-05 | -2.5351E-03 | 3.3300E+01 | 2.8307E+01 | 3.3300E-02 | -4.9935E+00 |
| Average deviation = 0.0082<br>Root mean square error = 0.018<br>Average absolute = 0.0093 | | | | | | | |
| 333.15 K | | | | | | | |
| 1.1300E-01 | 1.3602E-01 | 1.1300E-04 | 2.3022E-02 | 8.9000E+00 | 1.2618E+01 | 8.9000E-03 | 3.7184E+00 |
| 7.1000E-02 | 8.0091E-02 | 7.1000E-05 | 9.0910E-03 | 1.4000E+01 | 1.9603E+01 | 1.4000E-02 | 5.6026E+00 |
| 4.9500E-02 | 5.1771E-02 | 4.9500E-05 | 2.2712E-03 | 1.8800E+01 | 2.4887E+01 | 1.8800E-02 | 6.0865E+00 |
| 3.3000E-02 | 3.3147E-02 | 3.3000E-05 | 1.4749E-04 | 2.2900E+01 | 2.8368E+01 | 2.2900E-02 | 5.4681E+00 |
| 1.9500E-02 | 1.9360E-02 | 1.9500E-05 | -1.3995E-04 | 2.6700E+01 | 3.0833E+01 | 2.6700E-02 | 4.1333E+00 |
| 8.0000E-03 | 8.0080E-03 | 8.0000E-06 | 8.0123E-06 | 3.0400E+01 | 3.2684E+01 | 3.0400E-02 | 2.2838E+00 |
| 2.0000E-03 | 2.0161E-03 | 2.0000E-06 | 1.6073E-05 | 3.4000E+01 | 3.3593E+01 | 3.4000E-02 | -4.0705E-01 |
| 1.1500E-02 | 1.1168E-02 | 1.1500E-05 | -3.3249E-04 | 3.7200E+01 | 3.3046E+01 | 3.7200E-02 | -4.1536E+00 |
| 1.3100E-01 | 1.9824E-01 | 1.3100E-04 | 6.7236E-02 | 8.4000E+00 | 1.2475E+01 | 8.4000E-03 | 4.0752E+00 |
| 9.6500E-02 | 1.2943E-01 | 9.6500E-05 | 3.2932E-02 | 1.2000E+01 | 1.8018E+01 | 1.2000E-02 | 6.0183E+00 |
| 6.4500E-02 | 7.4499E-02 | 6.4500E-05 | 9.9988E-03 | 1.6700E+01 | 2.3782E+01 | 1.6700E-02 | 7.0820E+00 |
| 4.2500E-02 | 4.4391E-02 | 4.2500E-05 | 1.8910E-03 | 2.1700E+01 | 2.7618E+01 | 2.1700E-02 | 5.9176E+00 |
| 2.6500E-02 | 2.6523E-02 | 2.6500E-05 | 2.3363E-05 | 2.6000E+01 | 2.9989E+01 | 2.6000E-02 | 3.9887E+00 |
| 1.2500E-02 | 1.2432E-02 | 1.2500E-05 | -6.7594E-05 | 3.0000E+01 | 3.1958E+01 | 3.0000E-02 | 1.9578E+00 |
| 5.0000E-04 | 5.0509E-04 | 5.0000E-07 | 5.0880E-06 | 3.3900E+01 | 3.3685E+01 | 3.3900E-02 | -2.1540E-01 |
| 1.0500E-02 | 1.0291E-02 | 1.0500E-05 | -2.0856E-04 | 3.6600E+01 | 3.2845E+01 | 3.6600E-02 | -3.7548E+00 |
| 1.4850E-01 | 2.1097E-01 | 1.4850E-04 | 6.2472E-02 | 1.1600E+01 | 1.8451E+01 | 1.1600E-02 | 6.8512E+00 |
| 1.1000E-01 | 1.3805E-01 | 1.1000E-04 | 2.8048E-02 | 1.4800E+01 | 2.2387E+01 | 1.4800E-02 | 7.5867E+00 |
| 9.7500E-02 | 1.1482E-01 | 9.7500E-05 | 1.7324E-02 | 1.7100E+01 | 2.4104E+01 | 1.7100E-02 | 7.0037E+00 |
| 6.9500E-02 | 7.5161E-02 | 6.9500E-05 | 5.6606E-03 | 2.1200E+01 | 2.6743E+01 | 2.1200E-02 | 5.5429E+00 |
| 4.3500E-02 | 4.3762E-02 | 4.3500E-05 | 2.6156E-04 | 2.7400E+01 | 2.9242E+01 | 2.7400E-02 | 1.8423E+00 |
| 2.9000E-02 | 2.8427E-02 | 2.9000E-05 | -5.7312E-04 | 3.1800E+01 | 3.0611E+01 | 3.1800E-02 | -1.1893E+00 |
| 1.9000E-02 | 1.8495E-02 | 1.9000E-05 | -5.0489E-04 | 3.5300E+01 | 3.1629E+01 | 3.5300E-02 | -3.6706E+00 |
| 4.5000E-03 | 4.4850E-03 | 4.5000E-06 | -1.4989E-05 | 3.8400E+01 | 3.3365E+01 | 3.8400E-02 | -5.0349E+00 |
| Average deviation = 0.010<br>Root mean square error = 0.021<br>Average absolute = 0.010 | | | | | | | |
| 343.15 K | | | | | | | |
| 1.3850E-01 | 1.6990E-01 | 1.3850E-04 | 3.1397E-02 | 1.2000E+01 | 1.6664E+01 | 1.2000E-02 | 4.6636E+00 |
| 9.7500E-02 | 1.0798E-01 | 9.7500E-05 | 1.0475E-02 | 1.7300E+01 | 2.3262E+01 | 1.7300E-02 | 5.9618E+00 |
| 7.4500E-02 | 7.6143E-02 | 7.4500E-05 | 1.6432E-03 | 2.2100E+01 | 2.7889E+01 | 2.2100E-02 | 5.7889E+00 |
| 5.9000E-02 | 5.7189E-02 | 5.9000E-05 | -1.8115E-03 | 2.6400E+01 | 3.0885E+01 | 2.6400E-02 | 4.4850E+00 |
| 4.3500E-02 | 4.1120E-02 | 4.3500E-05 | -2.3803E-03 | 3.0100E+01 | 3.2882E+01 | 3.0100E-02 | 2.7821E+00 |
| 3.2000E-02 | 2.9965E-02 | 3.2000E-05 | -2.0350E-03 | 3.3900E+01 | 3.4274E+01 | 3.3900E-02 | 3.7447E-01 |
| 2.1500E-02 | 2.0325E-02 | 2.1500E-05 | -1.1750E-03 | 3.7600E+01 | 3.5280E+01 | 3.7600E-02 | -2.3199E+00 |
| 9.5000E-03 | 9.3168E-03 | 9.5000E-06 | -1.8319E-04 | 4.0700E+01 | 3.6163E+01 | 4.0700E-02 | -4.5366E+00 |
| 1.5650E-01 | 2.2946E-01 | 1.5650E-04 | 7.2964E-02 | 1.1500E+01 | 1.6651E+01 | 1.1500E-02 | 5.1507E+00 |

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**Table S3.**
Continued.

| $X_{MDEA(exp)}$ | $X_{MDEA(est)}$ | Standard deviation | Difference | $P_{exp}$ | $P_{est}$ | Standard deviation | Difference |
|---|---|---|---|---|---|---|---|
| 1.2600E-01 | 1.6270E-01 | 1.2600E-04 | 3.6700E-02 | 1.5600E+01 | 2.1956E+01 | 1.5600E-02 | 6.3560E+00 |
| 9.4000E-02 | 1.0704E-01 | 9.4000E-05 | 1.3043E-02 | 2.0400E+01 | 2.6730E+01 | 2.0400E-02 | 6.3299E+00 |
| 7.3500E-02 | 7.6435E-02 | 7.3500E-05 | 2.9355E-03 | 2.5600E+01 | 2.9962E+01 | 2.5600E-02 | 4.3625E+00 |
| 5.7500E-02 | 5.6789E-02 | 5.7500E-05 | -7.1117E-04 | 3.0000E+01 | 3.1928E+01 | 3.0000E-02 | 1.9279E+00 |
| 4.2000E-02 | 4.0282E-02 | 4.2000E-05 | -1.7177E-03 | 3.4000E+01 | 3.3379E+01 | 3.4000E-02 | -6.2085E-01 |
| 2.8500E-02 | 2.7063E-02 | 2.8500E-05 | -1.4368E-03 | 3.7900E+01 | 3.4524E+01 | 3.7900E-02 | -3.3758E+00 |
| 1.8000E-02 | 1.7262E-02 | 1.8000E-05 | -7.3785E-04 | 4.0700E+01 | 3.5344E+01 | 4.0700E-02 | -5.3559E+00 |
| 1.7850E-01 | 2.3698E-01 | 1.7850E-04 | 5.8483E-02 | 1.5200E+01 | 2.1783E+01 | 1.5200E-02 | 6.5829E+00 |
| 1.5000E-01 | 1.7908E-01 | 1.5000E-04 | 2.9080E-02 | 1.9500E+01 | 2.5312E+01 | 1.9500E-02 | 5.8122E+00 |
| 1.3600E-01 | 1.5580E-01 | 1.3600E-04 | 1.9797E-02 | 2.1700E+01 | 2.6736E+01 | 2.1700E-02 | 5.0365E+00 |
| 1.1200E-01 | 1.2028E-01 | 1.1200E-04 | 8.2848E-03 | 2.6200E+01 | 2.8981E+01 | 2.6200E-02 | 2.7809E+00 |
| 8.8000E-02 | 8.8869E-02 | 8.8000E-05 | 8.6879E-04 | 3.2700E+01 | 3.1140E+01 | 3.2700E-02 | -1.5601E+00 |
| 7.0500E-02 | 6.9180E-02 | 7.0500E-05 | -1.3197E-03 | 3.6900E+01 | 3.2308E+01 | 3.6900E-02 | -4.5924E+00 |
| 5.9500E-02 | 5.7341E-02 | 5.9500E-05 | -2.1592E-03 | 4.0400E+01 | 3.3071E+01 | 4.0400E-02 | -7.3290E+00 |
| 4.5000E-02 | 4.2826E-02 | 4.5000E-05 | -2.1742E-03 | 4.3600E+01 | 3.3840E+01 | 4.3600E-02 | -9.7602E+00 |

Average deviation = 0.011
Root mean square error = 0.023
Average absolute = 0.0126

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