Solubility of CO$_2$ in Ionic Liquids with Additional Water and Methanol: Modeling with PC-SAFT Equation of State

Xinyu Liao, Ke Zheng,* Gang Wang, Yong Yang, Yongwang Li, and Marc-Olivier Coppens*

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1. INTRODUCTION

Increasing concerns of global warming resulting from greenhouse gas emissions have stimulated the development of advanced technologies to mitigate carbon dioxide (CO$_2$) emissions.$^{1,2}$ Carbon capture and storage (CCS), which emphasizes the development of CO$_2$ capture methods and sequestration technologies, are primarily used to reduce CO$_2$ emissions from the use of fossil fuels. CCS involve CO$_2$ separation, capture, transportation, and storage in which CO$_2$ emissions from the use of fossil fuels. CCS involve CO$_2$ separation, capture, transportation, and storage in which CO$_2$ capture processes are the most important step.$^3$

The technologies for capture and separation of CO$_2$ include absorption, adsorption, membrane filtration, cryogenic separation, and microbial-based processes.$^4$ Among these technologies, absorption is the most commonly employed technology for CO$_2$ capture in large-scale applications. According to the type of solvent, absorption processes can be divided into two subcategories, including chemical absorption and physical absorption. Aqueous amines, due to their higher purification of the gases and a higher CO$_2$ absorption capacity, are widely used as the absorbent in carbon capture operations and commercialized for many decades.$^{4-6}$ Though there are many advantages, the inherent properties of amines also lead to some drawbacks, including low CO$_2$ loading capacity, high corrosion rate, high regeneration cost, and large equipment size.$^5$ The physical absorption process is superior to chemical absorption when the concentration of CO$_2$ is high due to their low regeneration cost and product loss.$^{5,7,8}$ However, due to the high vapor pressure and high volatility of the physical solvent, specifically for the Rectisol process with methanol as a solvent, the absorption process must be operated at a low temperature and high pressure to avoid significant solvent loss.$^{9,10}$ To be more cost-effective for large-scale industrial applications, much research has been devoted to finding or designing new solvents for CO$_2$ capture. Ionic liquids (ILs) have gained a lot of attention as potential alternatives to replace amine-based chemical absorbents and conventional physical absorbents in CO$_2$ absorption.$^{11-15}$ The exceptional physical and chemical properties, including high CO$_2$ solubility, high thermal stability, non-flammability, negligible volatility, tunability, and recyclability make ILs ideal solvents in CO$_2$ capture processes, because they are environmentally friendlier and cheaper for solvent regeneration. Furthermore, the tunability of ILs provides an...
extra degree of freedom for specific modifications to improve the CO$_2$ capacity and selectivity.\textsuperscript{14} On the other hand, the much higher viscosity of ILs than those of conventional solvents is one of the main limitations of their industrial application due to higher pumping and related operating costs. Furthermore, the high viscosity of ILs will significantly inhibit the mass transfer of CO$_2$ in ILs, resulting in a low absorption rate.\textsuperscript{16} To reduce the viscosity of ILs, a third solvent can be introduced as a co-solvent. Water and several organic solvents were found to reduce the viscosity of pure ILs sharply by adding them in small amounts as co-solvents.\textsuperscript{57–22}

The measurement and simulation of CO$_2$ solubility in pure ILs were systematically investigated by an enormous amount of research studies.\textsuperscript{23–30} The perturbed-chain statistical associating fluid theory (PC-SAFT) equation of state (EoS) derived using Wertheim’s first-order thermodynamic perturbation theory\textsuperscript{31–34} was recommended to model IL-containing systems due to their physical background.\textsuperscript{35–37} Baramaki et al. employed PC-SAFT to model CO$_2$ solubilities in 16 ILs using a temperature-independent interaction parameter, and excellent results were obtained.\textsuperscript{39} Solubility of acid gases (CO$_2$ and H$_2$S) in [C$_n$ mim][NTf$_2$] (where $n = 2, 4, 6$, and 8) ILs was modeled by Al-fnaish and Lue with PC-SAFT using two strategies and four different self-association schemes.\textsuperscript{30} They found that the four-site association scheme provided the best results for almost all of the investigated acid gases–IL binary systems and successfully represented solubility of CO$_2$ and H$_2$S in [C$_n$ mim][NTf$_2$]. Bülow et al. incorporated a concentration-dependent dielectric constant into electrolyte PC-SAFT to model liquid–liquid equilibria of binary mixtures containing water and commonly used hydrophobic ILs.\textsuperscript{37} This new approach was superior, with better correlation capability, compared to the original model whose dielectric constant was independent with concentration. Compared to popular studies on pure ILs, the effects of water and methanol as co-solvents on the solubility of CO$_2$ in ILs have been experimentally measured and theoretically modeled only by few researchers.\textsuperscript{38–44} Valuable experimental data and suitable thermodynamic models for selected systems can be obtained from these works; however, there is no thermodynamic model as yet that can predict the solubility of CO$_2$ in mixtures of ILs and co-solvents using only binary data for CO$_2$/ILs, CO$_2$/co-solvents, and ILs/co-solvents, which are more easily measured than data for ternary systems. The goal of this work is to develop a reliable thermodynamic model based on PC-SAFT that can predict the solubility of CO$_2$ in multi-component solvents containing ILs and water or methanol from a single set of binary interaction parameters only fitted to binary phase equilibria data. Due to the limitations of available experimental data for ternary mixtures and their binary sub-systems, the ILs under investigation contain only few common IL cations of [C$_n$ mim]$^+$ (where $n = 2, 4, 6$, and 8) and [Amin]$^+$ and IL anions of [PF$_6$]$^-$, [OTf]$^-$, and [TF$_2$N]$^-$. However, this is enough to validate the model.

2. THEORY

2.1. PC-SAFT and Modeling Strategies. The original PC-SAFT EoS developed by Gross and Sadowski\textsuperscript{35,46} and its extended version to electrolytes, referred to as electrolyte PC-SAFT (ePC-SAFT),\textsuperscript{37,47,48} were used to model thermodynamic properties for systems containing ILs. The expression of the model in terms of the residual Helmholtz free energy $a^{\text{res}}$ is as follows

$$a^{\text{res}} = a^{hc} + a^{\text{disp}} + a^{\text{assoc}} + a^{\text{ion}}$$

where $a^{hc}$ is the reference hard-chain contribution, $a^{\text{disp}}$ denotes the dispersion contribution, $a^{\text{assoc}}$ represents the association contribution referring to site–site association interactions, and $a^{\text{ion}}$ represents the electrolyte contribution caused by ion–ion interactions. Three pure-component parameters are used in PC-SAFT for non-associating components, including segment number ($m$), segment diameter ($\sigma$), and dispersion energy ($u/k$).

Two more associating parameters, association energy $\kappa^{\text{assoc}}$ and association volume $\kappa^{\text{vol}}$, are introduced to describe hydrogen-bonding interactions between associating molecules.

For mixtures, Berthelot–Lorentz combining rules are used

$$u_{ij} = \sqrt{u_iu_j(1 - k_{ij})}$$

$$\sigma_{ij} = \frac{1}{2}(\sigma_i + \sigma_j)(1 - l_{ij})$$

where $k_{ij}$ and $l_{ij}$ are binary interaction parameters (BIPs) to correct interactions of unlike molecules. An asymmetric BIP, $p_{ij}$, which had shown significant improvement to model correlations and predictions in previous works,\textsuperscript{49,50} was also introduced in this work.

Besides, $k_{ij}$ is defined as a temperature-dependent parameter

$$k_{ij} = a_0 + \frac{a_1T}{1000}$$

where $a_0$ are constants and $T$ is the temperature in kelvin.
Pure-component parameters for CO$_2$, water, and methanol are given in Table 1. The two-site association scheme was employed to model water and methanol. In this work, two strategies were adopted to model ILs. In the first strategy (i.e., original PC-SAFT, strategy 1), ILs were also considered as self-associating chain molecules with two association sites, and the association interaction was allowed between two different sites of two IL molecules or an IL with another association molecule. Pure-component parameters for ILs used in this work are summarized in Table 2. Parameters for [Amim][TF$_2$N] were regressed by fitting to liquid density data with 0.14% absolute average deviation (AAD) between theoretical and experimental values. The cross-association parameters can be determined from pure-component association parameters using combining rules

$$
\varepsilon_{A,B} = \frac{1}{2}(\varepsilon_{A} + \varepsilon_{B})
$$

$$
\kappa_{A,B} = \left[ \varepsilon_{A,B}^{1/2} \frac{2}{\varepsilon_0 \varepsilon_{A}^{1/2} \sigma_{A}} \right]^{3/2}
$$

In contrast, in the second strategy (i.e., ePC-SAFT, strategy 2), the ILs were treated as strong electrolytes that completely dissociate into cations and anions. IL ions were characterized by three adjustable pure-component parameters (\(m_i\), \(\sigma_i\), \(u_i/k\)) and their electric charge with parameter values summarized in Table 3. The AAD for [Amim][TF$_2$N] is 0.17% in this strategy. A concentration-dependent dielectric constant \(\varepsilon(x)\) was introduced to account for the influence of concentration on the dielectric constant in mixtures

$$
\varepsilon = \sum_i \varepsilon_i x_i
$$

The corresponding \(d_{i}^{\text{ion}}\) is shown as follows

$$
d_{i}^{\text{ion}} = -\frac{\kappa(x_i)}{12\pi k_B T} \sum_j q_j^i \chi_j \varepsilon_i(x)_j
$$

where \(x_i\) and \(q_i\) are the mole fraction and the charge for ion \(i\), respectively. The Debye screening length \(\kappa\) and the quantity \(\chi_i\) are expressed by

$$
\kappa = \frac{\rho_N}{k_B T \varepsilon_i(x)} \sum_i q_i^i \chi_i x_i
$$

$$
\chi_i = \frac{3}{(\kappa(x_i) \sigma_i)^3} \left[ \frac{3}{2} + \ln(1 + \kappa(x_i) \sigma_i) - 2\kappa(x_i) \sigma_i \right] + \frac{1}{2} \sigma_i^2
$$

Here, \(\rho_N\) is the number density of the system.

The derivative of the dielectric constant with respect to mole fraction, which is needed in the calculations of the chemical potential, is simplified to

$$
\frac{d\varepsilon}{dx_i} = \varepsilon_i \varepsilon_{r,i}
$$

where \(\varepsilon_0\) is the permittivity in vacuo and \(\varepsilon_{r,i}\) is the relative permittivity of the pure component. The relative permittivity for the investigated ILs was set to 1, according to Bålke et al., those for water and methanol were obtained from the literature, as shown in eqs 12 and 13 and for CO$_2$, it was set to unity.

$$
\varepsilon_{r,\text{water}} = 7.655618295 \times 10^{-1} T - 0.81783881423 T + 254.19616803
$$

$$
\varepsilon_{r,\text{methanol}} = -53.398 \ln(T) + 336.170
$$

---

**Table 3. Pure Parameters for IL Ions in Strategy 2**

<table>
<thead>
<tr>
<th>IL</th>
<th>(M_i) (g/mol)</th>
<th>(m)</th>
<th>(\sigma) (Å)</th>
<th>(u_i/k) (K)</th>
<th>refs</th>
</tr>
</thead>
<tbody>
<tr>
<td>[C$_2$mim]$^+$</td>
<td>111.168</td>
<td>1.4872</td>
<td>3.5926</td>
<td>206.4924</td>
<td>37</td>
</tr>
<tr>
<td>[C$_3$mim]$^+$</td>
<td>139.221</td>
<td>2.4605</td>
<td>3.6371</td>
<td>218.1441</td>
<td>37</td>
</tr>
<tr>
<td>[C$_4$mim]$^+$</td>
<td>167.275</td>
<td>3.4131</td>
<td>3.6781</td>
<td>230.0000</td>
<td>37</td>
</tr>
<tr>
<td>[C$_5$mim]$^+$</td>
<td>195.328</td>
<td>4.2977</td>
<td>3.7187</td>
<td>242.0000</td>
<td>37</td>
</tr>
<tr>
<td>[Amim]$^+$</td>
<td>123.155</td>
<td>1.7970</td>
<td>3.6245</td>
<td>226.9787</td>
<td>this work</td>
</tr>
<tr>
<td>[PF$_6$]$^-\text{[Amim]}$</td>
<td>144.973</td>
<td>4.2791</td>
<td>3.5889</td>
<td>492.2835</td>
<td>37</td>
</tr>
<tr>
<td>[OTf]$^-\text{[Amim]}$</td>
<td>149.070</td>
<td>3.7432</td>
<td>3.8771</td>
<td>519.3113</td>
<td>37</td>
</tr>
<tr>
<td>[TF$_2$N]$^-\text{[Amim]}$</td>
<td>218.145</td>
<td>6.0103</td>
<td>3.7469</td>
<td>375.6529</td>
<td>37</td>
</tr>
</tbody>
</table>

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**Figure 1.** Solubilities of CO$_2$ in (a) water (black for 323.2 K, red for 333.2 K, and blue for 353.1 K) and (b) methanol (black for 228.2 K, red for 243.2 K, blue for 258.2 K, and magenta for 273.2 K). Comparisons of PC-SAFT (lines) to experimental data (symbols).
2.2. Phase Equilibria. To calculate the solubility of CO$_2$ in water or methanol, equality of the fugacity for each component in the liquid and vapor phase should be achieved

$$x_i \phi_i^L(T, P, x_i) = \gamma_i \phi_i^V(T, P, \gamma_i)$$  \hspace{1cm} (14)

where $\phi_i^L$ and $\phi_i^V$ refer to the fugacity coefficients of component $i$ in either vapor or liquid phase and $x_i$ and $\gamma_i$ are mole fractions of component $i$ in liquid and vapor phases, respectively.

As ILs are nearly nonvolatile, eq 14 for vapor–liquid equilibria (VLE) involving ILs can be reduced to

$$x_i \phi_i^L(T, P, x_i) = \phi_i^V(T, P)$$  \hspace{1cm} (15)

For the calculation of liquid–liquid equilibria (LLE) for binary mixtures of water and ILs, the phase equilibrium equations become

$$x_i^{1L} \phi_i^{1L}(T, P, x_i^{1L}) = x_i^{2L} \phi_i^{2L}(T, P, x_i^{2L})$$  \hspace{1cm} (16)

where $\phi_i^{1L}$ and $\phi_i^{2L}$ denote the fugacity coefficients of ILs or water in liquid phases 1 and 2. $x_i^{1L}$ and $x_i^{2L}$ are the mole fractions of component $i$ in each liquid phase.

3. RESULTS AND DISCUSSION

The predictive capability is the most significant measure for the quality of a thermodynamic model. In this work, several ternary systems containing CO$_2$, ILs, and two important co-solvents (i.e., water and methanol) were selected to validate the predictive capability of PC-SAFT in IL-containing systems. BIPs were acquired by only fitting to binary VLE data or LLE data, and experimental data for ternary mixtures were never used for parameter estimation. The regressions were performed by the least-square method with the residuals defined as the difference between the calculated and experimental values of the CO$_2$ solubility. The values of the BIPs for the systems investigated are summarized in Tables S1–S5 in the Supporting Information.

3.1. Solubility of CO$_2$ in Pure Solvents. First, solubilities of CO$_2$ in water and methanol were calculated with PC-SAFT. The $P - x$ phase diagrams in Figure 1 illustrate that the calculations are in good agreement with the experimental values. The estimated BIPs and AAD (%) are represented in Table S1 in the Supporting Information.

The solubilities of CO$_2$ in ILs including [C$_{3}$ mim][PF$_6$], [C$_{2}$ mim][OTf], [C$_{4}$ mim][TF$_2$N], and [Amin][TF$_2$N] were modeled using two strategies. For strategy 1, the BIPs between CO$_2$ and ILs were obtained by simply fitting to the corresponding experimental solubility data for each pair. For strategy 2, the BIPs between CO$_2$ and IL cations or IL anions were the same for all ILs composed of these IL ions. Thus, the values of BIPs in strategy 2 were simultaneously obtained by fitting all of the experimental data together. The comparisons between results calculated using the two strategies are summarized in Figure 2. More details are listed in Tables S2 and S3. As can be observed in Figure 2, the theoretical calculations from both strategies exhibit excellent accuracy in calculations of CO$_2$ solubilities in ILs. The average AAD (%) for these two strategies are 4.35 and 2.75%, respectively. As a whole, strategy 2 is superior to strategy 1 for the selected cases except, perhaps, for the [C$_{3}$ mim][PF$_6$]-containing system, where the difference is insignificant. Figure 3 represents typical $P - x$ phase diagrams for the CO$_2$ solubility in ILs. It can be observed that strategy 2 performs better than strategy 1 in the range of high pressures, which are close to the critical region of CO$_2$. These results indicate that an ion-based model is more suitable to model CO$_2$ solubility in ILs, especially at high pressure.

3.2. Mutual Solubility of Water and ILs. Mutual solubilities of water and ILs, including [C$_{3}$ mim][TF$_2$N], [C$_{2}$ mim][PF$_6$], and [C$_{4}$ mim][OTf], were investigated. The AADs, along with BIPs used in both strategies, are depicted in Figure 4 and are listed in Tables S4 and S5, respectively. The BIPs were obtained with the same approach as for CO$_2$ and ILs. Collectively, correlations of both strategies match experimental values very well in all selected cases, with an average AAD below 5% for the solubility of water in ILs and the solubility of ILs in water. However, it is hard to distinguish which strategy is best, because both strategies show smaller deviations in some cases and larger ones in other cases. Typical results for mutual solubilities of water and [TF$_2$N]$^-$–based imidazolium ILs (with [C$_{3}$ mim]$^+$ as cations) are presented in Figure 5. Both water solubilities in the IL-rich phase and IL solubilities in the water-rich phase increase with temperature and decrease monotonically with the growth of the alkyl chain length (n) of [C$_{n}$ mim]$^+$; the solubilities of ILs in water decrease orders of magnitude as n increases, as shown in Figure 5b. Obviously, both strategy 1 and strategy 2 provide accurate correlation results in all cases, especially for the solubilities of ILs in water.

3.3. Solubility of CO$_2$ in Mixtures of ILs and Co-solvents. The ultimate goal of this work is to develop a thermodynamic model that can predict CO$_2$ solubility in multicomponent fluids containing ILs and co-solvents. Experimental data of binary mixtures are ideal for parameter estimation of the model, while the ternary experimental data present an excellent opportunity to test its predictive capability. Thus, VLE of four ternary systems, including CO$_2$, ILs, and water or methanol, were predicted with the two described strategies. It should be emphasized that all parameters in this section were obtained by fitting to binary data and that ternary data were never used for parameter estimation. The detailed validation results are summarized in Figure 6 and Table S6. BIPs for [C$_{2}$ mim][OTf] and water in strategy 1 were set equal to 0;
BIPs for ILs and methanol or for IL ions and methanol, as used in, respectively, strategies 1 and 2, were also set equal to 0, due to the scarcity of binary data.

For ternary systems, where data for the corresponding three binary sub-systems can be found to regress the BIPs, such as CO$_2$/[C$_4$mim][PF$_6$]/water, both strategies show similar prediction accuracy, as shown in Figure 6 and the $P - x$ phase

Figure 3. Solubilities of CO$_2$ in (a) [C$_4$mim][PF$_6$], (b) [C$_2$mim][OTf], (c) [C$_2$mim][TF$_2$N], (d) [C$_6$mim][TF$_2$N], (e) [C$_8$mim][TF$_2$N], and (f) [Amim][TF$_2$N]. Comparisons of strategy 1 (solid lines) and strategy 2 (dashed lines) to experimental data $^{39-42,55}$ (symbols).
diagrams in Figure 7. The deviations obtained with strategy 2 are slightly larger mainly because the simplified version of the dielectric constant derivative with respect to the mole fraction, as given in eq 11, is inaccurate in the water-containing ternary systems. The solubilities of CO$_2$ in mixtures of [C$_{4}$mim][PF$_6$] and water tend to decrease first and then increase with increasing water fraction, as shown in Figure 7. However, none of the two strategies can capture this trend, instead showing a monotonous decrease with increasing water fraction. The advantage of strategy 2 is that BIPs between IL ions and water can be used directly to predict phase equilibria of mixtures involving ILs composed of these IL ions. For systems of CO$_2$/[C$_2$mim]-[OTf]/water, the most appropriate BIPs for [C$_2$mim]$^+$ and [OTf]$^-$ with water cannot be obtained directly, because of lacking experimental data for mixtures of [C$_2$mim][OTf] and water. However, a set of relatively reliable BIPs can be indirectly obtained from LLE data of [C$_2$mim][TF$_2$N]/water for strategy 2 (solid lines) and strategy 2 (dashed lines) to experimental data$^{35}$ (symbols).

Figure 4. AAD (%) of mutual solubilities of (a) water in ILs and (b) ILs in water (green for strategy 1 and gray for strategy 2).

Figure 5. Mutual solubilities of (a) water in ILs and (b) ILs in water for [C$_2$mim][TF$_2$N] (black), [C$_4$mim][TF$_2$N] (red), [C$_6$mim][TF$_2$N] (blue), and [C$_8$mim][TF$_2$N] (magenta). Comparison of strategy 1 (solid lines) and strategy 2 (dashed lines) to experimental data$^{56}$ (symbols).
Figure 6. AAD (%) of CO₂ solubilities in mixtures of ILs and co-solvents (green for strategy 1 and gray for strategy 2).

Figure 7. Solubilities of CO₂ in the binary solvent of [C₄mim][PF₆] and water with different mass fractions of water (black for 0%, red for 0.15%, blue for 0.89%, and magenta for 1.6%) at (a) 313.15 K, (b) 323.15 K, and (c) 333.15 K. Comparisons of strategy 1 (solid lines) and strategy 2 (dashed lines) to experimental data 39 (symbols).

Figure 8. Mutual solubilities of [C₄mim][OTf] and water: black for solubility of water in IL and red for solubility of IL in water. Comparison of strategy 2 (dashed lines) to experimental data 58 (symbols).
The presence of water seriously abates the CO$_2$ solvation energy for the ILs by weakening interactions between anions and CO$_2$. The interactions between IL ions and methanol are not as strong as those between IL ions and water because of relatively weaker hydrogen-bonding interactions; thus, excellent predictions are obtained, even without any BIPs for IL ions and methanol. Typical predictions for CO$_2$/[C$_8$mim][Tf$_2$N]/methanol are given in Figure 9. With increasing mass fraction of [C$_8$mim][Tf$_2$N] added to methanol, the solubility of CO$_2$ increases gradually, and the prediction lines provided by the two strategies are in good agreement with experimental data.

4. CONCLUSIONS

In this work, the solubility of CO$_2$ in pure solvents, including ILs, water, and methanol, as well as binary mixtures of ILs and water or methanol was investigated. Two strategies based on PC-SAFT EoS were developed to model solubility, VLE, and LLE for these binary and ternary mixtures. In the first strategy, ILs were considered as self-associating chain molecules with two association sites. In the second strategy, they were considered to dissociate into anions and cations. BIPs used in the first strategy were obtained by fitting to the corresponding binary experimental data, while BIPs used in the second strategy were obtained by fitting to all of the binary data together to ensure the uniformity of BIPs for different ILs composed of the same cations and anions. Ternary data were employed to verify the predictive capability.

It was found that both strategies provide accurate correlations in modeling CO$_2$ solubilities in ILs and LLE of IL/water systems. In the verification stage, both strategies can provide accurate predictions for CO$_2$ solubilities in a binary mixture of water and [C$_4$mim][PF$_6$]. However, larger deviations were obtained for a ternary mixture of CO$_2$/[C$_2$mim][OTf]/water due to the scarcity of experimental data to regress the BIPs for ILs or IL ions with water. The second strategy performs better than the first one because BIPs for IL ions with water can be obtained by fitting to data of other ILs composed of the same cations and anions, which is an advantage. For cases of CO$_2$/ILs/methanol, accurate predictions were obtained with both strategies, even without any BIPs used to correct interactions between ILs or IL ions and methanol. More experimental data for binary mixtures of ILs and co-solvents are necessary for PC-SAFT EoS to serve as an ideal tool for phase equilibria.
calculations in CO₂ capturing processes with IL-based solvents as absorbers.

■ ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/10.1021/acs.iecr.2c02778.

Deviations and binary interaction parameters for CO₂ solubilities in water and methanol; CO₂ solubilities in ILs; mutual solubilities of ILs and water; and prediction results for ternary systems (PDF)

■ AUTHOR INFORMATION

Corresponding Authors
Ke Zheng — Beijing Key Laboratory of Coal to Cleaning Liquid Fuels, National Energy R & D Center for Coal to Liquid Fuels, Synfuels China Co., Ltd., Beijing 101400, P. R. China; Centre for Nature-Inspired Engineering, Department of Chemical Engineering, University College London (UCL), London WC1E 7JE, U.K.; orcid.org/0000-0001-7429-6420; Email: zhengke13@mails.ucas.edu.cn
Marc-Olivier Coppens — Centre for Nature-Inspired Engineering, Department of Chemical Engineering, University College London (UCL), London WC1E 7JE, U.K.; orcid.org/0000-0002-1810-2537; Email: m.coppens@ucl.ac.uk

Authors
Xinyu Liao — Centre for Nature-Inspired Engineering, Department of Chemical Engineering, University College London (UCL), London WC1E 7JE, U.K.
Gang Wang — Beijing Key Laboratory of Coal to Cleaning Liquid Fuels, National Energy R & D Center for Coal to Liquid Fuels, Synfuels China Co., Ltd., Beijing 101400, P. R. China; orcid.org/0000-0003-1222-2893
Yong Yang — Beijing Key Laboratory of Coal to Cleaning Liquid Fuels, National Energy R & D Center for Coal to Liquid Fuels, Synfuels China Co., Ltd., Beijing 101400, P. R. China; orcid.org/0000-0002-9220-6166
Yongwang Li — Beijing Key Laboratory of Coal to Cleaning Liquid Fuels, National Energy R & D Center for Coal to Liquid Fuels, Synfuels China Co., Ltd., Beijing 101400, P. R. China

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.iecr.2c02778

Notes
The authors declare no competing financial interest.

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■ REFERENCES

(25) Polishuk, I. Wide-ranging prediction of phase behavior in complex systems by CP-PC-SAFT with universal kij values. I. Mixtures
of non-associating compounds with [C2mim][EtSO4], [C4mim]-


