Article:
Moss, G., Campbell, K.L.S. and Howse, J.R. (2026) Conductivity and bonding structures of aqueous solutions of imidazolium and choline ionic liquids. Journal of Ionic Liquids, 6 (1). 100191. ISSN: 2772-4220

https://doi.org/10.1016/j.jil.2026.100191

Journal of Ionic Liquids 6 (2026) 100191

Contents lists available at ScienceDirect

Journal of Ionic Liquids

journal homepage: www.elsevier.com/locate/jil

Conductivity and bonding structures of aqueous solutions of imidazolium and choline ionic liquids

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ARTICLE INFO

Keywords:
1-Butyl-3-methylimidazolium chloride
Betaine hydrochloride
Metal chloride
Hydrogen bond
Conductivity
IR spectroscopy

ABSTRACT

Ionic liquids (ILs) have gained recognition as a potential alternative to traditional solvents due to their environmentally friendly properties. The ease of handling and higher conductivities of aqueous solutions of ILs has also stimulated research of these green solvents. While 1-butyl-3-methylimidazolium chloride ([C_4mim]Cl) is a commonly researched IL, it is more expensive than traditionally used solvents. Whilst [C_4mim]Cl is less toxic than many traditional solvents, it is still hazardous to both human and marine life. Betaine hydrochloride (BetHCl) presents a cheaper, low toxicity alternative primarily due to the similar aggregation and bonding exhibited by the pure ILs, and their aqueous solutions. This study investigates the conductivity of aqueous solutions of [C_4mim]Cl and BetHCl. We examined these properties across varying water content and temperature, and with added metal salts ($CuCl_2$ and $ZnCl_2$), to develop insights into aggregation, bonding, and IL-metal interactions. Fourier Transform Infrared (FTIR) spectroscopy was used to further explore aggregation and bonding. The conductivity data displayed good agreement with Walden's rule and the Vogel-Fulcher-Tammann (VFT) equation. Results indicate non-linear relationships between water content and conductivity for both ILs, whereas, near-linear relationships were found between temperature and conductivity. Different interactions were observed between the two metal chlorides and the ILs. Gradual changes in conductivity with varying water content, rather than a step change, suggest the formation of hydrogen bonding networks in aqueous solutions of both [C_4mim]Cl and BetHCl. The similar trends exhibited by the ILs imply the potential of BetHCl as a viable, cost-effective alternative to traditional solvents.

1. Introduction

Ionic liquids (ILs) are salts often consisting of a bulky organic cation and an organic or inorganic anion. Typically, they have melting points below 100°C due to their loosely packed structures. Discovered by Walden in 1914, the use of ILs has gained traction in the last few decades due to their environmentally benign properties, for example low vapor pressure, high thermal stability and absence of volatile organic compounds (Walden, 1914; Seddon et al., 2000; Bailey et al., 2024; Clarke et al. 2018; James et al. 2003; Pinkert et al., 2009; Rogers and Seddon, 2003). Due to the vast number of potential cations and anions, the tunability and versatility of ILs make them appealing for many applications including solvent replacement, polymer and catalytic chemistry, separations, biomass pretreatment and lubricants (Seddon et al., 2000; Malaret et al., 2020; Visser et al., 2001; Wasserscheid and Keim, 2000; Ye et al., 2001). Additionally, ILs have demonstrated significant potential for enhancing reaction rates, yields, and selectivities (Forsyth et al., 2004; García-Verdugo et al., 2015).

Research of aqueous solutions of ILs has been stimulated recently due to the high conductivity values reported by several authors, in contrast to traditional solvents (Anouti et al., 2012; Bonhôte et al., 1996). Authors have demonstrated that IL concentration, temperature and chemical structure, including cation functional group, are key factors in the conductivity of aqueous solutions of ILs (Niazi et al., 2013; Chang et al., 2008; Bobrova et al., 2016; Widegren and Magee, 2007; Vieira et al., 2015; Fan et al., 2022). Therefore, further investigation of their physical properties is vital to enable large-scale deployment of ILs and to meet environmental and economic targets. Comprehensive understanding of solution conductivity is essential to facilitate the large-scale deployment of ILs, and their aqueous solutions, as conductivity directly impacts solvent performance in many applications, such as electrochemical processes (Chen et al., 2022).

The low flammability, low volatility, and air and water stability of imidazolium-based ILs has made them one of the most widely

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https://doi.org/10.1016/j.jil.2026.100191
Received 5 August 2025; Received in revised form 9 December 2025; Accepted 28 January 2026
Available online 30 January 2026

researched classes of IL (Tokuda et al., 2005; Huddleston et al., 2001). This prevalence in research has made them less expensive than most ILs, although they remain more expensive than traditional solvents. The four-part alkyl chain and chloride anion of 1-butyl-3-methylimidazolium chloride ([C_4mim]Cl) has made this IL an attractive option for industry and research due to the high solubility of the IL and hydrophilic nature of the anion (Fig. 1). The melting point of [C_4mim]Cl (70°C) makes aqueous solutions of the IL more attractive in low temperature applications. Although, [C_4mim]Cl is less toxic than traditional solvents, the IL remains hazardous to human and marine life (Safety data sheet - 1-Butyl-3-methylimidazolium chloride' 2025).

The bonding structures of imidazolium-based ILs, both pure and aqueous, are the source of much disagreement. Although authors agree that aqueous solutions of 1-alkyl-3-methylimidazolium chloride, with varying chain length, display self-aggregation behavior, the chain length required is a source of debate. Some authors report a critical aggregation point in aqueous solutions of [C_4mim]Cl, other authors report longer alkyl chain lengths are required (Singh and Kumar, 2007; Blesic et al., 2007). Several other authors report a more gradual change in bonding structures in aqueous solutions of [C_4mim]Cl (Niazi et al., 2013; Chang et al., 2008). These latter studies suggest the development of a hydrogen bonding network rather than the existence of a critical aggregation point.

Betaine hydrochloride (BetHCl) is an IL from the choline cation group, a family of ILs well researched across a variety of industrial sectors (Panda et al., 2018; Gadilohar and Shankarling, 2017). Many choline-based systems are classified as deep eutectic solvents (DES), solvents formed from a hydrogen bond donor and hydrogen bond acceptor, which have melting points significantly lower than their constituents (Li and Row, 2016; Paiva et al., 2014; Yeow et al., 2024). Recent publications have shown they are a versatile alternative to ILs, predominantly due to the ease of synthesis, availability of the components and biodegradability of the DES (Yeow et al., 2024; Abbott et al., 2004; Zhang et al., 2018).

The higher melting point of BetHCl (241°C) excludes it from the traditional definition of an IL (melting temperature < 100°C). However, recent publications have designated higher melting point salts as ionic liquids due to their structural similarities and analogous properties (Naserifar et al., 2023; Wang et al., 2023; Dai et al., 2024; Arkhipova et al., 2021; Mital et al., 2022). Additionally, these newly classified ILs often contain a functional group common in IL cations, such as imidazolium, phosphonium or choline (Arkhipova et al., 2021; Mital et al., 2022). BetHCl is not a DES as its melting point is not considerably lower than that of both its constituents (betaine = 293°C, hydrochloric acid = -35°C for a 37 mass% aqueous solution). For these reasons, BetHCl has been classified as an IL in this study, however, it is important to acknowledge this categorization may be debated by those who strictly adhere to the conventional melting point criterion. Betaine is an oxidized derivative of choline consisting of a hydrophobic quaternary ammonium salt head and a hydrophilic carboxylic acid tail (Fig. 1) (Ueland, 2011). This amphoteric structure leads to self-aggregation in the liquid phase, similar to imidazolium-based ILs (Panda et al., 2018; Rengstl et al., 2013). Furthermore, BetHCl is not classified as hazardous to human or marine life, and its commercial use in pharmaceuticals has made it less expensive than many ILs, including [C_4mim]Cl (Safety data sheet - Betaine hydrochloride 2024; Guilliams and Drake, 2020).

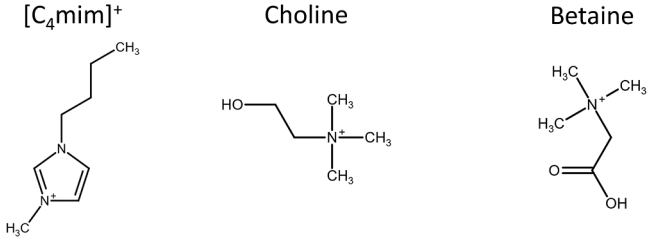

Fig. 1. Chemical structure of 1-butyl-3-methylimidazolium ([C_4mim]$^+$), choline, and betaine cations.

Despite the aprotic nature of [C_4mim]Cl and protic nature of BetHCl, it has been hypothesized that choline ILs exhibit similar bonding structures in aqueous solutions due to the similarities in self-aggregation behavior of pure choline and imidazolium-based ILs. Additionally, authors have reported, both, IL-IL aggregation and IL-IL hydrogen bonding in aqueous solutions of BetHCl (Nockemann et al., 2006; Amorim Da Costa and Leite, 2001). If their bonding regimes are analogous, solvent performance may be similar, despite the differences in protic and aprotic structures, making BetHCl a potential alternative to the more expensive and higher toxicity [C_4mim]Cl.

Corrosion studies using aqueous solutions of ILs have reported corrosion inhibition using dilute solutions and exacerbated corrosion in concentrated solutions (Verma et al., 2017; Zhang and Hua, 2009; Qiang et al., 2017; Cai et al., 2011; Uerdingen et al., 2005). This difference in behavior, with varying water content, highlights the need for further investigation into IL-metal interactions.

In this study, we investigated the influence of water content (93–99.5 mol% H_2O) and temperature (15–55 °C) on the aggregation and bonding of aqueous solutions containing [C_4mim]Cl and BetHCl. The solubility range of BetHCl in water was found to be significantly smaller than [C_4mim]Cl, with incomplete dissolution of BetHCl at water contents lower than 93 mol%, or 60 mass%, H_2O at 25°C. To enable a true comparison of the aqueous solutions of ILs, the concentration range was chosen to reflect the full solubility range of BetHCl. Conductivity measurements of aqueous solutions of [C_4mim]Cl at lower water contents are reported by several authors (Mao and Zhu, 2013; Yang et al., 2013; Liu et al., 2008). Additionally, the impact of $CuCl_2$ and $ZnCl_2$ (added at concentrations of 1000–9000 ppm), was explored. To assess these effects and characterize changes in solution structure, measurements of solution conductivity and Fourier Transform Infrared (FTIR) spectroscopy were conducted. By providing additional insight into the bonding structures of aqueous solutions of ILs, this work paves the way for the large-scale application of ILs.

2. Methodology

2.1. Solution preparation

Ionic liquids ([C_4mim]Cl ≥ 98.0 %, BetHCl ≥ 99.0 %) were purchased from Sigma Aldrich, and used as received. Aqueous solutions of [C_4mim]Cl and BetHCl with water content between 93–99.5 mol% were prepared with deionized water (Purite HPA 30, quality = 16.0 MΩ) and incubated for 24 h at 40°C and 150 rpm to ensure full dissolution. The purity of the ILs resulted in an uncertainty of 0.13 mol% in the aqueous solutions of [C_4mim]Cl and 0.065 mol% in aqueous solutions of BetHCl (Supporting information). $CuCl_2$ (97 %, Sigma Aldrich) or $ZnCl_2$ (97 %, Sigma Aldrich), used as received, was then added to solutions with water contents of 93, 96 and 99.5 mol% to reach metal salt contents of 1000–9000 ppm and incubated for a further 24 h, uncertainty is reported in the supporting information. This concentration range was

Table 1
Details and compositions of IL solutions studied.

Water content (mol%)	IL content (mol%)	IL content (mass%)	$CuCl_2$ content (ppm)	$ZnCl_2$ content (ppm)	Temperature (°C)
93–99.5	7–0.5	42–4	0	0	15–55
93	7	42	1000–9000	0	25
96	4	28	1000–9000	0	25
99.5	0.5	4	1000–9000	0	25
93	7	42	0	1000–9000	25
96	4	28	0	1000–9000	25
99.5	0.5	4	0	1000–9000	25

chosen to correlate with the observed solubility range of BetHCl in water at 25°C. A description of all solutions studied can be found in Table 1.

2.2. Conductivity measurements

Conductivity measurements were conducted at 10°C intervals from 15–55°C for aqueous solutions of [C_4mim]Cl and BetHCl. Measurements were repeated at 25°C for aqueous solutions of [C_4mim]Cl and BetHCl with metal chlorides. Measurements were conducted in triplicate using the Mettler Toledo SevenCompactDuo and InLab 731-ISM probe. The probe was calibrated using a 12800 μS/cm calibration standard, purchased from Hanna Instruments, prior to measurement. The uncertainty of reported conductivity measurements is 0.67 mS/cm (a full description of the calculation of uncertainty is available in the supporting information). A jacketed vessel and water bath (Grant Instruments ecocool 150) were employed to control temperature within 0.1°C.

3. Results

3.1. Binary ILs

The conductivity of aqueous solutions of [C_4mim]Cl and BetHCl demonstrate a significant deviation from that expected of traditional salts at all temperatures explored (15–55°C, Fig. 2). A second order polynomial was fitted to the data in Fig. 2 to guide interpretation, a coefficient of determination (R^2) > 0.97 was calculated for each data series. Aqueous solutions of both ILs with high water content exhibit low conductivities. As water content decreases, conductivity increases to a maxima. As water content decreases further, past the maxima, conductivity begins to sharply decrease. The conductivity of binary systems of [C_4mim]Cl reach a maxima at 94 mol% water, whereas, binary systems of BetHCl reach a maxima at 95 mol% water. This indicates a reduced number of free moving ions and implies a reduced ion mobility, possibly due to the formation of aggregates or the development of a hydrogen bonding network. An increase in conductivity is found with increased temperature, with the effect amplified at lower water content.

3.2. Binary ILs with metal chlorides

Fig. 3 depicts the conductivity of aqueous solutions of [C_4mim]Cl and BetHCl with metal chlorides. A second order polynomial was fitted to the data in Fig. 3 to guide interpretation, a R^2 > 0.99 was calculated for each data series. The addition of $CuCl_2$ increases conductivity in every case, suggesting the electrolytic behavior of $CuCl_2$ in both ILs. The conductivity of both ILs increased with the addition of $ZnCl_2$ at high water concentration, but decreased at low concentrations. This indicates the possible formation of IL-zinc complexes at low water contents and electrolytic behavior of $ZnCl_2$ at high water contents. Solutions of [C_4mim]Cl with high water content and $ZnCl_2$ concentrations ≥ 5000 ppm displayed higher conductivities than lower water content solutions. This trend was not replicated by solutions of BetHCl, suggesting that $ZnCl_2$ may be in excess in aqueous solutions of [C_4mim]Cl with high water content and $ZnCl_2$ concentrations. Slight differences between the ILs originate mainly from the increased conductivity of binary solutions of BetHCl.

4. Discussion

The pronounced impact of water content on the conductivity of ILs is evident in the temperature dependence of conductivity measurements. Conductivity is significantly influenced by increasing temperature at low water contents. As water content increases, this influence diminishes. Elevated temperatures increase ion diffusivity in all solutions due to increased thermal energy. However, the greater number of ions present in low water content solutions, results in conductivity increases substantially larger than observed at higher water contents.

4.1. Effect of water content

The comparable conductivity trends displayed by the ILs may suggest similar bonding structures within solutions. In micellar solutions, a step change is observed as the critical micelle concentration is reached, resulting in a significant change in trend. The absence of a significant step change shown in Fig. 2, indicates a more gradual shift in trend. This behavior appears reflective of the development of a hydrogen bonding network as water concentration decreases and suggests a gradual shift from mobile ions showing electrolytic behavior to a more aggregate-like behavior. In this state, the fully solvated cations and anions begin to favor IL-IL bonds, which could lead to the formation of large IL structures in solution. These results are in agreement with molecular dynamics simulation data, reported by other authors (Niazi et al., 2013; Chang et al., 2008). Additionally, results for aqueous solutions of

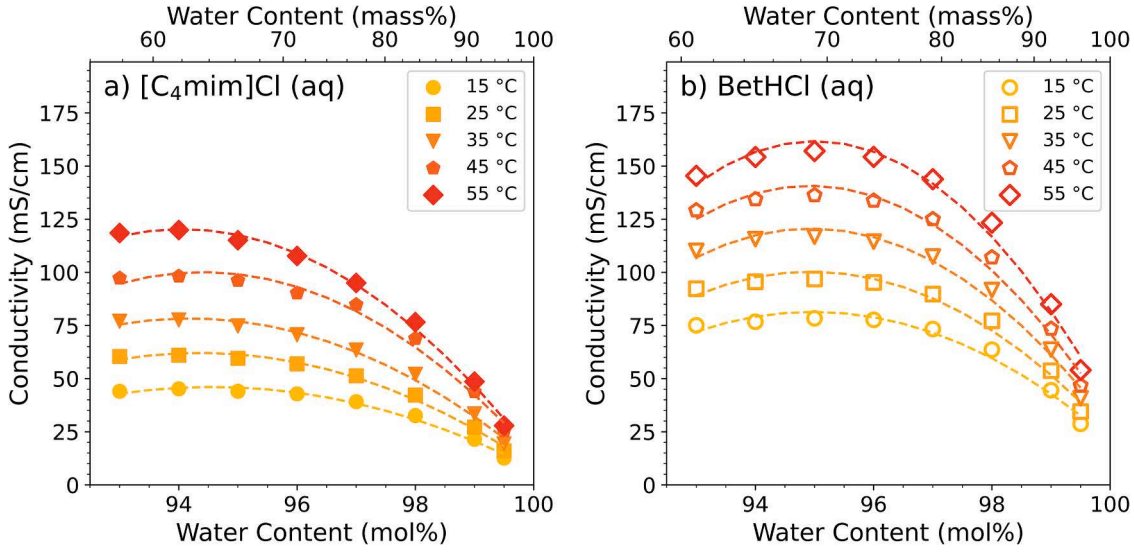

Fig. 2. Conductivity of aqueous solutions of [C_4mim]Cl (filled symbols, left) and BetHCl (open symbols, right) with varying water content. Error bars are present but obscured by data points, quantitative uncertainty numbers can be found in Supporting information. A 2nd order polynomial was fitted to the data for ease of interpretation (R^2 > 0.97). (●) 15°C, (■) 25°C, (▼) 35°C, (⬟ 45°C and (◆) 55°C.

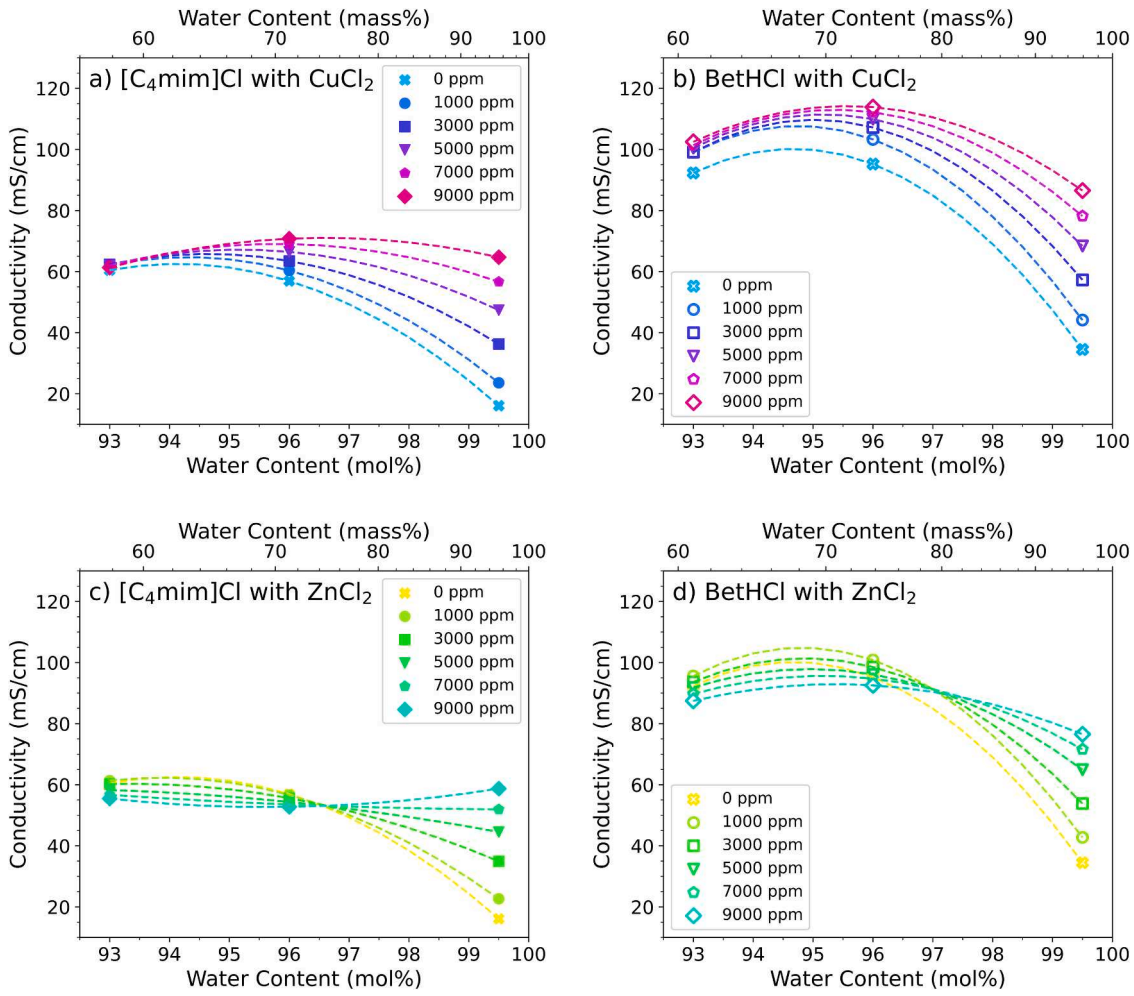

Fig. 3. Conductivity of aqueous solutions of $[C_4mim]Cl$ (filled symbols, left) and BetHCl (open symbols, right) with $CuCl_2$ (top) or $ZnCl_2$ (bottom). Error bars are present but obscured by data points, quantitative uncertainty numbers can be found in Supporting information. A 2nd order polynomial was fitted to the data for ease of interpretation ($R^2 > 0.99$). (✖) 0 ppm of $CuCl_2$, (●) 1000 ppm of $CuCl_2$, (■) 3000 ppm of $CuCl_2$, (▼) 5000 ppm of $CuCl_2$, (⬟) 7000 ppm of $CuCl_2$ and (◆) 9000 ppm of $CuCl_2$. (✖) 0 ppm of $ZnCl_2$, (●) 1000 ppm of $ZnCl_2$, (■) 3000 ppm of $ZnCl_2$, (▼) 5000 ppm of $ZnCl_2$, (⬟) 7000 ppm of $ZnCl_2$ and (◆) 9000 ppm of $ZnCl_2$.

$[C_4mim]Cl$ are analogous with experimental data reported by several authors at ambient pressure and 25°C (Figure S1) (Yang et al., 2013; Passos et al., 2014). Minimal differences in conductivity values are most likely the result of differences in experimental procedure and reported uncertainties.

The reported maxima in conductivity at water contents of 94 and 95 mol% for solutions of $[C_4mim]Cl$ and BetHCl, respectively, may be indicative of the solution progressively being dominated by growing IL-IL hydrogen bonding networks in solution as water content decreases. At high water contents, the limited concentration of ions in solution are free moving, leading to a marked increase in conductivity compared to that of pure water. As water content decreases, a higher concentration of ions are present, increasing conductivity. At the same time, the hydrogen bonding network between the ions is believed to be increasing in strength, restricting the free movement of the new ions. When the concentration reaches the observed maxima, the hydrogen bonding network appears to begin to dominate the solution and aggregate-like behavior is observed. As water content decreases further, the hydrogen bonding network reduces both the number of free moving ions and their ability to move through the solution.

Despite the proposed similar bonding regimes, solutions of BetHCl display consistently higher conductivities than solutions of $[C_4mim]Cl$. The asymmetrical $[C_4mim]^+$ cation, with its longer alkyl chain and bulky imidazolium ring, reduces the ion diffusivity more than the smaller $BetH^+$ cation. The effect of alkyl chain length on conductivity of aqueous solutions of $[C_nmim]Cl$ can be found in Figure S2 (Mao and Zhu, 2013; Alopina et al., 2017; Alopina et al., 2023). Similarly, a comparison of the effect of bulky anions, using experimental data reported by other authors, is presented in Figure S3 (Chaudhary et al., 2014; Stoppa et al., 2009; Andanson et al., 2014).

Walden's rule is an empirically derived relation that suggests that the product of the molar conductivity (Λ) and dynamic viscosity (η) is constant for a solute with varying solution concentration, as given by Equation 1 (Walden, 1906). It was originally based on the properties of dilute aqueous solutions, but has since been applied to the properties of non-aqueous electrolyte solutions, molten salts and ILs, however limitations in the application of Walden's rule to pure and aqueous solutions of ILs have been reported (Pereiro et al., 2012; Adams and Laidler, 1968; Ueno et al., 2010; Yoshizawa et al., 2003; Angell et al., 2007).

$$\Lambda \approx constant\left(\frac{1}{\eta}\right) \quad (1)$$

Fig. 4 shows a Walden plot of aqueous solutions of [C4mim]Cl and BetHCl with varying water content. The experimental dynamic viscosity data measured and methodology used are presented in the Supporting information (Reviakine et al., 2011). The diagonal line in Fig. 4 represents the behavior of a 0.01 mol/L aqueous solution of KCl, where the ions are known to be fully dissociated, with equal mobility (Angell et al., 2007). The deviation from the ideal line (ΔW) is described as the vertical distance measured from the ideal line to the data point of the aqueous solution of IL. The deviation can be used to calculate the ionicity, a quantitative measure of ion dissociation, Equation 2 (Thawarkar et al., 2016; Wang et al., 2020).

$$Ionicity\ (\%) = 10^{-\Delta W} * 100 \quad (2)$$

Table 2 presents the deviation and ionicity data of aqueous solutions of the ILs at water contents of 93 mol%. The ΔW and ionicity data are reported in full in Table S7.

Fig. 4 and Table 2 show that aqueous solutions of [C4mim]Cl show larger deviations from the ideal behavior, while aqueous solutions of BetHCl show good agreement with the reference line. Aqueous solutions of [C4mim]Cl show lower ionicity, indicating that ion pairing may be present. These results may show that the ions in [C4mim]Cl are not fully dissociated in aqueous solutions, due to the formation of hydrogen bonding networks, ion pairing and other ion interactions. Interestingly, the Walden plot of aqueous solutions of [C4mim]Cl and BetHCl show greater agreement than would be expected when comparing a protic and aprotic IL, respectively. Thawarkar *et al.* report that protic and aprotic ILs occupy distinct areas of the Walden plot, with protic ILs recorded near the origin and aprotic ILs observed further from the origin (Thawarkar et al., 2016). This result is not replicated in this study, with aqueous solutions of the aprotic [C4mim]Cl displaying conductivity and viscosity results closer to the origin. This may indicate that aqueous solutions of [C4mim]Cl and BetHCl show greater similarities than their aprotic and protic structures suggests. Although Table 2 indicates that BetHCl in aqueous solution may display greater ion dissociation, the Walden rule has important limitations that must be considered.

The most commonly cited limitation of the use of the Walden rule with ILs, and their aqueous solutions, is that it does not consider ion size (MacFarlane et al., 2009; De Sarkar et al., 2020). Increases in ion size, such as increasing alkyl chain length, can introduce new intermolecular interactions that can significantly influence the dynamic viscosity, in turn affecting the relationship between conductivity and viscosity. Additionally, it has been shown that large differences in the size of the anion and cation can introduce unbalanced coulombic interactions, increasing ΔW and decreasing ionicity (Wang et al., 2020). Macfarlane *et al.* report that some ILs with strong electrostatic bonding, such as hydrogen bonding, can exhibit very small deviations from the reference line, despite low ion dissociation, due to the ions in solution being influenced equally by neighboring ions (MacFarlane et al., 2009). Therefore, the combined impact of ion size, cation-anion size disparity, and the influence of neighboring ions are likely to impact the ΔW and ionicity of the aqueous solutions of [C4mim]Cl and BetHCl, potentially obscuring ion pairing or viscosity-governed effects seen in strong electrolytes. Finally, the use of 0.01 mol/L aqueous solution of KCl as the ideal Walden line has been critically challenged, potentially limiting the accuracy of the Walden rule in describing the behavior of aqueous solutions of ILs (Schreiner et al., 2010). These factors limit the findings of the Walden rule for aqueous solutions of [C4mim]Cl and BetHCl.

Table 2
Walden rule deviations (ΔW) and ionicity of aqueous solutions of [C4mim]Cl and BetHCl at water content of 93 mol%.

Temperature (°C)	ΔW (S.cm^2/mol)		Ionicity (%)	
	[C4mim]Cl	BetHCl	[C4mim]Cl	BetHCl
15	0.130	0.027	74.15	94.03
25	0.156	0.057	69.90	87.73
35	0.173	0.055	67.21	88.12
45	0.163	0.066	68.68	85.90
55	0.178	0.113	66.45	77.13

Attenuated Total Reflectance (ATR) FTIR spectra were obtained in the range of 800–4000 cm^{-1}. Fig. 5 displays the FTIR spectra of aqueous solutions of [C4mim]Cl and BetHCl in the hydrogen bonding region (2400–4000 cm^{-1}) (the full spectra can be found in the supporting information). Below wavenumbers of approximately 2400 cm^{-1}, the spectra display clear differences due to the differences in the chemical structures of the ILs. Greater similarities can be seen when examining the broad O-H peak (2800 – 3700 cm^{-1}), however some differences due to chemical structure remain. As water content decreases, aqueous solutions of [C4mim]Cl and BetHCl demonstrate a decrease in O-H peak intensity and a peak shift to higher wavenumbers. This may be due to the disruption of water-water hydrogen bonds and the formation of IL-water

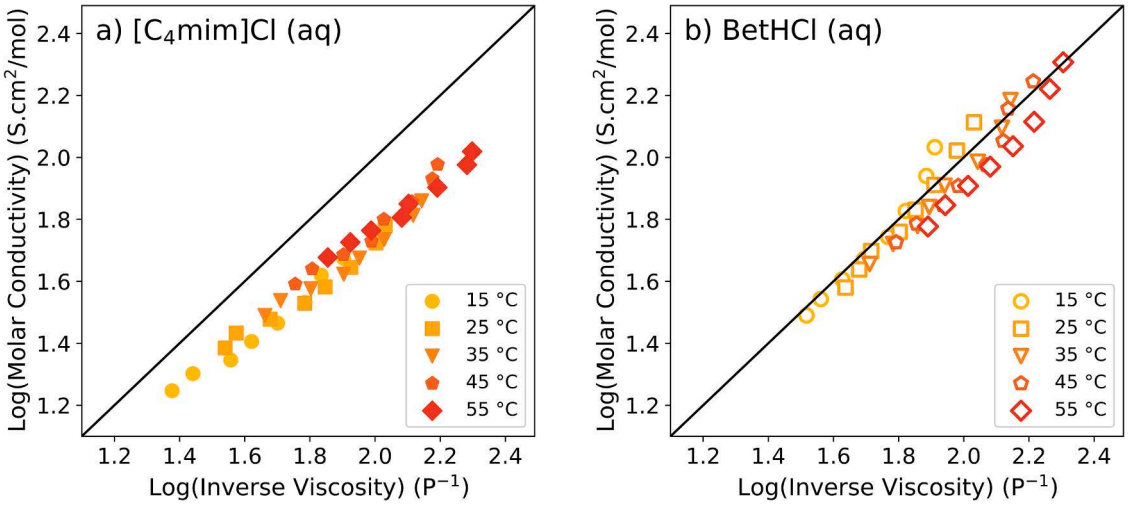

Fig. 4. Walden plot of aqueous solutions of [C4mim]Cl (filled symbols, left) and BetHCl (open symbols, right) with a 0.01 mol/L aqueous solution of KCl as reference (solid line). (●) 15°C, (■) 25°C, (▼) 35°C, (⬟) 45°C and (◆) 55°C.

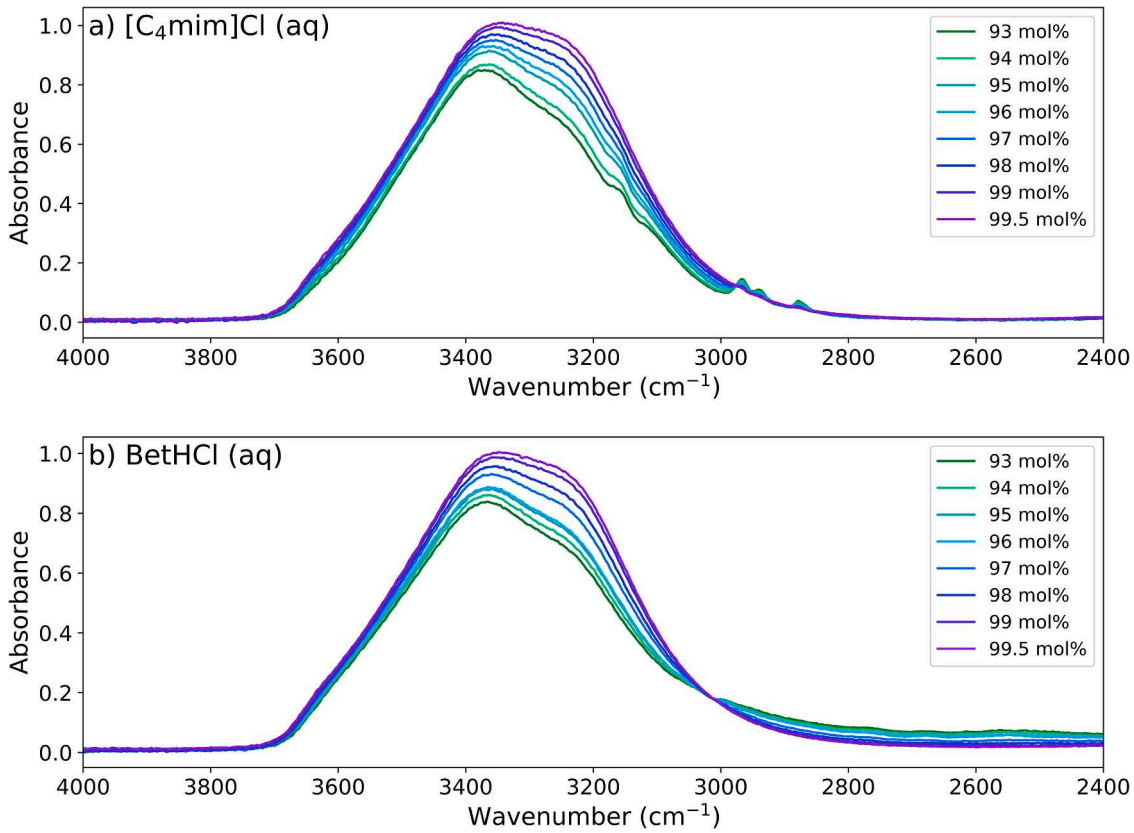

Fig. 5. ATR-FTIR spectra of aqueous solutions of [C_4mim]Cl (top) and BetHCl (bottom) at 25°C and wavenumbers 2400–4000 cm^{-1}. For each spectrum obtained, a total of 64 scans were recorded at a resolution of 4 cm^{-1}.

and IL-IL hydrogen bonds (Ren et al., 2019; Pensini et al., 2024). As water content decreases in aqueous solutions of [C_4mim]Cl, C-H peaks are seen at 3100–3200 cm^{-1} and 2700–3100 cm^{-1} due to C-H stretching in the imidazolium ring and the aliphatic alkyl chain in the imidazolium cation, respectively (Li and Kobayashi, 2016). Additionally, Fig. 5 shows a broadening of the O-H peak in aqueous solutions of BetHCl with decreasing water content due to the addition of the carboxylic acid functional group of the betaine cation, as seen in pure BetHCl (Pensini et al., 2024). The trends observed in Fig. 2 and the peaks displayed in Fig. 5 may suggest that the formation of hydrogen bonding networks occurs in aqueous solutions of [C_4mim]Cl and BetHCl as water content decreases.

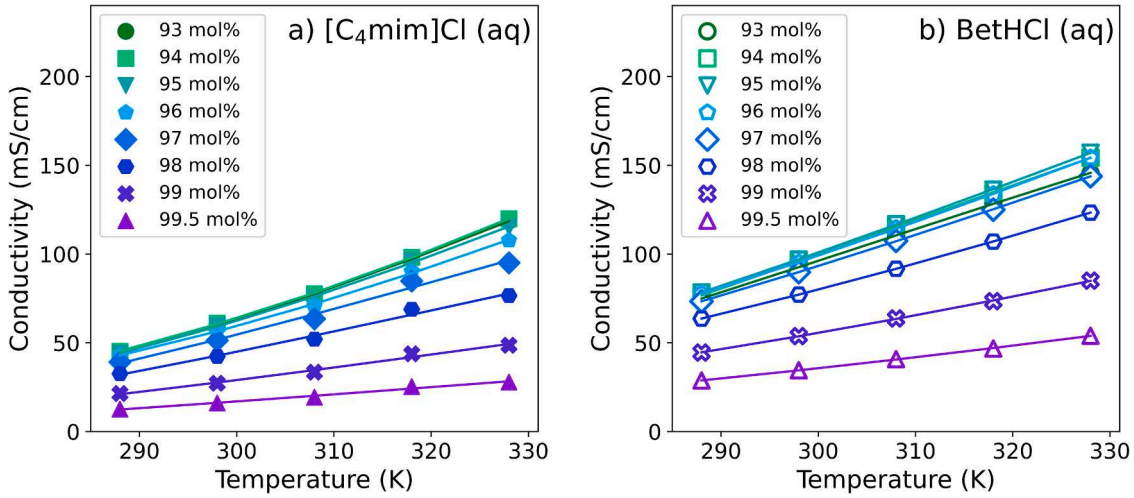

Fig. 6. Conductivity of aqueous solutions of [C_4mim]Cl (filled symbols, left) and BetHCl (open symbols, right) with varying temperature and Vogel–Fulcher–Tammann fitting ($R^2 > 0.98$). Error bars are present but obscured by data points, quantitative uncertainty numbers can be found in Supporting information. (●) 93 mol%, (■) 94 mol%, (▼) 95 mol%, (⬟) 96 mol%, (◆) 97 mol%, (⬢) 98 mol%, (✖) 99 mol% and (▲) 99.5 mol%.

4.2. Effect of temperature

Fig. 6 shows an equivalent trend for both ILs across the examined temperature range, with the maxima remaining consistent for each IL. This result is unexpected, given the known temperature dependence of hydrogen bonding. Elevated temperatures increase thermal energy in solutions, which in turn disrupts the electrostatic attraction of hydrogen bonds. This disruption is evident from the larger change in conductivity between water contents of 93–97 mol%, at high temperatures. It is, therefore, possible that subtle variations in the maxima occur between the examined data points as temperature increases. The Vogel-Fulcher-Tammann (VFT) equation is used to express the effect of temperature on the conductivity of the aqueous solutions of ILs in Fig. 6 (Equation 3) (Vogel, 1921; Fulcher, 1925; Tammann and Hesse, 1926).

$$\sigma = \sigma_{\infty} exp\left(\frac{-B}{T - T_0}\right) \quad (3)$$

Where σ is ionic conductivity and T is temperature in kelvin. σ_{∞}, B and T_0 are adjustable parameters, where σ_{∞} is conductivity at infinite temperature, B is the pseudoactivitation energy and T_0 is the Vogel temperature (Green et al., 2011; Rault, 2000). The values of σ_{∞}, B, T_0 and R^2 are listed in Table S8.

Marginally higher R^2 values were calculated for aqueous solutions of BetHCl, this may be due to the smaller, more symmetrical cation (Yadav and Pandey, 2014). However, while the R^2 values indicate a good quality fit using the VFT equation for all concentrations of the aqueous solutions of ILs, there is no consistent change of σ_{∞}, B and T_0 across the concentration range for either IL. This is likely due to the limited temperature range of the data. Thus, a larger temperature range should be explored, a higher compositional resolution may be required, or alternatively a concentration based model may be more appropriate for these solutions.

4.3. Effect of metal chlorides

Fig. 3 details similarities between the ILs with the addition of $CuCl_2$ or $ZnCl_2$. Slight variations in trend were observed due to the differing maxima in Fig. 2. The conductivity of BetHCl solutions remained higher than [C_4mim]Cl solutions in every instance, likely due to ion size and shape. The restriction of ion diffusivity results in a lowered conductivity, as found with the binary ILs.

The addition of $CuCl_2$ appeared to have a comparable effect on binary solutions of both ILs. At low water contents, the IL hydrogen bonding network is believed to be highly developed and may greatly limit diffusivity, which could explain the negligible increase in conductivity. As water content increases, the hydrogen bonding network is suggested to decrease in size and strength, thereby allowing greater ion diffusivity and increasing conductivity. The observed pattern is consistent with the expected electrolytic behavior of $CuCl_2$ when added to aqueous solutions of [C_4mim]Cl and BetHCl at all water contents in this study.

The suggested electrolytic behavior of $CuCl_2$ is supported by the observed colour change of solutions. All solutions of binary ILs were clear. A colour change to green was seen with the addition of all concentrations of $CuCl_2$ to 93 mol% water solutions of both ILs, indicating the formation of $[CuCl_4]^{2-}$ complexes (Sasmal et al., 2013). Solutions with water content of 99.5 mol% water were blue in colour, demonstrating the presence of $[Cu(H_2O)_6]^{2+}$ complexes (Naskar et al., 2002). 96 mol% water solutions were blue-green, indicating a mix of both $[CuCl_4]^{2-}$ and $[Cu(H_2O)_6]^{2+}$ complexes were present (Sasmal et al., 2013; Naskar et al., 2002).

Differences between the behavior of $CuCl_2$ and $ZnCl_2$ in aqueous ILs are observed. The metal-water interactions are likely to remain consistent, which is reflected in the potential formation of small $[ZnCl_4]^{2-}$ and $[Zn(H_2O)_6]^{2+}$ complexes (Maeda et al., 1996). However, the formation of larger IL-Zn complexes is also a well documented possibility. Several authors have reported the formation of [C_4mim][$ZnCl_3$] and [C_4mim][Zn_2Cl_5] complexes in aqueous solutions of [C_4mim]Cl with $ZnCl_2$ (Xu and Zhang, 2017; Zhang and Wei, 2008; Wang et al., 2014). Additionally, Qu *et al* and Xiao-Ming *et al* have demonstrated the existence of dichlorobis(betaine)zinc(II) monohydrate complexes in aqueous solutions of BetHCl with $ZnCl_2$ (Qu et al., 2023; Xiao-Ming and Mak, 1991).

Aqueous solutions of both ILs with water content of 93 mol% displayed a small increase in conductivity upon the addition of a low concentration of $ZnCl_2$, likely reflecting the electrolytic behavior of the Zn^+ and Cl^- ions. However, as the concentration of $ZnCl_2$ increased, the conductivity decreased. This may suggest the potential formation of larger IL-Zn complexes, which could limit diffusivity and the number of available, charge carrying, free ions.

As water content increases to 96 mol%, differences between the ILs emerge, likely due to the strength of bonding in solution and the stoichiometry of the proposed IL-Zn complexes. The addition of $ZnCl_2$ to [C_4mim]Cl reduces the conductivity in every case explored. This result would be consistent with the formation of IL-Zn complexes, favored over the IL-IL hydrogen bonds present in the binary solutions, as well as the smaller $[ZnCl_4]^{2-}$ and $[Zn(H_2O)_6]^{2+}$ complexes formed in ternary solutions. Aqueous solutions of BetHCl at water contents of 96 mol% demonstrate the same trend shown at 93 mol%. This is likely due to the stronger IL-IL hydrogen bonds present in aqueous solutions of BetHCl requiring higher $ZnCl_2$ concentrations for dissolution than [C_4mim]Cl IL-IL hydrogen bonds. In both [C_4mim][$ZnCl_3$] and [C_4mim][Zn_2Cl_5] complexes, one [C_4mim]Cl molecule is required, whilst two BetHCl molecules are required to form one dichlorobis(betaine)zinc(II) monohydrate complex. Consequently, $ZnCl_2$ may be in excess in solutions of BetHCl at 96 mol%, but not in equivalent solutions of [C_4mim]Cl. Excess $ZnCl_2$ ions would form $[ZnCl_4]^{2-}$ and $[Zn(H_2O)_6]^{2+}$ complexes, significantly smaller complexes than IL-Zn complexes, with greater mobility, creating a further increase in conductivity, similar to the trend seen in Fig. 3.

The addition of $ZnCl_2$ to aqueous solutions of both ILs with water contents of 99.5 mol% had a significant impact on the conductivity. The low molar concentration and limited number of IL ions present likely prevent the formation of IL-IL bonding networks and greatly limit potential IL-Zn complexation. Therefore, the addition of $ZnCl_2$ most likely results in the formation of $[ZnCl_4]^{2-}$ and $[Zn(H_2O)_6]^{2+}$ complexes. These smaller complexes combined with the limited number of ions in the binary solutions would allow for the greatest increase in conductivity upon the addition of $ZnCl_2$.

The conductivity of 99.5 mol% water content solutions of [C_4mim]Cl with $ZnCl_2$ concentrations $\geq$ 5000 ppm are higher than the corresponding solutions with water content of 96 mol%. This trend is not replicated by BetHCl solutions or [C_4mim]Cl with $CuCl_2$. This trend is possibly a result of the higher $ZnCl_2$ concentration leading to an excess of the metal salt in solution, potentially creating a heterogeneous solution. Researchers must consider this trend during process design and determine if it is replicated in other IL solutions with high concentrations of metal salts. As this trend is not replicated by aqueous solutions of BetHCl, these results provide initial evidence that BetHCl may not be a suitable alternative to [C_4mim]Cl in all applications.

5. Conclusions

In this study, the bonding structures of aqueous solutions of [C_4mim]Cl and BetHCl with and without metal chlorides have been investigated using conductivity and FTIR measurements. Water content and cation size appear to be the preeminent factors affecting the conductivity of aqueous solutions of ILs. Although the conductivity of ILs can vary significantly, the conductivity of aqueous solutions of [C_4mim]Cl and BetHCl demonstrate clear similarities. While conductivity values for aqueous solutions of BetHCl were higher than [C_4mim]Cl, the same trend was exhibited by both ILs. A gradual change in conductivity with

changing water content was observed, which suggests the gradual formation of a hydrogen bonding network rather than the existence of a critical aggregation point. These findings were supported by FTIR spectra which showed changes in hydrogen bonding as water content decreased. The results in this study are helpful for informing the development of the large-scale application of ILs. This work contributes to the existing body of knowledge by providing additional insight into the bonding structures of aqueous solutions of imidazolium and choline-based ILs, with and without metal salts, as well as expanding the known physical property data of ILs. However, this study is limited by the small scope of the dataset. Future research should aim to extend this dataset, to include similar ILs, which may help to ensure successful large-scale deployment of ILs across several industrial sectors. Future research should also focus on a more expansive study of IL-metal interactions by exploring wider metal salt and IL concentration ranges, as well as the use of further analytical techniques, such as spectroscopy, to examine the potential formation of IL-metal complexes. Future studies would further benefit from a wider examination, and comparison, of the physical properties of other protic and aprotic ILs. By enabling the enhanced understanding of aqueous solutions of [C_4mim]Cl and BetHCl, as well as IL-metal interactions in aqueous solutions, this work helps to pave the way for the industrial use of aqueous solutions of ILs. Additionally, it points toward the possibility that higher industrial impact may be obtained by directing research efforts to explore the potential of similar ILs.

CRediT authorship contribution statement

Georgina Moss: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Kyra L.S. Campbell:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Jonathan R. Howse:** Writing – review & editing, Visualization, Validation, Supervision.

Declaration of competing interest

All authors declare no conflict of interest.

Acknowledgements

This work was funded by an EPSRC Doctoral Training Partnership (DTP) Scholarship.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jil.2026.100191.

Data availability

Data will be made available on request.

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