

Physicochemical Properties of Choline Chloride/Acetic Acid as a Deep Eutectic Solvent and Its Binary Solutions with DMSO at 298.15 to 353.15 K

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Cite This: <https://doi.org/10.1021/acsomega.3c07739>

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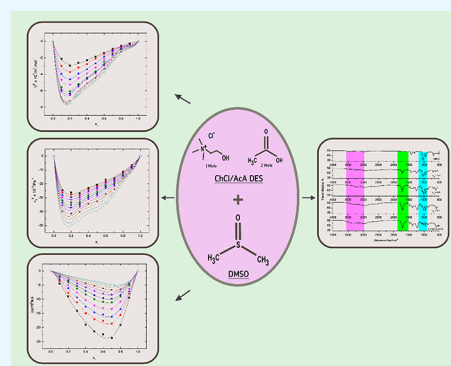


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ABSTRACT: Deep eutectic solvents (DESs) are considered to play an important role in green chemistry and other technological fields as an alternative to organic solvents. The present study reports measurements of density (ρ), speed of sound (u), dynamic viscosity (η), and electrical conductivity (κ) and investigates physicochemical properties of choline chloride/acetic acid (ChCl/AcA DES) and its binary mixtures with dimethyl sulfoxide (DMSO) over the entire composition and temperature (298.15–353.15 K) range. The density data are well fitted by a second-degree polynomial equation in T . DES/DMSO mixtures exhibit negative excess molar volume and isentropic compressibility deviation with a minimum in respective curves at $x_1 \approx 0.15$ (x_1 is the mole fraction of DES in the mixture), which became deeper with increasing temperature. The ChCl/AcA DES and DMSO curves for excess partial molar volume cross each other at $x_1 \approx 0.15$, showing that the packing effect is dominant over specific interactions. A similar behavior is observed for excess molar viscosity, showing the minima at $x_1 \approx 0.62$, and substantiates volumetric results. The temperature dependence of viscosity and conductivity is well described by the Vogel–Fulcher–Tammann (VFT) equation.



1. INTRODUCTION

A safe and healthy environment is a necessity of every society. There is a growing concern as to how to protect the environment against deterioration emerging from the large-scale use of organic solvents. This has led to an increasing demand for the application of ecofriendly processes within the framework of green and sustainable chemistry. The recognition of beneficial and interesting properties of deep eutectic solvents (DESs) has created an interest in using them and also their predecessor ionic liquids (ILs) as substitutes for conventional organic solvents.^{1–3}

The terms DESs and ILs are interchangeably used in the literature owing to the similarity in physicochemical properties. However, they differ fundamentally from each other: DESs are systems produced by the interaction of a hydrogen bond donor (HBD) and hydrogen bond acceptor (HBA) that self-associate at a certain mole ratio resulting in a eutectic phase.⁴ On the other hand, ILs are formed via a neutralization reaction and, thus, consist of ions. The eutectic phase is characterized by a melting point lower than either of the individual components, usually below 100 °C. Commonly desirable characteristics for selection of constituting components of DES are that they should have low toxicity and low cost and be biodegradable. On this basis, choline chloride, a quaternary ammonium salt, along with carboxylic acids and sugars, forms reasonable starting materials for synthesizing deep eutectic solvents. DESs

have emerged as a feasible alternative to ILs, although their “greenness” has been questionable at times in the context of their biocompatibility, biodegradability, and sustainability.^{5,6} DESs can be produced at a cheaper cost and with practical ease, which makes them promising potential candidates for industrial applications compared to their traditional predecessors ILs.³ They find applications in a wide variety of fields that include redox flow batteries,^{7–9} chemical and biosensors,^{10,11} separation processes,¹² drug solubility,¹³ environmentally benign solvents in chemical synthesis, CO₂ capture to moderate emerging risk of climate change,¹⁴ and biomass processing.^{15,16} Although there are many combinations possible out of a large store of hydrogen bond acceptors (HBAs) and hydrogen bond donors (HBDs) for the preparation of DES, the lack of understanding of the structure–property relationship imposes a limitation on predictive designing of task-specific DESs. Choline and its derivatives are often used as HBAs due to their low toxicity and wide availability. This quaternary ammonium salt can form

Received: October 5, 2023**Revised:** December 11, 2023**Accepted:** December 18, 2023

Table 1. CAS Registry Number, Source, and Purity of the Chemicals

chemical	CAS reg. no.	source	purity	purification method
choline chloride	67-48-1	Sigma-Aldrich	≥99%	used as received
acetic acid	64-19-7	Sigma-Aldrich	≥99%	used as received
dimethylsulfoxide	67-68-5	Sigma-Aldrich	≥99%	used as received (dried by 4 Å activated molecular sieves)

Table 2. Density ($\rho/\text{kg m}^{-3}$) of ChCl/AcA DES and ChCl/AcA DES–DMSO Mixtures in the Temperature Range $T = 298.15$ to 353.15 K ($x_1 = \text{Mole Fraction of ChCl/AcA DES}$) and Pressure $p = 0.1$ MPa^a

x_1/T (K)	$\rho/\text{kg m}^{-3}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	1100.36	1094.11	1087.69	1081.72	1075.47	1070.25	1065.17	1059.36	1054.44	1049.52	1045.09	1040.05
0.10	1104.96	1100.29	1095.61	1090.94	1086.27	1081.60	1076.92	1072.24	1067.55	1062.86	1058.62	1053.77
0.20	1107.48	1102.97	1098.47	1093.97	1089.46	1084.97	1080.46	1075.96	1071.45	1066.94	1062.87	1058.30
0.30	1108.14	1103.68	1099.26	1094.90	1090.59	1086.07	1081.22	1077.34	1072.29	1067.82	1064.77	1060.48
0.40	1108.92	1104.55	1100.33	1096.16	1091.82	1087.49	1083.20	1079.02	1074.84	1070.77	1067.07	1063.01
0.50	1109.44	1105.07	1101.07	1097.17	1093.08	1088.99	1084.90	1081.14	1077.09	1073.22	1069.68	1065.60
0.60	1109.96	1105.66	1101.62	1097.77	1093.70	1090.09	1086.26	1082.44	1078.66	1074.97	1071.41	1067.83
0.70	1110.39	1106.42	1102.33	1098.43	1094.56	1090.96	1087.30	1083.40	1079.81	1076.19	1072.79	1069.20
0.80	1110.99	1107.26	1103.45	1099.75	1096.02	1092.46	1088.95	1085.24	1081.88	1078.53	1075.22	1071.93
0.90	1111.74	1108.27	1104.69	1101.20	1097.64	1094.23	1090.81	1087.33	1084.00	1080.60	1077.39	1074.09
1.00	1112.45	1109.12	1105.78	1102.48	1099.18	1095.89	1092.62	1089.35	1086.10	1082.84	1079.71	1076.52

^aStandard uncertainties (u) in T , x_1 , p , and ρ are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(p) = 10$ kPa, and $u_r(\rho) = 0.003$.

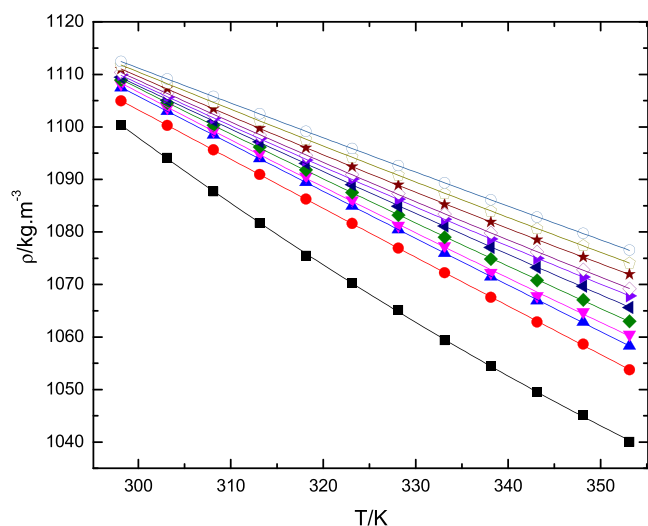


Figure 1. Temperature (T) dependence of the density (ρ) of ChCl/AcA DES–DMSO mixtures for different mole fractions of ChCl/AcA DES; $x_1 = \blacksquare$: 0.0; \bullet : 0.1; \blacktriangle : 0.2; \blacktriangledown : 0.3; \blacklozenge : 0.4; \blacktriangleleft : 0.5; \blacktriangleright : 0.6; \blacklozenge : 0.7; \star : 0.8; \triangleleft : 0.9; \circ : 1.00.

eutectic mixtures by forming complexes with hydrogen bond donors that lower the freezing point and lattice energy.^{17,18}

They are biocompatible and degrade completely under aerobic conditions.¹⁹ Acetic acid is the most important and simplest carboxylic acid and is a byproduct of fermentation. Vinegar, a food additive, is a dilute solution of acetic acid produced by the fermentation and oxidation of natural carbohydrates.²⁰ It has many industrial applications (preparation of metal acetates, plastics, cellulose acetate etc.). Acetic-acid-based DESs are extensively used in extraction and biomass pretreatment.^{21,22} Choline-chloride-based NADES have higher levels of polarity and are therefore suitable solvents for solubilizing lignocellulose. They also have the potential to replace harsh chemicals like HCl and NaOH for the pretreatment of lignocellulosic agro-residues.²³

In many cases, the relatively high viscosity of DES is an impediment in their large-scale applications.^{24,25} There is no established approach for tuning and tailoring of viscosity and other physicochemical properties entirely based on HBA and HBD chemistry.

Addition of molecular solvents (water, dimethyl sulfoxide, alcohols, and alkanes) to DES significantly decreases its

Table 3. Parameters a_ρ , b_ρ , and c_ρ Obtained by Fitting Density Data According to Eq 1 and Associated Standard Deviations

x_1	$a_\rho/\text{kg m}^{-3}$	$b_\rho/\text{kg m}^{-3} \text{ K}^{-1}$	$c_\rho/\text{kg m}^{-3} \text{ K}^{-2}$	r^2
0.00	$1820.67 \pm 2.75 \times 10^1$	$-3.53 \pm 1.69 \times 10^{-1}$	$3.75 \times 10^{-3} \pm 2.60 \times 10^{-4}$	0.9998
0.10	$1405.87 \pm 1.14 \times 10^1$	$-1.07 \pm 7.03 \times 10^{-2}$	$2.21 \times 10^{-4} \pm 1.07 \times 10^{-4}$	0.9997
0.20	$1405.73 \pm 1.09 \times 10^1$	$-1.08 \pm 6.74 \times 10^{-2}$	$2.96 \times 10^{-4} \pm 1.03 \times 10^{-4}$	0.9997
0.30	$1459.05 \pm 4.66 \times 10^1$	$-1.42 \pm 2.87 \times 10^{-1}$	$8.51 \times 10^{-4} \pm 4.40 \times 10^{-4}$	0.9993
0.40	$1445.92 \pm 1.62 \times 10^1$	$-1.37 \pm 1.00 \times 10^{-1}$	$8.26 \times 10^{-4} \pm 1.53 \times 10^{-4}$	0.9999
0.50	$1419.13 \pm 1.43 \times 10^1$	$-1.24 \pm 8.85 \times 10^{-2}$	$6.92 \times 10^{-4} \pm 1.35 \times 10^{-4}$	0.9999
0.60	$1444.68 \pm 1.14 \times 10^1$	$-1.42 \pm 7.03 \times 10^{-2}$	$1.02 \times 10^{-3} \pm 1.08 \times 10^{-4}$	0.9999
0.70	$1444.79 \pm 1.08 \times 10^1$	$-1.43 \pm 6.70 \times 10^{-2}$	$1.06 \times 10^{-3} \pm 1.02 \times 10^{-4}$	0.9999
0.80	$1440.15 \pm 7.21 \times 10^0$	$-1.43 \pm 4.43 \times 10^{-2}$	$1.11 \times 10^{-3} \pm 6.81 \times 10^{-5}$	0.9999
0.90	$1377.54 \pm 5.35 \times 10^0$	$-1.06 \pm 3.29 \times 10^{-2}$	$5.81 \times 10^{-4} \pm 5.05 \times 10^{-5}$	0.9999
1.00	$1341.26 \pm 2.37 \times 10^0$	$-0.86 \pm 1.68 \times 10^{-1}$	$3.21 \times 10^{-4} \pm 2.58 \times 10^{-5}$	1

viscosity.^{26–28} Evaluation of thermodynamic properties of binary mixtures of DES with molecular solvents in itself is an important exercise for their use in industrial processes.^{29–31} Dimethyl sulfoxide (DMSO) is a polar aprotic solvent of considerable commercial utility because of its dissolving power and structure that comprises two hydrophobic methyl groups and a strongly polar sulfoxide group.³² The relative inertness, stability at high temperature, environmental compatibility, and low toxicity of DMSO make it a very good cosolvent apart from water, which is undoubtedly the greenest and most ideal. DMSO molecules interact with other substances through van der Waals forces and hydrogen bonding.^{33,34} The presence of water in DES increases its mobility while maintaining its uniqueness in properties. The other important property is electrical conductivity, which is greatly affected by the presence of water content in DES.³⁵ For practical applications, the study of DES–water mixture compositions is desirable for a general understanding of their nature, optimization of performance, and knowledge of physicochemical and thermodynamic properties. Several publications on DES/water mixtures have been reported in the literature that evaluate the effect of water, but similar studies with DMSO are few.^{36–38}

In the present study, ChCl/AcA DES is prepared from choline chloride (ChCl) and acetic acid (AcA) in mole ratio 1:2, and volumetric (density, speed of sound) and transport (dynamic viscosity, electrical conductivity) properties of its binary mixtures with DMSO are investigated in the 298.15 to 353.15 K temperature range at ambient pressure. The VFT equation is employed to fit the viscosity and electrical conductivity data, and thermodynamic properties (excess molar volume, isentropic compressibility deviation, and intermolecular free length) are evaluated from volumetric data. The functional dependence of the volumetric and transport properties on temperature and composition is discussed.

2. EXPERIMENTAL SECTION

2.1. Materials. Details of chemicals used in this work are listed below with their CAS numbers, sources, and purity (Table 1).

2.2. Methods. The ChCl/AcA DES was prepared following the method already reported.^{3,39–41}

Choline chloride (HBA) and acetic acid (HBD) were mixed together in a 1:2 molar ratio in a round-bottom flask with constant stirring. The flask was fitted with a condenser, at the end of which a drying tube filled with silica gel was provided. Stirring was continued for 3 h at 323.15 to 333.15 K until a homogeneous, colorless, and transparent liquid of ChCl/AcA DES was formed. The liquid was allowed to cool to room temperature before storing it in a sealed bottle for further work. The moisture content of the final product (ChCl/AcA DES) was determined with coulometric Karl Fischer titration equipment (Mettler Toledo, V10S) using the Karl Fischer reagent (CombiNorm5 and methanol), which was found to be 0.06%. The moisture content of the solvent was also determined and was found to be 0.015%. The FTIR spectra of choline chloride, ChCl/AcA DES, and acetic acid were recorded on an FTIR Spectrometer (ALPHA, Bruker II). The ¹H NMR spectrum was recorded on a Bruker Advance 300 MHz spectrometer. These spectra are shown in Figures S1 and S3 (Supporting Information).

Binary solutions of ChCl/AcA DES and DMSO over the entire composition range were prepared by weighing DES in

airtight glass vials using an analytical balance (Shimadzu AUW220D, precision ±0.01 mg).

The FTIR spectra of ChCl/AcA DES, DMSO, and their binary mixtures at mole fractions 0.3, 0.5, and 0.7 were also recorded and produced in Figure S2 (Supporting Information).

A density and sound velocity meter (DSA 5000, Anton Paar) was used for density (ρ) and speed of sound (u) measurements in the 298.15 to 353.15 K range at 5 K intervals. Calibration of the instrument was checked with air and water before measurements were made. The measuring tube was thoroughly cleaned with ultrapure water, rinsed with ethanol, and dried using a blower before injecting samples.

Viscosity measurements were performed with a viscosity module (Lovis 2000, Anton Paar) attached to a DSA 5000. The stated temperature uncertainty and repeatability factor of DSA 5000 and Module 2000 were 0.01, 0.02, 0.001, and 0.005 K, respectively. Viscosity measurements were made with capillary tubes of diameter 1.59, 1.8, and 2.5 mm (Anton Paar), which were calibrated with viscosity standards (S3, S6, N100, and N415) also from Anton Paar. Electrical conductivity was measured with a conductivity meter (Mettler Toledo Seven Compact S 230). The temperature of the conductivity cell was maintained within ±0.1 K with a thermostated oil bath (Sci Finetech, FTCOB-501). The standard uncertainties in mole fractions, density, speed of sound, dynamic viscosity, and conductivities were $u(x_1) = 0.01$, $u(\rho) = 3 \times 10^{-3} \text{ kg m}^{-3}$, $u(u) = 0.50 \text{ m s}^{-1}$, $u(\eta) = 1.0\%$, and $u(\kappa) = 0.5\%$, respectively.

3. RESULTS AND DISCUSSION

3.1. Density. The measured experimental densities ($\rho/\text{kg m}^{-3}$) of ChCl/AcA DES and its binary solutions with DMSO are given in Table 2. Experimental values are fitted to a second-degree polynomial in T with eq 1, and the corresponding plots of ρ vs T are shown in Figure 1.

$$\rho = a_\rho + b_\rho T + c_\rho T^2 \quad (1)$$

The three fitting parameters a_ρ , b_ρ , and c_ρ with their standard deviation are summarized in Table 3.

The solid lines are the best fit representations of eq 1.

ρ decreases steeply with increasing temperature as a result of thermal expansion for all compositions and with increasing content of DMSO in the solution. In the last column of Table 3, r^2 values are produced that are obtained by fitting the experimental density vs temperature data with a second-order polynomial equation. The r^2 values indicate the success of the second-degree polynomial model used in the present case. Statistically, r^2 values represent the goodness of fit, and the best fit results using a quadratic function of temperature have also been reported by other authors.^{30,31}

The isobaric thermal expansion coefficient α_p is calculated using eq 2^{42,43} and tabulated in Table S2. Plots of α_p vs x_1 are shown in Figure S4.

$$\alpha_p = -\rho^{-1} \left(\frac{\partial \rho}{\partial T} \right)_p \quad (2)$$

The results show that between $0.1 \leq x_1 \leq 0.5$, α_p increases with increasing temperature, and then with further mole fractions, it decreases. However, beyond $x_1 = 0.5$, the trend in α_p values is not regular. For pure components (DES and DMSO), values of α_p decrease with increasing temperature. α_p measures the extent to which a component expands with

Table 4. Excess Molar Volume ($V^E/\text{m}^3 \text{mol}^{-1}$) of ChCl/AcA DES–DMSO Mixtures at Different Temperatures^a

x_1/T (K)	$V^E \times 10^7/\text{m}^3\text{mol}^{-1}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	0	0	0	0	0	0	0	0	0	0	0	0
0.10	-2.133	-2.982	-3.949	-4.664	-5.564	-5.847	-6.046	-6.703	-6.802	-6.904	-7.002	-7.066
0.20	-2.951	-3.721	-4.598	-5.248	-6.061	-6.330	-6.523	-7.125	-7.230	-7.337	-7.463	-7.592
0.30	-2.589	-3.174	-3.899	-4.453	-5.183	-5.294	-5.477	-5.960	-6.140	-6.198	-6.301	-6.474
0.40	-2.301	-2.760	-3.399	-3.897	-4.405	-4.502	-4.571	-5.019	-5.098	-5.259	-5.438	-5.648
0.50	-1.848	-2.111	-2.690	-3.188	-3.659	-3.779	-3.851	-4.410	-4.456	-4.637	-4.837	-4.886
0.60	-1.406	-1.515	-1.861	-2.206	-2.478	-2.792	-2.915	-3.248	-3.355	-3.535	-3.616	-3.895
0.70	-0.916	-1.048	-1.145	-1.271	-1.477	-1.661	-1.770	-1.853	-1.974	-2.073	-2.173	-2.287
0.80	-0.540	-0.647	-0.729	-0.809	-0.910	-0.984	-1.071	-1.111	-1.272	-1.452	-1.513	-1.715
0.90	-0.277	-0.371	-0.412	-0.461	-0.475	-0.522	-0.543	-0.571	-0.631	-0.651	-0.691	-0.741
1.00	0	0	0	0	0	0	0	0	0	0	0	0

^aStandard uncertainties (u) in T , x_1 , p , and ρ are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(p) = 10$ kPa, and $u(\rho) = 0.003$.

temperature, which has practical applications in various engineering processes.⁴⁴

Excess molar volume V^E calculated with eq 3 is an important parameter that provides an understanding of intermolecular forces operative within the binary system.

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \quad (3)$$

where x_1 and x_2 are mole fractions, ρ_1 and ρ_2 are densities, and M_1 and M_2 are molar masses, respectively, of DES and DMSO.⁴⁵ The molar mass of DES was calculated from eq 3a.^{46,47}

$$M_1 = M_{\text{ChCl}}x_{\text{ChCl}} + M_{\text{AcA}}x_{\text{AcA}} \quad (3a)$$

M_{ChCl} and M_{AcA} are molar masses of choline chloride and acetic acid, and x_{ChCl} and x_{AcA} are their mole fractions.

The calculated V^E values are listed in Table 4, and the corresponding plots vs x_1 are given in Figure 2.

The solid lines are the best fit representation of eq 5.

For ideal mixing of components in a binary mixture, excess molar volume V^E is expected to be close to zero, for example, as observed in the case of DMSO–*n*-propyl alcohol mixture.⁴⁸ In the present study, the DES–DMSO binary system exhibits negative excess molar volume for all compositions, which demonstrates volume contraction on mixing the components.

Generally, excess property (V^E , κ_s^E , $\Delta\eta$) values reflect the strength of intermolecular forces between the components, which could be physical (dipole–dipole interaction and dispersion forces), chemical (specific), or geometric (structural) in nature.^{31,49,50} Physical interactions are weak forces, whereas charge transfer and hydrogen bonding interactions are stronger and lead to more efficient packing in the system and volume contraction. Structural effects arise as a result of interstices within the components producing a compact structure.^{51,52} These contributions have complex and even opposing effects on one another.³⁸

The variation of V^E with x_1 produces a minimum located at $x_1 \approx 0.15$ that lies in the region rich in DMSO (83.7 wt %). The magnitude of $|V^E|$ decreases with the increase in the DES content for all compositions and temperatures investigated.

Recent studies have shown the existence of clusters in DMSO liquid and the presence of eight types of noncovalent interactions that stabilize these clusters. The CH \cdots O hydrogen bonding interaction is the strongest, and the H \cdots H bonding type is the weakest. Another study also proposes an SO \cdots S

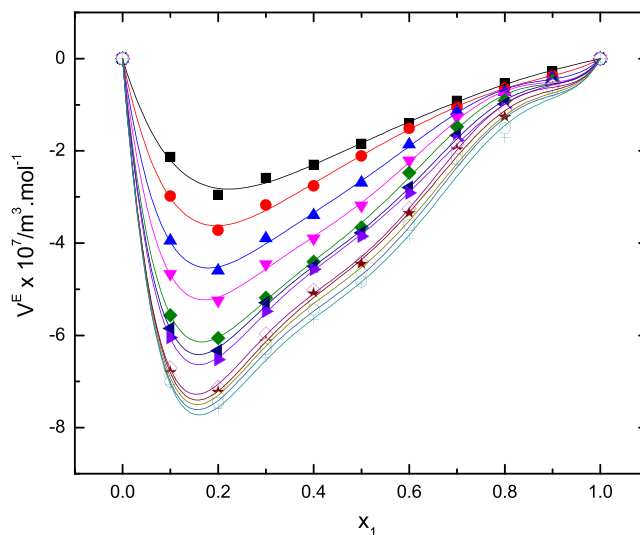


Figure 2. Variation of excess molar volume (V^E) of ChCl/AcA DES–DMSO mixtures with the mole fraction of ChCl/AcA DES (x_1) in the temperature range $298.15 \leq T/\text{K} \leq 353.15$ (■(black): 298.15 K; ●(red): 303.15 K; ▲(blue): 308.15 K; ▼(magenta): 313.15 K; ◆(olive): 318.15 K; ◀(navy): 323.15 K; ▶(violet): 328.15 K; ◇(purple): 333.15 K; ★(wine): 338.15 K; ◇(dark yellow): 343.15 K; ○(grey blue): 348.15 K; +(teal): 353.15 K).

type interaction in linear tetramers and large clusters and SO \cdots OS type in dimers and trimers.^{53–56}

These clusters should play an important part in the mixed state. Initially, when the DMSO concentration is high in the mixture, addition of DES produces additional strong H-bonds, leading to a more efficient packing of components in the mixed system. The other factor that gives negative values of V^E is accommodation of DES molecules into interstitial spaces among DMSO clusters particularly in the DMSO-rich region, which also produces compaction in the mixed state. As the DES content increases, DMSO structures in the mixture are disrupted. As a result, the DES–DMSO interaction is weakened, decreasing the absolute values of V^E .⁵⁷ The effect of increasing temperature on V^E is somewhat complex. $|V^E|$ increases with increasing temperature, more at a low concentration of DES than at high concentrations. This could likely be explained by considering that thermal expansion creates more and larger cavities leading to more efficient accommodation of the components into each other.⁴⁶

Table 5. Fit Parameters (A_j) of Redlich–Kister Eq 5 for the Excess Molar Volume (V^E) of ChCl/AcA DES–DMSO Mixtures^a

T/K	A_0	A_1	A_2	A_3	A_4	$\sigma \times 10^{-17}$
298.15	$-7.329 \times 10^{-7} \pm 2.220 \times 10^{-8}$	$9.389 \times 10^{-7} \pm 7.266 \times 10^{-8}$	$-7.688 \times 10^{-7} \pm 2.427 \times 10^{-7}$	$6.488 \times 10^{-7} \pm 1.892 \times 10^{-7}$	$-3.419 \times 10^{-7} \pm 4.264 \times 10^{-7}$	6.477
303.15	$-8.402 \times 10^{-7} \pm 1.860 \times 10^{-8}$	$1.174 \times 10^{-6} \pm 6.087 \times 10^{-8}$	$-1.051 \times 10^{-6} \pm 2.033 \times 10^{-8}$	$1.043 \times 10^{-6} \pm 1.585 \times 10^{-7}$	$-9.061 \times 10^{-7} \pm 3.573 \times 10^{-7}$	4.546
308.15	$-1.059 \times 10^{-6} \pm 2.068 \times 10^{-8}$	$1.434 \times 10^{-6} \pm 6.767 \times 10^{-8}$	$-7.509 \times 10^{-7} \pm 2.260 \times 10^{-7}$	$1.589 \times 10^{-6} \pm 1.762 \times 10^{-7}$	$-2.235 \times 10^{-6} \pm 3.972 \times 10^{-7}$	5.619
313.15	$-1.249 \times 10^{-6} \pm 2.343 \times 10^{-8}$	$1.592 \times 10^{-6} \pm 7.668 \times 10^{-8}$	$-4.396 \times 10^{-7} \pm 2.561 \times 10^{-7}$	$2.040 \times 10^{-6} \pm 1.997 \times 10^{-7}$	$-3.314 \times 10^{-6} \pm 4.500 \times 10^{-7}$	7.213
318.15	$-1.421 \times 10^{-6} \pm 2.438 \times 10^{-8}$	$1.790 \times 10^{-6} \pm 7.980 \times 10^{-8}$	$-5.736 \times 10^{-7} \pm 2.665 \times 10^{-7}$	$2.639 \times 10^{-6} \pm 2.078 \times 10^{-7}$	$-3.906 \times 10^{-6} \pm 4.684 \times 10^{-7}$	7.813
323.15	$-1.490 \times 10^{-6} \pm 1.788 \times 10^{-8}$	$1.647 \times 10^{-6} \pm 5.851 \times 10^{-8}$	$-5.824 \times 10^{-7} \pm 1.954 \times 10^{-7}$	$3.190 \times 10^{-6} \pm 1.524 \times 10^{-7}$	$-4.170 \times 10^{-6} \pm 3.434 \times 10^{-7}$	4.201
328.15	$-1.521 \times 10^{-6} \pm 1.630 \times 10^{-8}$	$1.631 \times 10^{-6} \pm 5.336 \times 10^{-8}$	$-8.110 \times 10^{-7} \pm 1.782 \times 10^{-7}$	$3.407 \times 10^{-6} \pm 1.390 \times 10^{-7}$	$-4.023 \times 10^{-6} \pm 3.132 \times 10^{-7}$	3.493
333.15	$-1.717 \times 10^{-6} \pm 2.802 \times 10^{-8}$	$1.770 \times 10^{-6} \pm 9.171 \times 10^{-8}$	$-2.927 \times 10^{-7} \pm 3.063 \times 10^{-7}$	$3.866 \times 10^{-6} \pm 2.388 \times 10^{-7}$	$-5.330 \times 10^{-6} \pm 5.382 \times 10^{-7}$	10.33
338.15	$-1.742 \times 10^{-6} \pm 2.853 \times 10^{-8}$	$1.765 \times 10^{-6} \pm 9.338 \times 10^{-8}$	$-5.969 \times 10^{-7} \pm 3.119 \times 10^{-7}$	$3.884 \times 10^{-6} \pm 2.432 \times 10^{-7}$	$-4.994 \times 10^{-6} \pm 5.481 \times 10^{-7}$	10.69
343.15	$-1.810 \times 10^{-6} \pm 3.825 \times 10^{-8}$	$1.715 \times 10^{-6} \pm 1.251 \times 10^{-7}$	$-5.401 \times 10^{-7} \pm 4.181 \times 10^{-7}$	$4.009 \times 10^{-6} \pm 3.260 \times 10^{-7}$	$-5.126 \times 10^{-6} \pm 7.347 \times 10^{-7}$	19.22
348.15	$-1.876 \times 10^{-6} \pm 4.042 \times 10^{-8}$	$1.740 \times 10^{-6} \pm 1.322 \times 10^{-7}$	$-3.893 \times 10^{-7} \pm 4.418 \times 10^{-7}$	$4.024 \times 10^{-6} \pm 3.445 \times 10^{-7}$	$-5.406 \times 10^{-6} \pm 7.764 \times 10^{-7}$	21.46
353.15	$-1.936 \times 10^{-6} \pm 4.609 \times 10^{-8}$	$1.731 \times 10^{-6} \pm 1.508 \times 10^{-7}$	$-6.264 \times 10^{-7} \pm 5.039 \times 10^{-7}$	$4.027 \times 10^{-6} \pm 3.929 \times 10^{-7}$	$-5.042 \times 10^{-6} \pm 8.854 \times 10^{-7}$	27.92

^aThe fourth-degree Redlich–Kister polynomial equation satisfactorily reproduces V^E values calculated using experimental density data as shown by the solid lines in Figure 2. Thus, a fairly good correlation is achieved between the theoretical and experimentally determined values.

Table 6. Speed of Sound ($u/m \text{ s}^{-1}$) of ChCl/AcA DES–DMSO mixtures as a Function of Temperature $T = 298.15$ to 353.15 K and Pressure $p = 0.1 \text{ MPa}$ ^a

x_1/T (K)	$u/(m \text{ s}^{-1})$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	1455.1	1438.4	1421.6	1404.9	1388.0	1371.3	1354.5	1337.7	1320.9	1304.1	1287.4	1270.6
0.10	1514.9	1498.6	1482.4	1466.2	1450.1	1433.9	1417.9	1401.9	1386.0	1370.2	1354.3	1338.4
0.20	1536.0	1520.1	1504.3	1488.5	1472.6	1456.8	1441.1	1425.3	1409.7	1394.0	1378.4	1363.0
0.30	1558.0	1542.1	1526.4	1510.7	1494.9	1479.1	1463.5	1447.8	1432.2	1416.7	1401.2	1385.8
0.40	1570.9	1555.4	1539.8	1524.4	1508.9	1493.4	1477.8	1462.3	1446.9	1431.6	1416.3	1401.1
0.50	1587.9	1572.9	1557.8	1542.8	1527.9	1513.3	1498.2	1483.7	1470.0	1455.5	1442.8	1428.6
0.60	1602.4	1587.7	1573.0	1558.3	1543.5	1528.8	1514.3	1500.7	1487.9	1475.3	1460.6	1446.0
0.70	1623.5	1609.2	1595.0	1581.0	1566.8	1552.8	1538.6	1524.5	1510.3	1496.3	1482.6	1470.0
0.80	1636.5	1622.7	1608.8	1594.9	1581.0	1567.2	1553.3	1539.4	1525.3	1511.4	1497.4	1483.4
0.90	1649.0	1636.1	1622.9	1609.5	1596.1	1583.1	1570.1	1556.8	1543.5	1530.0	1516.6	1503.3
1.00	1658.1	1645.1	1632.3	1619.3	1606.2	1593.3	1580.3	1567.3	1554.3	1541.4	1528.3	1515.4

^aStandard uncertainties (u) in T , x , P , and u are $u(T) = 0.01 \text{ K}$, $u(x_1) = 0.01$, $u(P) = 10 \text{ kPa}$, and $u_r(u) = 0.5$.

In the understanding of the solute–solute and solute–solvent interaction behavior in solutions, partial molar properties are important parameters. The calculation of the apparent molar volume $V_{\phi,i}$ (eq 4) and partial molar volume \bar{V}_i (eq 6) for individual components of the binary system is evaluated [$i = 1$ (ChCl/AcA DES), $i = 2$ (DMSO)].

$$V_{\phi,i} = V_{m,i} + \frac{V^E}{x_i} \quad (4)$$

$V_{m,i}$ is the molar volume of the pure component i . Tables S3 and S4 (Supporting Information) summarize the calculated apparent molar volumes of the components (DES and DMSO) at different temperatures. The values of $V_{\phi,i}$ are positive for both DES and DMSO, indicating strong solvent–solute interactions in binary mixtures. A representative plot of $V_{\phi,1}$ values of ChCl/AcA DES + DMSO mixtures as a function of x_1 is shown in Figure S5 (Supporting Information).

The excess molar volume V^E data are fitted to the Redlich–Kister polynomial (eq 5).^{58,59} The excess molar property ($Y^E = V^E, \kappa_s^E, \Delta\eta$) in the combined Redlich–Kister (CNIBS/R-K) model at constant temperature can be expressed by eq 5.

$$Y^E = x_{\text{DES}}x_{\text{DMSO}} \sum_{j=0}^k A_j (x_{\text{DES}} - x_{\text{DMSO}})^j \quad (5)$$

where A_j 's are coefficients that are adjustable parameters obtainable by regression analysis and collected in Table 5 and j is the degree of polynomial equation.

The partial and excess partial molar volumes \bar{V}_i 's of individual components were calculated using eqs 6 and 7, respectively,²⁶ and the results are listed in Tables S5–S8 (Supporting Information). As an example, Figure S6 (Supporting Information) shows the \bar{V}_i^E of ChCl/AcA DES and DMSO vs x_1 at 298 K. It is interesting to note that the two curves cross each other at $x_1 \approx 0.15$, where V^E exhibits its minimum. The temperature dependence of \bar{V}_i^E suggests that the packing effect is dominant over specific interactions.

$$\bar{V}_i = V_{m,i} + V^E + (1 - x_i) \left(\frac{\partial V^E}{\partial x_i} \right)_{P,T} \quad (6)$$

$$\bar{V}_i^E = \bar{V}_i - V_{m,i} \quad (7)$$

The solute–solvent interactions fade at the limit of infinite dilution. The calculation of partial molar properties at infinite dilution provides useful information about solute–solvent interactions independent of the composition effect. The partial molar volume at infinite dilution for ChCl/AcA DES (\bar{V}_1^∞) and DMSO (\bar{V}_2^∞) can be calculated by using eqs 8 and 9, respectively.

$$\bar{V}_1^\infty = V_{m,1} + \sum_{i=1}^n A_i (-1)^i \quad (8)$$

$$\bar{V}_2^\infty = V_{m,2} + \sum_{i=2}^n A_i \quad (9)$$

The calculated values from eqs 8 and 9 are listed in Table S9 (Supporting Information). The partial molar volume at infinite dilution supports the conclusion that the packing effect is dominant over specific interactions.

3.2. Speed of Sound. Apart from volumetric properties, acoustic properties of binary solutions also contribute toward investigating thermophysical properties and in the understanding of intermolecular interactions forces between components.^{60,61} The speed of sound, u , of DES and its mixtures with molecular liquids can be determined experimentally with precision with DSA5000. The measurement of the property has its significance as it relates to other properties such as density, thermal conductivity, isentropic and isothermal compressibility, and heat capacity.⁶² Experimentally determined values of u are given in Table 6 and plotted as a function of T in Figure 3.

The speed of sound decreases with increasing temperature due to the slowing down of the sound waves in the medium that has become less dense.^{63–65} The effect of increasing the DES content in the mixture results in increasing u because the medium has now become denser. The effects of varying

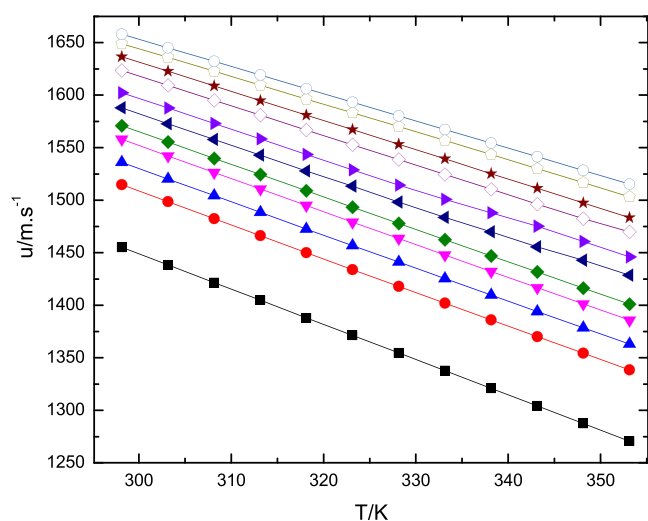


Figure 3. Temperature dependence of speed of sound (u) of the binary mixture ChCl/AcA DES + DMSO with varying x_1 = (■(black), 0.0; ●(red), 0.1; ▲(blue):0.2; ▼(magenta): 0.3; ◆(olive), 0.4; ◀(navy), 0.5; ▶(violet), 0.6; ◇(purple), 0.7; ★(wine), 0.8; ◊(dark yellow), 0.9; ○(grey blue): 1.00).

temperature and composition on the speed of sound and density are analogous.

The Newton–Laplace equation (eq 10) enables the calculation of isentropic compressibility, κ_s , of the binary mixture from the density and speed of sound data.⁶⁶ The results are reported in Table 7 as functions of temperature and compositions. The “compressibility” parameter reflects the relative change in volume of the fluid as a result of the corresponding change in pressure. The information obtainable from the experimentally determined isentropic compressibility is about the available free space in the liquid structure.⁶⁷

$$\kappa_s = \frac{1}{\rho \cdot u^2} \quad (10)$$

κ_s is considered to have contributions from solvent intrinsic and solute intrinsic isentropic compressibilities. The κ_s value of neat DMSO is $429.2 \times 10^{-12} \text{ Pa}^{-1}$ at 298.15 K. At $x_1 = 0.1$, there is large decrease of $45.1 \times 10^{-12} \text{ Pa}^{-1}$ in κ_s value compared to neat DMSO, which indicates the tightening of the DES–DMSO structure in the mixture.⁴⁰ The decrease in κ_s values continues with increasing DES content for all compositions and temperatures. The increase in κ_s with increasing temperature over the entire range suggests the loosening of the tight structure.⁶⁸ The temperature dependence of κ_s for ChCl/AcA DES is shown in Figure 4.

The isentropic compressibility deviation, κ_s^E can be calculated from the following eq 11:

$$\kappa_s^E = \kappa_s - \sum_i x_i \kappa_{s,i} \quad (11)$$

where x_i and $\kappa_{s,i}$ are mole fractions and isentropic compressibility of component i , respectively.

The calculated values of κ_s^E are collected in Table 8 as functions of composition and temperature. The plot of κ_s^E vs x_1 is shown in Figure 5. The κ_s^E parameter provides insight into the nature of the interaction within the binary liquid system. The calculated κ_s^E values are all negative over the entire range of temperatures and compositions investigated. The negative values of κ_s^E are attributed to specific and structural interactions in the liquid mixture.^{69,70} The shapes of κ_s^E vs x_1 (Figure 5) and V^E vs x_1 (Figure 2) are very similar in nature. The minimum in Figure 5 occurs at $x_1 = 0.15$,⁶⁵ which is exactly the same as found in the V^E vs x_1 plot. Moreover, the absolute values of κ_s^E decrease at all compositions and temperatures. Further, as the temperature is increased, absolute κ_s^E values increase, again more at low concentrations of DES than at high concentrations. These trends are like those observed for the excess molar volume property and deserve the same explanation.

The isentropic compressibility deviation (κ_s^E) is fitted with Redlich–Kister eq 5, and the fitting parameters are tabulated in Table 9.

The solid lines are the best fit representation of eq 5.

Intermolecular free length, L_f is calculated from eq 12. This acoustic property is used to study intermolecular interaction and its influence on structural arrangement.⁷¹ It specifies the distance between the surfaces of neighboring molecules.⁷²

$$L_f = \frac{K}{\rho u^{0.5}} \quad (12)$$

where K is Jacobson’s constant,⁷³ ρ (kg m^{-3}) is density, and u (m s^{-1}) is the speed of sound. The calculated values of L_f are shown in Table 10 and plotted in Figure 6 against x_1 . L_f decreases with an increasing concentration of DES but

Table 7. Isentropic Compressibility (κ_S/Pa^{-1}) of ChCl/AcA DES–DMSO Mixtures as a Function of Temperature (T)^a

x_1/T (K)	$\kappa_S \times 10^{12}/\text{Pa}^{-1}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	429.2	441.8	454.9	468.4	482.6	496.9	511.7	527.5	543.5	560.3	577.3	595.6
0.10	394.4	404.7	415.3	426.4	437.8	449.7	461.9	474.5	487.6	501.1	515.0	529.8
0.20	382.7	392.4	402.3	412.6	423.2	434.3	445.7	457.5	469.7	482.3	495.2	508.6
0.30	371.8	381.0	390.4	400.2	410.3	420.9	431.8	442.8	454.6	466.6	478.3	491.0
0.40	365.4	374.2	383.3	392.6	402.3	412.3	422.7	433.4	444.4	455.7	467.2	479.2
0.50	357.5	365.8	374.2	382.9	391.9	401.0	410.6	420.2	429.7	439.8	449.1	459.8
0.60	350.9	358.8	366.9	375.2	383.8	392.5	401.5	410.2	418.8	427.4	437.5	447.9
0.70	341.7	349.0	356.6	364.2	372.1	380.1	388.5	397.2	406.0	415.1	424.1	432.9
0.80	336.1	343.0	350.1	357.5	365.0	372.7	380.6	388.9	397.3	405.9	414.8	423.9
0.90	330.8	337.1	343.7	350.6	357.6	364.6	371.9	379.5	387.2	395.3	403.5	412.0
1.00	327.0	333.1	339.4	345.9	352.7	359.5	366.5	373.7	381.1	388.7	396.6	404.5

^aStandard uncertainties (u) in T , x , P , and u are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(P) = 10$ kPa, and $u_r(u) = 0.5$.

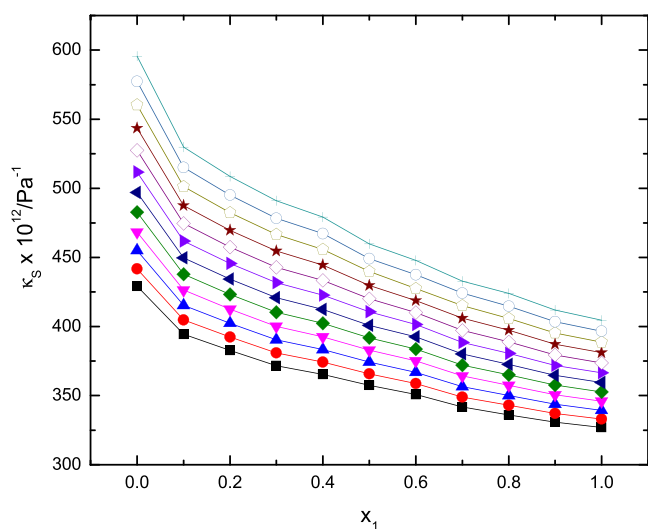


Figure 4. Isentropic compressibility ($\kappa_S \times 10^{12}/\text{Pa}^{-1}$) of ChCl/AcA DES–DMSO mixtures as a function of x_1 in the temperature range $298.15 \leq T/\text{K} \leq 353.15$ (■(black): 298.15 K; ●(red): 303.15 K; ▲(blue): 308.15 K; ▼(magenta): 313.15 K; ◆(olive): 318.15 K; ◀(navy): 323.15 K; ▶(violet): 328.15 K; ◇(purple): 333.15 K; ★(wine): 338.15 K; ◊(dark yellow): 343.15 K; ○(grey blue): 348.15 K; + (teal) : 353.15 K).

increases as the temperature is increased. The decreasing L_f values indicate the formation of more compact structures as

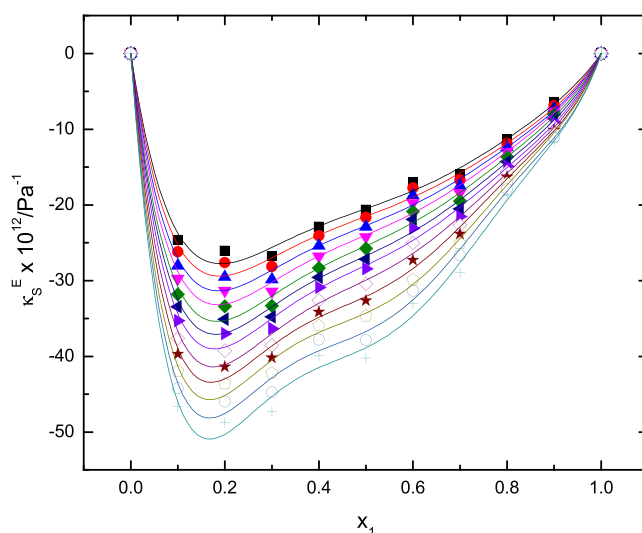


Figure 5. Isentropic compressibility deviation ($\kappa_S^E \times 10^{12}/\text{Pa}^{-1}$) of ChCl/AcA DES–DMSO mixtures as a function of x_1 in the temperature range $298.15 \leq T/\text{K} \leq 353.15$ (■(black): 298.15 K; ●(red): 303.15 K; ▲(blue): 308.15 K; ▼(magenta): 313.15 K; ◆(olive): 318.15 K; ◀(navy): 323.15 K; ▶(violet): 328.15 K; ◇(purple): 333.15 K; ★(wine): 338.15 K; ◊(dark yellow): 343.15 K; ○(grey blue): 348.15 K; + (teal) : 353.15 K).

Table 8. Isentropic compressibility deviation ($\kappa_S^E/\text{Pa}^{-1}$) of ChCl/AcA DES–DMSO Mixtures at Different Temperatures (T)^a

x_1/T (K)	$\kappa_S^E \times 10^{12}/\text{Pa}^{-1}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	0	0	0	0	0	0	0	0	0	0	0	0
0.10	-24.64	-26.20	-28.03	-29.75	-31.83	-33.47	-35.31	-37.61	-39.71	-41.96	-44.21	-46.67
0.20	-26.04	-27.64	-29.54	-31.32	-33.39	-35.09	-37.00	-39.28	-41.37	-43.63	-46.00	-48.74
0.30	-26.78	-28.16	-29.84	-31.44	-33.33	-34.79	-36.32	-38.56	-40.18	-42.20	-44.76	-47.26
0.40	-22.90	-24.09	-25.42	-26.81	-28.36	-29.58	-30.89	-32.60	-34.16	-35.96	-37.81	-39.95
0.50	-20.64	-21.67	-22.93	-24.22	-25.75	-27.20	-28.45	-30.44	-32.65	-34.67	-37.86	-40.23
0.60	-17.00	-17.77	-18.77	-19.75	-20.86	-21.93	-23.08	-25.02	-27.30	-29.89	-31.36	-33.04
0.70	-15.96	-16.70	-17.49	-18.42	-19.50	-20.54	-21.55	-22.67	-23.86	-25.12	-26.69	-28.98
0.80	-11.33	-11.88	-12.41	-12.94	-13.63	-14.27	-14.90	-15.61	-16.29	-17.10	-17.92	-18.77
0.90	-6.397	-6.903	-7.285	-7.623	-8.044	-8.553	-9.099	-9.594	-10.12	-10.51	-11.12	-11.64
1.00	0	0	0	0	0	0	0	0	0	0	0	0

^aStandard uncertainties (u) in T , x , P , and u are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(P) = 10$ kPa, and $u_r(u) = 0.5$.

Table 9. Fit Parameters (A_i) of the Redlich–Kister Equation (Eq 5) for the Isentropic Compressibility Deviation (κ_S^E) of ChCl/AcA DES–DMSO Mixtures

T/K	A_0	A_1	A_2	A_3	A_4	$\sigma \times 10^{-24}$
298.15	$-8.200 \times 10^{-11} \pm 3.038 \times 10^{-12}$	$4.401 \times 10^{-11} \pm 9.945 \times 10^{-12}$	$-7.921 \times 10^{-11} \pm 3.322 \times 10^{-11}$	$1.162 \times 10^{-10} \pm 2.590 \times 10^{-11}$	$-8.821 \times 10^{-11} \pm 5.837 \times 10^{-11}$	1.213
303.15	$-8.607 \times 10^{-11} \pm 3.121 \times 10^{-12}$	$4.713 \times 10^{-11} \pm 1.021 \times 10^{-11}$	$-8.208 \times 10^{-11} \pm 3.411 \times 10^{-11}$	$1.226 \times 10^{-10} \pm 2.660 \times 10^{-11}$	$-1.016 \times 10^{-10} \pm 5.994 \times 10^{-11}$	1.280
308.15	$-9.095 \times 10^{-11} \pm 3.184 \times 10^{-12}$	$5.061 \times 10^{-11} \pm 1.042 \times 10^{-11}$	$-8.534 \times 10^{-11} \pm 3.481 \times 10^{-11}$	$1.330 \times 10^{-10} \pm 2.714 \times 10^{-11}$	$-1.144 \times 10^{-10} \pm 6.116 \times 10^{-11}$	1.332
313.15	$-9.594 \times 10^{-11} \pm 3.318 \times 10^{-12}$	$5.335 \times 10^{-11} \pm 1.085 \times 10^{-11}$	$-8.844 \times 10^{-11} \pm 3.627 \times 10^{-11}$	$1.435 \times 10^{-10} \pm 2.828 \times 10^{-11}$	$-1.249 \times 10^{-10} \pm 6.373 \times 10^{-11}$	1.447
318.15	$-1.016 \times 10^{-10} \pm 3.535 \times 10^{-12}$	$5.634 \times 10^{-11} \pm 1.157 \times 10^{-11}$	$-9.257 \times 10^{-11} \pm 3.864 \times 10^{-11}$	$1.561 \times 10^{-10} \pm 3.013 \times 10^{-11}$	$-1.379 \times 10^{-10} \pm 6.790 \times 10^{-11}$	1.642
323.15	$-1.069 \times 10^{-10} \pm 3.646 \times 10^{-12}$	$5.738 \times 10^{-11} \pm 1.193 \times 10^{-11}$	$-9.276 \times 10^{-11} \pm 3.986 \times 10^{-11}$	$1.673 \times 10^{-10} \pm 3.108 \times 10^{-11}$	$-1.541 \times 10^{-10} \pm 7.003 \times 10^{-11}$	1.747
328.15	$-1.119 \times 10^{-10} \pm 3.633 \times 10^{-12}$	$5.887 \times 10^{-11} \pm 1.189 \times 10^{-11}$	$-9.601 \times 10^{-11} \pm 3.971 \times 10^{-11}$	$1.798 \times 10^{-10} \pm 3.097 \times 10^{-11}$	$-1.688 \times 10^{-10} \pm 6.978 \times 10^{-11}$	1.735
333.15	$-1.197 \times 10^{-10} \pm 3.617 \times 10^{-12}$	$6.073 \times 10^{-11} \pm 1.183 \times 10^{-11}$	$-9.143 \times 10^{-11} \pm 3.954 \times 10^{-11}$	$1.974 \times 10^{-10} \pm 3.083 \times 10^{-11}$	$-1.944 \times 10^{-10} \pm 6.947 \times 10^{-11}$	1.719
338.15	$-1.283 \times 10^{-10} \pm 3.304 \times 10^{-12}$	$5.820 \times 10^{-11} \pm 1.081 \times 10^{-11}$	$-7.563 \times 10^{-11} \pm 3.611 \times 10^{-11}$	$2.210 \times 10^{-10} \pm 2.816 \times 10^{-11}$	$-2.347 \times 10^{-10} \pm 6.346 \times 10^{-11}$	1.434
343.15	$-1.371 \times 10^{-10} \pm 3.083 \times 10^{-12}$	$5.612 \times 10^{-11} \pm 1.009 \times 10^{-11}$	$-6.765 \times 10^{-11} \pm 3.370 \times 10^{-11}$	$2.463 \times 10^{-10} \pm 2.628 \times 10^{-11}$	$-2.616 \times 10^{-10} \pm 5.922 \times 10^{-11}$	1.249
348.15	$-1.468 \times 10^{-10} \pm 3.933 \times 10^{-12}$	$5.993 \times 10^{-11} \pm 1.287 \times 10^{-11}$	$-5.685 \times 10^{-11} \pm 4.300 \times 10^{-11}$	$2.582 \times 10^{-10} \pm 3.353 \times 10^{-11}$	$-2.941 \times 10^{-10} \pm 7.556 \times 10^{-11}$	2.033
353.15	$-1.554 \times 10^{-10} \pm 4.463 \times 10^{-12}$	$6.038 \times 10^{-11} \pm 1.460 \times 10^{-11}$	$-6.589 \times 10^{-11} \pm 4.879 \times 10^{-11}$	$2.798 \times 10^{-10} \pm 3.805 \times 10^{-11}$	$-2.977 \times 10^{-10} \pm 8.574 \times 10^{-11}$	2.618

Table 10. Intermolecular Free Length (L_f /m) of ChCl/AcA DES–DMSO Mixtures as a Function of Mole Fraction (x_1)^a

x_1/T (K)	$L_f \times 10^{11}/m$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	4.076	4.171	4.270	4.370	4.474	4.578	4.685	4.796	4.908	5.024	5.142	5.264
0.10	3.907	3.992	4.080	4.169	4.261	4.355	4.451	4.549	4.649	4.752	4.856	4.965
0.20	3.849	3.931	4.015	4.101	4.189	4.280	4.372	4.466	4.563	4.662	4.762	4.865
0.30	3.793	3.874	3.956	4.039	4.125	4.213	4.303	4.394	4.489	4.585	4.680	4.780
0.40	3.761	3.839	3.919	4.001	4.084	4.170	4.258	4.347	4.438	4.531	4.625	4.722
0.50	3.720	3.796	3.873	3.951	4.031	4.112	4.196	4.280	4.364	4.451	4.535	4.626
0.60	3.685	3.759	3.834	3.911	3.989	4.069	4.149	4.229	4.308	4.388	4.476	4.565
0.70	3.637	3.708	3.780	3.854	3.928	4.004	4.082	4.161	4.242	4.324	4.407	4.488
0.80	3.607	3.675	3.746	3.818	3.891	3.964	4.040	4.118	4.197	4.277	4.358	4.441
0.90	3.578	3.644	3.711	3.780	3.851	3.922	3.994	4.068	4.143	4.220	4.299	4.378
1.00	3.557	3.622	3.688	3.755	3.824	3.894	3.964	4.037	4.110	4.185	4.261	4.338

^aStandard uncertainties (u) in T , x , P , and u are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(P) = 10$ kPa, and $u_r(u) = 0.5$.

the DES concentration is increased. The increase in L_f on increasing temperature is due to thermal agitation in the liquid mixture resulting in more free space in DES.⁵⁵ The variation of L_f with the temperature and DES concentration reinforces the results concluded from κ_S^E in terms of intermolecular interactions.

Acoustic impedance (Z) is the physical property that describes the resistance faced by an ultrasound beam while passing through the medium.⁷⁴ The value of Z is calculated by eq 13.

$$Z = \rho \cdot u \quad (13)$$

where ρ (kg m^{-3}) is density and u (m s^{-1}) is the speed of sound. The calculated values of Z (eq 13) are listed in Table 11 and plotted in Figure 7.

The dependence of Z and L_f parameters on DES concentration and temperature is reversed. The value of Z increases with increasing concentration of DES at all temperatures, indicating strong interactions among components of the liquid mixture.^{71,75}

3.3. Viscosity. The knowledge of viscosity (η) an important transport property of fluids is desirable because of its practical applications in designing process equipment, mass transfer, and reaction rate calculations.⁷⁶ Factors that influence η of DESs are temperature, mole ratio, water content and molecular weight.⁷⁷ Because most DESs are relatively more viscous than conventional organic solvents, it becomes necessary that their mixtures with molecular liquids (e.g., water, DMSO, alcohols) should be used in industrial processes.⁷⁸

In this study, the dynamic viscosity of ChCl/AcA DES and its binary mixtures with DMSO is measured in the 298.15 to 353.15 K range, and the values are reported in Table 12. Figure 8 shows the plot of $\ln \eta$ against T .

Table 12 shows that viscosity of the mixture increases with DES mole fraction at all temperatures, but it decreases as the temperature is increased for all compositions.⁷⁹

The Vogel–Fulcher–Tammann (VFT) model (eq 14) is applied to investigate the temperature dependence of transport

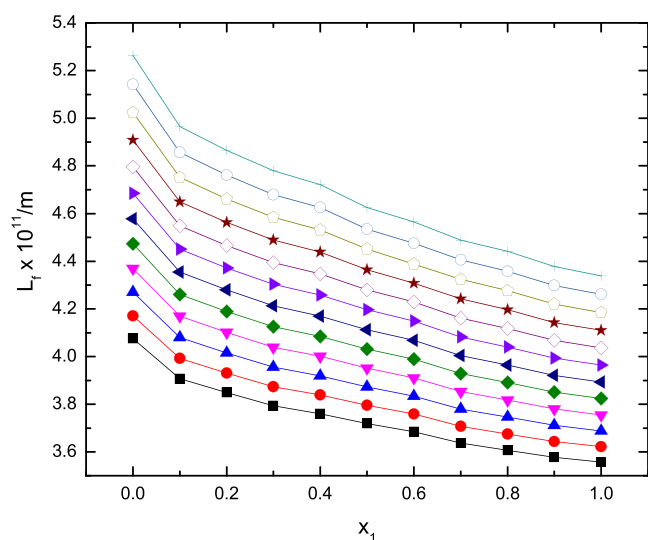


Figure 6. Inter-molecular free length ($L_f \times 10^{11}/\text{m}$) of ChCl/AcA DES–DMSO mixtures as a function of x_1 in the temperature range $298.15 \leq T/\text{K} \leq 353.15$ (■(black): 298.15 K; ●(red): 303.15 K; ▲(blue): 308.15 K; ▼(magenta): 313.15 K; ◆(olive): 318.15 K; ◀(navy): 323.15 K; ▶(violet): 328.15 K; ◇(purple): 333.15 K; ★(wine): 338.15 K; ◊(dark yellow): 343.15 K; ○(grey blue): 348.15 K; + (teal) : 353.15 K).

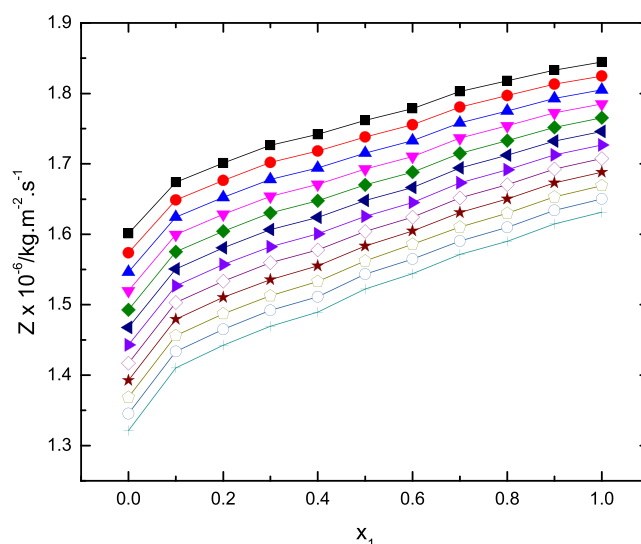


Figure 7. Acoustic impedance ($Z \times 10^{-6}/\text{kg m}^{-2} \text{s}^{-1}$) of ChCl/AcA DES–DMSO mixtures as a function of x_1 in the temperature range $298.15 \leq T/\text{K} \leq 353.15$ (■(black): 298.15 K; ●(red): 303.15 K; ▲(blue): 308.15 K; ▼(magenta): 313.15 K; ◆(olive): 318.15 K; ◀(navy): 323.15 K; ▶(violet): 328.15 K; ◇(purple): 333.15 K; ★(wine): 338.15 K; ◊(dark yellow): 343.15 K; ○(grey blue): 348.15 K; + (teal) : 353.15 K).

property, Y , of viscous liquids close to their glass transition temperature.^{26,80}

$$\ln\left(\frac{Y}{Y^0}\right) = \ln\left(\frac{A_Y}{Y^0}\right) + \frac{B_Y}{(T - T_{0,Y})} \quad (14)$$

where Y is viscosity η ($Y^0 = \eta^0 = 1 \text{ mPa}^{-1}$) or resistivity K^{-1} ($Y^0 = \frac{1}{\kappa^0} = 1 \text{ S}^{-1} \text{ m}$) and A_Y , B_Y , and $T_{0,Y}$ are adjustable parameters. The experimental data obtained at various temperatures are fitted with eq 14 by adjusting the VFT parameters.⁸¹ The VFT equation explains that the system's dynamics is slowed down on approaching the critical VFT (or Vogel) temperature, $T_{0,Y}$, which is usually 10–15 K below the calorimetric glass transition temperature of the system. A_Y represents the property Y (viscosity or resistivity) as $T \rightarrow \infty$, the quantity $E_a = B_Y \times R$ (where R is a general gas constant) is the pseudo activation energy, and $T_{0,Y}$ is the Vogel temper-

ature. The calculated values of these fitting parameters are listed in Table 13.

The solid lines are the best fit representation of eq 14.

The Arrhenius equation is also used to correlate the temperature dependence of viscosity (η) and electrical conductivity (κ) of ChCl/AcA DES and its binary mixtures with DMSO. The calculated parameters are shown in Tables S11 and S12 and plotted as a function of $1/T$ in Figures S7 and S8, respectively (Supporting Information).

The viscosity deviation, $\Delta\eta$, is calculated from eq 15 for the viscous fluids.

$$\Delta\eta = \eta - \sum_i x_i \eta_i \quad (15)$$

where x_i and η_i are mole fractions and dynamic viscosity of component i , respectively. Calculated values of $\Delta\eta$ are collected in Table 14, and its plot as a function of x_1 is shown in Figure 9 for all temperatures investigated.

Table 11. Acoustic Impedance ($Z/\text{kg m}^{-2} \text{s}^{-1}$) of ChCl/AcA DES–DMSO Mixtures as a Function of Mole Fraction (x_1)^a

x_1/T (K)	$Z \times 10^{-6}/\text{kg m}^{-2} \text{s}^{-1}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	1.601	1.574	1.546	1.520	1.493	1.468	1.443	1.417	1.393	1.369	1.345	1.321
0.10	1.674	1.649	1.624	1.600	1.575	1.551	1.527	1.503	1.480	1.456	1.434	1.410
0.20	1.701	1.677	1.652	1.628	1.604	1.581	1.557	1.534	1.510	1.487	1.465	1.442
0.30	1.726	1.702	1.678	1.654	1.630	1.606	1.582	1.560	1.536	1.513	1.492	1.470
0.40	1.742	1.718	1.694	1.671	1.647	1.624	1.601	1.578	1.555	1.533	1.511	1.489
0.50	1.762	1.738	1.715	1.693	1.670	1.648	1.625	1.604	1.583	1.562	1.543	1.522
0.60	1.779	1.755	1.733	1.711	1.688	1.667	1.645	1.624	1.605	1.586	1.565	1.544
0.70	1.803	1.780	1.758	1.737	1.715	1.694	1.673	1.652	1.631	1.610	1.590	1.572
0.80	1.818	1.797	1.775	1.754	1.733	1.712	1.691	1.671	1.650	1.630	1.610	1.590
0.90	1.833	1.813	1.793	1.772	1.752	1.732	1.713	1.693	1.673	1.653	1.634	1.615
1.00	1.845	1.825	1.805	1.785	1.765	1.746	1.727	1.707	1.688	1.669	1.650	1.631

^aStandard uncertainties (u) in T , x , P , and u are $u(T) = 0.01 \text{ K}$, $u(x_1) = 0.01$, $u(P) = 10 \text{ kPa}$, and $u(u) = 0.5$.

Table 12. Dynamic Viscosity ($\eta/\text{mPa s}$) of ChCl/AcA DES–DMSO Mixtures as a Function of Temperature $T = 298.15$ to 353.15 K and Pressure $p = 0.1$ MPa^a

x_1/T (K)	$\eta/\text{mPa s}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	1.856	1.751	1.650	1.510	1.390	1.280	1.190	1.110	1.030	0.948	0.901	0.857
0.10	3.030	2.707	2.433	2.198	1.997	1.820	1.674	1.544	1.433	1.336	1.244	1.141
0.20	4.119	3.637	3.237	2.895	2.609	2.365	2.154	1.970	1.805	1.660	1.533	1.406
0.30	4.596	4.123	3.696	3.339	3.038	2.779	2.557	2.366	2.199	2.053	1.924	1.806
0.40	5.228	4.693	4.260	3.886	3.589	3.286	3.025	2.829	2.642	2.456	2.323	2.220
0.50	8.476	7.067	6.075	5.127	4.503	3.985	3.633	3.374	3.190	2.956	2.850	2.726
0.60	13.00	11.03	9.150	7.420	6.235	5.243	4.579	4.080	3.776	3.512	3.299	3.180
0.70	17.34	14.52	12.13	10.17	8.646	7.296	6.149	5.352	4.740	4.309	3.889	3.704
0.80	27.57	22.06	18.64	16.01	13.37	10.82	9.231	7.607	6.404	5.605	4.964	4.485
0.90	42.86	34.13	28.76	23.70	20.01	17.24	14.32	12.10	10.33	8.930	7.773	6.900
1.00	57.88	46.62	39.79	33.14	27.64	23.89	20.26	17.93	15.89	13.98	12.59	11.59

^aStandard uncertainties (u) in T , x , P , and η are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(P) = 10$ kPa, and $u_r(\eta) = 0.01$.

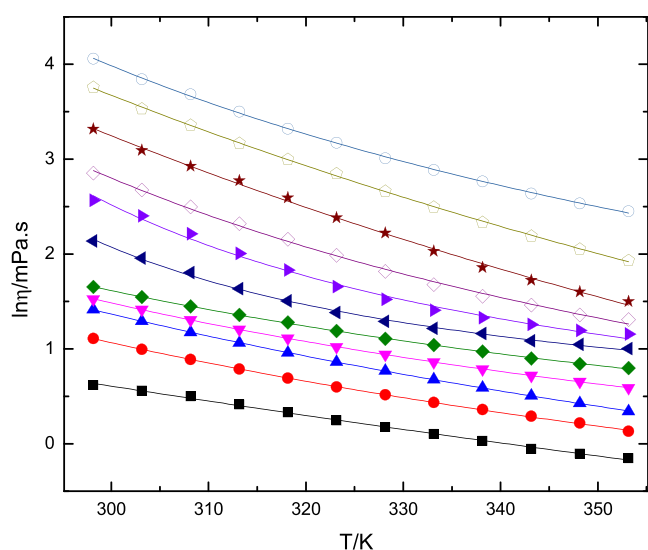


Figure 8. Temperature and composition dependence of viscosity η for pure ChCl/AcA DES, DMSO, and their binary mixtures measured at atmospheric pressure; $x_1 =$ (■(black): 0.0; ●(red): 0.1; ▲(blue): 0.2; ▼(magenta): 0.3; ◆(olive): 0.4; ◀(navy): 0.5; ▶(violet): 0.6; ◇(purple): 0.7; ★(wine): 0.8; ◊(dark yellow): 0.9; ○(grey blue): 1.00).

Table 13. VFT Parameters from Eq 14 for Dynamic Viscosity η and Associated Standard Deviations: $\eta^0 = 1$ mPa s

x_1	$\ln\left(\frac{A_\eta}{\eta^0}\right)$	B_η/K	$T_{0,\eta}/\text{K}$
0.00	-8.886 ± 5.682	5642.7 ± 7030.4	-294.35 ± 384.18
0.10	-2.889 ± 0.235	691.4 ± 93.21	125.15 ± 13.22
0.20	-3.384 ± 0.177	924.08 ± 77.14	105.51 ± 9.027
0.30	-1.734 ± 0.054	445.23 ± 17.38	161.72 ± 3.095
0.40	-1.380 ± 0.158	417.83 ± 51.54	160.46 ± 9.860
0.50	0.001 ± 0.094	99.65 ± 12.67	251.87 ± 3.971
0.60	-0.487 ± 0.298	180.49 ± 47.60	239.95 ± 9.997
0.70	-1.930 ± 0.550	522.41 ± 145.73	189.56 ± 18.08
0.80	-6.900 ± 2.414	2525.9 ± 1317.3	51.06 ± 70.86
0.90	-6.124 ± 1.033	2385.6 ± 552.54	56.50 ± 30.82
1.00	-1.103 ± 0.304	616.39 ± 87.25	178.84 ± 9.966

The $\Delta\eta$ values are negative for all compositions and temperatures and reflect the strength of interaction between components of the mixture.^{82,83}

Negative $\Delta\eta$ values are associated with differences in shape and size of the component molecules and loss of dipolar association, whereas specific interactions such as H-bonding and charge transfer complex formation led to positive contribution to $\Delta\eta$.⁷⁶

Figure 9 shows that a minimum occurs at about $x_1 \approx 0.62$, which lies in the region rich in DES. With increasing DES content, interaction forces are progressively weakening up to the minimum limit. The behavior is more prominent at lower temperatures, which could be correlated with the size and shapes of the component molecules. At $x_1 > 0.62$, absolute values of $\Delta\eta$ decrease, which indicate the dominance of specific forces in the system. Negative $\Delta\eta$ values support the presence of a weak interaction between DES and DMSO molecules and the breaking of self-associated structures of DMSO molecules. The calculated parameters from the Redlich–Kister equation for $\Delta\eta$ are listed in Table 15.

3.4. Electrical Conductivity. Knowledge of electrical conductivity and κ of DESs is required for their industrial applications in the design, control, and optimization of electrochemical processes. DESs, which are usually highly viscous fluids, exhibit low ionic conductivity ($\kappa < 2$ mS cm⁻¹ at room temperature).^{84,85} The κ increases with an increase in temperature due to the resulting decrease in viscosity. In a less viscous medium, the mobility of charged molecular species increases.

The κ of ChCl/AcA DES and its binary mixtures with DMSO was measured in the $T = 298.15$ to 353.15 K range over the entire composition range. Experimentally measured κ values are reported in Table 16, and the plots of $\ln \kappa^{-1}$ vs T and κ vs x_1 are shown in Figures 10 and 11, respectively.

The solid lines are the best fit representation of eq 14.

Table 16 shows that κ has increased with the increase in temperature for all compositions, which is attributed to the decrease in viscosity (Table 12). κ of the DES increased with increase in DES content in the mixture, which is in opposition to the observed increase in viscosity. The increase in conductivity following an increase in temperature is due to the greater mobility of charges as a result of the weakening of intermolecular forces. The concentration effect could be due to the increase in the number of charge species. The experimental

Table 14. Viscosity Deviation $\Delta\eta$ of ChCl/AcA DES–DMSO Binary Mixtures as a Function of ChCl/AcA DES Mole Fraction x_1 in the Temperature Range $298.15 \leq T/\text{K} \leq 353.15^a$

x_1/T (K)	$\Delta\eta/\text{mPa s}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.00	0	0	0	0	0	0	0	0	0	0	0	0
0.10	-4.428	-3.531	-3.031	-2.475	-2.018	-1.721	-1.423	-1.248	-1.083	-0.915	-0.826	-0.789
0.20	-8.942	-7.748	-6.041	-4.941	-4.031	-3.437	-2.850	-2.504	-2.197	-1.894	-1.706	-1.598
0.30	-14.07	-11.09	-9.396	-7.660	-6.227	-5.284	-4.354	-3.790	-3.289	-2.804	-2.483	-2.271
0.40	-19.04	-15.01	-12.65	-10.28	-8.301	-7.038	-5.793	-5.009	-4.333	-3.705	-3.253	-2.931
0.50	-21.39	-17.12	-14.65	-12.20	-10.01	-8.600	-7.092	-6.146	-5.271	-4.508	-3.895	-3.499
0.60	-22.47	-17.64	-15.38	-13.07	-10.91	-9.603	-8.053	-7.122	-6.171	-5.255	-4.614	-4.118
0.70	-23.73	-18.64	-16.21	-13.48	-11.12	-9.811	-8.391	-7.533	-6.693	-5.761	-5.193	-4.668
0.80	-19.11	-15.59	-13.52	-10.80	-9.020	-8.548	-7.216	-6.960	-6.515	-5.769	-5.287	-4.960
0.90	-9.418	-8.003	-7.216	-6.277	-5.005	-4.389	-4.034	-4.149	-4.080	-3.746	-3.648	-3.618
1	0	0	0	0	0	0	0	0	0	0	0	0

^aStandard uncertainties (u) in T , x , P , and η are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(P) = 10$ kPa, and $u_r(\eta) = 0.01$.

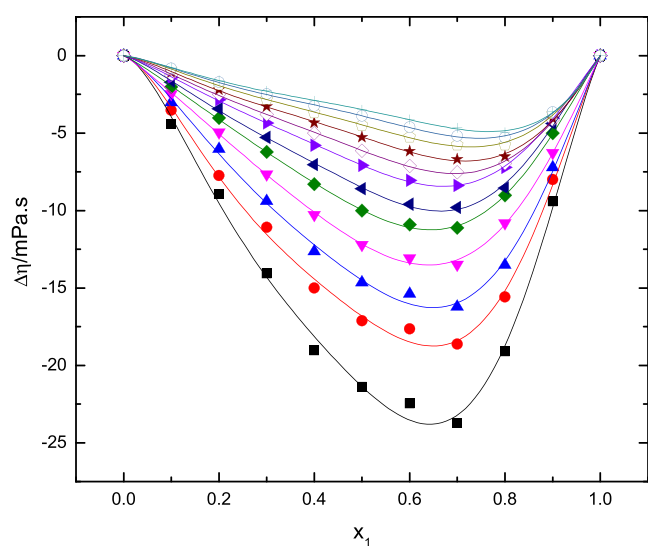


Figure 9. Viscosity deviation $\Delta\eta$ of ChCl/AcA DES–DMSO binary mixtures as a function of x_1 in the temperature range $298.15 \leq T/\text{K} \leq 353.15$ (■(black): 298.15 K; ●(red): 303.15 K; ▲(blue): 308.15 K; ▼(magenta): 313.15 K; ◆(olive): 318.15 K; ◀(navy): 323.15 K; ▶(violet): 328.15 K; ◇(purple): 333.15 K; ★(wine): 338.15 K; ◊(dark yellow): 343.15 K; ○(grey blue): 348.15 K; +(teal): 353.15 K).

κ data were fitted with the VFT eq 14, and the calculated fitting parameter are listed in Table 17.

The B_κ and $T_{0,\kappa}$ values in Table 16 show considerable scattering as with the corresponding values obtained for the viscosity data.

The dependence of κ on the composition of the mixture is explained by empirical Casteel–Amis eq 16.^{46,86,87}

$$\kappa = \kappa_{\max} \left(\frac{x_{\text{DES}}}{x_{\max}} \right)^a \exp \left[b(x_{\text{DES}} - x_{\max})^2 - a \frac{x_{\text{DES}} - x_{\max}}{x_{\max}} \right] \quad (16)$$

where x_{\max} is the mole fraction of DES at κ_{\max} , the maximum conductivity, and a and b are adjustable parameters without any specific meanings. Figure 11 shows that conductivity increases exponentially with an increase in mole fraction of DES particularly at higher temperatures. The resulting fitting parameters of eq 16 are listed in Table 18.

The solid lines are the best fit representations of eq 16.

The correlation between conductivity and viscosity is explained in terms of the (1) reduction in the number of charge carriers due to aggregate formation and (2) reduced mobility of charge carriers due to high viscosity. The former effect is the major factor for the variation of κ .^{88,89}

4. CONCLUSIONS

Choline chloride is a bifunctional quaternary ammonium salt consisting of choline cation and chloride as counterion.

Table 15. Fit parameters (A_j) of the Redlich–Kister Equation (Eq 5) for Viscosity Deviation $\Delta\eta$ of ChCl/AcA DES–DMSO Mixtures

T/K	A_0	A_1	A_2	A_3	A_4	σ
298.15	-85.46 ± 1.991	-57.17 ± 6.518	-36.66 ± 21.77	25.81 ± 16.97	79.36 ± 38.25	0.521
303.15	-67.30 ± 1.476	-42.88 ± 4.833	-34.41 ± 16.14	12.89 ± 12.59	59.74 ± 28.36	0.287
308.15	-58.02 ± 1.129	-40.23 ± 3.696	-22.82 ± 12.34	11.98 ± 9.628	38.42 ± 21.69	0.168
313.15	-48.64 ± 0.569	-34.59 ± 1.863	-8.821 ± 6.224	11.73 ± 4.853	14.98 ± 10.93	0.043
318.15	-39.88 ± 0.318	-30.62 ± 1.043	-9.727 ± 3.487	14.32 ± 2.719	17.78 ± 6.127	0.013
323.15	-34.07 ± 0.477	-29.87 ± 1.561	-18.04 ± 5.216	14.86 ± 4.067	27.65 ± 9.165	0.030
328.15	-28.29 ± 0.208	-25.66 ± 0.683	-15.91 ± 2.282	10.48 ± 1.779	19.89 ± 4.010	0.006
333.15	-24.50 ± 0.253	-23.24 ± 0.830	-19.20 ± 2.774	3.311 ± 2.163	16.18 ± 4.874	0.008
338.15	-20.99 ± 0.294	-20.32 ± 0.962	-21.38 ± 3.215	-2.467 ± 2.507	14.01 ± 5.649	0.011
343.15	-17.88 ± 0.276	-17.09 ± 0.904	-19.23 ± 3.022	-5.503 ± 2.357	9.764 ± 5.311	0.010
348.15	-15.55 ± 0.186	-14.92 ± 0.611	-19.45 ± 2.042	-8.345 ± 1.592	7.297 ± 3.589	0.005
353.15	-13.91 ± 0.178	-12.57 ± 0.585	-18.15 ± 1.956	-11.92 ± 1.525	2.024 ± 3.437	0.004

Table 16. Electrical Conductivity ($\kappa/S\text{ m}^{-1}$) of ChCl/AcA DES–DMSO Mixtures as a Function of Temperature $T = 298.15$ to 353.15 K and Pressure $p = 0.1$ MPa^a

x_1/T (K)	$\kappa/S\text{ m}^{-1}$											
	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.10	0.552	0.573	0.591	0.606	0.623	0.644	0.663	0.687	0.716	0.738	0.762	0.789
0.20	0.849	0.874	0.897	0.926	0.950	0.978	1.011	1.051	1.084	1.127	1.156	1.197
0.30	1.004	1.045	1.079	1.123	1.152	1.198	1.249	1.301	1.357	1.421	1.478	1.538
0.40	1.123	1.178	1.216	1.267	1.317	1.372	1.429	1.500	1.567	1.672	1.737	1.822
0.50	1.230	1.281	1.358	1.405	1.452	1.523	1.603	1.689	1.758	1.876	1.938	2.047
0.60	1.318	1.382	1.448	1.534	1.599	1.691	1.784	1.844	1.968	2.099	2.242	2.357
0.70	1.338	1.417	1.484	1.600	1.669	1.792	1.886	2.026	2.135	2.324	2.484	2.661
0.80	1.427	1.524	1.609	1.688	1.773	1.893	2.010	2.137	2.312	2.473	2.664	2.776
0.90	1.516	1.575	1.706	1.802	1.912	2.053	2.212	2.359	2.535	2.699	2.884	3.122
1.00	1.666	1.771	1.847	1.939	2.080	2.219	2.383	2.527	2.712	2.890	3.053	3.253

^aStandard uncertainties (u) in T , x_1 , p , and κ are $u(T) = 0.01$ K, $u(x_1) = 0.01$, $u(P) = 10$ kPa, and $u_r(\kappa) = 0.005$.

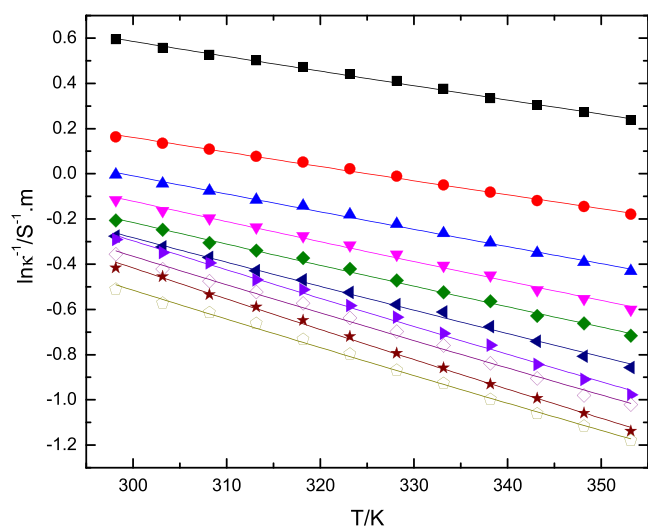


Figure 10. Temperature and composition dependence of the electrical conductivity κ of pure ChCl/AcA DES and its binary mixtures with DMSO; $x_1 =$ (■(black): 0.1; ●(red): 0.2; ▲(blue): 0.3; ▼(magenta): 0.4; ◆(olive): 0.5; ◀(navy): 0.6; ▶(violet): 0.7; ◇(purple): 0.8; ★(wine): 0.9; ◊(dark yellow): 1.00).

Choline cation occurs in nature. Choline chloride is a major industrial product with a market value of 662.1 million US dollars in 2022 that is projected to increase further in future. Acetic acid is a byproduct of fermentation. The present study reports the preparation of ChCl/AcA DES using choline chloride and acetic acid and investigation of its physicochemical properties along with its binary mixtures with DMSO in the temperature range $T = 298.15$ to 353.15 K for all compositions. The regression analysis of experimental density data is fitted with a second-degree polynomial in T . The statistical r^2 values of the fit are collected in Table 3, which shows the goodness of the fit and success of the model. Excess properties (V^E , κ_S^E , and $\Delta\eta$) calculated from the respective properties of the pure DES and its binary mixtures with DMSO for all temperatures and compositions are fitted with the Redlich–Kister polynomial equation. The negative values of V^E are attributed to the presence of specific interactions or the packing effect between components of the mixture. The minima in the V^E vs x_1 curves lie in the DMSO-rich region at $x_1 \approx 0.15$, which indicates stronger interaction between the components of the mixture at this composition. This behavior

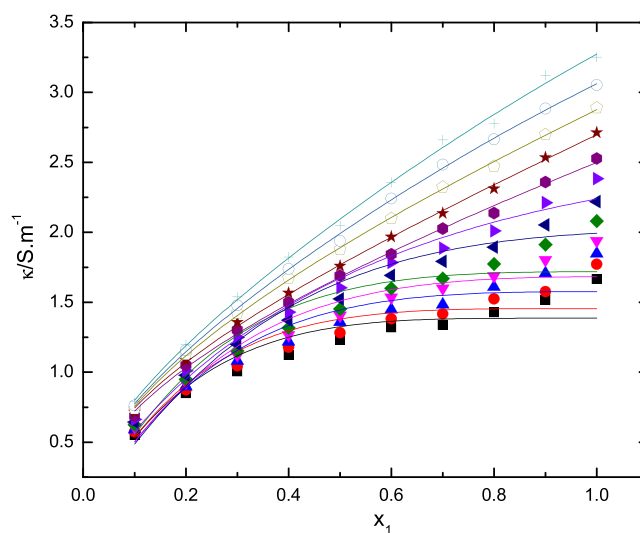


Figure 11. Variation of electrical conductivity κ of ChCl/AcA DES–DMSO mixtures with mole fraction of ChCl/AcA DES x_1 in the temperature range $283.15 \leq T/K \leq 353.15$ (■(black): 298.15 K; ●(red): 303.15 K; ▲(blue): 308.15 K; ▼(magenta): 313.15 K; ◆(olive): 318.15 K; ◀(navy): 323.15 K; ▶(violet): 328.15 K; ●(purple): 333.15 K; ★(wine): 338.15 K; ◊(dark yellow): 343.15 K; ○(grey blue): 348.15 K; +(teal): 353.15 K).

Table 17. VFT Parameters of Eq 14 for Electrical Conductivity κ and Associated Standard Deviations: $\kappa^0 = 1\text{ S m}^{-1}$

x_1	$\ln(A\kappa \times \kappa^0)$	B_κ/K	$T_{0,\kappa}/K$
0.10	-8.149 ± 13.34	11422 ± 35504	-1007.6 ± 2059.6
0.20	-8.675 ± 14.72	11946 ± 40498	-1052.1 ± 2320.9
0.30	-11.14 ± 19.92	15424 ± 56094	-1084.7 ± 2548.9
0.40	-13.26 ± 29.11	19017 ± 85591	-1147.4 ± 3294.5
0.50	-12.59 ± 20.06	15961 ± 52645	-988.95 ± 2154.9
0.60	-15.32 ± 28.94	20737 ± 81091	-1078.1 ± 2727.9
0.70	-18.35 ± 34.58	25250 ± 98296	-1098.4 ± 2755.1
0.80	-18.14 ± 36.81	24859 ± 104640	-1098.5 ± 2979.1
0.90	-17.98 ± 23.22	22327 ± 60097	-970.86 ± 1734.3
1.00	-16.90 ± 24.10	20841 ± 62407	-971.46 ± 1930.3

can be explained by considering the structure of DMSO in the liquid state and in solutions. The presence of dimeric and/or larger associations among DMSO molecules in the pure liquid state and in solutions is known. At higher mole fractions of

Table 18. Fit Parameters (κ_{\max} , x_{\max} , a , and b) of the Casteel–Amis Equation (Eq 16) for the Electrical Conductivity (κ) of ChCl/AcA DES–DMSO Mixtures

temperature T/K	κ_{\max} ($S\ m^{-1}$)	a	b	x_{\max}	σ
298.15	1.482	1.057	0.668	0.888	2.3×10^{-2}
303.15	1.453	1.072	0.636	0.917	2.6×10^{-2}
308.15	1.576	1.113	0.515	1.037	2.2×10^{-2}
313.15	1.684	1.109	0.442	1.117	2.0×10^{-2}
318.15	1.717	1.101	0.507	1.038	3.8×10^{-2}
323.15	2.018	1.101	0.272	1.419	2.0×10^{-2}
328.15	2.523	0.807	0.060	2.579	8.8×10^{-3}
333.15	74.03	0.499	−0.002	37.10	1.1×10^{-3}
338.15	89.37	0.496	−0.003	27.75	6.2×10^{-4}
343.15	24.92	0.547	−0.003	20.33	4.3×10^{-4}
348.15	4.166	0.526	−0.129	2.187	7.4×10^{-3}
353.15	4.483	0.524	−0.146	2.148	2.3×10^{-3}

DMSO, DES molecules are trapped in the DMSO clusters producing a compact structure in the mixed state. With increasing concentration of the DES, these clusters break, and the compact structure is progressively demolished. The negative $\Delta\eta$ values are smaller in magnitude than observed for V^E , and the minima in the $\Delta\eta$ vs x_1 curves also shift to $x_1 \approx 0.62$ in the region of higher DES concentration. These results show the progressive domination of weak interactions in the mixture and the breaking of self-associated structures of DMSO molecules. The observed κ_S^E values are also negative, exhibiting a minimum located at $x_1 \approx 0.15$ in the of κ_S^E vs x_1 plots. The Vogel–Fulcher–Tammann (VFT) model is reasonably applicable to variation of the transport properties (η and κ) with temperature compared to the Arrhenius model. The behavior observed in the variation of electrical conductivity κ with temperature is highlighted with the Casteel–Amis equation.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.3c07739>.

FTIR and 1H spectra of ChCl/AcA DES; spectral profile of ChCl/AcA DES, DMSO, and their binary mixtures; apparent and excess partial molar volumes of ChCl/AcA DES and DMSO; and Arrhenius equation (viscosity and electrical conductivity) (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors are thankful to the Department of Physics, Forman Christian College, A Chartered University, Lahore, Pakistan, and the Department of Chemistry, Quaid-i-Azam University, Islamabad, Pakistan, for providing technical support.

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