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Research Article

Effects of Molar Ratio, Temperature and Water Content on Acidity and Viscosity of Deep Eutectic Solvent Formed from Choline Chloride-Ethylene Glycerol

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Abstract: Deep eutectic solvents (DESs) become preferred solvents owing to unique features and promising environmentally friendly nature they have. Evaluation their properties is essential in order to use them effectively. Even though DESs are a good alternative to classic solvents, their viscosity is high, limiting their usage in industrial applications. However, this can be overcome by adding cosolvent or varying application conditions. In this study, the effect of temperature ranging from 20^{0}C to 60^{0}C (293.15-333.15 K), molar ratio (1:1 to 1:9) and cosolvent addition (water) on acidity and viscosity of DES were investigated. Overall, as a result of this study, operation conditions had a significant effect on acidity and viscosity of DESs. Acidity and viscosity of DESs were on decrease as molar ratio increased, and the increase in temperature negatively affected viscosity of DESs. Dynamic viscosity of DESs were temperature dependence, following Vogel-Fulcher-Tammann (VFT) equality. Cosolvent addition decreased viscosity of DESs. Addition of water higher than 25% did not have a significant effect on viscosity. In addition, water addition decreased acidity of DES up to 1:5 molar ratio, but no significant effect regardless of the amount of water addition on the acidity was determined. These results provide a new perceptive about the mechanism of DES designed for usage in food applications.

Keywords: deep eutectic solvent; temperature; physicochemical property; acidity; viscosity

Araştırma Makalesi

Mol Oranı, Sıcaklık ve Su İçeriğin Kolin Klorür-Etilen Gliserolden Üretilen Derin Ötektik Çözücülerin Asitliğine ve Viskozitesine Etkileri

Özet: Derin ötektik çözücüler (DÖÇ) kendilerine özgü özellikleri ve sahip oldukları doğaya dost yapıları her geçen gün daha çok tercih edilmelerini sağlamaktadır. Etkin kullanılmaları için özelliklerinin değerlendirilmesi önem arz etmektedir. Her ne kadar klasik çözücülere karşı iyi alternatif olmalarına karşın yüksek viskoziteleri bu çözücülerin endüstride kullanımlarını kısıtlamaktadır. Ama bu durum yardımcı çözücü katılarak veya operasyon şartlarının düzenlenmesiyle aşılabilinmektedir. Bu çalışmada, 20^{0}C- 60^{0}C arasında değişen sıcaklık (293.15-333.15 K), mol oranı (1:1- 1:9) ve yardımcı çözücü katımının derin ötektik çözücünün asitliği ve viskozitesi üzerindeki etkisi incelenmiştir. Genel olarak, bu çalışma sonucu olarak ortam şartları DÖÇ'lerin asitlikleri ve viskoziteleri üzerinde önemli etki göstermiştir. DÖÇ'lerin asitliği ve viskozitesi mol oranı arttıkça azalmış, sıcaklık artışı DÖÇ'lerin viskozitesini olumsuz olarak etkilemiştir. DÖÇ'lerin viskozitelerindeki sıcaklık bağlantılı değişim göstermiş, bu da Vogel-Fulcher-Tammann eşitliğine uyumluluk göstermektedir. Yardımcı çözücü (su) ilavesi DÖÇ'lerin viskozitelerini düşürmüştür. 25%'den fazla su ilavesi viskozitede önemli bir değişim oluşturmamıştır. Ayrıca 1:5 mol oranına dek ek çözücü ilavesi asitliği azaltmıştır, fakat ilave edilen su miktarı asitlik üzerinde anlamlı değişim oluşturmamıştır. Sonuçlar gıda endüstri uygulamalarında kullanılmak için tasarlanan DÖÇ mekanızması hakkında yeni bir bakış açısı sağlamaktadır.

Anahtar Kelimeler: derin ötektik çözücü; sıcaklık; fizikokimyasal özellik; asitlik; viskozite

1. Introduction

There is an enormous problem in chemical-based industries due to conventional solvent dependence. The conventional solvents are hazardous and flammable. Thus, replacing them with environmental-friendly solvents is a need for sustainable industrial applications [1]. Deep eutectic solvents, that are an emerging solvent, are liquid at room temperature and are recommended as a good alternative to conventional solvents in chemical-based industries in previous studies [2, 3]. Many recent studies have been performed to mainly identify aspects and applications of DESs in industry applications. The technology has various advantages, including low-cost production, environmentally friendly applications, biodegradability and increase in process efficiency [3].

DESs are recommended for use in chemical industry due to their non-toxic nature. They are produced from at least two components. Chemically, they can be formed from a HBA (hydrogen bond acceptor) and a HBD (hydrogen bond donor) compound, strong hydrogen bond interactions between them creates DESs. Melting point is another advantage of DESs. Because, DESs have much lower melting point than the components forming them [1]. The lower melting point is due to hydrogen bond interactions between HBD and HBA. The interactions can decrease electronic anion-cation strength [2].

The aspects of DESs vary depending on chemical properties of forming components (HBA: Hydrogen bond acceptor, HBD: Hydrogen bond donor). The most used HBA component in DES formation is Choline chloride (ChCI), that is cheap, biodegradable, hygroscopic and non-toxic. The DESs containing ChCI easily diffuse into the matrix, thus are effective in extraction of bioactive compounds in foods [4]. HBDs are generally comprised of metal halides, carbohydrates, amides, alcohols and carboxylic acid. Choosing an appropriate HBD has importance due to its strong effect on solubility. The HBD can increase the solubility due to surface complexation reactions in solvent [7, 34]. Ethylene glycerol is an emerging component in DES formation. Ethylene glycerol is most widely used in DES formation, because it is cheap and easy to diffuse [4]. Similar to ChCI, it is non-toxic and effective to diffuse into any matrix [5]. A previous study has reported the DES, formed from ChCI and ethylene glycerol, had the lowest freezing point and better diffusion efficiency than the DES formed from ChCI with different HBD components [6].

Suitable DES selection can be made for a specific industrial application identifying DES features. In the present study, the DES, that was synthesized from ChCI as HBA and ethylene glycerol as HBD, has been explored investigating the effect of temperature on viscosity and acidity of the DES. The impact of temperature on properties of DES has been rarely assessed so far. However, it is a fundamental aspect in evaluation of any solvent in industrial applications. Because it is effective in determination of molecular volume and activation energy. In addition, the importance of viscosity is related to chemical transformations of solution controlling mass transport [7]. The high viscosity makes DES application impossible in chemical-based industries [8]. It is the first time that a study reports the effect of temperature on viscosity and acidity of DES formed from choline chloride and ethylene glycerol at different molar ratio. Moreover, the effect of water addition on DES properties was investigated.

2. Materials and Methods

2.1. Materials

Choline chloride (98%) and Ethylene Glycerol were supplied from Sigma-Aldrich (St. Louis, Missouri, USA), and pH meter buffer solution powders were obtained from Duvin (DuvinDD, China).

2.2. Solvent preparation

The solvent was formed according to the method described by a previous study [12]. The corresponding components, namely choline chloride and ethylene glycerol, were mixed at different molar ratio ranging from 1:1 to 1:9 in a water bath (Wisebath, WSB series) at 70^0C for 30 min until the formation of a uniform colorless transparent liquid. Then, DESs were cooled down to room temperature and kept in sealed vessel until analyze.

2.3. Determination of physicochemical properties

The investigated physicochemical properties were viscosity and acidity at temperature ranging between 20^0C to 60^0C (293.15- 333.15 K), at molar ratio of 1:1 to 1:9, without and with different water addition content. Each measurement was performed three times using a thermostatic water bath.

The temperature degreees were chosen according to the result of previous studies [35, 38], reporting the conservation of the characteristic supramolecular interactions in DES up to 80^0. Moreover, water addition effect was investigated at the content ranging from 25% to 50%. These water content percentages were decided according to the finding of a previous study [38], stating that the increase in temperature linearly affects physicochemical properties of DES, formed from ChCI/Glycols, up to 25% water addition to DES formation, and the supramolecular structures of DES/water mixtures were affected differently from temperature after higher than 25% water content. Moreover, the molar ratios were determined according to the method of a previous study [37].

2.3.1. Viscosity

It was performed according to the method identified by a previous study [15] using Anton Paar MCR 301 (Anton Paar Gmbh, Graz, Austria) within temperature range of 20^0C- 60^0C (293.15- 333.15 K). The increase in temperature was at the rate of 20°C per min. The gap size was 1 mm and PP25 was used as the cone (24.981 mm in diameter). The cone and plate design were selected over parallel plate layout. 1 mL of sample was applied to the apparatus using a syringe. The sample was allowed to settle on the plate before the measurements.

Apparent viscosity was measured following the method identified by Dimante and Lan [9]. The average of 30 data points from 0 s^{-1} to 100 s^{-1} shear rate at temperature range of 20^0C to 60^0C was reported as the viscosity. Anton Paar Labor-software was used to evaluate the data.

2.3.2. Acidity

Acidity was measured with a digital pH meter (Thermo Scientific, Model: Orion 4-star) at temperatures ranging from 20^0C to 60^0C. The calibration was verified before the analyze with buffer solutions at 4.01, 6.86 and 9.18 pH.

2.4. Statistical analysis

Statistical evaluation was performed using SPSS version 29 software (statistical package for social science). Data was firstly applied to homogeneity test (Shapiro wilk). The result showed all the data homogenously distributed, thus parametric statistic methods were applied. ANOVA test with Duncan's multiple comparison was applied to the data for significance of differences with the significance level of $p<0.05$.

3. Results and Discussion

All deep eutectic solvents were prepared through mixing forming components, namely ChCI and ethylene glycerol, until a homogeneous liquid was formed. Evaluation physicochemical properties of the mixture was performed according to the change in viscosity and acidity in proportion to water content and molar ratio of DES under different temperature ranging between 20^0C to 60^0C. In previous studies, stronger hydrogen bond interactions between HBD and HBA was reported with water addition [36], and the molar ratio of the composition impacted the eutectic point of a solution [13, 14]. In addition, temperature can directly influence hydrogen bond interactions, resulting to the change in properties of DES [33].

3.1. Viscosity

Viscosity influences solvent application into chemical-based industries. Strong hydrogen bond interactions causes high viscosity, resulting to low molecular mobility and high fluid flow resistance [12]. Thus, better industrial applications require DESs to have low viscosity, that can save energy in manufacturing process and in application [11, 33]. The possible way to synthesise low viscosity DES is to add water to DES formation. A certain amount mole fraction of water was determined not to improve diffusion of ions, but significantly decreased viscosity, enhancing fluidity and mass transfer performance of solution. However, high molar fraction of water can form hydrogen bonds with anions, causing serious changes in DES structure [4, 17]. Thus, effect of water addtion to DES formation was investigated at the amounts of 25% and 50%.

Apparent viscosities at different temperatures and molar ratios are demosntrated in Table 1 for DESs, and Table 2 and 3 shows apparent visocsities of aqueous DESs. The viscous flow tests were shown in figure 3 for DESs, and in figure 4 for aqueous DESs.

Table 1. The values of apparent viscosity for the DES without water addition

Molar Ratios	**Apparent Viscosity, cP**			**Significant, *p***
Temperature	20^0C	40^0C	60^0C	
1:1	1610±20a	530±10a	170±10a	<0.001
1:2	1230±10b	410±10b	130±0b	<0.001
1:3	650±10c	240±10c	80±0c	<0.001
1:4	340±10d	130±10d	50±0d	<0.001
1:5	160±10e	70±0e	30±0e	<0.001
1:6	140±10f	60±0e,f	20±0f	<0.001
1:7	100±0g	50±0f	20±0f	0.100
1:8	60±0h	20±0g	10±0g	0.018
1:9	40±0j	20±0g	10±0g	0.018
Significant, *p*	<0.001	<0.001	<0.001	

Note: Different letters in the same column show significant difference ($p< 0.05$).
Mean value ± standard error (n=3)

Table 1 and figure 1 show apparent viscosity of DESs. There was a steadily decreasing trend in apparent viscosity as molar ratio and temperature were on rise. The decrease in viscosity can enhance DES transport properties, thus it can retrenchment cost of using this solvent into practice for industrial applications [10]. Molar ratio and temperature significantly influenced apparent viscosity of DESs, $p<0.05$. Similar finding was reported in previous studies, stating a decrease in viscosity depended on increasing content of HBD [13, 14] and temperature [31, 32]. In addition, forming components have also an essential effect on viscosity [3]. Jablonsky et al. [4] indicated that addition of HBD (Lactic acid) to the mixture containing ChCI, reduced viscosity, and HBD type can influence viscosity of DESs depending on its carbon chain length [15]. A similar phenomenon for HBA concentration was found at a fixed temperature [16].

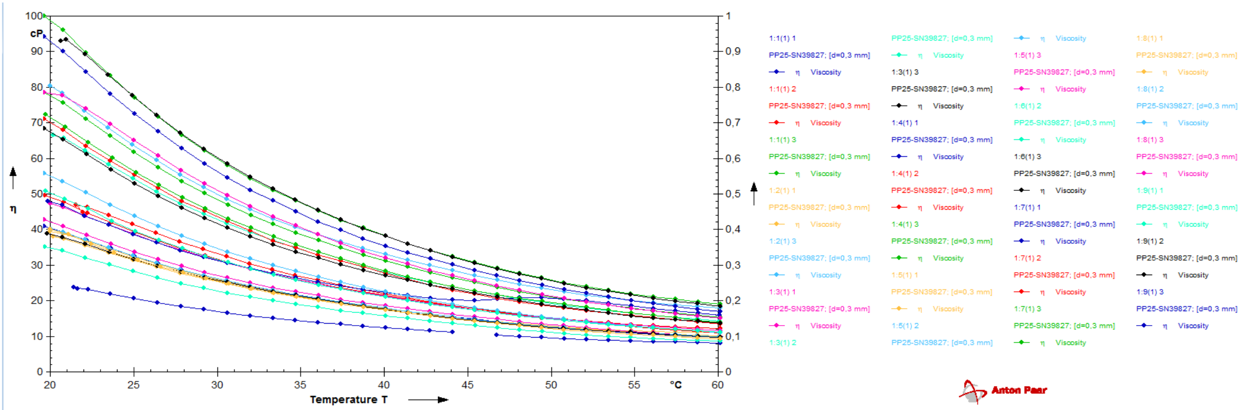

Figure 1. Apparent viscosity of deep eutectic solvents at the temperature ranging from 20⁰C to 60⁰C

Table 2 and figure 2 demonstrate apparent viscosity of the DES with cosolvent addition (water-25%) and table 3 is for the DES with cosolvent addition (water-50%). Water addition to DES formation significantly decreased the viscosity at the molar ratio lower than 1:8 under any ambient temperature, meaning even though low water addition does not affect ions diffusion, high amount of water addition forms hydrogen bonds with anions [17]. Similar result was reported by previous studies, stating high amount of water addition in DES formation causes the decrease in its viscosity [4,15,33]. In addition, water addition to DES formation affects H^+ bond interaction between HBA and HBD, thus can influence apparent viscosity [5]. The highest apparent viscosities among water-added DESs were measured at temperature of 20⁰C and the lowest were measured at temperature of 60⁰C regardless of molar ratio.

Table 2. The values of apparent viscosity for the DESs with 25% cosolvent addition (water)

Molar Ratios	**Apparent Viscosity, cP**			**Significant, *p***
Temperature	20⁰C	40⁰C	60⁰C	
1:1	60±4a	20±1a	10±0a	0.090
1:2	50±2a	20±1a	10±0a	0.021
1:3	70±2a	30±1a	10±0a	0.003
1:4	50±2a	20±1a	10±0a	0.021
1:5	40±1a	20±1a	10±0a	0.011
1:6	40±1a	20±1a	10±0a	0.011
1:7	60±1a	30±1a	10±0a	<0.001
1:8	60±2a	20±1a	10±0a	0.007
1:9	70±2a	30±1a	10±0a	0.003
Significant, *p*	0.548	0.649	1.000	

Note: Different letters in the same column show significant difference ($p < 0.05$).
Mean value ± standard error (n=3)

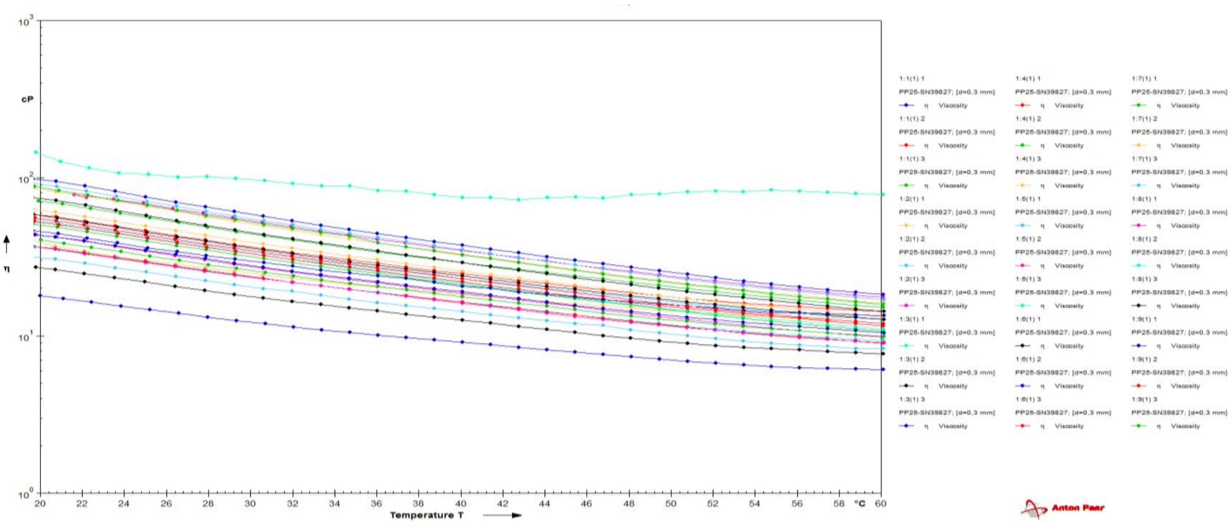

Figure 2. Apparent viscosity of the DESs with water addition (25%)

Temperature significantly changed viscosity of aqueous DESs, except for molar ratio of 1:1. Water can act not only as hydrogen-bond donor but also as acceptor in DES structure [22]. At low molar ratio, DES matrix adsorbed water molecules with forming hydrogen bonds with anions, thus the change in temperature did not influence viscosity [33]. Previous studies reported decrease in viscosity for aqueous DESs as temperature was on increase [18, 19, 20]. High temperature affected interactions between DES molecules, resulting to low viscosity [17]. Low viscosity enhanced solvent solubility and mass transfer, therefore low viscosity is desirable in industrial applications of DESs [21]. Regardless of water addition amount, viscosities of aqueous DESs at 40^0C and 60^0C didn't show significant difference between each other. However, viscosity of aqueous DESs at 20^0C was significantly higher than viscosities of aqueous DESs at 40^0C and 60^0C at any water addition amount. Similar result was stated by previous studies [21, 22, 31]. Moreover, molar ratio did not affect apparent viscosity of aqueous DESs regardless of addition amount, as stated by a previous study [3].

Table 3. The values of apparent viscosity for the DESs with 50% cosolvent addition (water)

Molar Ratios	**Apparent Viscosity, cP**			**Significant, *p***
Temperature	20^0C	40^0C	60^0C	
1:1	50±3a	20±1a	10±0a	0.082
1:2	60±2a	30±1a	10±0a	0.009
1:3	70±3a	30±1a	10±0a	0.018
1:4	60±3a	20±1a	10±0a	0.034
1:5	40±1a	20±1a	10±0a	0.011
1:6	40±1a	20±1a	10±0a	0.011
1:7	70±3a	30±1a	10±0a	0.018
1:8	90±6a	40±3a	30±2a	0.110
1:9	60±2a	30±1a	10±0a	0.009
Significant, *p*	0.251	0.615	1.000	

Note: Different letters in the same column show significant difference ($p < 0.05$).
Mean value ± standard error (n=3)

The amount of water addition did not influence apparent viscosity of DESs at any temperature, $p > 0.05$. Similar result was reported for viscosity of DESs produced from choline chloride [33]. In addition, 7-10% water addition in DES formation can halve viscosity [33]. In this study, higher amount (25 and 50%) than 7-10% water addition were evaluated, and no significant difference in viscosity was detected.

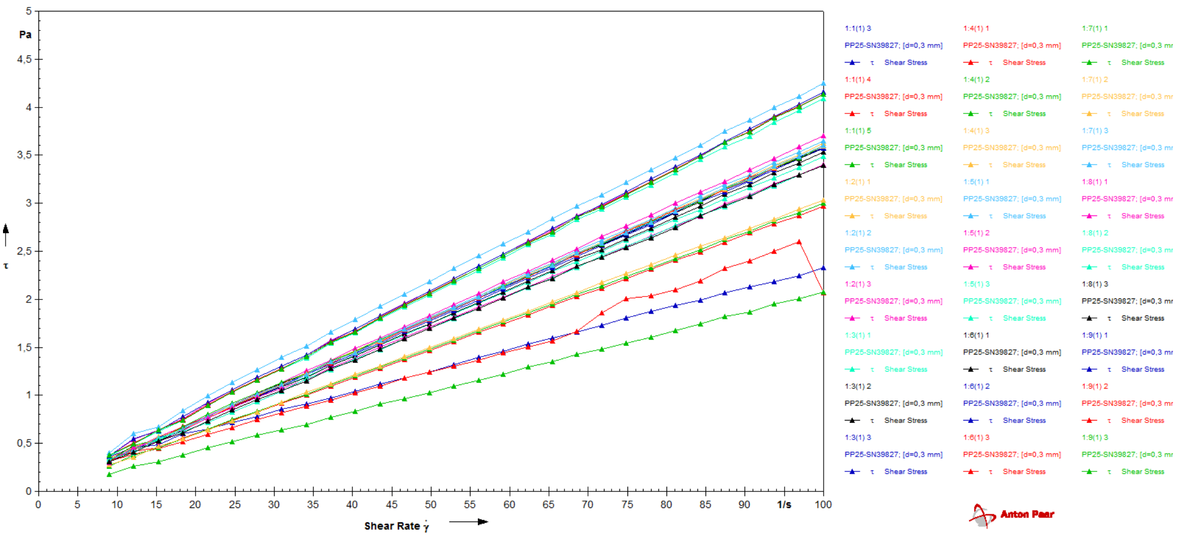

Figure 3. Viscous flow tests of the DESs without water addition

Figure 3 shows viscous flow tests of DESs, and figures 4 demonstrates viscous flow tests of aqueous DESs. Regardless of molar ratio and water addition amount, shear rate increased as shear stress was on rise. This exhibits Arrhenius behavior. However, even though shear rate increased with rise in shear stress for DES with 25% water addition, they showed dilatant fluid. This is due to effect of water addition to strong H^+ bond interaction between HBA and HBD [5, 31]. Similar effect of water addition to DES viscosity behavior was reported by previous studies [24, 33]. In addition, the interactions were stated to weaken with water addition, and they even disappear after 50% water addition (w/w) [33].

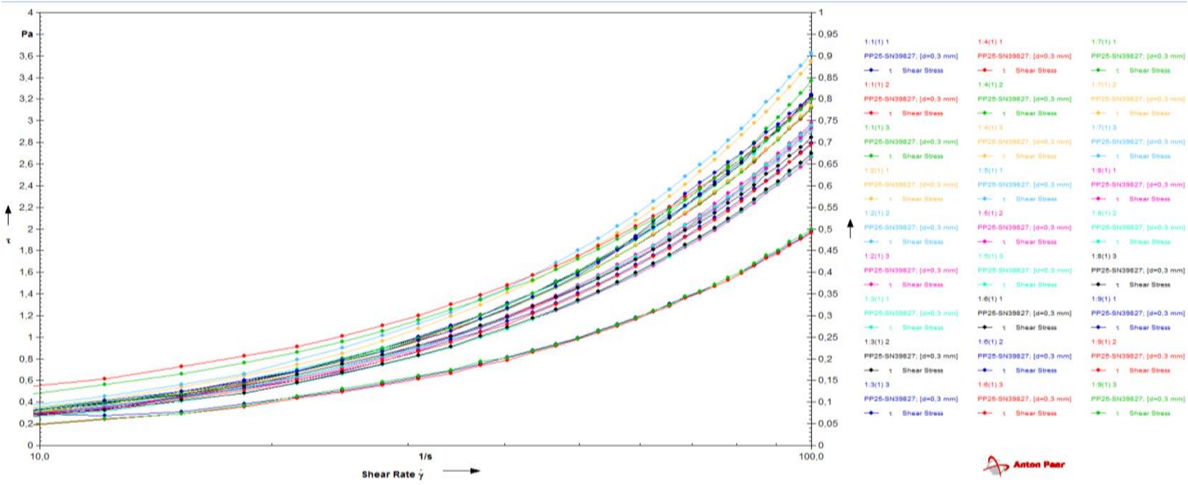

Figure 4. Viscous flow tests of the DESs with 25% water addition

Volume of salt ions and voids directly affects DES viscosity according to Eyring equation, meaning that electrostatic forces are more dominant than van der walls interactions [15]. The viscosity of all DESs and aqueous DESs mixtures decreased exponentially with increase in temperature, resulting to higher molecular mobility with reduction in DES interactions [12].

3.2. Acidity

Estimation pH of new solvents to comprehend their dissolution, catalytic, biomass delignification and other properties is useful for applications [23]. High acidity enables better delignification, while low acidity reduces the cost of putting food industrial applications of DES into practice due to its inhibitory effect against corrosion [10]. The pHs of all choline chloride/ethylene glycerol mixtures were found to be in acidic region (1.28 to 5.96) due to acidic hydrogens in ethylene glycerol structure [27]. Table 4 shows pH of DESs, and Table 5 and 6 demonstrate pH of aqueous DESs (25% and 50% water, w/w).

Table 4. The values of pH for DES without water

Molar Ratios	**Acidity, pH**			**Significant, *p***
Temperature	20^0C	40^0C	60^0C	
1:1	1.28±0.01a	1.70±0.01a	1.84±0.02a	<0.001
1:2	2.64±0.01b	2.86±0.02b	3.22±0.02b	<0.001
1:3	4.63±0.01c	3.95±0.01c	3.97±0.01c	0.123
1:4	4.49±0.00d	4.50±0.01d	4.51±0.01d	0.332
1:5	5.14±0.01e	4.84±0.01e	4.66±0.02e	0.049
1:6	5.24±0.01f	5.02±0.02f	4.76±0.01f	<0.001
1:7	5.47±0.01g	5.09±0.01g	4.98±0.02g	<0.001
1:8	5.84±0.02h	5.48±0.02h	5.35±0.03h	<0.001
1:9	5.96±0.02j	5.68±0.02j	5.58±0.02j	<0.001
Significant, *p*	<0.001	<0.001	<0.001	

Note: Different letters in the same column show significant difference ($p< 0.05$).
Mean value ± standard error (n=3)

Ions activity in DESs changes with temperature [12, 25]. Thus, pH of DESs significantly changed in proportion to temperature and molar ratio. DESs at 1:1 and 1:2 molar ratio were the most acidic solvent, pH<3. The pH increased as more HBD was added to mixtures. The similar result was stated by other studies [13, 24, 27, 30]. High concentration of anion halide salt (ChCI) doesn't bind to HBD (ethylene glycerol), these free ChCI in mixture decreases pH of DESs [13].

Water addition influences interactions between HBA and HBD [5], resulting to decrease in concentration of compounds (mainly HBD) [24]. There was a significant difference in pH of aqueous DESs at any molar ratio. Previous studies reported similar result, showing the effect of water addition to DES formation on pH [21, 24, 28]. Moreover, the amount of added water didn't significantly influence pH regardless of molar ratio and temperature. Similar finding was reported by previous studies [28, 29].

Table 5. The values of acidity for aqueous DESs with 25% cosolvent addition (water)

Molar Ratios	**Acidity, pH**			**Significant, *p***
Temperature	20^0C	40^0C	60^0C	
1:1	2.74±0.01a	2.36±0.01a	2.22±0.03a	<0.001
1:2	3.26±0.01b	2.71±0.02b	2.62±0.04b	<0.001
1:3	3.94±0.01c	3.28±0.01c	3.08±0.02c	<0.001
1:4	4.31±0.01d	3.77±0.01d	3.43±0.03d	<0.001
1:5	4.42±0.01f	3.83±0.01e	3.60±0.03e	<0.001
1:6	4.40±0.01e	3.92±0.02f	3.74±0.04f	<0.001
1:7	4.45±0.00g	4.00±0.01g	3.82±0.02g	<0.001
1:8	4.72±0.01h	4.08±0.02h	3.89±0.04h	<0.001
1:9	4.81±0.02j	4.15±0.01j	3.97±0.03j	<0.001
Significant, *p*	<0.001	<0.001	<0.001	

Note: Different letters in the same column show significant difference ($p< 0.05$).
Mean value ± standard error (n=3).

H^+ bond interaction between HBA and HBD influences pH of DESs [25]. Regardless of water addition and amount, pH of almost all DESs in this study significantly changed with temperature. Water addition regardless of amount decreased pH for DESs formed at higher molar ratio than 1:2, water addition amount didn't have a significant effect on pH at any temperature, p>0.05. The lowest pHs were measured at 60^0C for aqueous DESs while the lowest pHs were found at 20^0C for DESs. Similar effect of temperature on pH was reported in previous studies for DESs with or without water addition [26, 27].

Moreover, pH of DESs at 1:3, 1:4 and 1:5 molar ratios didn't significantly change with temperature while pH significantly changed in aqueous DESs at 1:3, 1:4 and 1:5 molar ratios. Previous studies stated similar result for DESs formed from different forming components, namely choline chloride-based DESs [27] and phosphonium-based DESs [32].

Table 6. The values of acidity for the DESs with 50% cosolvent addition (water)

Molar Ratios	**Acidity, pH**			**Significant, *p***
Temperature	20^0C	40^0C	60^0C	
1:1	2.49±0.02a	2.29±0.02a	2.03±0.01a	<0.001
1:2	3.16±0.02b	2.69±0.02b	2.50±0.00b	<0.001
1:3	3.76±0.02c	3.27±0.01c	3.05±0.00c	<0.001
1:4	4.10±0.01d	3.63±0.01d	3.36±0.00d	<0.001
1:5	4.26±0.00e	3.77±0.00e	3.47±0.00e	0.100
1:6	4.31±0.01f	3.81±0.01f	3.43±0.00f	<0.001
1:7	4.35±0.01g	3.90±0.02g	3.70±0.00g	<0.001
1:8	4.67±0.00h	3.98±0.00h	3.78±0.00h	0.018
1:9	4.73±0.00j	4.13±0.00j	3.82±0.00j	0.100
Significant, *p*	<0.001	<0.001	<0.001	

Note: Different letters in the same column show significant difference ($p< 0.05$).
Mean value ± standard error (n=3).

4. Conclusion and Suggestions

Acidity and viscosity for DESs formed from choline chloride/ethylene glicerol at different molar ratios were evaluated at temperature range of 20-60^{0}C (293.15- 333.15 K) in this study. In addition, change in these physicochemical properties with cosolvent addition was analysed for aqueous solutions (from 25% to 50%).

Apparent viscosity of DESs was on decrease as a function of molar ratio. The highest viscosity was determined for DES with 1:1 HBA: HBD molar ratio regardless of added water amount and temperature. Temperature significantly affected viscosity; the lowest viscosities were measured at 60^{0}C while the highest ones were found at 20^{0}C regardless of molar ratio. Water addition significantly reduced viscosity of DESs. The amount of added water didn't cause any significant difference in viscosity. Molar ratio and temperature had a significant effect on pH of all DESs, and all choline chloride/ethylene glycerol mixtures were found to be in acidic region. The lowest pHs was measured for 1:1 HBA: HBD molar ratio regardless of temperature and water added amount, and pH increased in propertion to molar ratio while pH changed associated with temperature. In addition, amount of water didn't cause any significant difference in pH of DESs.

Overall, temperature and molar ratio have an effect on DES acidity and viscosity. Thus, they should be taken into account before any DES application. Moreover, water addition modified DES properties for favorable manner, resulting to decrease in acidity and viscosity. The changes observed in DES properties can enable comprehension of solvent used for food applications and understanding mechanism of acidic DESs properties.

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Conflict of Interest

The Author reports no conflict of interest relevant to this article.

Research and Publication Ethics Statement

The author declares that this study complies with research and publication ethic.

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