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SUPPLEMENTARY MATERIAL TO

The physicochemical properties of the deep eutectic solvents with triethanolamine as a major component

BILJANA S. ĐORĐEVIĆ[1], DRAGAN Z. TROTER[1], VLADA B. VELJKOVIĆ[1,3], MIRJANA LJ. KIJEVČANIN[2], IVONA R. RADOVIĆ[2] and ZORAN B. TODOROVIĆ[1]*

[1]*University of Niš, Faculty of Technology, Bulevar Oslobođenja 124, 16000 Leskovac, Serbia,* [2]*Faculty of Technology and Metallurgy, University of Belgrade, Karnegijeva 4, 11120 Belgrade, Serbia and* [3]*Serbian Academy of Sciences and Arts, Knez Mihailova 35, 11000 Belgrade, Serbia*

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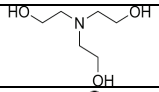
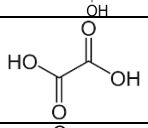
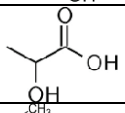
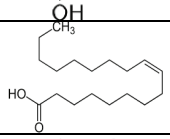
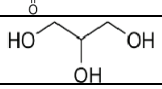
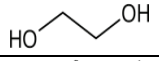
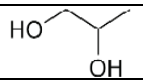
TABLE S-I. Information on the used chemicals

Chemical name	Abbreviation	Formula	Molar mass, g·mol^{-1}	Melting point, K	Chemical structure	CAS number
Triethanol-amine	TEOA	$C_6H_{15}NO_3$	149.19	294.75		102-71-6
Oxalic acid	OA	$C_2H_2O_4$	90.03	462		144-62-7
Glacial acetic acid	AA	$C_2H_4O_2$	60.05	289		64-19-7
L-(+)-Lactic acid	LA	$C_3H_6O_3$	90.08	291		50-21-5
Oleic acid	OLA	$C_{18}H_{34}O_2$	282.47	286		112-80-1
Glycerol	G	$C_3H_8O_3$	92.09	290.9		56-81-5
Ethylene glycol	EG	$C_2H_6O_2$	62.07	261.6		107-21-1
Propylene glycol	PEG	$C_3H_8O_2$	76.09	214		57-55-6

* Corresponding author. E-mail: ztodorovictfle@yahoo.com

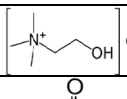
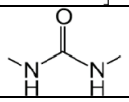
Choline chloride	ChCl	$C_5H_{14}ClNO$	139.62	573.15		67-48-1
1,3-Dimethylurea	DMU	$C_3H_8N_2O$	88.11	377.5		96-31-1

TABLE S-II. Prepared DESs

DES	Abbreviation	Mole ratio	Molar mass of DES[a], g·mol^{-1}	Water content (mass fraction)
Triethanolamine:oxalic acid	TEOA:OA	1:1	119.61	0.0007
Triethanolamine :acetic acid	TEOA:AA	1:1	104.62	0.0006
Triethanolamine:L-(+)-lactic acid	TEOA:LA	1:1	119.64	0.0004
Triethanolamine:oleic acid	TEOA:OLA	1:1	215.83	0.0007
Triethanolamine:glycerol	TEOA:G	1:2	110.93	0.0006
Triethanolamine:ethylene glycol	TEOA:EG	1:2	90.82	0.0005
Triethanolamine:propylene glycol	TEOA:PEG	1:2	100.21	0.0004
Choline chloride:triethanolamine	ChCl:TEOA	1:2	146.03	0.0006
Triethanolamine:1,3-dimethylurea	TEOA:DMU	1:2	108.27	0.0004

[a]The molecular mass (M_{DES}) for TEOA-based DESs is determined from Eq[1]:

$$M_{DES} = \frac{x_{HBA}M_{HBA} + x_{HBD}M_{HBD}}{x_{HBA} + x_{HBD}},$$

where M_{DES} is the molecular mass of DES in g·mol^{-1}, x_{HBA} and M_{HBA} are the mole ratio and molecular mass of the HBA in g·mol^{-1}, respectively; x_{HBD} and M_{HBD} are the mole ratio and molecular mass of the HBD in g·mol^{-1}, in order.

TABLE S-III. The physicochemical properties of TEOA:OA DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1362.2	0.7261	0.102	1.48016
303.15	1357.9	0.4869	0.120	1.47717
313.15	1350.8	0.2386	0.162	1.47391
323.15	1346.4	0.1088	0.251	1.47118
333.15	1342.4	0.0701	0.340	1.46816
343.15	1338.5	0.0441	0.467	1.46450
353.15	1334.7	0.0377	0.503	1.46219
363.15	1330.8	0.0363	0.564	1.45918

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u) η = 5% of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-IV. The physicochemical properties of TEOA:AA DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1182.9	1.2054	0.0189	1.47778
303.15	1181.7	0.8143	0.0202	1.47585
313.15	1179.4	0.4114	0.029	1.47365
323.15	1176.6	0.1944	0.046	1.47171
333.15	1174.8	0.0991	0.086	1.46978
343.15	1173	0.0651	0.111	1.46769
353.15	1171.3	0.0534	0.159	1.46574
363.15	1168.9	0.0511	0.22	1.46385

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5% of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-V. The physicochemical properties of TEOA:LA DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1265.4	1.7377	0.004	1.47999
303.15	1258.2	1.218	0.019	1.47874
313.15	1252.3	0.7572	0.026	1.47782
323.15	1244.6	0.2714	0.052	1.47647
333.15	1238.9	0.1922	0.095	1.47574
343.15	1230.2	0.1102	0.199	1.47482
353.15	1224.1	0.0406	0.356	1.47363
363.15	1216.9	0.0094	0.598	1.47291

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5 % of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-VI. The physicochemical properties of TEOA:OLA DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1114.9	39.76	0.00555	1.43984
303.15	1108.9	25.93	0.00682	1.43789
313.15	1102.9	12.64	0.0127	1.43372
323.15	1096.9	4.542	0.0327	1.42765
333.15	1090.9	0.1359	0.072	1.42499
343.15	1084.9	0.0949	0.121	1.41992
353.15	1080.1	0.0097	0.159	1.41785
363.15	1077.9	0.0046	0.1812	1.41374

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5 % of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-VII. The physicochemical properties of TEOA:G DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1249.2	0.3124	0.00056	1.46873
303.15	1242.7	0.1749	0.00089	1.46735
313.15	1234.4	0.0948	0.0013	1.46645
323.15	1230.9	0.0355	0.00251	1.46528
333.15	1224.2	0.0082	0.00446	1.46472
343.15	1218.7	0.0028	0.00667	1.46385
353.15	1212.5	0.0019	0.00824	1.46254
363.15	1206.7	0.0011	0.00945	1.46129

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5 % of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-VIII. The physicochemical properties of TEOA:EG DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1147.2	0.0795	0.00332	1.45585
303.15	1139.7	0.0542	0.00481	1.45536
313.15	1132.8	0.0369	0.00674	1.45472
323.15	1126.9	0.0238	0.00753	1.45421
333.15	1119.5	0.0112	0.00998	1.45387
343.15	1117.3	0.0056	0.01126	1.45325
353.15	1111.7	0.0025	0.01254	1.45282
363.15	1104.7	0.0008	0.01485	1.45234

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5 % of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-IX. The physicochemical properties of TEOA:PG DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1092.2	0.1027	0.00015	1.44928
303.15	1081.4	0.0657	0.0004	1.44869
313.15	1073.2	0.0386	0.00085	1.44788
323.15	1069.1	0.0138	0.00184	1.44599
333.15	1062.1	0.0086	0.00247	1.44501
343.15	1056.9	0.0048	0.00281	1.44422
353.15	1048.5	0.0023	0.0034	1.44356
363.15	1042.1	0.001	0.00699	1.44281

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5 % of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-X. The physicochemical properties of TEOA:ChCl DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1178.9	1.881	0.2196	1.4814
303.15	1173.8	0.704	0.301	1.4803
313.15	1168.6	0.501	0.5375	1.4789
323.15	1163.7	0.319	0.75	1.4781
333.15	1158.4	0.1742	1.56	1.4773
343.15	1153.3	0.0386	2.215	1.4764
353.15	1148.9	0.0272	4.773	1.4755
363.15	1145.2	0.0189	9.003	1.4743

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5 % of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

TABLE S-XI. The physicochemical properties of TEOA:DMU DES[a]

T / K	ρ / kg·m^{-3}	η / Pa·s	κ / S·m^{-1}	n_D
293.15	1182.6	7.244	0.0018	1.4991
303.15	1173.4	2.623	0.0029	1.494
313.15	1163.7	1.1921	0.0041	1.4886
323.15	1153.2	0.3502	0.0093	1.4839
333.15	1143.3	0.0952	0.0197	1.478
343.15	1129.1	0.0232	0.0319	1.4745
353.15	1116.9	0.00677	0.0405	1.4687
363.15	1113.2	0.00289	0.0447	1.4642

[a]Uncertainties (u): (u)T = ±0.005 K; (u)ρ = ±0.5 kg·m^{-3}; (u)η = 5 % of the measured value; (u)k = ± 0.0001 S·m^{-1}; (u)n_D = ±0.00005.

Effect of temperature on density of DESs

The density of the tested DESs slightly linearly decreases, which agrees with the previous reports.[2,3–5] The density-temperature correlation can be outlined by a linear equation (1):

$$\rho = a + bT \quad (1)$$

where ρ, T, a and b represent the density, the absolute temperature, the density at 0 K and the coefficient of volume expansion, respectively. The characteristic parameters a and b of Eq. (1), density ranges, describe the relative percent deviation (MRPD) and the coefficient of determination (R^2) are given in Table S-XII. A good linear dependence of density on temperature is confirmed by the low MRPD-values (<0.2 %) and the R^2-values close to unity (>0.990).

TABLE S-XII. Parameters of Eq. (1) (293.15-363.15) K

DES	Density range, kg·m^{-3}	a / kg·m^{-3}	b / kg·m^{-3}·K^{-1}	MRPD, %	R^2
TEOA:OA (1:1)	1362.2-1330.8	1492.6	-0.4485	0.07	0.990
TEOA:AA (1:1)	1182.9-1168.9	1242.9	-0.2036	0.08	0.995
TEOA:LA (1:1)	1265.4-1216.9	1468.7	-0.6929	0.04	0.999
TEOA:OLA (1:1)	1114.9-1077.9	1275.5	-0.5512	0.09	0.991

TEOA:G (1:2)	1249.2-1206.7	1423.6	-0.5980	0.05	0.997
TEOA:EG (1:2)	1147.2-1104.7	1316.9	-0.5850	0.10	0.990
TEOA:PEG (1:2)	1092.2-1042.1	1288.8	-0.6799	0.10	0.991
ChCl:TEOA (1:2)	1178.9-1145.2	1322.1	-0.4900	0.03	0.998
TEOA:DMU (1:2)	1182.6-1113.2	1491.5	-1.0500	0.15	0.993

Considering the values of the coefficient of volume expansion, the thermal sensitivity of these DESs is in the following order: TEOA:DMU > TEOA:LA > > TEOA:PEG > TEOA:G > TEOA:EG > TEOA:OLA > ChCl:TEOA > > TEOA:OA > TEOA:AA.

The molecular volume (V_m), the lattice energy (U_{pot}) and the heat capacity (C_p) were calculated by the means of Eqs. (2), (3), (4), (5) and (6)[2,6] and the values obtained at 313.15 K are listed in Table S-XIII.

$$V_m = \frac{M_{DES}}{N_A \rho} \tag{2}$$

where M_{DES} is the molecular mass of DES in g·mol^{-1} and N_A is Avogadro's constant.

$$U_{pot} = 1981.2\left(\frac{\rho}{M_{DES}}\right)^{1/3} - 103.8 \tag{3}$$

for TEOA:OA (1:1), TEOA:AA (1:1), TEOA:LA (1:1) and TEOA:OLA (1:1).

$$U_{pot} = 8375.6\left(\frac{\rho}{M_{DES}}\right)^{1/3} - 178.8 \tag{4}$$

for TEOA:G (1:2), TEOA:EG (1:2), TEOA:PEG (1:2) and TEOA:DMU (1:2).

$$U_{pot} = 6763.3\left(\frac{\rho}{M_{DES}}\right)^{1/3} - 365.4 \tag{5}$$

for ChCl:TEOA (1:2).

$$C_p = 1037 V_m + 45 \tag{6}$$

TABLE S-XIII. The values of molecular volume (V_m), lattice energy (U_{pot}) and heat capacity (C_p) for the tested DESs at 313.15 K

DES	V_m / nm^3	U_{pot} / kJ·mol^{-1}	C_p / J·mol^{-1}K^{-1}
TEOA:OA (1:1)	0.147	548	197
TEOA:AA (1:1)	0.147	548	198
TEOA:LA (1:1)	0.159	537	209
TEOA:OLA (1:1)	0.325	445	382
TEOA:G (1:2)	0.149	1691	200
TEOA:EG (1:2)	0.133	1764	183
TEOA:PEG (1:2)	0.155	1667	206
ChCl:TEOA (1:2)	0.208	1718	260
TEOA:DMU (1:2)	0.154	1670	205

Effect of temperature on viscosity of DESs

The viscosity–temperature relationship of the analyzed DESs can be described by the VTF equation:[7]

$$\eta = \eta_0 \exp\frac{B_\eta}{T - T_0} \quad (7)$$

where T, η_0, B_η, and T_0 represent the absolute temperature, the adjustable parameter, the factor related to the activation energy and the so-called ideal glass-transition temperature, respectively. The values of these parameters, along with the VTF equations, MRPD and R^2 are shown in Table S-XIV.

TABLE S-XIV. The parameters of the VTF equation, Eq. (7), for the tested DESs (293.15-363.15) K

DES	η / Pa s range	Parameters of VTF eq.*		η_0 / Pa s	B_η / K	T_0 / K	MRPD, %	R^2
		a	B					
TEOA:OA (1:1)	0.726-0.036	645.38	7.664	$4.696\cdot10^{-4}$	645.4	207	13.97	0.979
TEOA:AA (1:1)	1.205-0.051	995.25	8.773	$1.548\cdot10^{-4}$	995.3	184	29.61	0.977
TEOA:LA (1:1)	1.738-0.009	7161.3	23.983	$3.840\cdot10^{-11}$	7161.3	7	33.53	0.940
TEOA:OLA (1:1)	39.76-0.005	15048	46.791	$4.774\cdot10^{-21}$	15048	9	29.39	0.940
TEOA:G (1:2)	0.312-0.001	9343.1	32.746	$6.006\cdot10^{-15}$	9343.1	5	10.57	0.977
TEOA:EG (1:2)	0.080-0.001	6716.9	25.044	$1.329\cdot10^{-11}$	6716.9	8	7.80	0.941
TEOA:PEG (1:2)	0.103-0.001	7019.1	26.013	$5.043\cdot10^{-12}$	7019.1	11	4.29	0.985
ChCl:TEOA (1:2)	1.881-0.019	7134	23.681	$5.194\cdot10^{-11}$	7134	10	24.41	0.965
TEOA:DMU (1:2)	7.244-0.003	12191	39.425	$7.550\cdot10^{-18}$	12191	12	41.71	0.987

*$\ln \eta = a\cdot(T\text{-}T_0)^{-1} - b$

TABLE S-XV. The thermodynamic functions of activation of viscous flow, ΔH^*, ΔS^* and ΔG^*, and R^2 for the tested DESs at 313.15 K

DES	Parameters of Eyring's eq.*		R^2	ΔH^* / kJ·mol^{-1}	$T\Delta S^*$ / kJ·mol^{-1}	ΔG^* / kJ·mol^{-1}
	a	B				
TEOA:OA (1:1)	5003	5.143	0.960	41.59	13.34	28.20
TEOA:AA (1:1)	5355.9	5.774	0.962	44.52	15.03	29.49
TEOA:LA (1:1)	7427.1	11.921	0.930	61.75	31.04	30.71
TEOA:OLA (1:1)	15191.3	33.835	0.930	126.30	88.09	38.21
TEOA:G (1:2)	9354.9	20.364	0.972	77.78	53.02	24.76
TEOA:EG (1:2)	6707.3	12.734	0.930	55.76	33.15	22.61
TEOA:PEG (1:2)	6998.3	13.514	0.982	58.18	35.18	22.99
ChCl:TEOA (1:2)	7138.1	10.965	0.960	59.34	28.55	30.80
TEOA:DMU (1:2)	12252.8	27.011	0.985	101.87	70.32	31.54

*$\ln (\eta V/hN_A) = a\cdot T^{-1} - b$

The values of the molar enthalpy change of activation for the viscous flow are higher than the $T\Delta S^*$ values, implying that the energetic contribution corresponding to the molar enthalpy change of activation for the viscous flow is more important than the entropic contribution to the molar Gibbs energy change of activation.

Effect of temperature on the electrical conductivity of DESs

Analogous to the viscosity, the electrical conductivity–temperature relationship is also described by the VTF equation.[8]

$$\kappa = \kappa_0 \exp\frac{B_\kappa}{T - T_0} \tag{8}$$

where κ_0, B_k and T_0 represent the preexponential factor, a factor correlated to the activation energy and the ideal glass-transition temperature, respectively. The fitting parameters of the VTF equation for the electrical conductivity of the tested DESs are summarized in Table S-XVI. The preexponential factor κ_0 is correlated with the number of mobile charge carriers in the molecule. The highest κ_0 value of ChCl:TEOA can be explained by its enhanced ion dissociation, while the reason for the lowest κ_0 of TEOA:PEG is the formation of the polypropylene glycol that inhibits the ion mobility.

TABLE S-XVI. The parameters of the VTF equation, Eq. (7), for the electrical conductivity (κ) of the tested DESs (293.15-363.15) K

DES	Parameters of VTF eq.*		κ_0 / S·m^{-1}	$B\kappa$ / K	T_0 / K	MRPD, %	R^2
	a	*b*					
TEOA:OA (1:1)	-1258.3	4.4946	89.5	1258.3	110	7.47	0.979
TEOA:AA (1:1)	-4038.1	9.5925	14654.50	4038.1	11	3.72	0.975
TEOA:LA (1:1)	-3282.3	12.1660	192143.90	3282.3	105	4.31	0.986
TEOA:OLA (1:1)	-3621.5	11.0850	65186	3621.5	74	7.32	0.969
ChCl:TEOA (1:2)	-5607.1	17.3960	35891103.20	5607.1	10	23.5	0.979
TEOA:DMU (1:2)	-2390.9	6.5285	684.40	2390.9	109	3.89	0.979
TEOA:G (1:2)	-1647.2	2.5920	13.40	1647.2	131	1.82	0.985
TEOA:EG (1:2)	219.03	-2.7029	0.08	219.03	220	0.58	0.995
TEOA:PEG (1:2)	310.9	-2.6100	0.07	310.9	243	2.11	0.983

*$\ln \kappa = a(T\text{-}T_0)^{-1} + b$

Molar conductivity and viscosity relationship

The equation used for determining the molar conductivity (Λ) is:

$$\Lambda = \frac{\kappa M}{\rho} \tag{9}$$

where κ is the electrical conductivity, M is the molar mass and ρ is the density. The empirical VTF equation for the molar conductivity is as follows:

$$\Lambda = \Lambda_0 \exp\frac{-B_\Lambda}{T - T_0} \tag{10}$$

where Λ_0, B_Λ, T_0 are the fitting parameters. Their values are given in Table S-XVII.

TABLE S-XVII. The parameters of VTF equation, Eq. (10), for the molar conductivity (Λ) of the tested DESs over the temperature range (293.15-363.15) K

DES	Parameters of VTF eq.*		Λ_0 / S·m^2·mol^{-1}	B_Λ / K	T_0 / K	MRPD, %	R^2
	a	*b*					
TEOA:OA (1:1)	-1285.8	-4.7336	0.009	1285.8	109	0.76	0.975
TEOA:AA (1:1)	-4006.3	0.2439	1.276	4006.3	3	0.92	0.970
TEOA:LA (1:1)	-3339.8	3.1104	22.430	3339.8	104	0.97	0.984
TEOA:OLA (1:1)	-3566.3	2.5061	12.257	3566.3	77	1.78	0.964
TEOA:G (1:2)	-1529.2	-6.9835	0.001	1529.2	139	0.68	0.984
TEOA:EG (1:2)	-233.8	-12.0295	$5.97\cdot10^{-6}$	233.8	218	0.20	0.994
TEOA:PEG (1:2)	-332.5	-11.7204	$8.13\cdot10^{-6}$	332.5	241	0.80	0.981
ChCl:TEOA (1:2)	-5651.7	8.5519	5176.581	5651.7	1	1.93	0.971
TEOA:DMU (1:2)	-2737.9	-1.8599	0.156	2737.9	96	1.19	0.972

*$\ln \Lambda = a\cdot(T\text{-}T_0)^{-1} + b$

The Walden plot is useful for illustrating the conductivity-viscosity relationship for the pure ionic liquids.[9] It describes the connection between the mobility of ions and the fluidity of their surrounding medium, according to the following equation:[10]

$$\Lambda\eta = k \tag{11}$$

where Λ is the molar conductivity, η is the viscosity and k are a temperature-dependent constant. Therefore, the Walden plot is used for classifying the ionic liquids as "good", "poor", "superionic", *etc*.[2] The logarithmic plot of Λ, representing the ion mobility, versus the fluidity φ ($\varphi = \eta^{-1}$) is used for comparing the ions' formation ability in non-aqueous electrolyte solutions, molten salts and ionic liquids.[11] This so-called "fractional" Walden rule is written as follows:[11]

$$\log \Lambda = \log C + \alpha' \log \eta^{-1} \tag{12}$$

where C is the Walden product, and α' is the slope of the Walden plot line and reflects on the decoupling of the ions. The coefficients of the Walden equation for the DESs are given in Table S-XVIII.

TABLE S-XVIII. The coefficients of the Walden equation, Eq. (12), along with MRPD and R^2 for the tested DESs over the temperature range (293.15-363.15) K

DES	Parameters of Walden equation	α'	C	MRPD, %	R^2
TEOA:OA (1:1)	$\log \Lambda = 0.5722\cdot\log \eta^{-1} - 5.1623$	0.5722	$6.88\cdot10^{-6}$	0.51	0.988
TEOA:AA (1:1)	$\log \Lambda = 0.7397\cdot\log \eta^{-1} - 5.8138$	0.7397	$1.54\cdot10^{-6}$	1.32	0.947
TEOA:LA (1:1)	$\log \Lambda = 0.9027\cdot\log \eta^{-1} - 5.8043$	0.9027	$1.57\cdot10^{-6}$	3.21	0.897
TEOA:OLA (1:1)	$\log \Lambda = 0.3836\cdot\log \eta^{-1} - 5.2010$	0.3836	$6.3\cdot10^{-6}$	2.43	0.926
TEOA:G (1:2)	$\log \Lambda = 0.4939\cdot\log \eta^{-1} - 7.4646$	0.4939	$3.43\cdot10^{-8}$	0.64	0.982
TEOA:EG (1:2)	$\log \Lambda = 0.304\cdot\log \eta^{-1} - 6.7715$	0.304	$1.69\cdot10^{-7}$	1.06	0.878
TEOA:PEG (1:2)	$\log \Lambda = 0.7439\cdot\log \eta^{-1} - 8.3162$	0.7439	$4.83\cdot10^{-9}$	1.80	0.900
ChCl:TEOA (1:2)	$\log \Lambda = 0.7694\cdot\log \eta^{-1} - 4.4159$	0.7694	$3.84\cdot10^{-5}$	2.46	0.945
TEOA:DMU (1:2)	$\log \Lambda = 0.443\cdot\log \eta^{-1} - 6.3386$	0.4430	$4.59\cdot10^{-7}$	1.44	0.962

Effect of temperature on the refractive index of DESs

The parameters of the linear equations,[12] refractive index ranges, MRPD and R^2 are listed in Table S-XIX.

TABLE S-XIX. The parameters of the refractive index (n_D) equation for the tested DESs (293.15-363.15) K

DES	n_D range	Intercept	Slope	MRPD, %	R^2
TEOA:OA (1:1)	1.4802-1.4592	1.5684	-0.0003	0.03	0.999
TEOA:AA (1:1)	1.4778-1.4639	1.5363	-0.0002	0.01	0.999
TEOA:LA (1:1)	1.4800-1.4729	1.5094	-0.0001	0.02	0.996
TEOA:OLA (1:1)	1.4398-1.4137	1.5547	-0.0004	0.25	0.991
TEOA:G (1:2)	1.4687-1.4613	1.4980	-0.0001	0.02	0.992
TEOA:EG (1:2)	1.4559-1.4523	1.4705	-0.0001	0.01	0.997
TEOA:PEG (1:2)	1.4493-1.4428	1.4783	-0.0001	0.03	0.979
ChCl:TEOA (1:2)	1.4814-1.4743	1.5098	-0.0001	0.05	0.994
TEOA:DMU (1:2)	1.4991-1.4642	1.6451	-0.0005	0.03	0.999

Table S-XX contains the ranges of the phase velocity (υ), the molar refractivity (A) and the free volume (f_m) for the tested DESs, calculated as recommended elsewhere.[1,13]

The phase velocities are similar for all tested DESs, the highest being for the TEOA:OLA (1:1) DES due to its lowest refraction index in the applied temperature range. The molar refractivity, which is a measure of the polarizability of a substance, is mostly influenced by the molecular mass while the temperature, density and refraction index show a weak effect. A minor effect of the temperature on molar refractivity has already been reported.[1] The free volume increases with heating, as expected.[1] Among all tested DESs, TEOA:OLA (1:1) has the highest free volume due to the longest alkyl chain of oleic acid.[1]

At 313.15 K, the phase velocity of the tested DESs is in following order: TEOA:DMU < ChCl:TEOA < TEOA:LA < TEOA:OA < TEOA:AA < TEOA:G < < TEOA:EG < TEOA:PEG < TEOA:OLA.

TABLE S-XX. The phase velocity (υ), molar refractivity (A) and free volume (f_m) ranges for the tested DESs (293.15-363.15) K

DES	υ / 10^7 m·s^{-1}	A / 10^{-6} m^3 mol^{-1}	f_m / 10^{-6} m^3 mol^{-1}
TEOA:OA (1:1)	20.27-20.56	24.57-24.95	62.29-6289
TEOA:AA (1:1)	24.69-25.03	49.38-50.05	64.81-63.42
TEOA:LA (1:1)	20.27-20.37	26-85-27-57	67.85-70.74
TEOA:OLA (1:1)	20.84-21.22	50.01-51.00	142.58-150.22
TEOA:G (1:2)	20.43-20.53	24.72-25.23	64.08-66.69
TEOA:EG (1:2)	20.61-20.66	21-51-22.19	57.65-60.02
TEOA:PEG (1:2)	20.70-20.79	35.27-35.85	88.59-91.66
ChCl:TEOA (1:2)	20.25-20.35	26.84-26.88	64.66-70.41
TEOA:DMU (1:2)	20.01-20.49	32.33-35-76	79.71-89.58

FTIR analysis

FTIR spectra of the DESs and their individual components are shown in Fig. S-1. The FTIR spectra of the tested DESs show a very strong and broad band at 3200-3500 cm^{-1}, ascribed to ν(OH) stretching vibration, which confirms the existence of the hydrogen bonds in these mixtures. This band covers all bands belonging to the amine vibrations from TEOA and ChCl and has great intensity in every starting component of the DESs, except OLE, due to its long carbon chain that limits the formation of the intramolecular hydrogen bonds. The presence of ν(C-H) stretching bands at 2800-3000 cm^{-1} is obvious in the spectra of all DESs and their initial compounds,[14] except OA. The band at 1550-1690 cm^{-1} in all spectra belongs to δ(OH) vibrations, overlapping the $\delta(NH_3^+)$ bands at 1660 and 1646 cm^{-1} in the spectra of TEOA and ChCl. The bands at 1403-1419 cm^{-1} and 1000-1117 cm^{-1} are derived from δ(C-H) and ν(C-O) vibrations, respectively, are present in all spectra. The ν(C-N) bands at 1358,1350 and 1340 cm^{-1} present in the spectra of TEOA, ChCl and DMU are also present in all DESs that contain these components. In the spectra of AA, OA, LA and OLE, as well as of their DESs the ν(C=O) bands are at 1710-1730 cm^{-1}.[14] The FTIR analysis proved both the presence of the hydrogen bonds in these DESs and the characteristic functional groups of their constituents, showing no chemical changes (reactions) that occurred during their preparation.

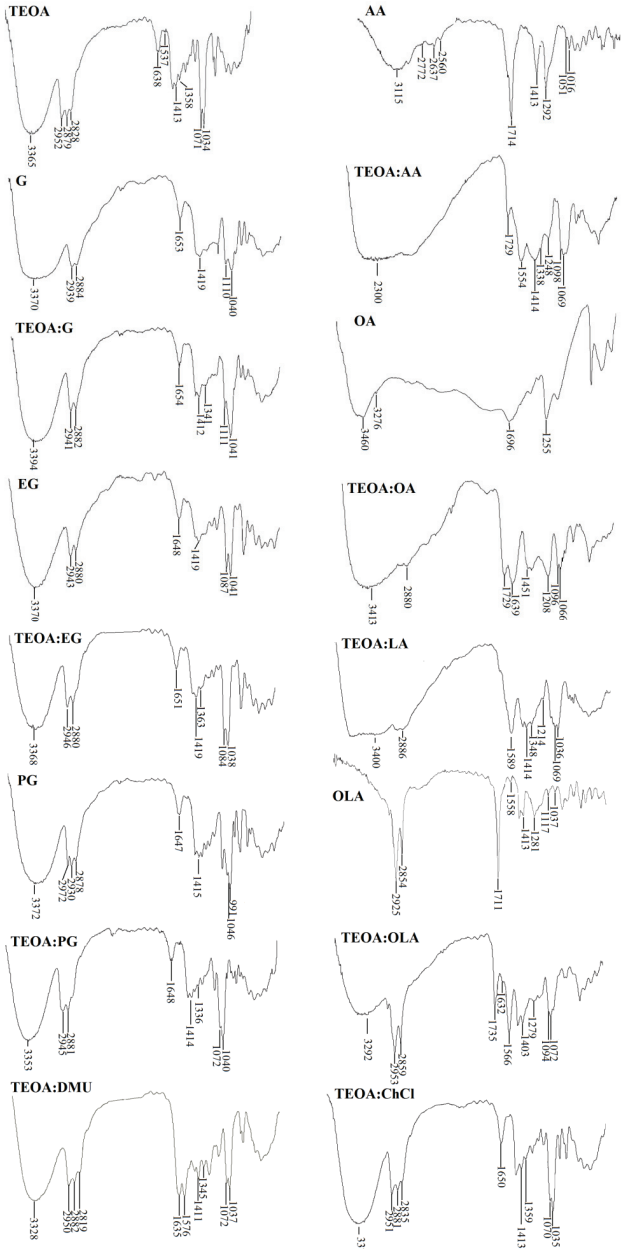


Fig. S-1. FTIR spectra of the DESs and their individual components at 25 °C in the region of 400–4000 cm^{-1}.

TGA and DSC analyses

The TGA and DSC curves of the selected TEOA- and ChCl-based DESs with the same donors are shown in Figs. S-2 and S-3. As can be seen in Figs. S-2 and S-3, the TGA and DSC curves do not show any surprising behaviour of the samples, since no characteristic peaks are present, except the certain mass loss evident from the TGA curves, within which one part refers to water evaporation certainly, explained by the higher upper limit of the temperature range.

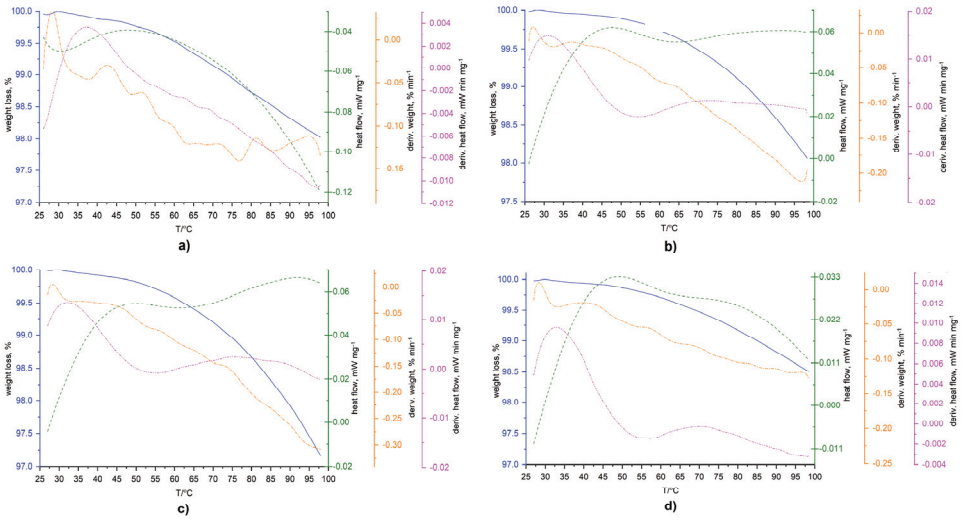


Fig. S-2. TGA and DSC curves recorded for: (a) TEOA:G, (b) TEOA:EG, (c) TEOA:PEG, (d) ChCl:TEOA 298.15-373.15 and 101.325 kPa. Note: – denotes TGA curve, –·– denotes the first derivative of the TGA function, – – denotes DSC curve and –··– denotes the first derivative of DSC function.

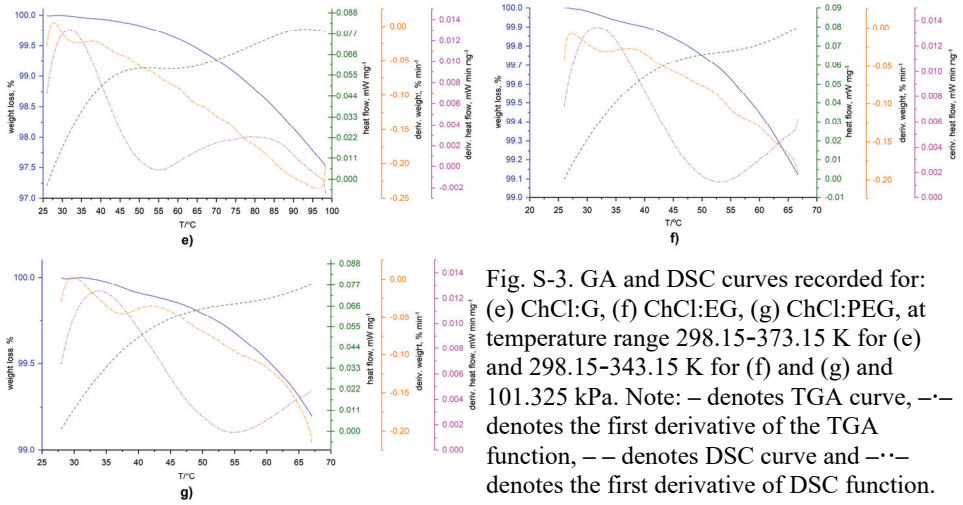


Fig. S-3. GA and DSC curves recorded for: (e) ChCl:G, (f) ChCl:EG, (g) ChCl:PEG, at temperature range 298.15-373.15 K for (e) and 298.15-343.15 K for (f) and (g) and 101.325 kPa. Note: – denotes TGA curve, –·– denotes the first derivative of the TGA function, – – denotes DSC curve and –··– denotes the first derivative of DSC function.

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