Supplementary Information (SI) for Physical Chemistry Chemical Physics.

Supplementary Information

Polyols-Based Deep Eutectic Solvents: Betaine *versus* Choline Chloride

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Figures

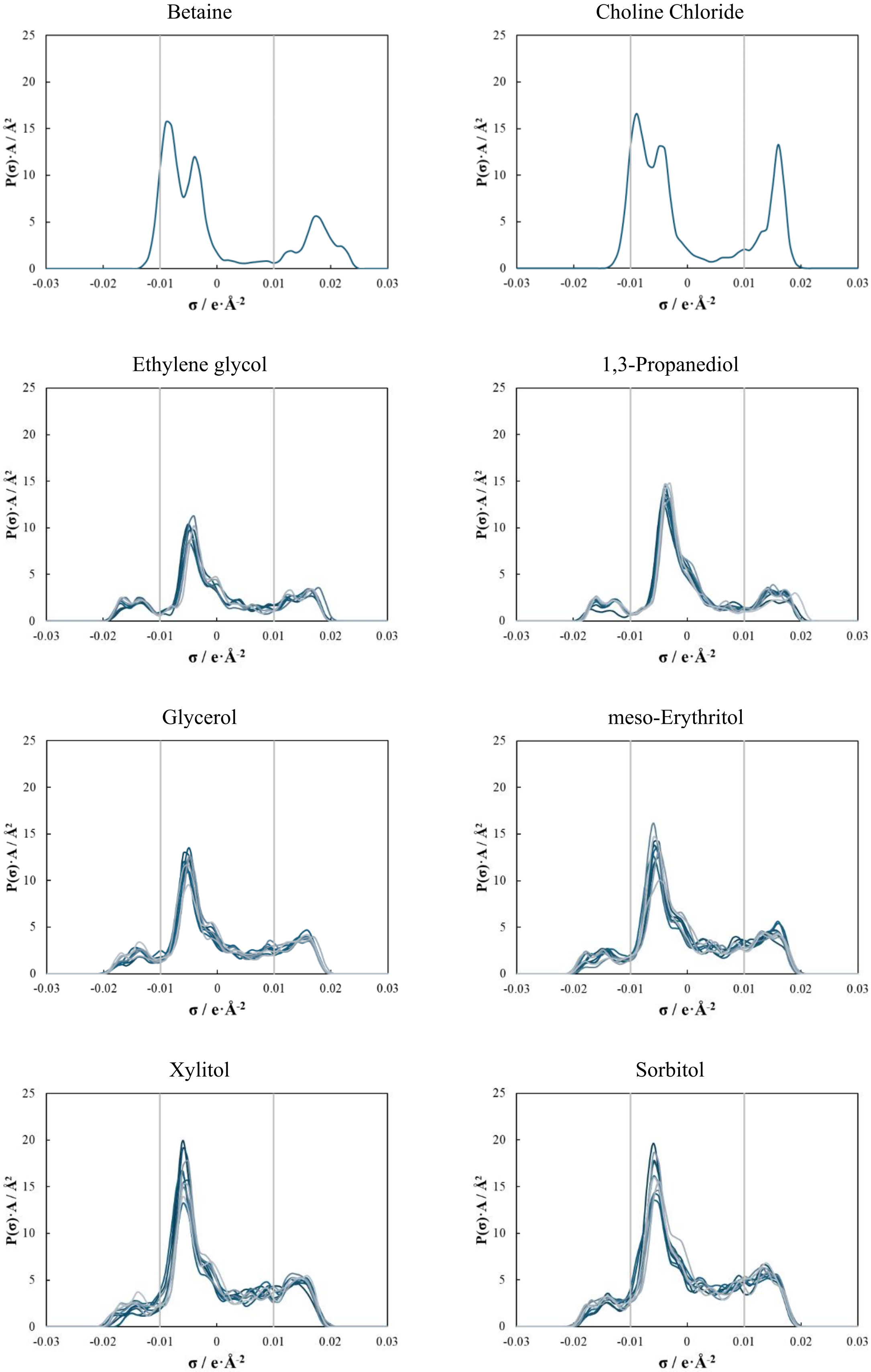

Figure S.1 - Sigma-profile using TZVP parametrization of the different conformers studied in this work.

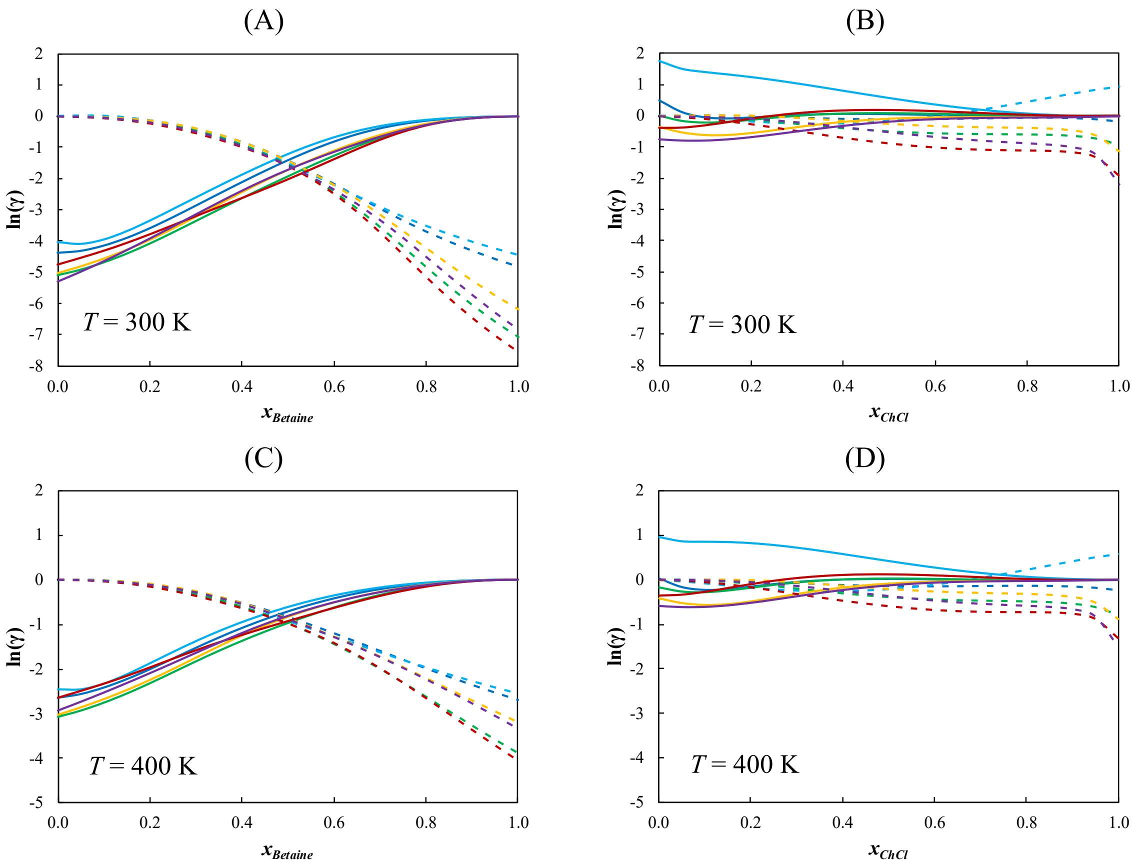

Figure S.2 - Isothermal activity coefficients predictions of COSMO-RS (TZVPD-FINE parametrisation) of the binary systems of betaine (A and C) or ChCl (B and D) with ethylene glycol (dark blue), 1,3-propanediol (light blue), glycerol (green), *meso*-erythritol (yellow), xylitol (red), and sorbitol (F) at 300 K (A and B) and 400 K (C and D). Lines represent the activity coefficients of either betaine or ChCl (solid lines), polyols (dashed lines), and $\ln(\gamma) = 0$ (dotted line).

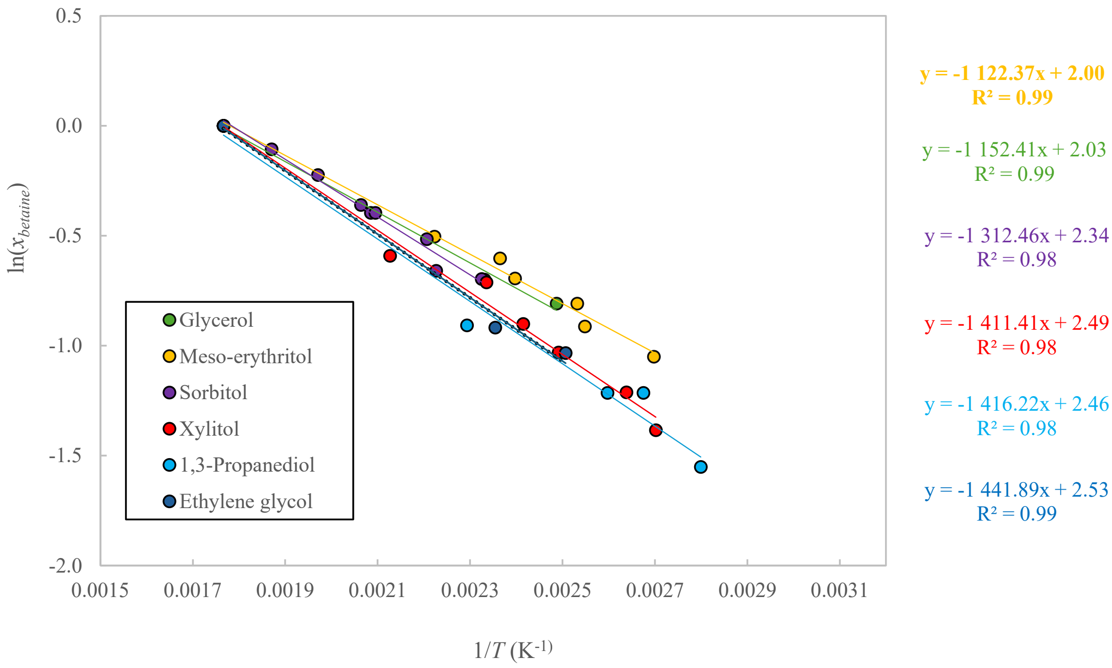

Figure S.3 – Representation of the ln($x_{betaine}$) as a function of 1/T of the binary mixtures of betaine and a polyol. Colour code for the polyol: 💧, 1,3-propanediol; 💧, ethylene glycol; 💧, glycerol; 💧, *meso*-erythritol; 💧, sorbitol; 💧, xylitol. The solid lines and equations correspond to the linear fits of each system. For the linear fit, only the first data points after the pure component ($x_{betaine}$ = 1, T = T_m = 566.2 K) were considered.

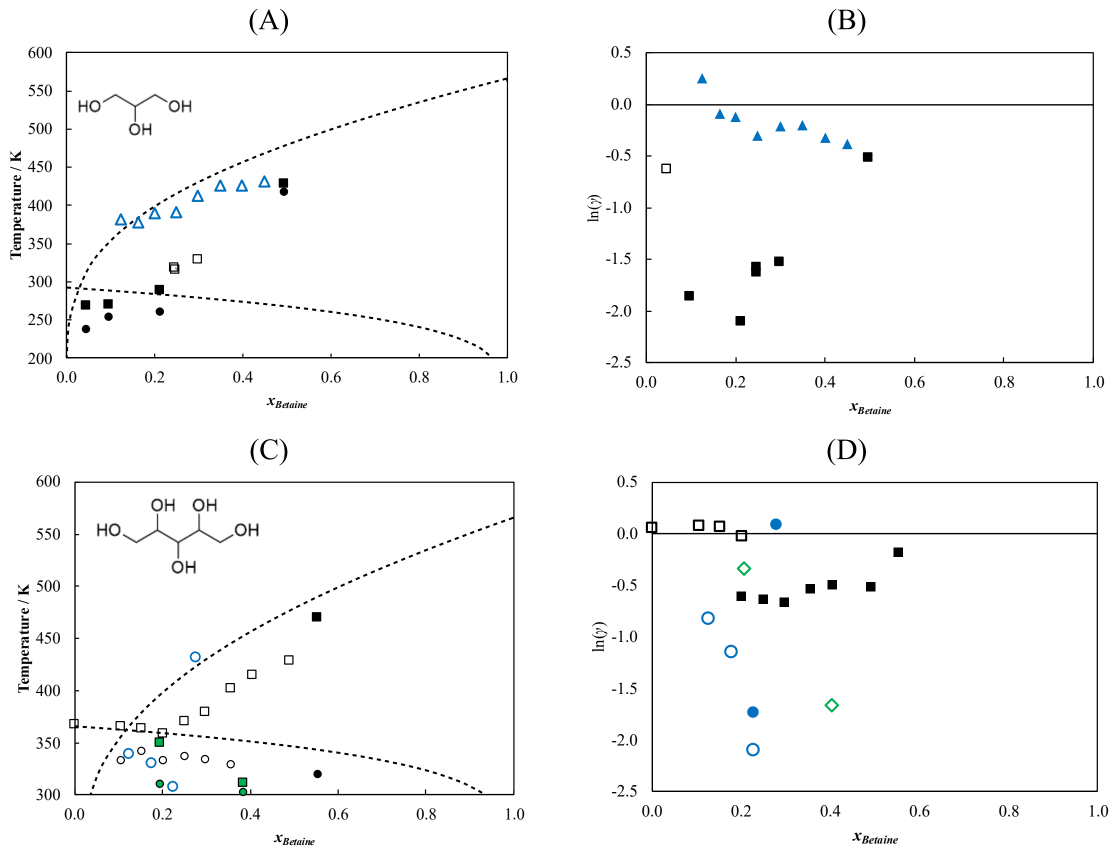

Figure S.4 - Solid-liquid phase diagrams of (A) betaine/GLY and (C) betaine/XYL, and respective non-isothermal activity coefficients (B and D). Symbols in (A) and (C) represent data of betaine systems without added water obtained by VM (white symbols) and DSC (black symbols), with 0.6 wt% of added water (green symbols), and data of betaine monohydrate systems from Zahrina et al. (blue triangles)[1] and Palmelund et al. (blue circles).[2] The x-axis is the mole fraction of betaine anhydrous. Lines in (A) and (C) represent the ideal model (dashed lines) and normal boiling temperature of GLY and in (B) and (D) represent the ideal behaviour, $\ln(\gamma_i) = 0$.

Tables

Table S.1 – Experimental solid-liquid equilibrium data (x, T_m, T_{per}, T_{eut}) for the system betaine/EG. The activity coefficients (γ) were obtained as per Eq. (2) using the melting properties reported in Table 1.

$x_{betaine}$	T_m / K	T_{per} / K	T_{eut} / K	$\ln(\gamma_{BET})$	$\ln(\gamma_{EG})$	Measuring method
0.05	252.8	-	-	-	-0.1	DSC
0.09	272.1	-	248.5	-	-	DSC
0.15	303.2	-	-	-	-	Heating Block
0.19	315.7	-	-	-	-	DSC
0.25	313.2	-	-	-1.7	-	Heating Block
0.25	309.8	-	265.7	-1.8	-	DSC
0.30	357.2	321.6	-	-1.0	-	DSC
0.30	348.5	-	-	-1.2	-	Heating Block
0.36	390.8	-	-	-0.7	-	Heating Block
0.36	398.9	328.3	-	-0.6	-	DSC
0.40	424.8	-	-	-0.4	-	Heating Block
0.40	-	315.7	-	-	-	DSC

Standard uncertainties, u, are $u_r(x)$ = 0.01, for the temperatures measured by the heating block $u(T)$ = 2 K, and by DSC $u(T)$ = 1 K.

Table S.2 – Experimental solid-liquid equilibrium data (x, T_m, T_{tr}, T_{eut}) for the system betaine/13PG. The activity coefficients (γ) were obtained as per Eq. (2) using the melting properties reported in Table 1.

$x_{betaine}$	T_m / K	T_{tr} / K	T_{eut} / K	$\ln(\gamma_{BET})$	$\ln(\gamma_{13PG})$	Measuring method
0.05	266.4	-	243.2	-1.4	0.4	DSC
0.21	357.2	345.9	-	-0.7	-	DSC
0.30	373.8	-	-	-0.8	-	Heating Block
0.30	385.0	366.7	-	-0.6	-	DSC
0.40	436.2	-	-	-0.2	-	Heating Block

Standard uncertainties, u, are $u_r(x)$ = 0.01, for the temperatures measured by the heating block $u(T)$ = 2 K, and by DSC $u(T)$ = 1 K.

Table S.3 – Experimental solid-liquid equilibrium data (x, T_m, T_{tr}, T_{eut}) for the system betaine/GLY. The activity coefficients (γ) were obtained as per Eq. (2) using the melting properties reported in Table 1.

$x_{betaine}$	T_m / K	T_{tr} / K	T_{eut} / K	$\ln(\gamma_{BET})$	$\ln(\gamma_{GLY})$	Measuring method
0.05	268.8	-	-	-	-0.6	DSC
0.10	270.1	-	253.9	-1.9	-	DSC
0.21	289.7	-	261.5	-2.1	-	DSC
0.25	317.2	-	-	-1.6	-	Heating Block
0.30	330.2	-	-	-1.5	-	Heating Block
0.50	429.0	418.7	-	-0.5	-	DSC

Standard uncertainties, u, are $u_r(x)$ = 0.01, for the temperatures measured by the heating block $u(T)$ = 2 K, and by DSC $u(T)$ = 1 K.

Table S.4 – Experimental solid-liquid equilibrium data (x, T_m, T_{eut}) for the system betaine/ME. The activity coefficients (γ) were obtained as per Eq. (2) using the melting properties reported in Table 1.

$x_{betaine}$	T_m / K	T_{eut} / K	$\ln(\gamma_{BET})$	$\ln(\gamma_{ME})$	Measuring method
0.09	386.9	-	-	0.0	Visual Method
0.15	385.2	367.9	-	0.0	Visual Method
0.18	375.7	-	-	-0.3	Visual Method
0.25	370.4	367.5	-	-0.4	Visual Method
0.30	374.6	-	-	-0.2	Visual Method
0.35	370.7	-	-	-0.2	Visual Method
0.40	-	363.3	-1.2	-0.4	Visual Method
0.45	380.3	-	-1.1	-	Visual Method
0.50	394.7	-	-1.0	-	Visual Method
0.55	422.9	-	-0.7	-	Visual Method
0.60	449.9	-	-0.5	-	Visual Method

Standard uncertainties, u, are $u_r(x)$ = 0.01, for the temperatures measured by the visual method: $u(T)$ = 2 K.

Table S.5 – Experimental solid-liquid equilibrium data (x, T_m, T_{eut}) for the system betaine/XYL. The activity coefficients (γ) were obtained as per Eq. (2) using the melting properties reported in Table 1.

$x_{betaine}$	T_m / K	T_{eut} / K	$\ln(\gamma_{BET})$	$\ln(\gamma_{XYL})$	Measuring method
0.11	364.8	332.3	-	0.1	Visual Method
0.15	363.0	341.2	-	0.1	Visual Method
0.20	358.5	333.2	-0.6	0.0	Visual Method
0.25	370.1	337.2	-0.6	-	Visual Method
0.30	379.1	334.2	-0.7	-	Visual Method
0.36	401.4	329.2	-0.5	-	Visual Method
0.41	414.2	-	-0.5	-	Visual Method
0.49	428.3	-	-0.5	-	Visual Method
0.55	470.2	320.1	-0.2	-	DSC

Standard uncertainties, u, are $u_r(x) = 0.01$, for the temperatures measured by the Visual Method $u(T) = 2$ K, and by DSC $u(T) = 1$ K.

Table S.6 – Experimental solid-liquid equilibrium data (x, w, T_m, T_{eut}) for the system betaine/XYL/water. The activity coefficients (γ) were obtained as per Eq. (2) using the melting properties reported in Table 1.

$x_{betaine}$	x_{water}	w_{water}	T_m / K	T_{eut} / K	$\ln(\gamma_{XYL})$	Measuring method
0.38	0.05	0.006	310.7	302.6	-1.7	DSC
0.20	0.04	0.006	349.6	310.0	-0.3	DSC
0.36	0.11	0.016	353.1	312.6	-	DSC

Standard uncertainties, u, are $u_r(x) < 10^{-5}$ and $u_r(w) = 0.002$, and for the temperatures measured by DSC $u(T) = 1$ K.

References

1 I. Zahrina, M. Nasikin, K. Mulia, M. Prajanto and A. Yanuar, Molecular interactions between betaine monohydrate-glycerol deep eutectic solvents and palmitic acid: Computational and experimental studies, *J Mol Liq*, 2018, 251, 28–34.

2 H. Palmelund, B. J. Boyd, J. Rantanen and K. Löbmann, Influence of water of crystallization on the ternary phase behavior of a drug and deep eutectic solvent, *J Mol Liq*, 2020, 315, 113727.