# Oil-in-water emulsions based on hydrophobic eutectic systems
## *Supporting Information*

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## Physicochemical properties

A recent investigation showed detailed physicochemical properties of the DES composed of 1-tetradecanol and menthol in a 1:2 molar ratio. [1] The degradation temperature of this DES is $113.1 \pm 8.4\,^{\circ}\mathrm{C}$. Before mixing with water, the density at $20\,^{\circ}\mathrm{C}$ is $872.1\,\mathrm{kg\,m^{-3}}$, while after mixing with water the density is $874.2\,\mathrm{kg\,m^{-3}}$. Here, the density is investigated as function of temperature to get some detailed insights. The correlation between the densities and temperature is found to be linear:

$$\rho = a + bT. \tag{1}$$

**Table 1:** Fitting parameters for the density as depicted in eq. (1).

| DES | $a$ [$\mathrm{kg\,m^{-3}}$] | $b$ [$\mathrm{kg\,m^{-3}\,K^{-1}}$] |
|---|---|---|
| Before mixing with water | 1081.44 | −0.71 |
| After mixing with water | 1085.31 | −0.72 |

As shown in fig. 1 this expression accurately describes the measured densities of the DES before and after mixing with water. The fit parameters $a$ and $b$ are presented in table 1.

We describe the viscosity using the Vogel–Fulcher–Tammann (VFT) expression:

$$\eta = A_\eta \cdot \exp\left(\frac{B_\eta}{T - C_\eta}\right). \tag{2}$$

The change in viscosity of the DES before and after mixing with water is given in fig. 2. The fit parameters for the VFT equation are given in table 2.

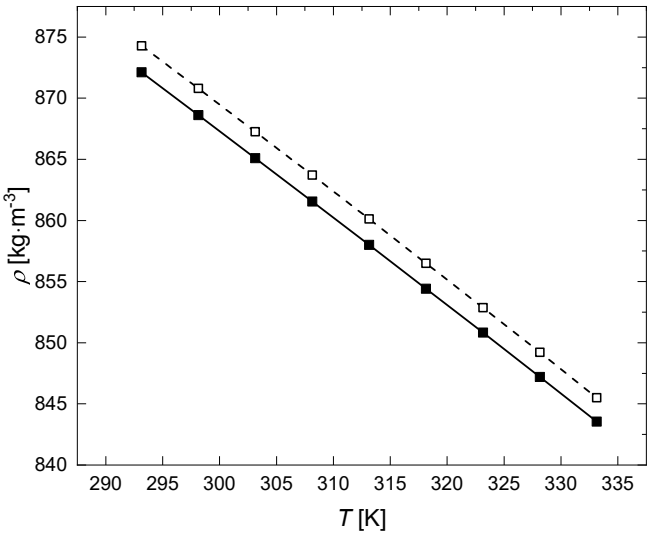


**Figure 1:** Temperature dependence for the density of 1-tdc:Men (1:2) before (closed symbols) and after (open symbols) mixing with water. The water content before mixing (closed symbols) is $289.0 \pm 1.0$ ppm, while after mixing (open symbols), this is $1.77 \pm 0.01$ wt%.

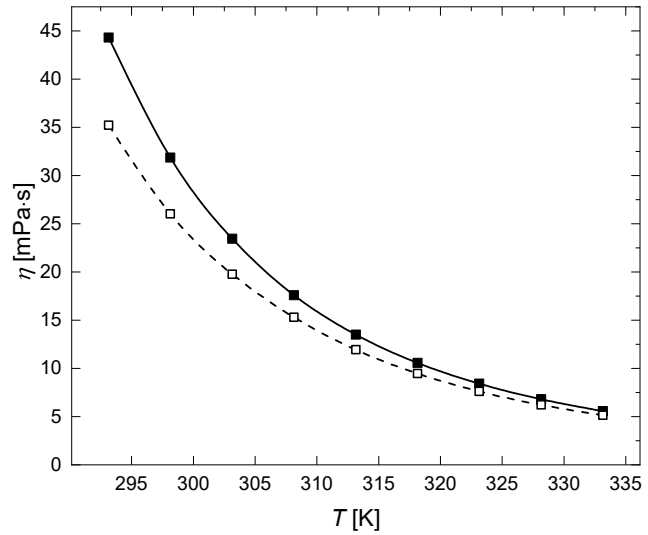


**Figure 2:** Viscosity as a function of temperature for 1-tdc:Men (1:2) before (closed symbols) and after (open symbols) mixing with water. The water content before mixing is $289.0 \pm 1.0$ ppm, while after mixing it is $1.77 \pm 0.01$ wt%.

**Table 2:** Fitting parameters of the Vogel–Fulcher–Tammann expression (eq. (2)).

| DES | $A_\eta$ [mPa s] | $B_\eta$ [K] | $C_\eta$ [K] |
|---|---|---|---|
| Before mixing with water | $9.797 \times 10^{-3}$ | 1025.63 | –171.30 |
| After mixing with water | $3.854 \times 10^{-2}$ | 715.51 | –188.27 |

Another physicochemical property of the DES 1-tdc:Men (1:2) that was measured is the surface tension between DES and air. The surface tension gives indirect information on the intermolecular forces within the liquid. The surface tension for the DES at 293.15 K is $29.33 \pm 0.06\,\mathrm{mN\,m^{-1}}$. This is slightly higher than decane, which has a surface tension of $24.47 \pm 0.06\,\mathrm{mN\,m^{-1}}$, [2] but also somewhat higher than most organic solvents. Most likely, the DES 1-tdc:Men (1:2) has stronger intermolecular attractions, such as hydrogen bonding between the alcohol groups, which induces the higher surface tension.

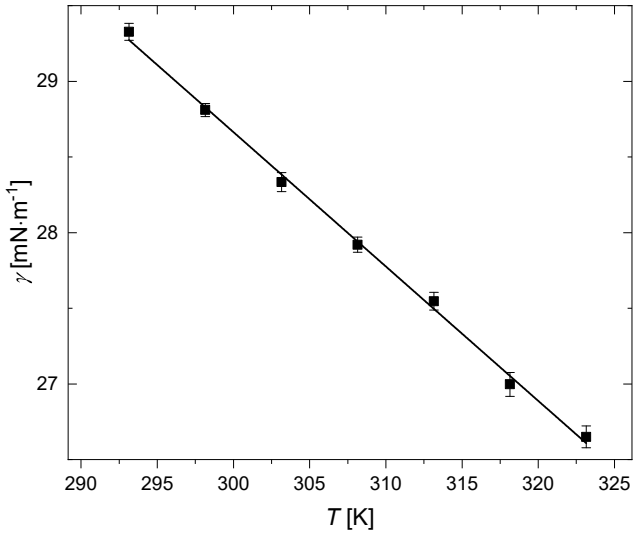


**Figure 3:** Temperature dependence of the air-DES surface tension with the fit $\gamma = H^\sigma - TS^\sigma$.

In fig. 3 we plot the temperature dependence of the surface tension. This enables to obtain the surface entropy $S^\sigma = -\mathrm{d}\gamma/\mathrm{d}T$ and surface enthalpy $H^\sigma$, which gives insight into the structure of the surface. We find $S^\sigma = 0.089\,\mathrm{mN\,m^{-1}\,K^{-1}}$ and $H^\sigma = 55.3\,\mathrm{mN\,m^{-1}}$. In literature for the hydrophobic solvents decane and hexadecane $S^\sigma = 0.094$ and $0.091\,\mathrm{mN\,m^{-1}\,K}$ and $H^\sigma = 52.0$ and $54.8\,\mathrm{mN\,m^{-1}}$, respectively, were found. [2] Thus, the surface entropy and enthalpy of the DES 1-tdc:Men (1:2) is similar to these conventional hydrophobic solvents.

## References

[1] D. J. G. P. van Osch, C. H. J. T. Dietz, J. van Spronsen, M. C. Kroon, F. Gallucci, M. van Sint Annaland, and R. Tuinier, "A search for natural hydrophobic deep eutectic solvents based on natural components," *ACS Sustainable Chemistry and Engineering*, vol. 7, no. 3, pp. 2933–2942, 2019.

[2] L. I. Rolo, A. I. Caco, A. J. Queimada, I. M. Marrucho, and J. A. P. Coutinho, "Surface tension of heptane, decane, hexadecane, eicosane, and some of their binary mixtures," *Journal of Chemical & Engineering Data*, vol. 47, pp. 1442–1445, 2002.