

Deep Eutectic Solvents: Similia Similibus Solvuntur?

Supporting Information

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LAMMPS¹ was employed to run a 10 ns NVT classical molecular dynamic simulation of reline for which the force field of Perkins *et al.*² was employed. The investigated system consists of 144 choline chloride ion pairs and 288 urea molecules. The box length of 3702.5 pm was obtained from a NPT simulation of 2 ns run time which was equilibrated 4 ns. The simulation temperature of 375 K was kept constant by a Nosé–Hoover chain thermostat.^{3–5} Lennard–Jones and Coulombic interactions were computed up to the cutoff radius of 1200 pm. Coulombic interaction energies beyond the cutoff were computed via the particle-particle particle-mesh solver.⁶ A time step of 0.5 fs was selected in all previous mentioned simulations.

Fig. 1 shows the dimer displacement distribution function for all investigated clusters in the classical molecular dynamic simulation. Similar as in the *ab initio* molecular dynamic simulations, indications for clusters migrating together significant longer than the cutoff value of the first solvation sphere could not be observed. We have employed the same cutoff criterion as in the *ab initio* molecular dynamic simulations.

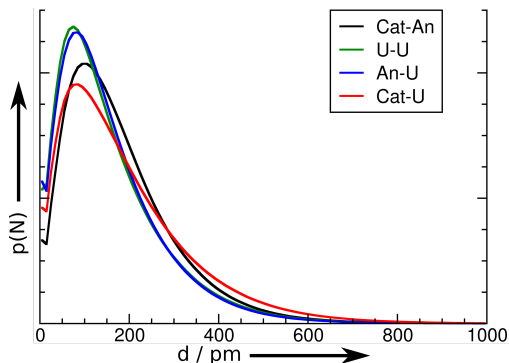


Figure 1: Normalized displacement function of clusters formed of one cation and one anion (*black*), two urea molecules (*green*), one anion and urea (*blue*), as well as one cation and one urea molecule (*red*). Values are added to the displacement function when the investigated cluster splits.

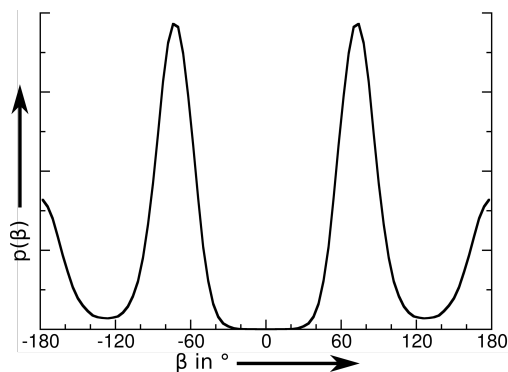


Figure 2: Dihedral angle distribution function of $N_{Ch}-C-C-O_{Ch}$ of choline (β) in the *ab initio* molecular dynamic simulations

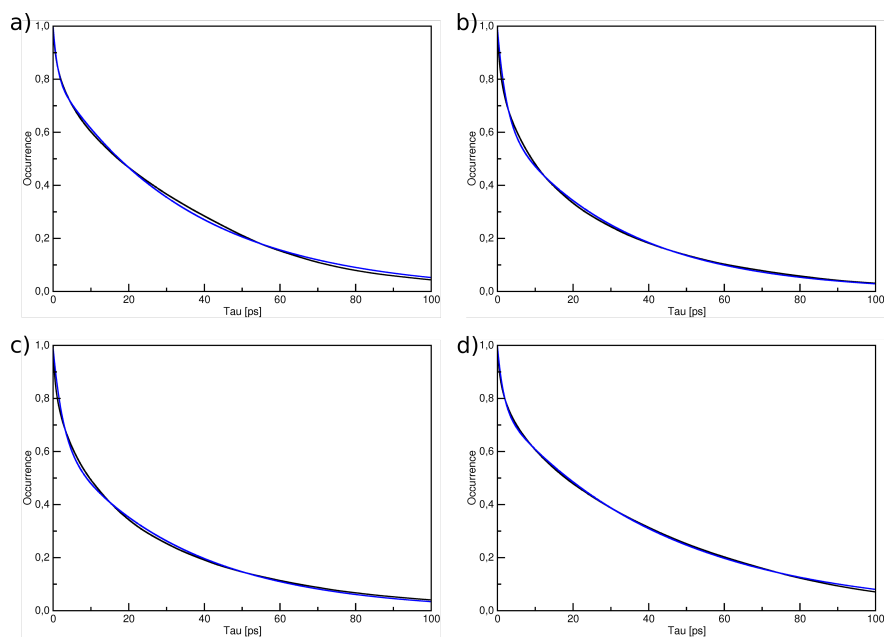


Figure 3: Intermittent autocorrelation function (black) and two exponential fit of the shown autocorrelation function (blue) for an ion pair (a), a cluster formed by two urea molecules (b), a cluster formed by one anion and one urea molecule (c), and a cluster formed by one cation and one urea molecule (d). In all cases, the correlation coefficient R of the two exponential fit was larger than 0.99.

References

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