First Principles Molecular Dynamics Study of a Deep Eutectic Solvent: Choline Chloride/Urea and Its Mixture with Water

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First principles molecular dynamics simulations in the canonical ensemble at temperatures of 333 and 363 K and at the corresponding experimental densities are carried out to investigate the behavior of the 1:2 choline chloride/urea (reline) deep eutectic solvent and its equimolar mixture with water. Analysis of atom–atom radial and spatial distribution functions and of the H-bond network reveals the microheterogeneous structure of these complex liquid mixtures. In neat reline, the structure is governed by strong H-bonds of the trans- and cis-H atoms of urea to the chloride ion. In hydrous reline, water competes for the anions, and the H atoms of urea have similar propensities to bond to the chloride ions and the O atoms of urea and water. The vibrational spectra exhibit relatively broad peaks reflecting the heterogeneity of the environment. Although the 100 ps trajectories allow only for a qualitative assessment of transport properties, the simulations indicate that water is more mobile than the other species and its addition also fosters faster motion of urea.