Supporting Information

Water Distribution at the Electrified Interface of Deep Eutectic Solvents

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S1. Influence of surface charge density on adsorption of ions and molecules of mixed water-reline system (choline chloride-urea +3 wt% water) on graphene.

Figure S1 shows the evolution of the normalized number density of choline (Ch⁺) and urea (URE) as a function of distance from the negatively charged (first column) and positively charged (second column) electrode for different surface polarization.

Figure S1. Normalized number density profile of choline cation (Ch⁺) (a and b) and urea molecule (URE) (c and d) as a function of distance from the graphene electrode for different surface polarization.
S2. Influence of surface polarization on the electrosorption of chloride anions and water molecules of mixed water-reline system (choline chloride-urea +3 wt% water) on graphene.

Figure S2 shows the evolution of the normalized number density of water ($\text{H}_2\text{O}$) and chloride (Cl$^-$) as a function of distance from the negatively charged (first column) and positively charged (second column) electrode for different surface polarization.

**Figure S2.** Influence of surface polarization on electrosorption of humid Reline species. The normalized number density of water molecules (a and b) and chloride (c and d) as a function of distance from the electrode surface.
S3. Intermolecular interactions in 1:2 molar ration of choline chloride-urea + 3 wt%.

Figure S3 shows the average strength of the intermolecular interactions between water molecules and Reline species (Urea-water, black bar; choline-water, red bar; chloride-water, green bar; and water-water, blue bar) at moderate surface polarizations.

Figure S3. The intermolecular interactions between water and urea (URE-H₂O, black bar), water and choline (Ch⁺-H₂O, red bar), water and chloride (Cl⁻-H₂O, green bar) and water and water (H₂O-H₂O, blue bar) at moderate polarizations.