Molecular Dynamics simulations of aqueous Deep Eutectic Solvents: foundations for Machine Learning screening of High Performance Electrolytes

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Recently, the field of chemistry is taking advantage of Machine Learning (ML) to design new materials. One fundamental problem that can be tackled with this approach is the screening of novel High Performance Electrolytes that surpass the current used ones in terms of stability and practicality [1].

In this context, we have put our attention on the promising concept of High Entropy Electrolytes (HEEs). HEEs are electrolytic solutions that contain a higher number of components than regular electrolytes, usually four or more. It has been proven that the effect of high entropy is correlated with improvements in electrochemical stability and ionic conductivity [2]. Unfortunately, data is still scarce on them, so the direct ML approach is not possible yet.

An alternative strategy is to explore a group of related systems, called Deep Eutectic Solvents (DESs), which are interesting as electrolytes because of their high electrochemical stability, and that can eventually be considered as High Entropy Liquids [3]. Data is abundant for DESs, and any lessons on how to model them should be valuable to study HEEs with the ML approach.

The main obstacle for modeling a mixture from properties of its individual components is capturing the non-ideal way in which they combine due to intermolecular interactions. Therefore, we aim to find microscopical effects that are strongly correlated with this non-ideal behavior and that should be considered within our future ML models.

For all of this, we have performed a detailed Molecular Dynamics study of reline (Figure 1), a well-known DES, and its mixture with water. We have characterized it both from the structural and dynamical point of view. This system strongly deviates from ideality around 80% of reline weight, as we have replicated in our simulations (Figure 2).

The main structural effect responsible for this behavior is a change in the hydrogen bond network which governs the intermolecular interactions of the system. We illustrate this network using the Spatial Distribution Function (Figure 3), which greatly aids in the visualization of the coordination spheres.

Among other conclusions, we suggest that ML models designed to predict how DES mixtures behave from its composition should factor in these HB networks within its architecture, and we shall continue on this line of work to ultimately apply it for novel HEEs in a similar fashion.

References

[1] Jagadeesan et al., Adv. Mater. Technol., 8 (2026) e02130.
[2] Ren et al., ACS Energy Lett., 9 (2024), 2960-2980.
[3] Yu Chen and Zhiwu Yu, Green Chem. Eng., 5 (2024) 409-417.

Figures

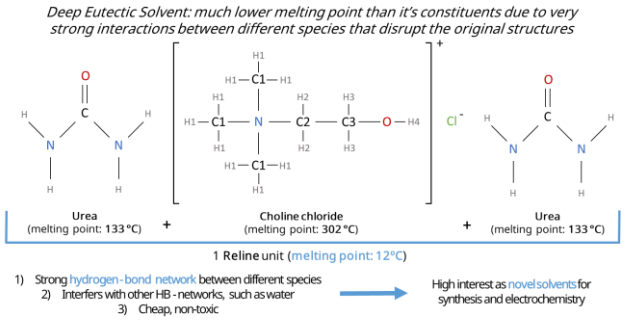

Figure 1. Scheme of reline as a DES and its main properties of interest.

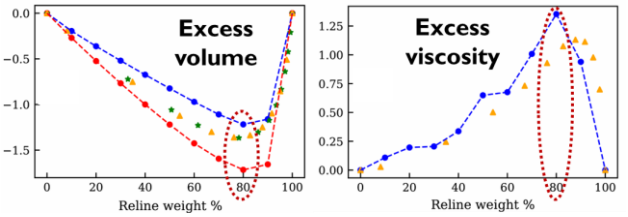

Figure 2. Excess volume (left) and logarithm of viscosity (right) of reline/water system as a function of reline weight percentage. Maximum deviations of ideality around 80% reline wt. are highlighted. Red and blue series are our simulated results, while green stars and yellow triangles are experimental data for validation purposes.

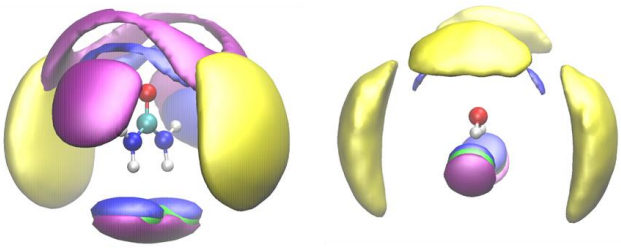

Figure 3. Spatial Distribution Functions of Centre of Mass with urea (left) and water (right) as central molecules for reline/water system at 50% reline wt. Isosurfaces represent the most probable regions in which other molecules are coordinated around the central molecule. Colour code: cholinium cathion (yellow), urea (purple), water (blue) and chloride anion (green).