

Molecular Resolution Nanostructure of Reline—Graphite Interface as a Function of Potential and Water content

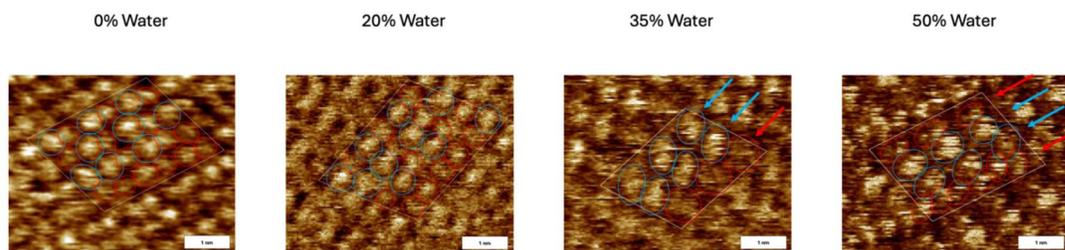
Ho Hong Chau,^a Hua Li,^{a,b} Rob Atkin^{a,*}

^aSchool of Molecular Sciences, The University of Western Australia, Perth, Australia

^bCentre for Microscopy, Characterisation and Analysis, The University of Western Australia, Perth, Australia

*Corresponding author's e-mail address: rob.atkin@uwa.edu.au

Deep Eutectic Solvents (DESs) have attracted increasing interest in electrochemistry as greener alternatives to Ionic Liquids (ILs) over the past decade. However, many DESs are hygroscopic, and previous studies suggest that presence of water would significantly influence DESs physicochemical properties and interfacial nanostructure¹. Furthermore, surface potential is also reported to control DES interfacial nanostructure². Therefore, understanding DES interfacial nanostructure as a function of both water content and potential is necessary for DESs design in electrochemistry.

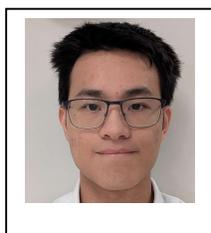


In our work, in situ amplitude modulated atomic force microscope (AM-AFM) is used to probe the Stern and near surface layer of DES Reline (1:2 ChCl:Urea) at highly orientated pyrolytic graphite (HOPG) surface as a function of both potential and water content. Our results shows that the Stern layer adopt choline-urea checkerboard pattern up to 20% water content, while transitioning to a choline double-row pattern with larger lattice angles at 35-50% water content. These Structural changes are consistent with AFM force curves and simulations from previous studies.

These outcomes provide valuable insights into DESs interfacial nanostructure and would benefit DESs application in electrochemistry, including electrodeposition, capacitor, electrolysis and sensors.

References

- (1) Hammond, O. S.; Bowron, D. T.; Edler, K. J. The Effect of Water upon Deep Eutectic Solvent Nanostructure: An Unusual Transition from Ionic Mixture to Aqueous Solution. *Angewandte Chemie* **2017**, *129* (33), 9914–9917. <https://doi.org/10.1002/ange.201702486>.
- (2) Freeman, J. S.; Mamme, M. H.; Ustarroz, J.; Warr, G. G.; Li, H.; Atkin, R. Molecular Resolution Nanostructure and Dynamics of the Deep Eutectic Solvent—Graphite Interface as a Function of Potential. *Small* **2023**, *19* (12). <https://doi.org/10.1002/sml.202204993>.



Mr. Ho Hong Chau obtained his Bachelor of Science with honours in chemistry from The University of Western Australia in 2024 and is now a PhD student in the University of Western Australia under supervision of Professor Rob Atkin and Doctor Hua Li. His research interest includes deep eutectic solvent and adsorbed structures at solid-liquid interfaces.