Modulating micelle morphology and electrostatic correlations through specific ion interactions in deep eutectic solvents

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In recent years, many studies into green solvents have been performed and deep eutectic solvents (DES) have emerged as environmentally friendly alternatives in many fields, such as separation processes, metal processing, biocatalysis and electrodeposition.[1] DES are green solvents obtained through the combination of cheap and simple organic compounds, where the interaction between the precursors promotes a depression in the melting point that allows the mixture to remain liquid at room temperature. Moreover, the combination of different precursors provides a certain control over the physicochemical properties of the solvent (e.g. solvent polarity and charge density). Thus, DES are regarded as task-specific "cocktails", where the properties of the solvent can be tuned to suit particular applications.

Recent developments have shown the ability of DES to support amphiphile self-assembly, bringing the possibility to develop sustainable alternatives for surfactant templating, drug delivery and biosensing, among others. Here we will explore the self-assembly of surfactants in DES as probed using small-angle neutron scattering. The increase of charge density in the solvent is shown to promote micelle growth through electrostatic screening in comparison to more neutrally charged DES environments.[2,3] In addition, the combined effect of hydrophobic and electrostatic interactions through the addition of hydrotropic salts leads to a profound change in micelle shape, resulting in the formation of worm-like micelles.[4] Finally, our latest results in the study of long-range colloidal interactions in DES will be presented. Aiming to gain a better understanding of the fundamental aspects of amphiphile self-assembly in these solvents, we will present details of the micellisation with varied physicochemical properties of the solvent, salt addition and counterion substitution.

[1] B.B. Hansen *et al.*, Deep Eutectic Solvents: A Review of Fundamentals and Applications, Chem. Rev. 121(3) (2021) 1232-1285.
[2] A. Sanchez-Fernandez *et al.*, Surfactant-Solvent Interaction Effects on the Micellization of Cationic Surfactants in a Carboxylic Acid-Based Deep Eutectic Solvent, Langmuir 33(50) (2017) 14304-14314.
[3] A. Sanchez-Fernandez *et al.*, Micellization of alkyltrimethylammonium bromide surfactants in choline chloride:glycerol deep eutectic solvent, Phys. Chem. Chem. Phys. 18(48) (2016) 33240-33249.
[4] A. Sanchez-Fernandez *et al.*, Complex by design: Hydrotrope-induced micellar growth in deep eutectic solvents, J. Colloid Interface Sci. 581(Pt A) (2021) 292-298.