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## A Multidisciplinary Approach to Unveil the Structural Arrangement of Deep Eutectic Solvents: from Local Order to Nano-scale Organization

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Deep eutectic solvents (DESs) deal with mixtures of two or more compounds which, for a well-defined molar ratio, display a unique and minimum melting point that is lower than the ideally predicted one, allowing the achievement of a liquid phase even from solid starting materials. This behavior relies on the extensive interplay among the components, so that it has become evident that DESs are high-entropic systems with tens of punctual interactions ruling the overall chemical-physical properties. This complexity makes the achievement of a clear picture about the structural arrangement of these materials a fundamental knowledge to understand their macroscopic behavior and ultimately promote their applications.

In this work, we demonstrate how a multidisciplinary approach combining X-ray absorption spectroscopy (XAS), small- and wide-angle X-ray scattering (SWAXS), UV-Vis, attenuated total reflection Fourier transform infrared (FTIR), and near-infra-red (NIR) spectroscopies with molecular dynamics (MD) and ab initio simulations is a powerful strategy to unveil the structural arrangement of DESs and their mixtures with co-solvents (i.e., water, methanol) ranging from short- to intermediate-scale levels. More specifically, we focus on the eutectic formed by choline chloride (ChCl) and sesamol in 1:3 molar ratio and on the metal-based deep eutectic solvent (MDES) formed by  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$  and urea in 1:3.5 molar ratio. In the former system, the employed techniques were able to detect a nano-phase segregation between water pools confining most of the ChCl and sesamol-rich domains (Fig. 1a), which is formed for high water contents [1], differently from methanol addition [2]. In the  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ :urea 1:3.5 MDES, we observed a close packing of  $\text{Ni}^{2+}$  ion clusters forming oligomeric agglomerates thanks to the mediation of bridging chloride anions and water molecules [3]. Conversely, urea acts as a sort of “inner solvent” owing to the formation of nanostructures intercalating the Ni-rich regions (Fig. 1b). This arrangement is disrupted upon the introduction of additional water, diluting the system up to an aqueous solution of the MDES constituents. In this way, our digression from shorter to larger lengths allowed the achievement of an all-round picture able to clarify the structural arrangement of these inherently complex systems.

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