



Formation of Ethanolamine-Mediated Surfactant-Free Microemulsions Using Hydrophobic Deep Eutectic Solvents

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Abstract

Hydrophobic deep eutectic solvents (HDESs) are emerging as versatile, relatively benign, and inexpensive alternatives to conventional organic solvents in a diverse set of applications. In this context, the formation of microemulsions with HDES replacing the oil phase has become an area of active exploration. Because of recent reports on the undesirable toxicity of many common surfactants, efforts are under way to investigate the formation of surfactant-free microemulsions (SFMEs) using HDES as an oil phase. We present SFME formation using HDESs constituted of *n*-decanoic acid and five (5) structurally different terpenoids [thymol, *l*(-)-menthol, linalool, β -citronellol, and geraniol] at a 1:1 molar ratio as the oil phase and water as the hydrophilic phase. Ethanolamine (ETA) exhibited the best potential as a hydrotrope among several other similar small molecules. Results showed a drastic increase in water solubility within the HDESs in the presence of ETA. ETA exerted its hydrotropic action at different extent for each DES system via chemical interaction with the H-bond donor (HBD) constituent of the HDES. The optimum hydrotropic concentration (minimum hydrotrope and maximum water retention, XETAOPT) assigned for each DES/ETA/water system and water loading are reported, and the trends are discussed in detail. Ternary phase diagrams are constructed using visual observation and the dye staining method. The area under the single- and multiple-phase regions (assigned in ternary phase diagrams) was estimated. “Pre-Ouzo” enforced by ETA was investigated using dynamic light scattering (DLS) of the DES/ETA/water systems at XETAOPT. A systematic growth in nanoaggregates was observed with the subsequent addition of water in DES/ETA systems while continuously changing the existing microstructure. The presence of a core (oil)–shell (water)-like structure as indicated by the fluorescence response of Nile red in the “pre-Ouzo” region is speculated. We were able to prepare a homogeneous solution of $[K_3Fe(CN)_6]$ salt in “pre-Ouzo” mixtures with no apparent deviation in the Beer–Lambert law.

Keywords: Hydrophobic Deep eutectic solvents, Microemulsion