

From Molecular Nano-Aggregates to Microemulsions: A Comprehensive Description of the Ternary Phase Diagram of Water and Mono-Alcohols mixtures

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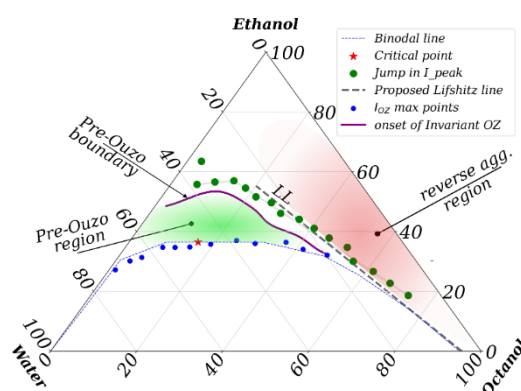
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The aim of our work is to understand how supra-molecular structuration drives nano-to-meso-to-macroscopic organization, phase transitions and affects the dynamics in ternary systems. To achieve this, we examined the ternary mixture of 1-octanol, ethanol, and water, in which several organized phases were identified, among which an Ouzo and pre-Ouzo region [1,2]. Using Small Angle X-rays Scattering, we mapped the entire composition range through successive auto-dilution lines (figure 1, left). By analyzing key structural parameters across the phase diagram, we provided a detailed description of organization at different scales. Beyond the transition from monophasic to biphasic states, we determine the full extension of distinct domains within the monophasic area. These range from hydrogen-bonded molecular clusters to mesoscopic structures characteristic of the pre-Ouzo domain, separated by a so-called Lifshitz line.

Next, using a combination of Neutron Scattering, NMR, rheology, and classical molecular simulations, we present a detailed description of molecular dynamics across the different structural domains. A subtle shift in individual dynamics appears upon crossing the Lifshitz line. Neutron Spin-Echo experiments reveal the collective nature of molecular dynamics, with a de Gennes narrowing effect slowing diffusion in the nano-structured octanol-rich region and a dramatic slowdown in droplet diffusion in the pre-Ouzo zone. We eventually discuss a method for the estimation of the droplets lifetime in such transient organisation, here close to 20 nanoseconds.

Keywords:

Ternary phase diagram, Microemulsions, Ouzo effect, SAXS, Neutron and X-ray scattering



Phase diagram of the 1-octanol, ethanol water ternary system. The blue points align on the binodal, the red and green aligns on the different estimations of the Lifshitz line. IN background, the autodilution lines followed by SAXS are drawn.

References:

- [1] T. Zemb et al., PNAS (2016) 113, 4260–4265
- [2] F. Malayil Kalathil, J. Mol. Liq., 432 (2025) 127684.
- [3] F. Malayil Kalathil, PhD thesis, 2025.

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