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## How the addition of salt destabilizes ionic liquid micellar suspension until phase separation

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Understanding the fundamental mechanisms of self-organization and phase separation in molecular solutions is of utmost importance from the extraction perspective. Aqueous biphasic systems have recently attracted a lot of attention because of their lowest toxicity (i.e. large water content) with respect to classical solvents. The ionic liquid tributyltetradecylphosphonium chloride (P4,4,4,14Cl), mixed in a salt or acid aqueous solution, is a biphasic system. In pure water, it forms spherical micelles with a very low CMC, below 1 wt%, dispersed in the solution thanks to electrostatic repulsion.

However, even large amounts of P4,4,4,14Cl in water do not induce phase separation while the ternary system P4,4,4,14Cl/NaCl/H<sub>2</sub>O is a biphasic system for a large enough salt content [1]. It moreover displays a LCST (Lower Critical Separation Temperature) behavior, being monophasic at low T for a low salt content and experiencing phase separation when T is increased [2]. We combined structural investigation by small angle neutron scattering and specific chloride electrode that allows the determination of chloride counterion adsorption as a function of salt content and temperature. The T-induced phase separation has been ascribed to the T-increased adsorption onto the micellar surface of these additional chloride ions, thus lowering repulsive interactions between micelles, finally allowing coalescence and thus phase separation [3]. The increase of salt content at constant temperature, instead, leads to a direct screening of the interactions between micelles, eventually to phase separation in a similar manner. The comparison with a regulation charge theory shows how specific chloride adsorption controls the stability of the micellar solution.

[1] N. Schaeffer, G. Perez-Sanchez, H. Passos, J.R.B. Gomes, N. Papaiconomou, J.A.P. Coutinho, , Phys. Chem. Chem. Phys. 21 (2019) 7462. <https://doi.org/10.1039/c8cp07750a>.

[2] N. Schaeffer, H. Passos, M. Gras, V. Mogilireddy, J.P. Leal, G. Pérez-Sánchez, J.R.B. Gomes, I. Billard, N. Papaiconomou, J.A.P. Coutinho, Phys. Chem. Chem. Phys. 20 (2018) 9838–9846. <https://doi.org/10.1039/c8cp00937f>.

[3] G. Meyer, R. Schweins, T. Youngs, J.F. Dufrêche, I. Billard, M. Plazanet, J. Phys. Chem. Lett. 13 (2022) 2731–2736. <https://doi.org/10.1021/acs.jpcllett.2c00146>.

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