

A stereoscopic scattering approach to elucidating molecular segregation in highly stable catanionic nanodiscs

Elucidating how multicomponent surfactant formulations segregate into stable nanodiscs requires correlating neutron, X-ray and light scattering with the underlying thermodynamics. In this work, multimodal small-angle scattering provides a quantitative description of molecular segregation and size control in crystalline catanionic nanodiscs. Using a formulation combining cetyltrimethylammonium hydroxide, stearic acid and Akypo® LF2, we show that introducing a highly hydrophilic surfactant provides a reliable lever to modulate nanodisc diameter, stacking, and thermal reversibility.

SANS profiles reveal that Akypo® LF2 segregates selectively to the semi-toroidal rims of nanodiscs, stabilizing high-curvature regions and suppressing the growth of large tactoids under cationic-rich conditions. Temperature cycling above the chain-melting transition demonstrates that this segregation is reversible: stacked nanodiscs melt into vesicles before recrystallizing into freely rotating discs whose diameters can be determined from Rayleigh-derived masses. In contrast, stearate-rich compositions display minimal segregation, preserving the behavior of the pseudo-ternary catanionic system.

The stereoscopic approach further highlights how trace amounts of hydrophobic aldehydes modulate edge-to-edge interactions: citronellal and nonanal reduce tactoid size and, at higher concentrations, trigger “house-of-cards” gelation at unexpectedly low additive levels (~150 ppm). Across all compositions, bilayer crystallinity and interdigitation remain intact, establishing these three-component assemblies as non-lamellar lipid-nanoparticle analogues with finely tunable interfacial organization.

Altogether, this work demonstrates how combining neutron and X-ray scattering with optical techniques and thermodynamic reasoning provides access to a physically interpretable description of self-assembled structures, beyond the limitations of single-profile analysis. The results exemplify the stereoscopic methodology pioneered by Thomas Zemb, linking scattering, segregation and entropy, to build predictive models for advanced colloidal materials and functional nanostructures.

Abstract Title

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