Data-driven density functional theory for phase coexistence & partitioning in multicomponent colloid–polymer mixtures

Vikki Anand Varma [1], Andrew J. Archer [2], and Alberto Scacchi [1]

[1] Department of Mechanical & Materials Engineering, University of Turku, Turku 20500, Finland
[2] Interdisciplinary Centre for Mathematical Modelling and Department of Mathematical Sciences, Loughborough University, Loughborough LE11 3TU, United Kingdom

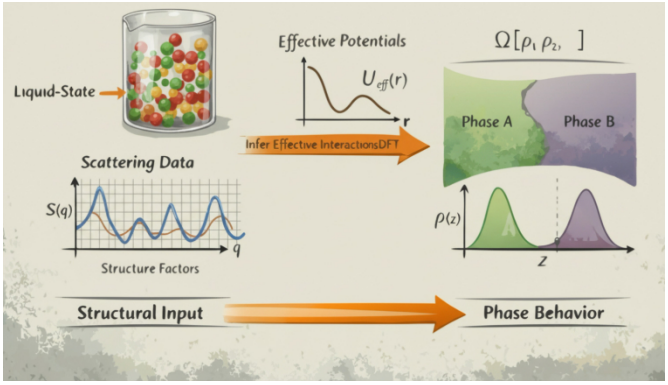
alberto.scacchi@utu.fi

Partitioning of biomolecules and colloids in multicomponent polymer mixtures underlies phenomena ranging from intracellular organization to separation technologies. Yet, predictive understanding of phase coexistence and interfacial structure remains limited, particularly in systems with many interacting species.

Here, we present a unified theoretical and computational framework to investigate phase behavior and partitioning in multicomponent colloid–polymer mixtures. Building on particle-based simulations of phase-separating polymer systems with embedded nanoparticles [1], we develop an improved classical density functional theory (DFT) capable of resolving phase coexistence in mixtures with multiple polymeric and colloidal components [2]. The framework combines refined free-energy functionals with an efficient scheme to determine equilibrium phase compositions and interfacial properties. Quantitative benchmarking against simulations shows very good agreement, while significantly extending the accessible parameter space.

To bridge theory and experiment, we introduce a data-driven extension of the DFT in which effective interactions are inferred from liquid-state structural input, such as partial structure factors. Using structure factors generated from independent simulations as a proxy for scattering data, we demonstrate how this approach enables predictive modeling of experimentally relevant systems without requiring prior knowledge of the underlying pair interactions, typically needed in both particle-based simulations and conventional DFT [3].

Overall, this work establishes a scalable and experimentally grounded framework to connect microscopic structure with macroscopic phase behavior in complex fluids.



Schematic overview of the conceptual framework presented in Ref. 3. AI-generated image.

References:

[1] A. Scacchi, C. Rigoni, M. P. Haataja, J. V. I. Timonen, M. Sammalkorpi, "A coarse-grained model for aqueous two-phase systems: Application to ferrofluids", *Journal of Colloid & Interface Science* **686** :1135-1146 (2025).
[2] V. A. Varma, A. Scacchi, "General approach for partitioning and phase separation in macromolecular coexisting phases", *arXiv preprint* :2509.14392 (2025).
[3] V. A. Varma, A. J. Archer, A. Scacchi, "Bridging structure and thermodynamics: a data-driven density functional approach to complex fluids", *in preparation* (2026).

Acknowledgements:
This work was supported by the Jane and Aatos Erkko Foundation under the grant No. 230052 (A.S.). Computational resources by CSC IT Center for Finland are also gratefully acknowledged. The work was conducted within the #SUSMAT profiling measure at the University of Turku, Finland.