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FROM DYNAMIC STRUCTURES TO FUNCTIONAL MATERIALS

Molecular architecture of bottlebrush polymers governs phase behaviour

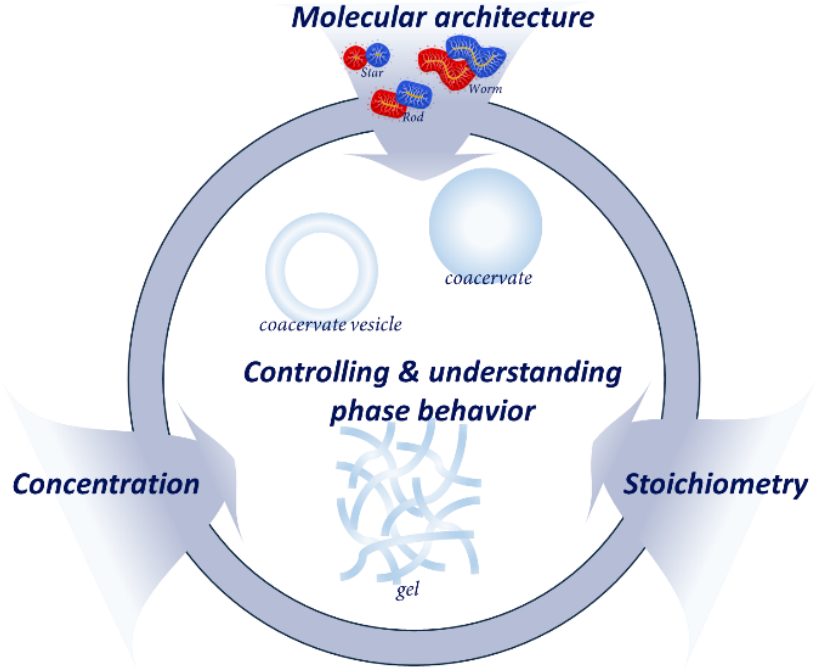
H. Duijs[1], B. Zhao[1], X. Chen[1], W. Noteborn[2], M. Barz[1], L. Su[1]

[1] Biotherapeutics, LACDR, Leiden University, the Netherlands
[2] NeCEN, LIC, Leiden University, the Netherlands

Cells are extremely crowded, yet maintain precise spatial organization through compartments with distinct material states, including liquid condensates, viscoelastic networks, and membrane-bound organelles. This complex organization is essential for cellular functioning, and its disruption is linked to diseases, such as neurodegeneration, cancer and viral infection.[1] The variety in material states arises from chemically similar biomolecules, suggesting an important role of molecular geometry on its mesoscale assembly and subsequent material state.

To address this question, we have synthesized oppositely charged bottlebrush polymers (BBPs) with high grafting density, forming stretched and stiff macromolecules. By varying the backbone length, while keeping the side chains constant, BBPs are created with distinct molecular geometries (spherical, rod-like or worm-like). Using this versatile platform, we systematically investigate the effect of BBP architecture on its phase separation and material properties.

Remarkably, the BBP architecture determines the formation of coacervates, coacervate vesicles or gel-like networks. Not only morphology is tuned in this way, but also the dynamics of these assemblies are drastically changed. Notably, anisotropic rod-like BBPs form liquid vesicles, likely driven by directional stacking interactions and slow relaxation. These findings demonstrate that the geometry of the molecular building block can encode several biologically relevant material states, providing mechanistic insights into the role of biomolecular architecture in cellular organization.



References
[1] B. Wang, *et al.*, *Sig Transduct and Target Ther.* **6**, 290 (2021).