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Polymer brush collapse under shear flow

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Shear is observed in many natural and technological systems, affecting their structure, dynamics, function and performance. Entangled polymers exhibit unique flow behaviours, as relaxation processes occur on time scales relevant to our daily lives, from milliseconds to hours or even days. Investigating the relation between out-of-equilibrium microscopic structure and dynamics of fluids and their macroscopic rheological response can enhance our understanding of viscoelastic flow, leading to improved material properties and applications.

This study combines neutron reflectometry (NR), rheology, and computer simulations to characterize the behaviour of polystyrene (PS) brushes under shear by an entangled PS semi-dilute solution. Two brushes with different chain lengths and grafting densities were used. NR reveals similar shear effects on both brushes restricted to the overlap region, causing a decrease in brush thickness and a sharper brush-bulk interface. In addition, the brush thickness returns to equilibrium upon cessation of shear, and the effect can be cycled many times over. The collapse of the brush occurs regardless of the type of brush used, indicating that the dynamics governing the structural change are determined by the free chains in solution rather than the brush itself. Coarse-grained computer simulations of the interfaces were in agreement with the experimental data.

We have recently developed a novel setup that enables the characterisation of sheared interfaces by combined NR and polarised infrared spectroscopy. The main novelty of this setup lies in the use of polarised infrared spectroscopy, which allows following any anisotropy due to shear stress appearing at the interface and, thus, determines molecular orientation. Additionally, NR allows elucidating the structure of these brushes perpendicular to the interface. The first results obtained from this new setup will be presented.

This research shows the feasibility of engineering shear-responsive polymer brushes in entangled polymer solutions, with potential applications in nanosensors and dynamic surface friction and adhesion control.

References

[1] Wolf, M. et al. Combined neutron reflectometry and rheology. J. Appl. Crystallogr. 2013, 46, 1729-1733.
[2] Korolkovas, A. et al. Polymer Brush Collapse under Shear Flow. Macromolecules 2017, 50, 1215-1224.

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Thin films and interfaces in soft matter and materials science

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