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Molecular Scope: watching macromolecular dynamic at solid-liquid interfaces at the single-chain level

The dynamics of macromolecules at solid-liquid interfaces is key to a number of surface phenomena such as functionalization, transport in porous media or friction. In the recent years, novel single-molecule microscopy techniques based on total internal reflection fluorescence microscopy have allowed to probe the in-plane equilibrium diffusion dynamic of macromolecules at the single chain level. These experiments revealed heterogeneous surface motion characterized by periods of immobilization at the surface followed by long desorption-mediated jumps [1, 2], in strong contrast with the traditional vision of Brownian-like macromolecular diffusion at interfaces.
Here, we couple microfluidics with our single-molecule set-up to bring these approaches towards out-of-equilibrium situations and probe the subtle interfacial couplings arising between flow and macromolecular transport at solid/liquid interfaces. We probe in particular the effect of solvent flow on the statistics of macromolecular trajectories at the interface. We observe an increasingly skewed distribution of interfacial displacements for increasing flow rates, due the symmetry-breaking effect of the flow. The effect appears particularly strong for the larger desorption-mediated surface displacements corresponding to transient excursions in the solvent, and we probe as well the effect of the flow on the desorption statistics of the chains. Comparing hydrophobic and hydrophilic surfaces, we evidence distinct flow-induced dynamics, which we ascribe to a strong effect of the finite slip length at hydrophobic silanized surfaces. Our approach will brings a new molecular vision of macromolecular friction at solid/liquid interface.

[1] M. J. Skaug, J. N. Mabry and D. K. Schwartz, J. Am. Chem. Soc., 136, 1327 (2014)
[2] C. Yu, J. Guan, K. Chen, S. C. Bae and S. Granick, ACS Nano, 7, 11, 9735 (2013)

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