Control and Probe Single-Molecule Dynamics of “Smart” Polymers in Solution and at Surface

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There has been growing interest in using stimuli-responsive polymers in solution and at surface as “smart” functional materials or coatings for broad applications from drug delivery to energy storage. Their equilibrium structures and assemblies have been much explored, yet the interfacial dynamics is not. In this talk, I will present our recent work on controlling and examining the conformational dynamics and molecular transport at stimuli-responsive polymer interfaces at a single-molecule level. The first project involves manipulating the conformational transition of polyelectrolyte chains in response to spatially uniform ac-fields. In stark contrast to an abrupt first-order coil-to-globule transition (CGT) by tuning solution pH, we have observed a gradual and hysteretic CGT within an optimum ac-frequency window and beyond a critical ac-field strength of molecular-weight dependence, at which the electrostatic energy barrier between coil and globule conformations are reduced below thermal fluctuation energy by ac-induced counterion migration and condensation. As we further investigate the interfacial dynamics of polymer chains tethered at surface, the second project I will talk about involves probing the diffusive dynamics of small molecules adsorbed at polymer brush interfaces. Much slower surface diffusion is observed with fluorescence probe molecules adsorbed on polymer brush surfaces than on methyl-terminated monolayers, despite much stronger surface adsorption of probe molecules in the latter case, suggesting the strong coupling of molecular thermal fluctuation and polymer chain dynamics. We have also observed an optimal range in grafted polymer chain thickness to facilitate molecular surface dynamics, where the polymer chains adopt the brush conformation with high polymer end-segment density.

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