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Impact of brush morphology on glass transition and glassy state properties of polymer thin films

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Polymer thin films with enhanced stability are highly desired for applications ranging from antifouling coatings for biomedical devices to organic optoelectronics. Depending on the grafting density (σ_p) of a polymer brush, end-grafted chains can adopt a stable arrangement, leading to enhanced thermal stability (i.e., increase in glass transition temperature, or T_g) regardless of polymer-substrate interactions. Confinement effects are expected to make such changes more drastic. Here, we systematically investigate the role of chain packing and confinement on overall film properties using a polymer brush as a model. We employ a synthetic protocol to form ultradense polystyrene (PS) brushes with a grafting density approaching the theoretical maximum limit of $\sigma_{p,max} = 1.45 \text{ nm}^{-2}$. We observe that a 5 nm PS brush ($\sigma_p = 1.23 \text{ nm}^{-2}$) exhibits a T_g that is $\sim 75 \text{ K}$ greater than a spin-coated film of similar thickness. In addition to studies on glass transition, we are exploring the impact of polymer brush morphology on glassy state properties, specifically physical aging and fragility, in thin films. Complementary techniques for these studies include ellipsometry and fast-scanning calorimetry. Ultimately, we aim to elucidate the complex relationship between confinement and chain packing to design highly stable polymer thin films.

Primary author: SRINIVASAN, Sneha (Princeton University)

Co-authors: XU, Quanyin (Zhejiang Sci-Tech University); Prof. ZUO, Biao (Zhejiang Sci-Tech University); Prof. PRIESTLEY, Rodney (Princeton University)

Presenter: SRINIVASAN, Sneha (Princeton University)

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