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Ph.D. DISSERTATION DEFENSE

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Title: Interfacial Design for Polymer Nanocomposites: From Miscibility to Macroscopic Properties

Chairperson: **Dr. Pinar Akcora**, Department of Chemical Engineering and Materials Science, School of Engineering and Science

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ABSTRACT

The design of mechanically adaptive polymer nanocomposites (PNCs) for next-generation materials relies critically on understanding the dynamics of the interfacial region, where polymer chains directly interact with nanoparticles (NPs). In this regime, chain mobility often deviates significantly from bulk behavior due to confinement effects. Because this interfacial region presents a high surface-area-to-volume ratio, optimizing its structure and dynamics is essential for enhancing overall material performance. Previously, we developed an asymmetric interphases by pre-coating high glass transition temperature (T_g) poly(methyl methacrylate) (PMMA) polymer onto silica NP surfaces and blending them with chemically distinct low-T_g poly(methyl acrylate) (PMA) matrix. This design enables thermally reversible modulation of viscosity and modulus, offering a promising platform for the development of high-temperature adhesives and structural materials. This system creates a dynamically heterogenous interface, and this dissertation systematically investigates the underlying molecular contribution to the microscale complex flow behavior by decoupling the blend interfacial behavior using quasi-elastic neutron scattering. Subsequently, we tuned the interphase architecture of the chains by synthesizing a polymer brush of PMMA on silica NPs using Radical Addition Fragmentation Chain Transfer polymerization. While this strategy helps to mitigate agglomeration and tune interfacial adhesion, we design a new chemistry that introduces a more nuanced topological constraint: *polymer loop brush*, where two chains are clicked on a single particle. This seemingly subtle change dramatically alters the entropic and enthalpic landscape at the NP interface. Molecular dynamics (MD) simulation in conjunction with experiment was used to differentiate loop and linear graft interfacial architecture, where the loop fraction acts as an extra parameter to improving particle dispersion and reinforcement, broadening the design space for tuning the mechanical response of PNCs.