Abstract Submitted
for the MAR16 Meeting of
The American Physical Society

Role of bound polymer mobility on multiscale dynamics of PEO in attractive nanocomposites ERKAN SENSES, Univ of Maryland-College Park NIST Center for Neutron Research, ANTONIO FARAONE, NIST Center for Neutron Research, PINAR AKCORA, Stevens Institute of Technology — We study intermediate and large scale chain dynamics in nanocomposites where particle-bound polymer (PMMA) and matrix (PEO) chains are chemically different, miscible, and have very large T_g difference ($\Delta T_g \approx 200$ K). These nanocomposites with dynamically asymmetric 'polymer blend interphases' were shown to exhibit an unusual reversible thermal-stiffening accompanied by vitreous-to-rubbery transition of PMMA.* Using quasi-elastic neutron scattering, this work examines the impact of mobility of the bound-polymer on segmental and collective dynamics of the matrix chains at sub-ns to 100 ns range. While bare silica particles appear to slow down the segmental relaxation, in the composites with PMMA coated particles the Rouse dynamics of PEO is identical to its bulk behavior, possibly due to the negligible enthalpic interaction inherent to this blend system. On larger scale, we observed ≈25% increase in the apparent tube diameter of PEO when PMMA is glassy. Remarkably, the tube size recovers its bulk value as PMMA softens at elevated temperatures. The resulting disentanglement-entanglement transition of PEO under hard and soft confinement well relates to the macroscopic softening-stiffening transition of these composites as evidenced from the bulk rheology. (*ACS Appl. Mater. Interfaces, 2015, 7 (27), pp 14682–14689)

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Date submitted: 06 Nov 2015 Electronic form version 1.4