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DEPOSITION DYNAMICS FOR PROCESSING FUNCTIONAL POLYMER NANOCOMPOSITE MEMBRANES

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Polymer functionalized nanoparticles have garnered strong research interests due to promising uses in a wide range of applications including drug delivery, tribology and stabilized composite solutions. Here we report on the synthesis of polymer nanocomposite thin films for applications related to membrane-based water treatment where the nanoparticle is functionalized for targeted sequestration or remediation of pollutants. The filler material for the model nanocomposite membranes are comprised of a ~10 nm silica core surrounded by chemically attached polymer “brushes”; these “hairy nanoparticles” (hNPs) are embedded in a polymer matrix to form the nanocomposite films.

Suspensions of highly uniform polyacrylic acid(PAA)-silica hybrid particles were studied to measure the dynamics of deposition and shear flow using quartz crystal microbalance with dissipation monitoring (QCM-D) and traditional rheometry respectively. Reversible addition-fragmentation chain transfer (RAFT) polymerization was used to synthesize polymer brushes on the nanoparticles. Polymer molecular weight, grafting density and the degree of ionization of the polymer were systematically varied on the nanoparticle. To create *grafting from* surfaces, RAFT-agent-decorated silica nanoparticles were introduced to a solution of monomer and initiator to create the hNPs. By varying the ratio of initiator to monomer, hNPs with a range of molecular weights were created. To control the grafting density, the RAFT agent was mixed with a propyl-terminated silane to create non-polymerized sites on the surface of the nanoparticle. The ratio of RAFT-silane to propyl-terminated silane determined the density of active chain-growth sites. From these experiments direct structure-processing relationships could be developed to inform processing of hNP based thin films.

The mechanism of hNP deposition and resulting particle adhesion on a substrate is influenced by both the material properties of the constituents and the shear processing conditions. The ability to measure deposition behavior of hNPs *in situ* will allow a direct measurement of the mechanism as a function of time. To this end, hNP suspensions were flowed across a QCM sensor under model processing conditions. As the hNPs deposit on the QCM sensor, the measured frequency will shift and dampen based on the properties of the deposited material.

Over the course of a QCM-D experiment, the total deposition mass and the instantaneous deposition rate is known across the time of the experiment. This information was used to construct an analytical kinetic model of deposition behavior. Furthermore, the adhesion behavior between model polyacrylic acid brush surfaces and inorganic nanoparticles decorated with polymer brushes or other surface modifiers was characterized *in situ* with QCM-D. The structural characteristics of the modified nanoparticles and the resulting nanostructure of the deposited nanoparticle thin films was correlated with the deposition conditions to reveal key structure-processing relationships required for the scalable processing of nanocomposite thin films and coatings.