123e Interfacial Engineering by Surface-Initiated Polymerization and Subsequent Modification

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We report two new methods that require only a single polymerization step to prepare ultrathin copolymer films on metal surfaces. In one case, we prepare copolymer films containing blocks with various fluorocarbon and hydrocarbon functionality by derivatizing side chains of surface-initiated poly(hydroxyethyl methacrylate) (PHEMA) films on gold through reaction with functional acid chlorides. We have previously demonstrated that fluorinated esters created in this manner may be hydrolyzed back to PHEMA by brief exposure to base. Controlled hydrolysis results in depth-dependent regeneration of PHEMA that can be subsequently refunctionalized to create a variety of copolymer films with tailored interfacial composition and barrier properties. In a second approach, we expose a PHEMA film to a mixture of alkyl and perfluoroalkyl acid chlorides to generate a copolymer film in a more random fashion. Phase separation of the pendant fluorocarbon side groups from the hydrocarbon side groups provides a film with nanoscale order in which the fluorocarbon groups dominate the outermost surface. Selective hydrolysis of the fluorocarbon chains yields a film with hydrophobic regions and hydrophilic pores.