476a Synthesis and Characterization of Mesoporous Silicate Films in Supercritical Carbon Dioxide through the Replication of Block Copolymer Templates

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Ordered mesoporous metal oxide films are of enormous interest due to their potential applications in separations, microfluidics, sensing and detection arrays, catalysis, and microelectronics. We have synthesized micron-thick highly-ordered mesostructured silica films by selective condensation of silicon alkoxides within the hydrophilic phase domain of poly(ethylene oxide-block-propylene oxide-block-ethylene oxide), PEO-b-PPO-b-PEO, triblock copolymer dilated with supercritical carbon dioxide. Subsequent calcination of this polymer-silica nanocomposite yields an ordered mesoporous material with a high surface area and regular pore order. First, we will discuss several strategies to reduce the structural contraction in these films, which is desirable for several applications. These strategies include infusion and condensation of the metal alkoxide precursor into the polymer template at elevated temperatures, ammonia exposure of the as-infused polymer-silica composite to further condense the silica network, and electron beam and ultraviolet irradiation of the composite films. We will discuss the specific structural and compositional changes in the films during these post-treatment steps. Second, we will discuss the use of novel, completely amorphous templates containing hydrophilic homopolymers such as poly(acrylic acid) (PAA) and poly(4-vinyl phenol) (PVPh) that offer several advantages. The absence of crystallinity affords better control over the block copolymer morphology and the presence of the homopolymer offers superior wetting properties, which enable high temperature infusions. Moreover, the silica films prepared using these templates exhibit a higher degree of order compared to the amorphous-crystalline templates. A higher degree of order translates into superior mechanical properties that are critical for device applications.