The effect of surface energy on the thin film structure of poly(ethylene oxide-b-styrene-b-isoprene) triblock copolymers with known bulk morphologies was investigated. Films with various polyisoprene (PI) contents, and with film thicknesses on the order of the bulk copolymer domain spacings (d₀), were analyzed on gradient substrates using atomic force microscopy (AFM), x-ray photoelectron spectroscopy (XPS), and near-edge x-ray absorption fine structure (NEXAFS). Surface energy libraries were generated on silicon wafers coated with a methyl-terminated self-assembled monolayer, accelerated under a UV/ozone lamp. Using this approach, surfaces exhibiting water contact angles ranging from 90° to 15° were probed on a single wafer. Surface energy and PI content influenced the polymer nanostructures and the film's terrace structures. Terrace heights typically corresponded to 1/2 d₀, and 3/2 d₀, indicating an asymmetric wetting layer in all cases. Indeed, the substrate surface energy influenced the structure of the wetting layer, which then affects the structure of the overlaying film. Several distinct polymer morphologies were identified, including a hexagonally-packed structure, a network-like structure, and planar lamella. For some triblock systems, the morphologies were surface energy dependent, and polymer morphology often varied from the 1/2 d₀ terrace to the 3/2 d₀ terrace. In addition, for low PI content materials and certain ranges of substrate surface energy, autophobic dewetting of material from the wetting layer was observed.