Conformation, temperature behavior and assembly of polymer chains in aqueous environment are largely dependent on competition of chain hydration and interpolymer binding. Lower critical solution (LCST) or upper critical solution temperature (UCST) behavior in polymer solutions resulting from such a competition has been used for the development of responsive polymer assemblies for a variety of applications, including control drug and/or small molecule delivery. In this talk, I will first review our recent results on responsive hydrogen-bonded layer-by-layer (LbL) films of upper critical solution temperature micelles (UCSTMs) and UCST-type star polymers. The emphasis will be on the role of assembly condition and ionization of the film components on film morphology and temperature response of such assemblies. I will then focus on the discussion of the role of mixed and hydrated salt solvents on the formation of hydrogen-bonded multilayers and temperature-responsive hydrogels for temperature-controlled shape retention of inorganic phase change materials.