This work aims to achieve inexpensive, reusable actinide- and lanthanide-specific binding sites on high surface area functionalized mesoporous materials with potential for use in separations, sensing, sequestration, and vitrification. Mesoporous silica type SBA-15 was chosen as a substrate based on its surface chemistry, stability, pore size, large surface area, and ease of production. Important features of the ligands grafted into a monolayer inside the SBA-15 pores, such as the functional groups and chelating ring size, were improved toward selective actinide complexation by systematic variation. Binding affinity was determined by batch complexation studies as a function of pH, measured using inductively coupled plasma-mass spectrometry (ICP-MS) and liquid scintillation counting (LSC). Eu(III), Ce(III), and Zr(IV) were used as initial test species before examining An(IV) and An(VI). The systems most promising in initial tests were further explored with respect to solution pH, analyte concentration, binding kinetics, and the presence of competing ions.