In large biological molecules, self-assembly and folding rely on various non-covalent interactions for generating functional higher-order structures. Mimicking this approach for the self-assembly of complex artificial supramolecular structures is still challenging. Building blocks with characteristics like low symmetry and flexibility, while common in biological systems, pose numerous problems for synthetic self-assembly. In this thesis, the research advanced knowledge of how non-covalent interactions designed into aromatic amide-based building blocks for self-assembly can be used to control conformation and generate complementary shapes for directing self-assembly of larger structures and through this, allowed us to develop an approach for the formation of more complex structures with interesting properties and reactivities. This work was divided into two different but complementary approaches: 1) Metal-based self-assembly of aromatic amide units and 2) The use of multiple helix formation in aromatic oligoamides as a driving force for self-assembly of larger systems. In chapter 3, a series of heteroaromatic amide-based ligands were synthesized and used for coordination-driven self-assembly of MnL2n polyhedral cages. The subsequent work in chapter 4 studied the more complex novel dual curvature ligands. Next, using the idea of the Diels-Alder reaction, we studied covalent post-modification in chapter 5 as a shorter alternative pathway to achieve similar complex structural and functional diversity. Finally, in chapter 6, we describe another important advance in this thesis, a proof of concept work on the use of multiple helix formation with aromatic oligoamide foldamers as a driving force for the self-assembly of higher-ordered architectures. Our design approach with non-covalent interactions and complementary shapes is of importance to advance knowledge and open up new design possibilities for controlling self-assembly of more complex systems, for instance, allowing isomer selectivity in coordination-driven self-assembly.