Proteins represent the most versatile building blocks available to living organisms or the laboratory scientist for constructing functional materials and molecular devices. Underlying this versatility is an immense structural and chemical heterogeneity that renders the programmable self-assembly of proteins a difficult design task. To circumvent the challenge of designing extensive non-covalent interfaces for controlling protein self-assembly, we have endeavored to use chemical bonding strategies based on fundamental principles of inorganic and supramolecular chemistry. These strategies have resulted in discrete or infinite, 1-, 2- and 3D protein architectures that display high structural order over many length scales yet are dynamic and stimuli-responsive while also possessing new physical and functional properties. In this talk, I will present some of the recent protein-based materials constructed in our laboratory.