Crystallization of organic or inorganic species is of paramount importance in many technological and natural processes, from the production of high-value added pigments, pharmaceutics, and novel nanomaterials, to the formation of shells, bones, and teeth. Crystals can be viewed as an example of self-assembly since they are ordered three-dimensional arrays of atoms/molecules. Crucial features (crystallographic orientation, morphology, size, polymorphism) of such an assembly process are defined at very early stages, and can be dramatically affected by a slight change in the interacting molecules or their molecular network motifs (by the presence of small impurities, macromolecules, etc.). This presentation will highlight our recent efforts toward the fundamental understanding of factors that control the nucleation and growth of important biominerals such as calcium carbonate, calcium oxalate, and calcium phosphate. We establish that macromolecules (nonionic polymers, polyelectrolytes, polypeptides, surfactants) can control the final morphology and polymorph by (i) face-selective adsorption on crystals (thermodynamic regime) or by (ii) temporarily stabilizing the primary nanoparticles formed thus leading to mesoscale crystallization (kinetic regime). We will also discuss the implications of our findings in the context of the design of crystal growth inhibitors and their role in the regulation of pathological biomineralization.