As an emerging tissue manufacturing technique, 3D bioprinting offers great precision and control of the internal architecture and outer shape of a scaffold, allowing for close recapitulation of complicated structures found in biological tissue. In addition, 4D bioprinting is a highly innovative additive manufacturing process to fabricate pre-designed, self-assembly structures with the ability to transform from one state to another directly off the bioprinter. The term “4D” refers to the time-dependent dynamic process triggered by specific stimulation according to predesigned requirements. However, current 3D/4D bioprinting based additive manufacturing technologies are hindered by the lack of advanced smart “inks”. Therefore, the main objective of our research is to develop novel biologically inspired nano or smart inks and advanced 3D/4D bioprinting techniques to fabricate the next generation of complex tissue constructs (such as vascularized tissue, neural tissue and osteochondral tissue). For this purpose, we designed and synthesized innovative biologically inspired nanomaterials (i.e., self-assembly materials, and conductive carbon nanomaterials) and smart natural materials. Through 3D/4D bioprinting in our lab, a series of biomimetic tissue scaffolds were successfully fabricated. Our results show that these bioprinted nano or smart scaffolds have not only improved mechanical properties but also excellent cytocompatibility properties for enhancing various cell growth and differentiation, thus promising for complex tissue/organ regeneration.