The introduction of nanoparticles into polymers constitutes a powerful strategy for enhancing their thermomechanical properties and for introducing new optical, electrical, and magnetic functionalities into the polymers. For many applications, such as plasmonics, the nanoparticles are required to be assembled into specific higher-order arrangements or structures. In this talk, I will discuss the crucial role played by molecular simulations in developing new strategies for assembling nanoparticles into anisotropic structures within polymers. I will first discuss our work on the assembly of polymer-grafted shaped nanoparticles that led to a simple strategy for creating tunable face-to-face or edge-to-edge plasmonic nanojunctions within a polymer film [1,2]. Next, I will discuss how 3-body effects arising from the deformability of polymer grafts lead to anisotropic interactions between uniformly-grafted spherical nanoparticles, which could under certain conditions stabilize a novel dimer phase [3]. Finally, I will discuss how one could exploit the surface tension between immiscible polymer layers to assemble nanoparticles into unique configurations, such as dimers with tunable tilt angle or ribbons with planar or zigzag topography.