Carbon and hydrogen, organized appropriately, have generated a massive global industry with products touching most aspects of modern life. Three quarters of a century later, polyethylene and other plastics are under a microscope due to the plastics waste problem. As part of the solution, we are searching for ways to craft polyethylene with new combinations of properties by carefully controlling the microstructure and functionality. Using this approach, all polyolefin articles can replace those with non-recyclable content, thus further enabling the circular economy. The catalysis to control both microstructure and functionality of polyethylene will be detailed, including some specific examples where new applications are enabled by these modified plastics.