

Abstract: Traditional applications of metal-organic frameworks (MOFs) are focused on gas storage and separation, which take advantage of the inherent porosity and high surface area of these materials. The MOFs' use in technologies that require charge transport have lagged behind, however, because MOFs are poor conductors of electricity. We show that design principles honed from decades of previous research in molecular conductors can be employed to produce MOFs with remarkable charge mobility and conductivity values that rival or indeed surpass those of common organic semiconductors and other coordination polymers. We expect that such high surface area, ordered, and crystalline conductors will be used for a variety of applications in thermoelectrics, rechargeable battery technologies, electrocatalysis, electrochromics, or new types of photovoltaics. Another virtually untapped area of MOF chemistry is related to their potential to mediate redox catalysis. We show that MOFs can be thought of as unique weak-field ligands that give rise to unusual molecular coordination environments where small molecules can be isolated and activated in a matrix-like environment, akin to the metal binding pockets of metalloproteins. By employing a mild synthetic method and a suite of spectroscopic techniques, we have undertaken forays into the redox chemistry of the MOFs' metal nodes, an exciting new area for small molecule activation.