Synthesis and Characterization of an organometallic polymer comprising of Fe4S4 clusters and Janus biscarbene linkers

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Abstract

Iron – sulfur (Fe-S) cluster assembly is an essential process for all cells. More than 120 types of Fe – S enzymes and proteins have been identified since 1960, and they are found to be involved in redox processes such as electron transfer and CO2 fixation. Thus, Fe – S clusters have become ideal candidates for use as building blocks in electronic materials due to their stability in numerous oxidation states and nuclearities. However, incorporating Fe – S clusters into crystalline solids remains a challenge. In this thesis, we report the assembly of bioinspired Fe4S4 clusters with poly(N-heterocyclic carbene)s (NHCs) as a route to new functional main-chain organometallic polymers (MCOP). More specifically, the mono-NHC ligated Fe4S4(Pri2NHCMe2)4 undergoes global substitution in the presence of a Janus biscarbene Et4BBNHC under solvothermal conditions in benzene. As a result, a blue, amorphous Fe4S4 – MCOP was formed. The UV-visible spectroscopy and NMR studies confirm the presence of Fe4S4 cluster in the polymer. The cyclic voltammetry (CV) shows that the polymer does not exhibit reversible redox process. Two new materials, including a Fe4S4•C60 cocrystal and cocrystal MCOP were also prepared, to investigate a templating strategy toward stabilized frameworks. The visible – near IR, cyclic voltammetry and energy dispersive analysis spectra support the presence of both Fe4S4 cluster and C60 in the cocrystal. Characterizations of the cocrystal MCOP are currently underway. These new bioinspired, electronic materials have potential applications in energy storage and catalysis.