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Title: COORDINATION-DRIVEN MULTI-COMPONENT SELF-ASSEMBLY OF DISCRETE AND

POLYMERIC COORDINATION ARCHITECTURES AND THEIR APPLICATIONS IN CATALYSIS,

CO 2 CAPTURE AND FIXATION, AND SENSING

Authors: Gupta, Vijay (/jspui/browse?type=author&value=Gupta%2C+Vijay)

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Abstract:

Rapid advances and widespread interest in the study of macromolecules of nanoscopic dimensions, has resulted a fascinating growth in the field of supramolecular chemistry over the past few decades. The discovery of "crown ethers", "cryptands", and "spherands" by Charles John Pedersen. Donald James Cram and Jean-Marie Lehn in early 1960s, has established the importance of supramolecular chemistry in recognition process via noncovalent interactions such as hydrogen bonding, charge-charge, donor-acceptor, □ - □ and van der Walls etc. Synthetic organic chemists enjoy the luxury of having a large collection of reliable reactions for preparing such programmed molecules ranging from small molecules to mesoscopic and polymeric structures. Inspiration from the natural ability of the simple molecular precursors to self-assemble into extremely complex structures via the above mentioned weak interactions, has led to an exponential growth in the field of supramolecular chemistry with numerous examples reported every year. However as the scale and degree of complexity of desired molecules increase, the assembly of small molecular units into large, discrete supramolecules becomes a difficult task. Following Alfred Werner's discovery on the geometric aspects of how ligands bind to metal ions, there was a rapid development of coordination chemistry aided by the growing understanding of coordinative bonding in the realm of both discrete and extended structures. After the pioneering work of Fujita et al. for the synthesis of discrete molecular squares and Robson et al. for the synthesis of extended metal organic structures (also referred to as coordination networks or coordination polymers), coordination driven self-assemblies have gained considerable attention for the synthesis of discrete and polymeric structures with desired structural topologies. This thesis work presents the strategic design of such architectures via multicomponent self-assembly of metal ion, neutral polypyridyl ligands and anionic carboxylate linkers in a one pot self-assembly procedure without the need of any predesigned precursors. The solid state structures of the obtained coordination architectures (discrete or polymeric) have been established by single crystal X-ray diffraction analysis and their bulk phase purity has been supported by various analytical techniques like powder X-ray diffraction, FT-IR, UV-Vis spectroscopy, thermo gravimetric and elemental analysis. The structural diversity of these coordination architectures has been systematically studied by changing one variable at a time and this understanding has been further utilized in the tailor made synthesis of coordination architectures for targeted application. The utility of these materials in different areas, particularly in the field of (i) heterogeneous catalysis for some well-known organic transformations (Knoevenagel reaction, Strecker reaction and Biginelli reaction), (ii) selective carbon dioxide capture, separation and its chemical conversion into value added cyclic carbonates by porous metal organic frameworks (MOFs) and (iii) sensing of nitroaromatic compounds (by discrete molecular squares or by luminescent MOFs) have been explored to further support the underlying strategy designed for the construction of these coordination architectures.

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