

Abstract

Crystal engineering is a branch of science deals with understanding intermolecular interactions leading to design various supra molecular solid materials. Research into the rational design and synthesis of extended materials has grown considerably over the last 25 years, with these materials finding applications in areas such as gas storage, catalysis and drug delivery. The Cambridge Structural Database (CSD) contains over 880,000 small molecule crystal structures, including tens of thousands of metal-organic coordination networks (MOCNs) and other extended materials. In this context, designing metal nodes and organic ligands and/ or linkers are the keys for tremendous synthesis to construct coordination polymers (CPs) with desired structural topologies and physicochemical properties. In this thesis work various unprecedented flexible and semirigid bis(tridentate) pyridyl, pyridyl-amide and pyridyl-carboxylate ligands have been synthesized by strategic design and have been extensively characterized. The utility of neutral bis(tridentate) pyridyl-amide ligands as cofomers to form a cocrystal with Theophylline (THP) for (a) demonstrating the presence of the rare amide-pseudo amide hydrogen bonding motif in the cocrystal and identify further structural features (unit cell packing and weak intermolecular interactions) by single crystal X-ray diffraction, and (b) establishing relevant physicochemical properties of the cocrystal through a comparison with its composite by numerous analytical techniques. Based on neutral and anionic nature of bis(tridentate) ligand, two different (two and three components) self-assembly reaction strategies have been utilized for the synthesis of various MOCNs in good yields and high purity. The structural diversity has been controlled *via* various parameters such as nature of metal atoms, counter anions, solvent or guest molecules and reaction conditions etc. Their solid state structures have been established by single crystal X-ray diffraction analysis and their bulk phase purity has been validated by various analytical techniques like powder X-ray diffraction, FTIR, UV-Vis spectroscopy, thermo gravimetric and elemental analysis. The multifunctional bis(tridentate) ligand based MOCNs have been used as heterogeneous catalysts by considering their Lewis acidic, Brønsted basic and hydrogen bond donating capability for various C-C bond forming organic transformations (Cyanosilylation reaction, Henry reaction, Friedel-Crafts alkylation reaction, Michael addition reaction, Knoevenagel-Intramolecular Cyclization reaction and Knoevenagel Condensation reaction).