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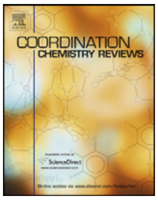

Review

Coordination and supramolecular aspects of the metal complexes of chiral N-salicyl-β-amino alcohol Schiff base ligands: Towards understanding the roles of weak interactions in their catalytic reactions

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ABSTRACT

Chiral N-salicyl-β-amino alcohol Schiff bases are important ligands widely employed in asymmetric catalysis. These are referred to as 'tridentate salen ligands' and are considered as a member of the well-known 'privileged ligands' group. The desire to understand the roles of weak non-covalent interactions in catalysis has greatly expanded the scope of structural studies on catalytic complexes. This review focuses on the structural and supramolecular features of the metal complexes derived from chiral N-salicyl-β-amino alcohol Schiff base ligands. Metal complexes of varying nuclearities, such as, mono-, di-, tri- and tetra-nuclear complexes, have been reported from this group of ligands. The geometries of the metal centers present in these complexes vary from tetrahedral, square-planar, square-pyramidal, octahedral to trigonal prismatic. The available structural data clearly indicate the coordination versatility of this group of ligands towards various metal ions. In addition, these complexes exhibit a variety of

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Crystal structure
Supramolecular chemistry

weak bonding interactions leading to the formation of supramolecular structures such as helices, 1-D chains, 3-D networks, etc. in solid state. The articles discussed here highlight the competence of chiral N-salicyl-β-amino alcohol Schiff base ligands to support various structural and supramolecular features in their complexes, but the role of these features in determining their catalytic efficiencies have not yet been studied in detail. We believe that the present review may inspire many such studies in future.

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1. Introduction

Chiral coordination compounds are important in multidisciplinary research areas including asymmetric catalysis [1], metallo-supramolecular chemistry [2], materials sciences [3] and bio-inorganic modeling studies [4]. The development of chiral complexes is a challenging task requiring considerable synthetic effort. The use of chiral ligands is an important strategy towards the development of asymmetric catalysts and chiral metallo-supramolecular systems [1,5]. Chiral N-salicyl-β-amino alcohol Schiff base ligands, derived from chiral β-amino alcohols and salicylaldehyde derivatives, comprise of a group of ligands that are widely employed in asymmetric transformations such as asymmetric oxidations [6], asymmetric alkynylations [7] and enantioselective trimethylsilylcyanations [8]. These ligands are referred to as 'tridentate salen ligands' and are often considered as a member of the well-known 'privileged ligands' group [9]. The attractions of this group of ligands include their structural and electronic fine-tunability, low molecular weights and extremely simple synthetic conditions. In addition, their precursor, viz., chiral β amino alcohols, can be readily synthesized from naturally available chiral amino acids [10].

Understanding the coordination behaviors of chiral ligands is important in asymmetric catalysis towards the development of efficient catalytic systems. Properties, such as, the coordination number, geometry, oxidation state, stability, etc. of the metal center present in a catalyst can strongly affect its catalytic performance [1]. Despite the importance of chiral N-salicyl-β-amino alcohol Schiff base ligands in asymmetric catalysis [11], only a limited number of studies are reported so far describing the structural and supramolecular features of their metal complexes.

Meanwhile, non-covalent interactions play significant roles in asymmetric catalysis [12]. It has been suggested that multiple non-covalent interactions operating in concert can induce sufficient conformational constraint, required for the stereo-induction in asymmetric transformations [13]. Attractive non-covalent interactions also play role in lowering the kinetic barriers of reactions by stabilizing their transition states [14]. Many asymmetric catalytic processes involving transition metal catalysts invoke attractive non-covalent interactions as elements of stereo-control [12,15]. Therefore, detailed structural analyses of the catalytic complexes, revealing their structural and supramolecular features, could be useful towards the development of optimized transition metal catalysts intended for specific applications.

In this review, the structural, supramolecular and coordination chemistry aspects of the metal complexes of chiral N-salicyl-β-amino alcohol Schiff base ligands have been discussed in detail. It is expected that a proper appreciation of the coordination and supramolecular features of the metal complexes of this group of ligands may inspire further studies in this area correlating their structural and supramolecular features with their catalytic properties.

2. Syntheses and supramolecular significances of chiral N-salicyl-β-amino alcohol Schiff base ligands

Chiral β-amino alcohols can be synthesized starting from chiral amino acids [10]. They are traditionally used as chiral auxiliaries in asymmetric catalysis [11]. The presence of amino and alcoholic functional groups facilitates the derivatization of β-amino alcohols leading to new classes of compounds. Chiral β-amino alcohols undergo Schiff base reaction with salicylaldehyde derivatives yielding N-salicyl-β-amino alcohol Schiff bases readily. The synthetic conditions, employed for chiral N-salicyl-β-amino alcohol Schiff bases, vary from report to report [16–20]. Most of the syntheses have been carried out in dry alcohols under ambient or reflux conditions [16]. In few cases, use of dry benzene [17] or mixture of solvents, such as, methanol and dichloromethane [18a] have also been reported. The reaction time for the synthesis of chiral N-salicyl-β-amino alcohol Schiff bases vary between 0.5 and 24 h [19,20]. Some synthetic procedures also involve the use of dehydrating agents, such as, anhydrous Na_2SO_4 [21], molecular sieves [22], etc. The condensation of aldehydes with β-amino alcohols lead to the formation of 1,3-oxazolidines besides imines in some cases [19,23]. The chiral N-salicyl-β-amino alcohol Schiff base imines can be reduced to amines using appropriate reducing agents [19].

Chiral N-salicyl-β-amino alcohol Schiff bases contain prominent hydrogen bonding functional groups such as alcoholic and phenolic OH groups. Due to the presence of these functional groups, metal complexes of chiral N-salicyl-β-amino alcohol Schiff bases are capable of exhibiting multiple attractive non-covalent interactions in their catalytic reactions and one can expect these interactions influencing their catalytic performances. This is because, some organo-catalysts, such as, β-amino alcohols, bearing catalytically similar active sites, mediate a variety of asymmetric transformations via noncovalent bifunctional activation of the reagents [24]. In this context, more dedicated studies are required to understand the roles of weak supramolecular interactions, exhibited by the complexes of chiral N-salicyl-β-amino alcohol Schiff bases, in defining their catalytic properties.

Metallo-supramolecular systems are of current interest due to their potential applications in bio-inorganic modeling, inclusion phenomena, guest exchange and catalytic reactions [25]. Chiral N-salicyl-β-amino alcohol Schiff bases are useful ligands in metallo-supramolecular chemistry due to their weak bonding functional groups. Moreover, the inherent chirality of these ligands induces dissymmetry in their complexes, which is beneficial for the effective modeling of metal centers present in chiral environments of biological proteins [26].

This review focuses mainly on the crystallographically characterized metal complexes of chiral N-salicyl-β-amino alcohol Schiff base ligands. Reports on the structural characterization of chiral N-salicyl-β-amino alcohol Schiff base ligands alone [19,27] are not discussed in this review. A listing of all the ligands, presented in this review, is given in Scheme 1.

3. Titanium complexes

Complexes derived from titanium alkoxide and chiral N-salicyl-β-amino alcohol Schiff base ligands are well studied with respect to their catalytic behavior towards a number of asymmetric reactions such as trimethylsilylcyanation [28], pinacol coupling [29], Strecker reactions [30], etc. N. Oguni and co-workers extensively used such complexes for the asymmetric cyanosilylation of aldehydes [28a,b]. Despite their importance as asymmetric catalysts,

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