

Review

Hydrogen-bond supramolecular structure of group 13 Schiff base complexes

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Abstract

The structural features of the group 13 element complexes of general formula R₂M(O,N) and RM(O,N)₂ (R = alkyl, M = B, Al, Ga, In), supported by bidentate and multidentate Schiff base ligands are discussed and considered in relation to the hydrogen-bond networks. Detailed structural analysis was performed for crystal structures of complexes retrieved from the Cambridge Structural Database (CSD) (version 5.25) and compounds recently characterized by our group. It was demonstrated that the intra- and intermolecular non-covalent interactions like the C–H_{imino}···O, C–H_{aryl}···O, C–H_{aliph}···O, and C–H···π hydrogen bonds, and π-stacking appear very frequently. Various structural motifs for the group 13 Schiff base complexes were delineated ranging from monomeric species, hydrogen bond dimers, linear polymers, helicates, layers to 3D network architectures. Such diversity in the supramolecular architecture arises from the ligands identity and both the coordination geometry and the nature of the metal center.

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1. Introduction

In Schiff base metal complexes, the environment at the coordination center can be modified by attaching different substituents to the ligand, which provides a useful range of steric and electronic properties essential for the fine-tuning of structure and reactivity. Therefore, Schiff base ligands are among the most fundamental chelating systems in coordination chemistry [1,2] and complexes of both transition and p-block metals based on this type ligands have been shown to catalyze a wide variety of reactions [3,4]. For instance, the aluminum complexes, which are of particular relevance to this paper, have been used as catalysts in polymerization of ethylene [5], methacrylate [6], lactide [7] and other heterocyclic monomers [8]. However, the vast majority of investigations focus on the metal first-coordination sphere, while the ligand frameworks have various donor and acceptor sites capable to participate in an internal hydrogen bond to an adjacent metal-bound ligand or an incoming substrate, and the fundamental structural consequences exhibited by the second-sphere hydrogen bonding interactions on the reactivity and properties of these type complexes are not a well-developed area. The importance of hydrogen bonding may lie in the numerous supporting roles for processes taking place at metal sites as well as in the rapidly developing field of crystal engineering and material chemistry. In our opinion the cooperation between the coordination center and non-coordinating active-site residues plays very often important (thought often yet unrevealed) role in the molecular recognition and activation processes involving catalysts supported by Schiff bases as well as it may be of fundamental importance in the design of well-organized solid-state materials with desired properties involving Schiff base ligands. The latter expectation is supported by recent studies, which have highlighted the potential importance of hydrogen bonding in tuning the properties of materials based on Schiff base metal complexes [9,10]. Undoubtedly, apart from wide application of Schiff base metal complexes in various fields of chemistry, exploitation of non-covalent interactions inherently exhibited by this group of compounds is an emerging area of research.

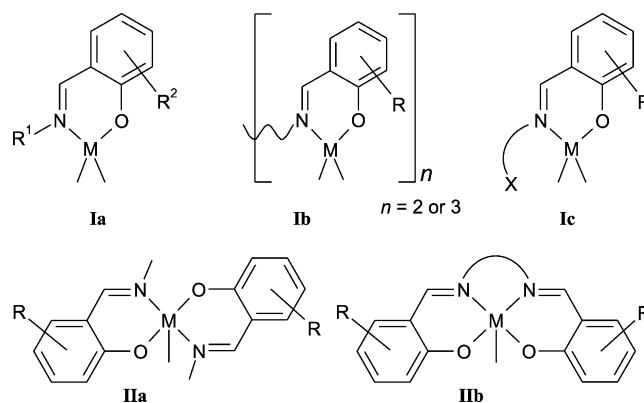
In the past few years, we have been engaged in an investigation of relationships between intra- and intermolecular secondary donor–acceptor interactions and hydrogen bonding using the group 13 alkoxides [11] and carboxylates as model complexes [12]. These studies revealed that minor differences in the subunit structure could have a profound effect on the molecular and crystal structure. An integral part of this work has been a study of the structure of group 13 organometallic chelate complexes ($M = B, Al, Ga$ and In) with the salicylideneimine anion as the O,N -bidentate ligand [13,14]. The latter compounds were found to be a good basic model system for exploiting the role of hydrogen bonding on the supramolecular structure of Schiff base metal complexes. The purpose of this contribution is to present extended studies on the identification and characterization of supramolecular interactions in the group 13 Schiff base complexes based on de-

tailed structural analysis of crystal structures retrieved from the Cambridge Structural Database (CSD) and recently published by our group. In particular, our crystal structure analyses focus on the identification of the hydrogen bond preferred modes for Schiff base metal complexes as well as an understanding their relative importance on the supramolecular architecture. It is also pertinent to note that a significant part of the revealed structural motifs and supramolecular structures had been not considered by the original authors.

1.1. Method of database analysis

The Cambridge Structural Database (CSD, version 5.25, April 2004) [15] was interrogated to extract all reliable structures of the group 13 element Schiff base complexes. The searches were performed for four-coordinate and five-coordinate complexes containing the $C_2M(O,N)$ (**I**) and $CM(O,N)_2$ (**II**) central core, respectively (where $M = B, Al, Ga$ or In , and O,N denotes the skeleton of a salicylideneimine ligand). The basic structural types **I** and **II** were further classified into distinct categories (Scheme 1). The category **Ia** contains mononuclear tetrahedral $R_2M(O,N)$ molecules. Bi- and tri-nuclear compounds with two or three tetrahedral $R_2M(O,N)$ units joined by an aliphatic backbone were categorized separately as **Ib** due to the distinguished supramolecular structure. Mononuclear derivatives of potentially tridentate salicylideneimine ligands with the terminal Lewis base termini attached to the imine nitrogen constitutes a particular case and were categorized as **Ic**. On the other hand, the monomeric five-coordinate monoalkyl $RM(O,N)_2$ or $RM(O_2N)_2$ complexes, i.e., derivatives of O,N -bidentate salicylideneimine or O,O,N,N -tetradentate Salen-type ligands, are denoted as **IIa** and **IIb**, respectively (Scheme 1).

The CSD searches were restricted to non-charged compounds and entries with the R -factor greater than 0.08 and those with an unresolved error were rejected (with one exception for NIXTOD because of its structural importance). Details concerning the data retrieval from the CSD file, and the list of CSD ref. codes with references and selected geo-



Scheme 1. Schematic representation of the structural types of the group 13 element Schiff base complexes analyzed in this work.

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