Status of Inorganic Chemistry Research in India

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The current status of research in the area of Inorganic chemistry in India is discussed. The article focuses on various aspects of inorganic chemistry being pursued in the country such as main-group chemistry, coordination chemistry, supramolecular chemistry, molecular materials and bio-inorganic chemistry. The strengths of the subject in the country and the possible future directions are discussed.

Keywords: Inorganic Chemistry; Modern Chemistry; Chemical Research; Mercurous Nitrite

Introduction

Modern chemistry research in India began with Acharya Prafulla Chandra Ray in Kolkata. The fact that he was an inorganic chemist by training is incidental. His influence on chemical research in India is profound (Chakraborty, 2014). His most important contribution is the preparation of mercurous nitrite, Hg₂(NO₃)₂, whose molecular structure was established several years later (Samanta et al., 2011).

The head start given by Ray was picked up, in various parts of the country, particularly after the expansion of Universities and Institutes, post-independence. Inorganic chemistry and analytical chemistry were taught both from a theoretical and experimental perspectives. Unfortunately, much of the progress in inorganic chemistry research depended on modern instrumental methods and since these were not available in many Institutions in India, research in Inorganic Chemistry lagged in terms of both quality and quantity. Similarly, inadequate laboratory equipment to handle air- and moisture sensitive compounds has hampered (and continues to hinder) research in organometallic chemistry. Compounding these difficulties, funding for scientific research in the western world, post-world War 2, increased enormously which continues unabated. In India, formal mechanisms for funding research were not available for a long time and the amount of funding was substantially lower. It is only in the last two decades that reasonable funding became available for researchers across the country and this is reflected in the number of publications resulting from the country. The increased funding has resulted in higher number of publications. By a search of web-of-science using the key words “India” and “Address” (in topics search) the publications in the decades 1980-2019 is shown in Fig. 1 for the journal of Inorganic Chemistry which clearly shows an increase in research activity in the years after 2000. It is important that the level of support for research increases continuously and remains robust, keeping pace with the increase in the number of higher educational research institutes of the country if we must compete with the West and China.

With this background, we will discuss the current status of Inorganic Chemistry research in India. For convenience this is divided in the following sub-topics. A) Main-group Chemistry b) Coordination Chemistry c) Supramolecular Chemistry d) Molecular Materials e) Bio-inorganic Chemistry. We will not be discussing organometallic chemistry and solid-state chemistry as these have been dealt in separate articles in this issue. Unfortunately, research in analytical chemistry in the country has not received adequate support and this
needs to be corrected to give the importance of this subject and its applications in chemical industry.

**Main-group Chemistry**

Research in the chemistry of main-group elements in India is picking up pace. A very good foundation of non-metal chemistry particularly of sulfur (Vasudeva Murthy, 1951), silicon (Padma et al., 1979) and fluorine (Padma et al., 1979) was laid at Bangalore. Research in silicon chemistry at the Indian Institute of Science also led to efforts in developing silicon technology by partnership with industry (Gopala Krishna Murthy, 2014). Mehrotra and co-workers have pioneered the alkoxide chemistry of various main-group elements and transition metals (Bradley and Mehrotra, 1978), (Bradley et al., 2001).

Inorganic heterocyclic rings such as cyclophosphazanes and cyclophophazenes (Fig. 2) were studied at Bangalore (Cameron et al., 1975) (Krishnamurthy and Sau, 1978) (Krishnamurthy, 1994). In particular, the reactions of various nucleophiles with \( \text{N}_4\text{P}_4\text{Cl}_8 \) were studied and the reaction mechanisms delineated. A highlight of this work was the isolation of the bicyclic phosphazene, \( \text{N}_4\text{P}_4(\text{NHEt})(\text{NMe}_2)_5(\text{NET}) \) (Cameron et al., 1975) (Fig. 2). The mechanism of polymerization of \( \text{N}_3\text{P}_3\text{Cl}_6 \) to the \( [\text{PCl}_2=\text{N}]_n \) was investigated and the crucial role of Lewis acids and water to initiate the polymerization was established (Ganapathiappan et al., 1987).
The research groups of Krishnamurthy and Chandrasekhar (Krishnamurthy, 1992) (Chandrasekhar et al., 2001) (Chandrasekhar et al., 2002) (Chandrasekhar et al., 2004) have shown that cyclophosphazenes can be utilized to prepare novel multi-site coordination ligands for the preparation of transition metal complexes, an example of which is shown in Figure 3. The use of cyclophosphazenes as scaffolds for anchoring macrocycles such as porphyrins was revealed recently (Rao et al., 2009). Studies were carried out in developing the chemistry of fluorinated cyclophosphazenes (Elias and Shreeve, 2001), which were extended to making organometallic derivatives of cyclophosphazenes (Kumar et al., 2014).

Penta-coordinated (Kumara Swamy and Kumar, 2006) (Fig. 4), and hexa-coordinated (Said et al., 1996) phosphorus compounds have been investigated and the established notions of site preferences among the former have been challenged (Kumara Swamy and Kumar, 2006). Phosphorus intermediates in important reactions such as the Mitsunobu reaction have been identified (Kumaraswamy et al., 2009). Organophosphorus chemistry involving reactions of allenylphosphonates and allenylphosphine oxides have been probed particularly with a view to exploring the allene reactivity (Kumara Swamy et al., 2017).
Singh and co-workers successfully used the synthetic strategy of using intramolecular E–X (E = Se, Te; X = N, O) coordination for isolating several novel and interesting organochalcogen species including triselenides, selenenyl iodides, selenenyl esters, ebselen derivatives and chalcogen containing macrocycles (Mugesh and Singh, 2002) (Mukherjee et al., 2010).

Low-valent chemistry of main-group elements and compounds containing multiple bonds among main-group elements are receiving attention in recent years. Some of the examples of the compounds isolated in recent years are shown in Fig. 5 (Sen et al., 2012), catalytic C-H arylation of arene and heteroarenes at ambient temperatures (Sreejyothi et al., 2018). Furthermore, an NHC-borane adduct was also used for the reduction of CO\textsubscript{2} captured from air into methoxyborane (Sau et al., 2019).

**Coordination Chemistry**

Animesh Chakravorty and his research group have contributed enormously to the coordination chemistry of transition metal ions probing the structural chemistry and the electrochemical behaviour of the complexes. He pioneered the use of modern instrumental methods such as X-ray crystallography, NMR, EPR and cyclic voltammetry in addressing contemporary problems in Inorganic Chemistry (Datta and Chakravorty, 1983) (Bhattacharya et al., 1984) (Ray and Chakravorty, 1988). This school contributed significantly to the development of new types of ligands such as those containing the azo groups/oxime groups and the reactivity of the complexes formed from them. Thus, trans-bis(phenylazoacetaldoximato)platinum(II) was shown to undergo hydroxylation of the aromatic ring (Fig. 6) (Bandyopadhyay et al., 1983). His research group also contributed significantly to the

**Fig. 5: Recent examples of main-group compounds reported from India**

(Pal et al., 2016), (Sinha babu et al., 2016), (Majumdar et al., 2018) (Dhara et al., 2018) (Pahar et al., 2018).

Main-group element-containing rings and cages containing Sn-O (Chandrasekhar et al., 2007), Te-O (Srivastava et al., 2010) and Sb-O (Kishore et al., 2014) (Srungavruksham et al., 2015) bonds have been studied.

Mandal and co-workers have been involved in work on main-group organometallic compounds such as a phenalenyl based aluminium compound for
specific problems. For example, his group, contributed to the coordination chemistry of de-protonated pyridine amide ligands (Mishra et al., 2007), the understanding of spin-equilibria in Fe(II)N₆ complexes (Mishra et al., 2009) and the stabilization of mixed Ni(III)/Ni(I) complexes (Rajput and Mukherjee, 2013). Bhattacharyya and his co-workers have used the aryl azo group of ligands and studied their complexation with various metal ions (Acharyya et al., 2004).

The chemistry of porphyrins and crown-ethers as ligands has been explored considerably. Thus, for example, octabromotetraphenylporphyrin and its metal complexes were extensively studied and the role of the bromo substituents in tuning the electronic properties of the porphyrin skeleton was elucidated (Fig. 6) (Bhyrappa et al., 1991). In an interesting synthetic approach, porphyrins were appended to crown ethers and the metalation was studied (Thanabal and Krishnan, 1982). Expanded porphyrins with diverse structures have been studied by T. K. Chandrashekar and his research group (Misra et al., 2006), (Ghosh et al., 2016) (Fig. 7). Some members of this family such as metalated smaragdyrins were shown to possess interesting NLO properties while some others were shown to demonstrate Möbius aromaticity (Ghosh et al., 2016) and Baird (Cha et al., 2017) aromaticity (Fig. 7). Aza homo-porphyrin both in its free-base and complexes forms was shown to have a Möbius topology (Anju et al., 2017). Partial core-modification of a porphyrin was employed to synthesize a 20π antiaromatic isophlorin. It was

![Fig. 6: (A) Bis (phenylazoacetaldoximato) platinum(II) (Bandyopadhyay et al., 1983); (B) Octabromotetraphenylporphyrin and its metal derivatives. (Bhyrappa and Krishnan, 1991)](image)

![Fig. 7: Line diagram of expanded porphyrins with (A) Structural diversity, (B) Baird aromaticity, (C) Möbius aromaticity. (Ghosh et al., 2016; Ghosh et al., 2016; Cha et al., 2017)](image)
shown that a monopyrrole porphyrin exhibits antiaromatic characteristics (Panchal et al., 2016).

Corrole derivatives, a sub-family of the porphyrin systems are receiving attention. Thus a corrole containing CCNN in the core was shown to stabilize Cu(III) (Adinarayana et al., 2015) while a corrole with a NNNN in the core was used for selective binding with Zn(II) (Adinarayana et al., 2016) (Fig. 8). Porphyrins containing heteroatoms have been state and time-resolved fluorescence studies were used to demonstrate unidirectional energy flow from one porphyrin subunit to another (Chatterjee et al., 2017). They have also been involved in the preparation of new types of carbaporphyrinoids (Kumar et al., 2018).

Phosphorus-based ligands such as cyclophosphazene-supported ligands (Chandrasekhar et al., 2007), phosphonates (Chandrasekhar et al., 2011) (Goura and Chandrasekhar, 2015) and phosphates (Murugavel et al., 2006) have been used to prepare multi-metallic architectures. Cyclodiphosphazanes have been used as ligands for preparing metal complexes, interesting examples being a diamondoid Cu(I) coordination polymer (Siddiqui et al., 2015) and a Rh(I) metallopentagon (Siddiqui et al., 2015).

Polyimido anions of the type

$$\text{[PE(NHR)}_x(NR)_{3-x}]_3^-(E = \text{O, S, NR})$$

have been used to build polyhedral and hierarchical cage assemblies (Yadav et al., 2017) (Rajasekar et al., 2018).

Metal peroxo complexes such as diperoxovanadate (Ravishankar et al., 1994) have been studied in inducing oxygen exchange reactions in vanadyl sulfate. Metal fluoro complexes have been studied from the point of view of their synthesis, structure and reactivity (Bhattacharjee et al., 1989).

Samar Das and co-workers have been working on polyoxometalates and complexes locked in a confined space for various purposes including electrocatalytic water oxidation (Manna et al., 2016), (Mukhopadhyay et al., 2018).
Non-innocent ligands (Fig. 9) (that are themselves redox-active) have been investigated in combination with various metal ions (Ali et al., 2015), (Ali et al., 2016) (Khan et al., 2018), (Chatterjee et al., 2018). Complexes of Ru(II) (Das and Bhattacharjee, 2012) (Singh et al., 2008) and Rh(III) (Baksi et al., 2007) with various types of ligands have been studied.

Studies on boranes and synthesis and structural characterization of metallaboranes have been pursued (Mondal et al., 2018). An interesting outcome of such an investigation is the isolation of an early transition metal-guarded heptaborane, [(Cp₂M)₂B₉H₁₁] (M = Zr/Hf) (Fig. 10) (De et al., 2018).

**Supramolecular Chemistry**

Cryptands have been used as ligands to bind to metal ions in a specific manner. Bharadwaj and co-workers have contributed extensively to the chemistry of cryptands (Fig. 11). For example, it has been shown that interaction of Ni(II), Cu(II) and Zn(II) with a cryptand enhances its fluorescence intensity (Ghosh and Bharadwaj, 1996). They were able to prepare cryptand-based amphiphiles which were shown to form vesicular structures or Langmuir-Blodgett Films (Das et al., 2000).

P Ghosh and co-workers have developed molecular receptors for anion recognition and anion sensing (Dutta and Ghosh, 2015) (Chakraborty et al., 2019). Their contributions include developing platforms containing suitable binding sites for the selective recognition anions of various sizes and topologies. They have also developed ligands for sensing metal ions such as Hg²⁺, Cu²⁺ or Zn²⁺ (Chowdhury et al., 2014). Molecular probes for monitoring bio-chemical transformations induced by endogenous enzymes as well for monitoring the concentrations of important biomarkers have been developed by Amitava Das and co-workers (Anila et al., 2016), (Agarwalla et al., 2018). Pullarao and co-workers have used calixarene-based ligands and carbohydrate-based ligands for the binding and sensing of metal ions. Such chemosensors were also used in cell imaging and anti cancer activity (Pathak et al., 2012; Areti et al., 2016). Mukherjee and co-workers from Bangalore have been designing ligands that can interact with metal ions to afford complexes with specific molecular topologies. Thus, his group has recently reported an octanuclear Pd(II) water soluble molecular barrel which could stabilize transient merocyanines under visible light. The molecular barrel could store transient merocyanine forms for months without any degradation (Howlader et al., 2018).

D K Chand and his group have contributed to the areas of supramolecular catalysis, supramolecular coordination chemistry and nanoscience. Interesting examples of the supramolecular chemistry of the Chand group includes the assembly of a double-decker coordination cage (Bandi et al., 2014) and a molecular star (Prusty et al., 2017).

**Molecular Materials**

Molecular magnetism in general and single molecular magnets in particular has picked up as a research activity in the country over the last decade. Chandrasekhar and his group have made several contributions (Chandrasekhar et al., 2007; Chandrasekhar et al., 2013; Bag et al., 2014; Biswas et al., 2016; Bar et al., 2018) including the first Co(II)
containing single-molecule magnet, \([\text{L}_2\text{Co}_2\text{Gd}]\)\([\text{NO}_3]_22\text{CHCl}_3\) (Chandrasekhar et al., 2007) (Fig. 12). Murugavel and co-workers have reported an air-stable single ion magnet (Fig. 13) with a pseudo \(D_{5h}\) symmetry that showed a high blocking temperature of magnetization (Gupta et al., 2016).

Rajaraman and co-workers have been active in carrying out theoretical studies on molecular magnets with a view to understand the factors that contribute to molecular anisotropy and on design of new molecular magnets (Singh et al., 2015) (Singh et al., 2016). Other groups have also been active in preparing new molecular magnets (Khatua et al., 2016), (Vaidya et al., 2018).

Thilagar’s research group at Bangalore has been working on boron-based molecular materials and medicinal aspects of boron compounds (Fig. 14). They have established the design and development of multiple emissive molecular polyads, aggregation induced emissive small molecules, strongly luminescent organic/organometallic solids and external stimuli responsive materials (Swamy et al., 2013). The concepts of BN/CC isosterism and topochemistry was utilized to assemble deep-blue delayed fluorescent materials (Neena et al., 2018).

Boomi Shankar and his group have developed new ferro-electric metal-organic materials from di- and tripodal ligands derived from pyridyl-functionalized P(V) ligands (Vijayakanth et al., 2018). Due to their pseudo-\(C_3\) symmetric or \(C_3\) symmetric nature such ligands (Fig. 15) were successfully deployed to prepare complexes which would crystallize in non-centrosymmetric space groups to display ferroelectric properties (Yadav et al., 2018).

Goswami and co-workers have shown that the bis-azoaromatic pincer ligand (Fig. 15) is capable to reversibly form azo-hydrazo (2e+ 2H\(^+\)) couple, enabling this group to design electro-protic sponges \([\text{M(H}_2\text{L}_3)]^{2+}\) from \([\text{M(L}_3]^{2+}\) (M = Fe and Co) (Pramanick et al., 2018). This group also showed that transition metal complexes of azo-aromatic ligands (Fig. 15 (B)) are useful in designing memristors.
Research in molecular approaches towards porous solids in the area of metal-organic frame works (MOFs) has been vigorously pursued in various laboratories in India. The primary focus of these efforts have been in trying to use MOFs for gas separation, for storage of gases, in catalytic applications, as sensors for toxic analytes, for magnetic properties and drug delivery. (Ghosh and Bharadwaj, 2004), (Aijaz et al., 2010) (Ramaswamy et al., 2010), (Hazra et al., 2011), (Kanoo et al., 2012), (Sarma et al., 2012), (Mallick et al., 2015), (Asha et al., 2016), (Samanta et al., 2018), (Khatua et al., 2016) and (Singh et al., 2019). This topic is being dealt in another article in this special issue and hence we will not go into further details.

Bio-Inorganic Chemistry

The groups of P T Manoharan, V Krishnan and S Mitra laid the foundations for research in the area of bio-inorganic chemistry in the country. Manoharan and co-workers have contributed to the area of reconstituted hemoglobins which were studied by spectroscopy (Manoharan et al., 1989) (Balagopalkrishna et al., 1996). Krishnan and his co-workers have studied porphyrin chemistry with a view to understand the role of this important ligand and its metal derivatives in biological systems (Maiya and Krishnan, 1985) (Droupadi and Krishnan, 1985). Mitra and co-workers have worked extensively on heme enzymes (Mazumdar et al., 1988).

Mazumdar and co-workers have worked on various metalloenzymes including horseradish peroxidaes (Das and Mazumdar, 1995) and cytochrome P-450 (Goyal et al., 2011) (Goyal et al., 2012). Aspects of research on these metalloenzymes include structure and mechanism of function, enzyme kinetics, protein stability etc.

Sarkar and co-workers have contributed to the understanding of model bio-inorganic chemistry by preparing synthetic compounds that can both mimic structurally and functionally the active sites of metalloenzymes that are broadly called as oxido-reductases. These include sulphite oxidase (Das et al., 1994), tungsten-containing enzymes (Das et al., 1996) (Yadav et al., 1997) and a nitrate reductase (Majumdar et al., 2006).

Mukherjee and his group have contributed to the model bio-inorganic chemistry of methemerythrin, (Mukherjee et al., 2003), the structural modelling of photosystem II (Lal and Mukherjee, 1998) and the functional modelling of tyrosinase (Mandal et al., 2012) involving aromatic hydroxylation.

Chakravarty and co-workers have contributed
extensively to the area of metal-based photo-chemotherapeutic agents using various types of complexes such as boron dipyrromethane (BODIPY)-based photosensitizers for generating reactive oxygen species (ROS) using low-energy light (Raza et al., 2017). A photocytotoxiciron(III) complex was used as a cellular imaging agent (Basu et al., 2012). Chakravarty and his group have also contributed to the evaluation of the therapeutic potential of curcumin, the active ingredient of turmeric (Banerjee and Chakravarty, 2015). Chandrashekar and co-workers have shown the utility of novel porphyrin derivatives for photo dynamic therapy applications (Thomas et al., 2012).

Palaniandavar and his group have contributed to the interaction of small molecule metal complexes with DNA leading to work on metal-based anti-cancer drugs (Ramakrishnan et al., 2011). Their group has also contributed to the model bioinorganic chemistry of non-heme enzymes (Sankaralingam et al., 2017). Rath and co-workers have investigated the bioinorganic chemistry of multi-heme proteins and enzymes using synthetic models (Sil et al., 2016, (Kumar et al., 2019). Paine and co-workers have been involved in model bio-mimetic chemistry involving iron and cobalt (Chatterjee et al., 2018). Singh and co-workers have contributed to selenoenzymes and glutathione peroxidase (GPx) mimetics. They have demonstrated that dichalcogenides having a weak donor group (O, N) in proximity exhibit excellent GPx-like activity (Selvakumar et al., 2011). Mugesh and co-workers have been involved in the understanding of the anti-oxidant activity of synthetic compounds in mammalian cells, the thyroid hormone metabolism and thyroid related disorders, and the mechanism of activity of organoselenium compounds to affect thyroid hormones (Mondal et al., 2016, Ungati et al., 2018, Ghosh et al., 2018).

Somdatta Ghosh Dey and her group have been investigating the hypothesis of the relevance of heme in Alzheimer’s disease (AD). They have not only successfully defined the heme-binding site of amyloid beta (Ab) peptides involved in AD but also demonstrated that heme once bound to these peptide fragments can exhibit the reactivity that matches the cytopathologies of AD (Pramanik and Dey, 2011) (Ghosh et al., 2015). Abhishek Dey and his group have been studying emulating the reactivity of redox active metallo-enzymes using small molecule analogues that include the primary and the second sphere interactions important for enzymatic activity. Using in-situ interrogation techniques they have achieved unprecedented reactivity and stability from molecular systems inspired by natural enzymes that act as electrocatalysts for processes important for clean energy and environment (Blunia et al., 2018), (Rana et al., 2019).

Outlook

Research in inorganic chemistry, in all its aspects, ranging from main-group chemistry to bio-inorganic chemistry, from coordination chemistry to supramolecular chemistry and molecular materials has been extremely actively pursued in the country and we have been competitive with the rest of the world. However, in order to sustain this activity continuous large infusion of research grants is necessary both in the public/private-funded Universities/Institutions. Like certain areas in physical chemistry, research in inorganic chemistry depends crucially on state-of the art instrumentation and if this is not sustained, we will lag behind very quickly.

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