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Review

Supramolecular self-assembly of nucleotide–metal coordination complexes: From simple molecules to nanomaterials

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ABSTRACT

Nucleotides are essential components of DNA and RNA, and their functions depend mainly on the participation of metal ions. Thus, research into nucleotide–metal coordination complexes, including the affinities of different coordination donors for metal ions, molecular and crystal structures, supramolecular assembly, and functional nanoparticles, will contribute to the interdisciplinary field of chemistry,

Abbreviations: $NP^{2-/3-/4-}$, 5′-phosphate nucleotide; $dNP^{2-/3-/4-}$, 2′-deoxy 5′-phosphate nucleotide; M(NMP), metal coordination complex of 5′-monophosphate nucleotide; $M(NTP)^{2-}$, metal coordination complex of 5′-triphosphate nucleotide; $H(NTP)^{3-}$, protonated 5′-triphosphate nucleotide at the terminal γ-phosphate group; NMP^{2-}, deprotonated 5′-monophosphate nucleotide; $H(NDP)^{2-}$, protonated 5′-diphosphate nucleotide at the terminal β-phosphate group; $O(P_\beta)$, oxygen atom of β-phosphate group; $O(P_\gamma)$, oxygen atom of γ-phosphate group; pKa, the acidity constant; K_M, the stability constants; $R\text{-}MP^{2-}$, monophosphate compound with different substituent groups (R); Guo, guanine; Cyt, cytimidine; Ado, adenine; Purine-NTPs, 5′-triphosphate nucleotide with purine nucleobase residues; Pyrimidine-NTPs, 5′-triphosphate nucleotide with pyrimidine nucleobase residues; purine-NDPs, 5′-diphosphate nucleotide with purine nucleobase residues; PPi, pyrophosphoric acid; AMP, adenine monophosphate nucleotide; GMP, guanosine monophosphate nucleotide; IMP, inosine monophosphate nucleotide; CMP, cytidine monophosphate nucleotide; HMQC, heteronuclear multiple quantum coherence; NMR, nuclear magnetic resonance; MS, mass spectrum; EPR, electron paramagnetic resonance; TEM, transmission electron microscopy; CNPs, coordination polymer nanoparticals; BSA, bovine serum albumin; STEM-HAADF, a scanning TEM with a high-angle annular dark-field; SPR, surface plasmon resonance.

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biology, and materials. Numerous achievements have been reported in this area but few comprehensive reviews have considered nucleotide–metal complexes from the viewpoints of crystallography and supramolecular chemistry, or aspects of their chirality and chirality delivery. In this review, we describe the coordination ability of nucleotide ligands, the structures and properties of nucleotide–metal coordination complexes, and supramolecular assemblies. We review mononuclear complexes, multinuclear complexes, 1D and 2D coordination polymers, and 3D supramolecular assemblies in terms of their structures, mainly based on their X-ray single crystal diffraction data. In particular, we highlight the chirality of nucleotide–metal complex, including their molecular chirality, supramolecular helical chirality, and extended axial chirality. Furthermore, we summarize the functional properties of nucleotide–metal nanomaterials, such as their luminescence, magnetism, and adaptive inclusion properties. We discuss the future challenges and opportunities of research into nucleotide–metal complex.

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1. Introduction

Nucleotides are essential components of DNA and RNA, which play important roles in the storage and transfer of genetic information and protein biosynthesis in living organisms [1]. They exhibit rich structural diversity with multiple functional groups, such as nucleobases, phosphate groups, and hydroxyls in the sugar moiety, which determine the complexity and diversity of their coordination chemistry, as well as their supramolecular chemistry [2]. In addition, nucleotides are environmentally friendly ligands due to their water solubility, biocompatibility, and chirality, which makes them ideal candidates when mimicking biological systems and designing functional materials [3,4]. Thus, there is now much research into nucleotide–metal complexes and supramolecular assembly, as well as nanomaterials. Some important and interesting results have been obtained in these areas in recent years. Therefore, a comprehensive review of research into nucleotide–metal complexes is necessary, which will stimulate further research in the areas of coordination chemistry, supramolecular chemistry, crystal engineering, medicine, and material sciences related to nucleotides. In this review, we consider research into nucleotide–metal complexes during the last 40 years.

Several recent reviews have considered the coordination activities of nucleobases, nucleotides, and nucleotide analogs using a variety of spectroscopic technologies and molecular simulations [5–11], but few have summarized the bonding properties and supramolecular assembly rules of nucleotides based on their crystal structures, which can provide direct information about the coordination sites and conformation. In addition, no previous reviews have addressed the chirality of nucleotide complexes, although nucleotides are inherently chiral ligands. In this review, we provide a critical overview of structural analysis and controllable synthetic approaches, ranging from simple nucleotide coordination complexes to supramolecular assembly based on crystal structures and supramolecular features. We also consider the chirality of nucleotide–metal complexes and chirality delivery from the synthon level to supramolecular assembly. Extended axial chirality, a new type of chirality induced by chiral nucleotide ligands, is highlighted based on crystal structures and solid-state circular dichroism (CD) spectroscopy. In addition, nucleotides can serve as molecular building blocks for nanoscale materials if they interact with suitable counterparts. We summarize recent progress in nanomaterials based on nucleotide–lanthanide coordination complexes, which fully exploit the benefits of using biomolecules and lanthanide ions in water, where they exhibit excellent fluorescence, adaptive inclusion properties, molecular recognition, chiral resolution, and other properties. We describe a simple approach for designing multifunctional nanomaterials with nucleotide–metal complexes.

2. Structures and properties of nucleotide ligands

The family of nucleotide ligands includes natural nucleotides and modified nucleotides, where these types of molecules possess important biological functions and they carry an abundance of genetic information [12]. Their attractive properties such as multiple metal bonding sites, pH-sensitivity, and inherent chirality can be responsible for their biological functions [13–17]. In this section, we summarize the structural features and properties of nucleotide ligands based on previous studies, thereby providing the requisite background for the following sections.

2.1. Structural diversity of nucleotide ligands

The structural diversity of nucleotide ligands can be understood at three levels (Fig. 1). First, nucleobases are nitrogen-containing heterocyclic compounds and the key components of nucleotide ligands, which can be divided into purine derivatives and pyrimidine derivatives. Different substituent groups such as exocyclic amino groups or carbonyl groups in different sites cause structural differences in nucleobases. In addition, the methylation (modified bases) and tautomerism of nucleobases can also enrich their chemical structures [18,21d]. The natural nucleotides comprise purine nucleotides, i.e., adenine and guanine nucleotides, and pyrimidine nucleotides, i.e., uracil, cytosine, and thymine nucleotides. The differences in nucleobases affect the coordination behaviors of nucleotide ligands. Furthermore, the roles of these nucleobases during the formation of hydrogen bonding and stacking interactions [19–22] are the forces that drive the assembly of nucleotide–metal complexes into supramolecular devices, as discussed in Sections 3 and 4. Second, nucleotides with the same types of nucleobases can be classified as mono-, di-, and tri-phosphate nucleotides. The phosphate groups can bind with one or two carbon atoms at the 2′-, 3′-, and 5′- positions in pentose, thereby yielding various nucleotides species and their C-analogs [23], which exhibit slight differences in their structure and biochemical functions. In this review, we mainly consider the most important and well known 5′-nucleotides. Third, nucleotide ligands can be categorized as ribonucleotides or deoxyribonucleotides based on the protonation/deprotonation of the hydroxyl groups in the sugar rings [24a]. Most of the sugar moieties that occur in nature comprise ribose and 2′-deoxyribose residues [24]. Consequently, the nucleotide building blocks yield two types of nucleic acids: ribonucleic acids (RNA) and 2′-deoxyribonucleic acids (DNA). The latter lack a 2′-hydroxy group and they are less sensitive to hydrolysis of the phosphate-diester backbone than RNA [25]. The presence or absence of the 2′-OH group affects the acidic/basic properties of the nucleotide, where 2′-deoxy 5′-phosphate nucleotides ($dNP^{2-/3-/4-}$) are slightly more basic than their 5′- counterparts ($NP^{2-/3-/4-}$) [26]. Different adenine nucleotide

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