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ORIGINAL ARTICLE

Synthesis, characterization and anti-bacterial activity of divalent transition metal complexes of hydrazine and trimesic acid

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KEYWORDS

Transition metal complexes; Hydrazine; Trimesic acid; Spectral studies; Anti-bacterial activity **Abstract** Transition metal complexes of trimesic acid and hydrazine mixed-ligands with a general formula $M(Htma)(N_2H_4)_2$, where, M = Mn, Co, Ni, Cu and Zn; $H_3tma = trimesic$ acid, have been prepared and characterized by elemental, structural, spectral and thermal analyses. For the complexes, the carboxylate v_{asym} and v_{sym} stretchings are observed at about 1626 and 1367 cm⁻¹ respectively, with Δv between them of ~260 cm⁻¹, showing the unidentate coordination of each carboxylate group. The hydrazine moieties are present as bridging bidentates. Electronic and EPR spectral studies suggest an octahedral geometry for the complexes. All these complexes show three steps of decomposition in TGA/DTA. SEM images of CuO and MnO residues obtained from the complexes show nano-sized clusters suggesting that the complexes may be used as precursors for nano-CuO and nano-MnO preparation. The antimicrobial activities of the prepared complexes, against four bacteria have been evaluated.

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1. Introduction

Investigation of coordination complexes has lured the attention of many researchers world-wide because of their interesting molecular assemblies held together by forces, like, metal-ligand coordination, π - π stacking interactions and hydrogen bonding [1,2]. These coordination compounds are

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expected to find utilities as new materials with fascinating photophysical, photochemical, catalytic, sensing, magnetic and electronic properties [3,4]. Hydrazine, the humblest diamine even after getting protonated as $N_2H_5^+$, shows its ability to coordinate with several metal ions and forms several salts with mineral as well as carboxylic acids. The chemistry of hydrazine is significant due to its ability to form various mixed-ligand complexes with transition metals, whose stability and other properties change dramatically depending upon the cations as well as the anions [5,6]. Thermal decomposition reactions of metal carboxylates with hydrazine as co-ligand are gaining the attention of researchers, because, they serve as precursors for the preparation of nanoparticle oxide materials and metal carbonates by simple pyrolysis reaction [7–9]. Neutral hydrazine coordinates to the metal ions mostly in two different

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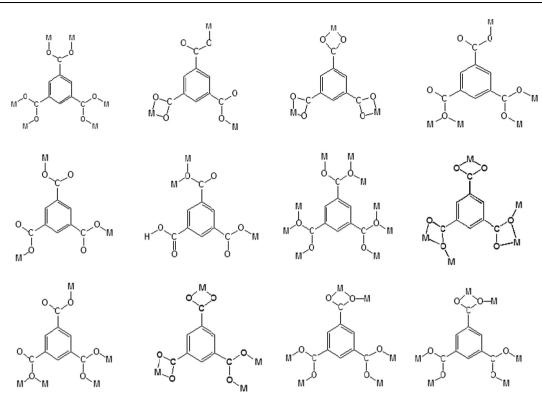


Fig. 1 Coordination fashions of trimesic acid (TMA).

| Compound | Molecular weight | Color | Found (calculated) % | | | | |
|---|------------------|-------------|----------------------|----------|------------|------------|------------|
| | | | Carbon | Hydrogen | Nitrogen | Metal | Hydrazine |
| Co(Htma)(N ₂ H ₄) ₂ | 331.15 | Light red | 32.4(32.6) | 3.8(3.6) | 16.4(16.9) | 17.2(17.7) | 18.1(19.3) |
| Cu(Htma)(N ₂ H ₄) ₂ | 335.77 | Pale blue | 30.1(30.4) | 4.3(4.5) | 15.5(15.7) | 17.5(18.9) | 18.4(19.0) |
| $Ni(Htma)(N_2H_4)_2$ | 330.91 | Green | 30.7(30.9) | 4.9(4.5) | 16.2(16.0) | 16.6(17.7) | 18.5(19.3) |
| $Mn(Htma)(N_2H_4)_2$ | 327.16 | Dirty white | 32.7(33.01) | 3.1(3.6) | 17.8(17.1) | 15.5(16.7) | 19.1(19.5) |
| $Zn(Htma)(N_2H_4)_2$ | 337.63 | Colorless | 31.4(31.9) | 3.5(3.6) | 17.2(16.5) | 18.2(19.3) | 17.9(18.9) |

modes, viz., either monodentate or bidentate bridging ligand (not simple bidentate to a metal ion). In the presence of carboxylic acids, hydrazine may coordinate with metal ions as hydrazinium cation since hydrazine exists as hydrazinium cation in acidic conditions. The nature of the complex formed (neutral hydrazine or hydrazinium ion) depends upon the concentration of hydrazine, pH of the reaction mixture and property of the metal ion. Hydrazinium (mono-protonated hydrazine) complexes are thermally less stable than neutral hydrazine complexes, but they are structurally interesting.

Trimesic acid, also known as benzene-1,3,5-tricarboxylic acid, is a benzene derivative with three carboxylic acid groups. Trimesates of Y(III) and lanthanides(III) were been prepared and characterized [10]. Two novel transition metal–organic frameworks of Zn and Cd with benzene-1,3,5-tricarboxylic acid as well as Tb-TMA (Trimesic acid) luminescent complex were also been prepared and characterized [11,12]. Three novel metal-trimesate compounds viz., $[Cd_3(TMA)_2(H-PRZ) (H_2O)_3(OH)].H_2O$ (1), $Cd_2Na_2(TMA)_2(H_2O)_4$ (2) and $[Cd_2Co(TMA)_2(H_2O)_4].2H_2O$ (3) exhibiting strong fluores-

cence emissions in the solid state at room temperature, due to diverse coordination architectures, were reported [13]. All the above facts prompted us to synthesize new complexes of some divalent transition metals (Co, Ni, Cu, Mn and Zn) with 1,3,5-benzenetricarboxylic acid (Trimesic acid, H_3 tma) and hydrazine. The possible diverse coordination fashions of benzene tricarboxylic acid with metal ions are shown in Fig. 1.

2. Experimental

2.1. Synthesis of $M(Htma)(N_2H_4)_2$, where M = Mn, Co, Ni, Cu and Zn

Benzene-1,3,5-tricarboxylic acid (0.722 g, 0.0035 mol) was added to 50 mL of aqueous hydrazine solution containing 0.172 mL (0.0035 mol) pure (99.5%) hydrazine hydrate and heated over water bath at 90 °C, with stirring, to get clear solution. It was added slowly to an aqueous solution of the corresponding metal nitrate (0.0035 mol, pH 5) with constant

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