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Coordination diversity in mono- and oligonuclear copper(II) complexes of pyridine-2-hydroxamic and pyridine-2,6-dihydroxamic acids.

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Solution and solid state studies on Cu(II) complexes of pyridine-2-hydroxamic acid (**HPicHA**) and pyridine-2,6-dihydroxamic acid (**H_2PyDHA**) were carried out. The use of methanol/water solvent allowed us to investigate the Cu(II)–**HPicHA** equilibria under homogeneous conditions between pH 1 and 11. In agreement with ESI-MS indication, the potentiometric data fitted very well with the model usually reported for copper(II) complexes of α-aminohydroxamate complexes ($[CuL]^+$, $[Cu_5(LH_{-1})_4]^{2+}$, $[CuL_2]$, $[CuL_2H_{-1}]^-$), however with much higher stability of the 12-MC-4 species. A series of copper(II) complexes has been isolated in the solid state and characterized by a variety of spectroscopic methods, X-ray structure analysis, and magnetic susceptibility measurements. The ligands show the tendency to form bi- and trinuclear species with copper(II) ions due to the {(N,N′); (O,O′)} bis-(bidentate) chelating-and-bridging mode involving (O,O′)-hydroxamate chelate formation combined with (N,N′) chelating with participation of the pyridine and hydroxamic nitrogen atoms, so that the hydroxamate groups play a μ_2-(N,O)-bridging role. Molecular and crystal structures of three synthesized complexes $[Cu_3$(**PicHA**-H$)_2$(dipy$)_2]$ $(ClO_4)_2 \cdot 4/3DMSO \cdot 2/3H_2O$ (**1**), $[Cu_2$(**PyDHA**) (dipy$)_2(ClO_4)_2] \cdot DMF \cdot H_2O$ (**4**), and $[Cu_3$(**PyDHA**-2H)(tmeda$)_3]$ $(ClO_4)_2$ (**5**) (dipy, 2,2′-dipyridyl; tmeda, N,N,N′,N′-tetramethyl-1,2-diaminoethane) have been determined by single crystal X-ray analysis. In **1**, two trans-situated doubly deprotonated hydroxamic ligands play a {(O,O′)(N,N′)}–(bis)bidentate-bridging function forming bridges between the medial, Cu(2) (CuN_4), and the terminal, Cu(1) and Cu(3) (CuN_2O_2), copper(II) ions; the chelating dipy ligands are coordinated to the latter. In **4**, the ligand is coordinated in a classical (O,O′)-hydroxamate chelating mode with the help of two separate hydroxamic groups while the central tridentate donor compartment remains vacant. In **5**, the hydroxamate ligand is coordinated by the {(O,O′);(N,N′,N″);(O″,O‴)}-tridentate-(bis)bidentate mode, bridging three copper(II) ions, while the chelating tmeda ligands are coordinated to all three copper(II) ions. Magnetic susceptibility measurements (1.7–300 K) of powdered samples of the trinuclear complexes **1** and **5** revealed strong antiferromagnetic coupling between the copper(II) ions mediated by the hydroxamate bridges.

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