

A systematic exploration of nickel(II)/acetate/Di-2-pyridyl ketone chemistry: neutral and cationic tetranuclear clusters, and a novel mononuclear complex.

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Abstract

The further use of di-2-pyridyl ketone [(2-py)₂CO] in nickel(II) acetate chemistry has been investigated. Various synthetic procedures have led to the synthesis of complexes [Ni₄(O₂CMe)₂{(2-py)₂C(OH)O}₄(H₂O)₂](ClO₄)₂ (**1**), [Ni(O₂CMe){(2-py)₂C(OH)₂}{(2-py)₂CO}](ClO₄)·H₂O (**2**·H₂O), [Ni₄(O₂CMe)₃{(2-py)₂C(OH)O}₄](ClO₄)·2H₂O·2EtOH (**3**·2H₂O·2EtOH), [Ni₄(O₂CMe)₄{(2-py)₂C(OH)O}₄]·2MeCN (**4**·2MeCN), and [Ni₄(O₂CMe)₃{(2-py)₂C(OH)O}₄](O₂CMe)·6H₂O·MeCN (**5**·6H₂O·MeCN). The Ni^{II}-mediated hydrolysis of (2-py)₂CO to give the coordinated molecule (2-py)₂C(OH)₂ or the monoanion (2-py)₂C(OH)O⁻ involves nucleophilic attack by H₂O on the carbonyl group. The Ni^{II} ion in **2**·H₂O is coordinated by one monodentate acetate, one *N,N',O*-tridentate chelating (2-py)₂C(OH)₂ molecule, and one *N,N'*-bidentate chelating (2-py)₂CO ligand. The tetranuclear cluster cation of **1** has a cubane {Ni₄(μ₃-OR)₄}⁴⁺ core with Ni^{II} ions and deprotonated oxygen atoms occupying alternate vertices. The [Ni₄(O₂CMe)₃{(2-py)₂C(OH)O}₄]⁺ cations present in the complexes **3**·2H₂O·2EtOH and **5**·6H₂O·MeCN have almost identical cubane structures that are different from the structure of the cation of **1**. The four metal ions in the centrosymmetric molecules of **4**·2MeCN are located at four vertices of a defect dicubane (with two missing vertices) and are bridged by six oxygen atoms. Characteristic IR bands are discussed in terms of the known structures of **1–5**. The magnetic data for the Ni^{II} cubane **1** has been modeled with two *J* values, and shows that the coupling consists of two ferromagnetically coupled pairs that are antiferromagnetically coupled to give a diamagnetic ground state. The magnetic properties of **4** have been modeled with three *J* values, and reveal competing antiferromagnetic and ferromagnetic coupling between the four Ni^{II} ions.

Słowa kluczowe

Acetate ligands, Di-2-pyridyl ketone, Magnetic properties, Nickel (II) cubanes, Nickel (II) defect dicubanes, X-ray diffractions

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