

Author's Accepted Manuscript

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PII: S0022-4596(16)30044-5
DOI: <http://dx.doi.org/10.1016/j.jssc.2016.02.018>
Reference: YJSSC19265

To appear in: *Journal of Solid State Chemistry*

Received date: 16 November 2015
Revised date: 31 January 2016
Accepted date: 13 February 2016

Cite this article as: Yayong Sun, Yingxia Zong, Haoran Ma, Ao Zhang, Kang Liu, Debao Wang, Wenqiang Wang and Lei Wang, Design and Syntheses of Hybrid metal-organic materials based on $K_3[M(C_2O_4)_3] \cdot 3H_2O$ [M(III)=Fe, al cr] metallotectons, *Journal of Solid State Chemistry*, <http://dx.doi.org/10.1016/j.jssc.2016.02.018>

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Design and Syntheses of Hybrid Metal-Organic Materials Based on $K_3[M(C_2O_4)_3] \cdot 3H_2O$ [M(III) = Fe, Al, Cr] metallotectons

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Abstract

By using $K_3[M(C_2O_4)_3] \cdot 3H_2O$ [M(III) = Fe, Al, Cr] ($C_2O_4^{2-}$ = oxalate) metallotectons as the starting material, we have synthesized eight novel complexes with formulas $[Fe(C_2O_4)_2(H_2O)_2]_2 \cdot (H-L_1)_2 \cdot H_2O$ 1, $[Fe(C_2O_4)Cl_2] \cdot (H_2-L_2)_{0.5} \cdot (L_2)_{0.5} \cdot H_2O$ 2, $[Fe(C_2O_4)_{1.5}Cl_2]_2 \cdot (H-L_3)_4$ 3, $[Fe_2(C_2O_4)Cl_8] \cdot (H_2-L_4)_2 \cdot 2H_2O$ 4, $K[Al(C_2O_4)_3] \cdot (H_2-L_5) \cdot 2H_2O$ 5, $K[Al(C_2O_4)_3] \cdot (H-L_6)_2 \cdot 2H_2O$ 6, $K[Cr(C_2O_4)_3] \cdot 2H_2O$ 7, $Na[Fe(C_2O_4)_3] \cdot (H-L_6)_2 \cdot 2H_2O$ 8 (with L_1 = 4-dimethylaminopyridine, L_2 = 2,3,5,6-tetramethylpyrazine, L_3 = 2-aminobenzimidazole, L_4 = 1,4-bis-(1H-imidazol-1-yl)benzene, L_5 = 1,4-bis((2-methylimidazol-1-yl)methyl)benzene, L_6 = 2-methylbenzimidazole). Their structures have been determined by single-crystal X-ray diffraction analyses, elemental analyses, IR spectra and thermogravimetric analyses. Compound 3 is a 2D H-bonded supramolecular architecture. Others are 3D supramolecular structures. Compound 1 shows a $[Fe(C_2O_4)_2(H_2O)_2]^-$ unit and 3D antionic H-bonded framework. Compound 2 features a $[Fe(C_2O_4)Cl_2]^-$ anion and 1D iron-oxalate-iron chain. Compound 3 features a $[Fe_2(C_2O_4)_3Cl_4]^{4+}$ unit. Compound 4 features distinct $[Fe_2(C_2O_4)_3Cl_8]^{4+}$ units, which are mutual linked by water molecules to generated a 2D H-bonded network. Compound 5 features infinite ladder-like chains constructed by $[Al(C_2O_4)_3]^{3-}$ units and K^+ cations. The 1D chains are further extended into 3D antionic H-bonded framework through O-H...O H-bonds. Compounds 6-8 show 2D $[KAl(C_2O_4)_3]^{2-}$ layer, $[KCr(C_2O_4)_3]^{2-}$ layer and $[NaFe(C_2O_4)_3]^{2-}$ layer, respectively.

Keywords: metallotectons; oxalate; supramolecular structures

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