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FULL PAPERS

一种大环化合物的晶体结构及其双核铜配合物的酪氨酸酶活性研究

周红[1]，潘志权[a,*]，罗勤慧[a, b]，梅光全[2]，龙德良[3]，陈久桐[3]

[1]武汉化工学院，湖北省新型反应器与绿色化学工艺重点实验室，武汉 430073
[2]南京大学，配位化学所，配位化学国家重点实验室，南京 210093
[3]中国科学院福州结构化学国家重点实验室，福州 350002

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摘要 通过2, 6-二甲酰氧化吡啶与二乙烯三胺缩合合成了一种结构大环化合物29,30-二氧-3,6, 9,17,20,23,29,30-八氮五环[23,3,1,1^{11,15},0^{2,6},0^{16,20}]三十烷1(28),9,11(12),13,15(30),23,25(29),26-八烯（L）。研究了它的自组装行为，通过分子间氢键和π-π堆积得到一种多孔的网状结构。制备了[$Cu_2L(MeOH)_2$]$(BF_4)_2$·2H_2O和[$Cu_2L(MeOH)_2$]$(ClO_4)_4$·2H_2O两种配合物。研究了它们在4:1的甲醇/乙腈溶液中催化氧化酚类底物(氢醌、2-甲基苯酚、2,6-二特丁基苯酚和2,6-二甲基苯酚)的反应。证明了配合物的氧合物是催化氧化的活性物种。

关键词 双核铜配合物,收缩大环化合物,晶体结构,载氧配合物,催化氧化

分类号

Study on the Crystal Structure of a Macrocycle and Tyrosinase Activity of Its Dinuclear Copper Complexes

ZHOU Hong[1], PAN Zhi-Quan[*,a], LUO Qin-Hui[a,b], MEI Guang-Quan[2], LONG De-Liang[3], CHEN Jiu-Tong[3]

[1]Hubei Key Laboratory of Novel Chemical Reactor and Green Chemical Technology, Wuhan Institute of Chemical Technology, Wuhan, Hubei 430073, China
[2] Coordination Chemistry Institute, State Key Laboratory of Coordination Chemistry, Nanjing University, Nanjing, Jiangsu 210093, China
[3] Fuzhou State Key Laboratory of Structure Chemistry, Fuzhou, Fujian 350002, China

Abstract A ring-contracted form macrocycle, 29,30-dioxo-3,6,9,17,20,23,29,30-octaazapentacyclo 23,3,1,1^{11,15},0^{2,6},0^{16,20}]- triacontaneocta-1(28),9,11(12),13,15(30),23,25(29),26-ene (L) was synthesized by condensation of diethyltriamine with pyridine-1-oxide-2,6-dicarboxaldehyde. A porous three-dimensional layer structure in its crystal was formed by self-assembly through hydrogen bonds and π-π interaction. Its dinuclear copper(I) complex [$Cu_2L(MeOH)_2$]- $(BF_4)_2$•2H_2O and dinuclear-copper(II) complex [$Cu_2L(MeOH)_2$]$(ClO_4)_4$•2H_2O were obtained and could oxidize catalytically four phenolic substrates hydroquinone, 2-methyl-hydroquinone, 2,6-di-*tert*-butylphenol and 2,6-di-methylphenol, in a mixture of methanol and acetonitrile (*V*：*V*, 4：1). The copper(I) complex reacted with dioxygen to form an oxygenated species as an initial active intermediate for oxidation of the phenols. Oxidation of the substrates by the copper(II) complex produced a copper(I) complex and the oxidation products of the substrates.

Key words dinuclear copper complex ring-contracted macrocycle crystal structure dioxygen complex catalytic oxidation

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通讯作者 潘志权 zhiqpan@163.com