四种含苯并咪唑基双核Cu(I)配合物的合成、表征和氧合性能

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摘要 本文合成了四种具苯并咪唑基配体:邻-二[N,N'-二(2'-苯并咪唑甲基]氨基]-反式-环己烷(OCTB) 及其三种系列物,分别与[Cu^I (CH~3CN)~4]ClO~4]反应制得了相应的双核Cu(I)配合物1-4。经元素分析、红外光谱和核磁共振等表明自由配体及其相应配合物均符合组成。紫外-可见光谱表明配合物1-4具有氧合性能,它们的无色DMF溶液吸氧后呈绿色,在可见区700nm附近出现宽吸收峰,用抗环血酸还原成无色,如此可循环3-4次。用气体吸收测量方法算得配合物1-4于DMF中氧合反应平衡常数,△H°和△S°。研究表明,配体苯并咪唑N上空间位阻大不利于氧合;斥电性取代基可增加配位原子的电负性,使中心原子Cu(I)上电子密度增加,有利于氧合。在不同溶剂中氧合速度大小为: DMF>DMSO>Py。

关键词 血蓝蛋白 双核配合物 氧载体 氧合性能

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Syntheses, characterization and oxygenation properties of four benzimidazolyl-containing dicopper (I) complexes

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Abstract This paper reports the syntheses of four benzimidazolyl-containing ligands: ortho-bis[N, N'-bis(2'-benzimidazolyl-methyl) amino]-trans-cyclohexane (OCTB) and its three alkylsubstituted derivatives. Dicopper(I) complexes 1-4 were respectively prepared through the reactions of the ligands with [Cu(CH~3CN)~4]ClO~4. Elemental analysis, IR spectra and ^1H NMR show that the compositions of the ligands and the complexes are the same as desired. UV-vis spectra indicate the complexes 1-4 can combine with dioxygen. The colorless solutions of the complexes in DMF show an uptake of oxygen and give green solutions with a wide absorption peak about at 700nm. Addition of ascorbic acid (AA) results in decolorization of the solutions and the cycle can be repeated three or four times. Equilibrium constants, $\triangle H^\circ$ and $\triangle S^\circ$ of oxygenation reactions of the complexes in DMF are obtained by manometric oxygen-uptake measurements. We consider that the steric hindrance on N atom of benzimidazole is unfavourable to oxygenation. The electron-donating substituents on N atom would increase the electron density of coordination atom and further the central ion Cu(I) and make the oxygenation more easier. The oxygenation rates in different solvents are in the order of DMF>DMSO>Py.

Key words HEMOCYANIN DINUCLEAR COMPLEX CARRIER OF OXYGEN

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