Copper(II) complexes of terminally free alloferon mutants containing two histidyl binding sites inside peptide chain structure and stability.

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Streszczenie

Mononuclear and polynuclear copper(II) complexes of alloferon 1 with point mutations, H1A/H12A H_2N-$A^1GVSGH^6GQH^9GVA^{12}G$-COOH, H1A/H9A H_2N-$A^1GVSGH^6GQA^9GVH^{12}G$-COOH, and H1A/H6A H_2N-$A^1GVSGA^6GQH^9GVH^{12}G$-COOH, have been studied by potentiometric, UV-visible, CD, and EPR spectroscopy, and mass spectrometry (MS) methods. Complete complex speciation at different metal-to-ligand molar ratios ranging from 1 : 1 to 3 : 1 was obtained. Over a wide 6–8 pH range, including physiological pH 7.4, and a 1 : 1 metal-to-ligand molar ratio, the peptides studied formed a $CuH_{-1}L$ complex with the $4N\{NH_2,N^-,2N_{Im}\}$ coordination mode. The presence of the 4N binding site for the $CuH_{-1}L$ complexes prevented the deprotonation and coordination of the second amide nitrogen atom to copper(II) ions ($pK_{-1/-2}$ 7.83–8.07) compared to that of pentaGly (6.81). The amine nitrogen donor and two imidazole nitrogen atoms (H^6H^9, H^6H^{12} and H^9H^{12}) can be considered to be independent metal-binding sites in the species formed. As a consequence, di- and trinuclear complexes for the metal-to-ligand 2 : 1 and 3 : 1 molar ratios dominate in the solution, respectively. For the Cu(II)-H1A/H9A and Cu(II)-H1A/H12A systems, the $Cu_3H_{-9}L$ complexes are likely formed by the coordination of amide nitrogen atoms towards C-termini with ring sizes (7,5,5).

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