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Article Author: Lin, G. D.; Zhang, H

Vol/Year/Pages: v.186, page: 16-INOR

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ABSTRACTS OF PAPERS

186th ACS NATIONAL MEETING

ISBN 8412-0771-2

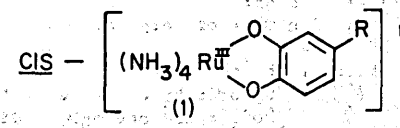
AMERICAN CHEMICAL SOCIETY

WASHINGTON, D.C.

August 28–September 2, 1983

15. SYNTHESIS AND BIODISTRIBUTION OF NEW CATECHOL AND CATECHOLAMINE COMPLEXES OF RUTHENIUM. S. Pell and M.A. Davis, Department of Chemistry, Worcester Polytechnic Institute, Worcester, MA 01609; R.B. Salmonsen, M.J. Clarke, and A. Abelleira, Department of Chemistry, Boston College, Chestnut Hill, MA 02167. Exploratory work performed at Department of Medicinal Chemistry, Northeastern University in cooperation with the Radiation Control Unit of Children's Hospital Medical Center, Boston, MA 02115

Cis- diolatotetraammineruthenium(III) complexes of the form 1 have been prepared in high yield and characterized by elemental analysis, cyclic voltammetry, ESR, ^{1}H NMR, and UV-visible spectroscopy.



R = H, alkyl, or amino acid, n = 1+
R = carboxylate, n = 0
R = protonated amine, n = 2+

Typically, these complexes are dark blue, paramagnetic, substitution inert, and air-stable at pH 7. They are readily isolated by ion exchange chromatography and their purity verified by reverse phase HPLC. The biodistributions of representative complexes labeled with Ru-103 have been investigated in normal mice. The results are discussed with regard to achieving tissue specificity in the design of Ru-97 diagnostic radiopharmaceuticals.

16. STUDY OF ATP-DRIVEN ELECTRON TRANSPORT IN N_2ASE REACTIONS WITH MODEL SYSTEMS Guo-dong Lin, Hong-tu Zhang, Hong-bo Chen, Ye-huan Wu, K. R. Tsai

Addition of MgATP(t) to $(Fe_4S_4L_4)^{-2}$ (L = $-SC_6H_5$; $-SCH_2C_6H_5$) in DMF-H_2O has been found to shift polarographic half-wave potential to more negative value, to suppress electronic absorption peaks of $(Fe_4S_4L_4)^{-2}$ to cause appreciable broadening and shifting of the ^{31}P NMR signals of the α,β,γ $^4PO_4$, of t. Redox-reaction rates of $(Fe_4S_4L_4)^{-2}$ with methylene blue or indigo carmine were enhanced by separate addition of t(no appreciable ATP hydrolysis attaining the redox reaction), $Na_2HPO_4(P_i)$,MgADP(d), or AMP, to decreasing extents: t≥P_i>d>m≫ClO_4^->H_2O, indicating that, in DMF-H_2O, t probably coordinated to $(Fe_4S_4L_4)^{-2}$as a monodentate ligand. However,in view of Mortenson's observation that complexation of t with reduced Fe-protein(2_s) shifted the ^{31}P NMR signals of theα,β,γ PO_4 of t, we infer that t probably coordinates as a bidentate ligand through a pair of O of theβ PO_4 to a pair of $Fe^{(II,III)}$ of the $(Fe_4S_4L_4)^{-2}$of (2_s), thereby increases the ligand field to drive the electron transfer from (2_s) to semi-reduced MoFe-protein (1_s), which in turn catalyzes hydrolysis of the liganted t. With additional assumptions that complexation of (2_s^{tt}), (2_s^{td}),(2_s^{dd}),(2_s^{tt}),or ($2_s^{t\phi}$), with (1_o) is rate controlling and requisite for driving electron transfer from (1_o) to coordinated substrate(without ATP hydrolysis), and that complexation of (1_o) with (2_o^{dd}) will lead to electron back-flow, it can be explained,diagrammatically, changes in EPR signals of N_2ase system with diffrent supplies of t and $S_2O_4^{-2}$, and changes in ATP/2e ratio with different t/d, or (1)/(2) ratios as observed by Mortenson et al.

17. LIGATIONAL EFFECTS ON REDUCTION OF MYOGLOBIN AND HORSERADISH PEROXIDASE BY INORGANIC REAGENTS. R. J. Balahura, Department of Chemistry, University of Guelph, Ontario, Canada N1G 2W1 and R. G. Wilkins, Department of Chemistry, New Mexico State University, Las Cruces, NM 88003.

Previous studies of the reduction of metmyoglobin (Mb^+) and adducts by dithionite have been extended to horseradish peroxidase (HRP^+). Second-order rate constants, ($M^{-1}s^{-1}$ at 25°C, pH=6.3, I=0.15M) for SO_2^- attack were 5.0×10^5 (HRP^+), 2.9×10^5 (HRP^+CN^-) and $\sim10^4$(HRP^+F^-). In addition, the reduction of Mb^+ and adducts, HRP^+, HIPIP and cyt-c(III) by a bulky reductant, the macrocycle cobalt(II) sepulchrate ($Cosep^{2+}$) was examined. Second-order rate constant were 3.5×10^3(Mb^+), 7.4×10^3(Mb^+CN^-)3.1×10^4(Mb^+imid)2.1×10^5 (HIPIP), 3.0×10^5(cyt-c(III)). Reduction of Mb^+F^- by $Cosep^{2+}$, $FeEDTA^{2-}$(and SO_2^-) only occurs via dissociation. Similar pattern of behavior for these reductants were thus observed and these are discussed. In the reduction of HRP^+ by $Cosep^{2+}$ three steps are observed. These are all first-order in HRP^+ with associated k_{obsd} values of (1) $7.0\times10^4[Cosep^{2+}][1+350[Cosep^{2+}]]^{-1}$ (2) $\sim1.5s^{-1}$ and (3) $0.011s^{-1}$. The last slow change is believed to represent an intramolecular electron transfer from cobalt(II) attached to the protein to the iron(III) center.