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Lubben, M.; Meetsma, Auke; Feringa, Bernard

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NON-SYMMETRIC DINUCLEATING LIGANDS FOR OXIDATION CATALYSTS TEMPLATES FOR REALISTIC DINUCLEAR METALLOENZYME MIMICS

<u>M. Lubben</u>, Auke Meetsma and Ben L. Feringa

Department of Organic and Molecular Inorganic Chemistry, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

Abstract. One of the most striking features in many dinuclear metalloenzymes is the different coordination environments for the two metal centers. Nevertheless most of the reported enzyme mimics which are small molecule analogues for the active sites of these complicated enzymes, are based on symmetric dinucleating ligands. These ligands generally result in the formation of dinuclear coordination complexes with identical coordination environments for the metal centers. However, a limited number of non-symmetric dinuclear complexes is known to be formed by spontaneous self-assembly in solution.

We present here a new entry into the synthesis of non-symmetric dinuclear transition metal complexes by making use of non-symmetric dinucleating ligands. The use of a non-symmetric dinucleating ligand will force the two metal ions in the resulting complex into different chemical or coordination environments. This will result in the formation of a more realistic enzyme mimic.

The synthesis of the non-symmetric ligands is based on the Bimolecular Aromatic Mannich reaction. Non-symmetric ligands can be obtained via two different routes. The first route comprises a sequential Mannich reaction on a phenol using two different secondary amines. An alternative route is a Mannich reaction using a secondary amine and a substituted salicylaldehyde followed by either condensation of the aldehyde functionality with a primary amine and subsequent reduction or reductive amination with a secondary amine.

The excellent ligating ability of these non-symmetric ligand systems is illustrated by the presentation of the crystal and molecular structure of a non-symmetric dicopper(II) complex.

Preliminary results on the use of non-symmetric dinuclear complexes in catalytic oxidations will be presented.

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