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## Synthesis and characterization of new Mannich base ligand. Structure of N, N-bis-(4,5-dimethyl-2-hydroxybenzyl)-N(2-pyridylmethyl)amine

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### Abstract

Examples of interesting ligands previously requiring lengthy synthetic procedures have been prepared by one step routes, opening the way to more extensive studies of their complexes and to possible applications. New amine bis-(phenolate) ligand with pendant arm on the amine nitrogen donor have been synthesized, via the Mannich condensation, between a disubstituted phenol, formaldehyde, and N(2-pyridylmethyl)amine

### Introduction

The chemistry of of amine bis-(phenolate) ligand has become a rapidly growing area of research, because of their importance in biomimetic studies of binuclear metalloproteins, their interesting catalytic properties, their ability to stabilize unusual oxidation states and mixed-valence compounds, and the possibilities for magnetic interaction between the two metal ions, leading to the design of molecular magnetic materials.<sup>1-4</sup> This kind of ligand provide efficient pathways for metal-metal interactions through bridging phenolic oxygen and nitrogen donor atoms of the binuclear complex with ligands having a tetradentate coordination sites are expected to be effective mimics of redox enzymes.<sup>5,6,7</sup>

In this work, we describe a simple 1-step procedure for the synthesis of N, N-bis-(4,5-dimethyl-2-hydroxybenzyl)-N(2-pyridylmethyl)amine compound based on the Mannich reaction (Figure 1). The synthesis and characterization of the dinuclear complexes with amine bis-(phenolate) ligand will be a subject of future studies.

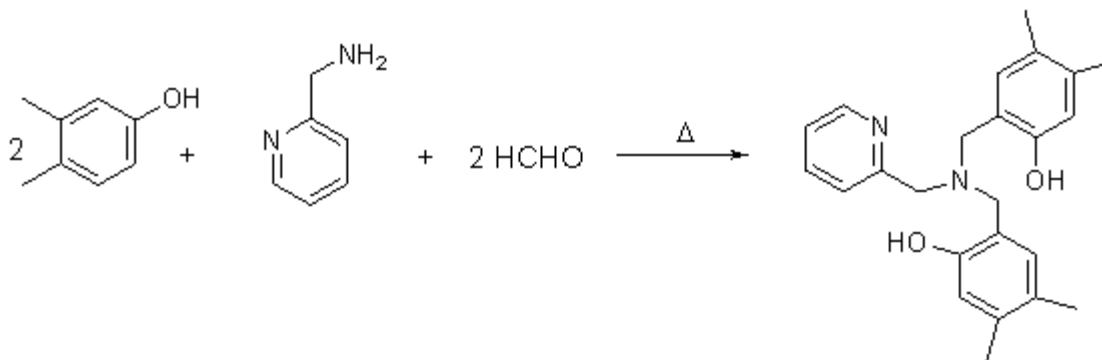


Figure 1

### Molecular structure

Now, we present the crystal structure of the N,N-bis-(4,5-dimethyl-2-hydroxybenzyl)-N(2-pyridylmethyl)amine compound Compound (I) (Fig. 2) which possesses normal geometric parameters. The hydrogen-bonding features an O—H<sup>1/4</sup>O interaction; inversion symmetry generates a dimeric association of molecules via two such bonds

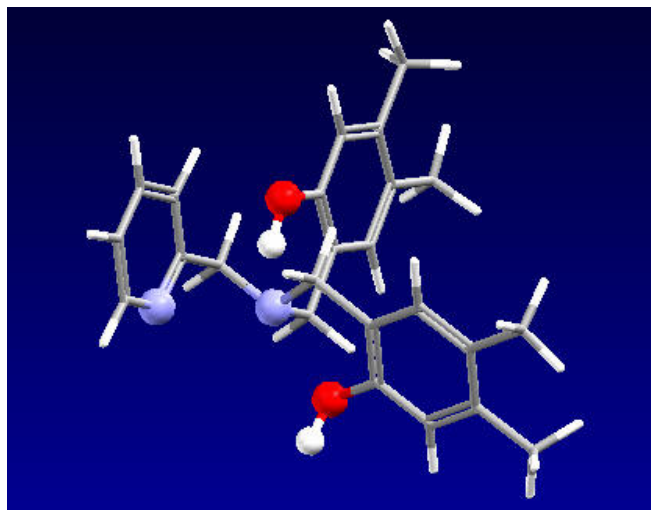


Figure 2

## Experimental Section

**General Considerations.** All solvents and 3,4-dimethylphenol, N(2-pyridylmethyl)amine, and 37% aq formaldehyde were commercial products (Aldrich) and were used as supplied. Microanalyses were performed using a Perkin-Elmer 240B microanalyzer. IR spectra were recorded on a Perkin-Elmer 1330 spectrophotometer.  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were recorded in deuterated  $\text{CDCl}_3$  with a Bruker WM 350 MHz spectrometer. The FAB mass spectra of the complexes were recorded on a Micromass Autospec instrument.

**Ligand Synthesis.** The ligand  $\text{H}_2\text{L}$  was prepared by condensation between N(2-pyridylmethyl)amine (2.6 mL, 25 mmol), formaldehyde 37% (3.7 mL, 50 mmol), and 3,4-dimethylphenol (6.10 g, 50 mmol) in methanol (250 mL). The solution was heated under reflux for 5 h and then cooled (4 °C). The resulting white precipitate was filtered off and recrystallized (dichloromethane/methanol) to give white crystals of  $\text{H}_2\text{L}$ . Anal. Found

(Calcd) for  $\text{C}_{24}\text{H}_{28}\text{N}_2\text{O}_2$ : C, 76.99 (76.56); H, 7.50 (7.50), N, 7.57 (7.44)%.  $^1\text{H}$  NMR ( $\text{CDCl}_3$ , ppm) 10.4 (s br, 2H, OH), 8.6, 7.7, and 7.1 (4H pyridine) 6.79 (s, 2H, phenyl), and 6.69 (s, 2H, phenyl), 3.86 (s, 2H,  $\text{CH}_2$ ), 2.57 (s, 4H,  $\text{CH}_2$ ), 2.17 (s, 6H,  $\text{CH}_3$ ) and 2.14 (s, 6H,  $\text{CH}_3$ )  $^{13}\text{C}$  NMR ( $\text{CDCl}_3$ , ppm) 155.3 pyridine, 148.4 (C-OH), 138-118 (phenyl, pyridine), 56.1 and 55.7 (N- $\text{CH}_2$ ), 19.8 and 18.9 ( $\text{CH}_3$ ). IR (KBr,  $\text{cm}^{-1}$ ): 736(vw), 768(m), 823(m), 869(m), 951(m), 971(m), 1002(m), 1019(m), 1096(s), 1108(m), 1149(m), 1174(s), 1242(w), 1250(m), 1266(m), 1295(vs), 1342(s), 1375(s), 1384(m), 1462(s), 1468(s), 1580(s), 1596(vs), 1630(w), 2762(w), 2821(m), 2848(w), 2919(vw), 2938(w), 3072(vbr), 3088(vbr). EI MS;  $m/z$ : 376

( $\text{M}^+$ , 100%).

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