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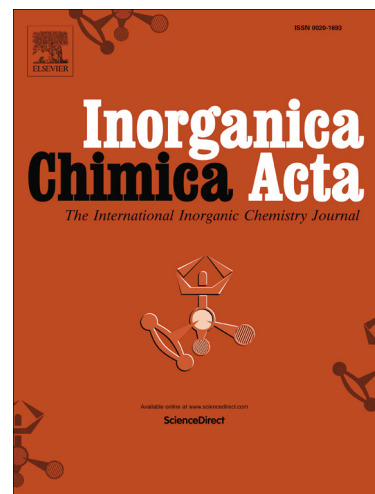
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# Push-Pull Ligands to Enhance the Oxygen Activation Step in Catalytic Oxidation with Platinum Complexes

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## ABSTRACT

The ligands RN(CH<sub>2</sub>-2-C<sub>5</sub>H<sub>4</sub>N)<sub>2</sub>, **L1**, R = CH<sub>2</sub>C<sub>6</sub>H<sub>4</sub>-2-OH; **L2**, R = CH<sub>2</sub>C<sub>6</sub>H<sub>3</sub>-4-Me-2-OH; **L3**, R = CH<sub>2</sub>C<sub>6</sub>H<sub>3</sub>-5-Cl-2-OH; **L4**, R = CH<sub>2</sub>C<sub>6</sub>H<sub>3</sub>-5-NO<sub>2</sub>-2-OH; give dimethylplatinum(II) complexes that activate dioxygen in the absence of a protic solvent. The initial reaction with [Pt<sub>2</sub>Me<sub>4</sub>(SMe<sub>2</sub>)<sub>2</sub>] gives [PtMe<sub>2</sub>(κ<sup>2</sup>-N,N'-L)], and these react with oxygen to give [Pt(OH)Me<sub>2</sub>(κ<sup>3</sup>-N,N',N''-L-H)], which exist as a mixture of isomers. The factors influencing reactivity and mechanism in these reactions are elucidated, and the presence of both a free pyridyl donor (push group) and a free hydroxyl (pull group) is suggested to give a synergy for dioxygen activation. Some platinum(II) complexes and trimethylplatinum(IV) complexes with these ligands are also reported.

## 1. Introduction

A major challenge in the use of dioxygen as a selective oxidant in the catalysis of hydrocarbon oxidation is to control the selectivity and reactivity of the reactions [1]. One approach to improving this form of catalysis is to study individual steps in the proposed catalytic reactions and, from the better fundamental understanding that this enables, to develop optimum conditions for selective reaction under mild conditions. Electron-rich dimethylplatinum(II) complexes with nitrogen-donor ligands are among the most reactive complexes towards oxidative addition, yet they are typically stable to air in the solid state or in non-protic organic solvents [2-4]. However, they can be oxidized by dioxygen in protic solvents [5]. The reaction of dimethylplatinum(II) complexes (**A**, Scheme 1, NN = bidentate nitrogen donor ligand such as 2,2'-bipyridine) with dioxygen in methanol occurs in two steps, each of which can be considered as a proton coupled 2-electron transfer, first to give a hydroperoxide and then a hydroxide complex (Scheme 1, **B**, **C**) [5]. It has also been shown that ligands with an additional Lewis base

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