

Rhodium assisted C–H activation of *N*-(2'-hydroxyphenyl)benzaldimines. Synthesis, structure and electrochemical properties of a group of organorhodium complexes

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Abstract

Reaction of a group of *N*-(2'-hydroxyphenyl)benzaldimines, derived from 2-aminophenol and five *para*-substituted benzaldehydes (the *para* substituents are OCH₃, CH₃, H, Cl and NO₂), with [Rh(PPh₃)₃Cl] in refluxing toluene in the presence of a base (NEt₃) afforded a family of organometallic complexes of rhodium(III). The crystal structure of one complex has been determined by X-ray crystallography. In these complexes the benzaldimine ligands are coordinated to the metal center, via dissociation of the phenolic proton and the phenyl proton at the *ortho* position of the phenyl ring in the imine fragment, as dianionic tridentate C,N,O-donors, and the two PPh₃ ligands are *trans*. The complexes are diamagnetic (low-spin d⁶, S = 0) and show intense MLCT transitions in the visible region. Cyclic voltammetry shows a Rh(III)–Rh(IV) oxidation within 0.63–0.93 V vs SCE followed by an oxidation of the coordinated benzaldimine ligand. A reduction of the coordinated benzaldimine is also observed within –0.96 to –1.04 V vs SCE. Potential of the Rh(III)–Rh(IV) oxidation is found to be sensitive to the nature of the *para*-substituent.

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1. Introduction

Utilization of transition metals in promoting useful chemical transformations of organic substrates has been of considerable current interest [1]. Such transformations usually proceed via a C–H activation of the organic substrate [2], leading to the formation of a reactive organometallic intermediate, which then undergoes further reactions to yield the final product. Transition metal mediated C–H activation of organic molecules is therefore of significant importance and the present work has originated from our interest in this area [3]. For the present study, a group of five Schiff base ligands, viz. *N*-(2'-hydroxyphenyl)benzaldimines (**1**), derived from *para*-substituted benzaldehydes

and 2-aminophenol, have been selected as the target molecules for C–H activation and rhodium has been selected as the transition metal for promoting the C–H activation. These ligands have two potential donor sites, the phenolate-oxygen and the imine-nitrogen, and hence are expected to bind to metal ions, via dissociation of the phenolic proton, as bidentate N,O-donors forming five-membered chelate ring (**2**). In view of the closeness of the pendent phenyl ring to the metal center in **2**, C–H activation at the *ortho* position of the phenyl ring leading to the formation of a cyclometallated species (**3**) appears to be a possibility. With the intention of inducing the said C–H activation of the *N*-(2'-hydroxyphenyl)benzaldimines (**1**), their reaction has been carried out with the Wilkinson's catalyst, viz. [Rh(PPh₃)₃Cl]. This particular complex has been picked up as the rhodium starting material for this reaction because of its demonstrated ability to promote C–H

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