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# Ruthenium(II) Pentamethylcyclopentadienyl Half-Sandwich Carbene Complexes with Polypyridyl Ligands

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**Abstract:** Polypyridyl complexes  $[\text{RuCl}(\eta^5\text{-C}_5\text{Me}_5)(\text{N-N})]$  (**1–4**) [ $\text{N-N} = 2,2'$ -bipyridine (bpy) (**1**), 4,4'-dimethyl-2,2'-bipyridine (4,4'-Me<sub>2</sub>bpy) (**2**), 1,10-phenanthroline (phen) (**3**) and 4,7-dimethyl-1,10-phenanthroline (4,7-Me<sub>2</sub>phen) (**4**)] were prepared by reacting  $[\text{RuCl}_2(\eta^5\text{-C}_5\text{Me}_5)]_2$  with Zn dust in the presence of polypyridyl ligands. Unlike the related  $[\text{RuCl}(\eta^5\text{-C}_5\text{Me}_5)(\text{L}_1)(\text{L}_2)]^+$  [ $\text{L}_1, \text{L}_2 = \text{P}(\text{OR})_3, \text{PPh}_3$ ] derivatives with P-donor ligands, treatment of chloro compounds **1–4** with diazoalkanes  $\text{Ar}_1\text{Ar}_2\text{CN}_2$  afforded polypyridyl carbene complexes  $[\text{Ru}(\eta^5\text{-C}_5\text{Me}_5)(=\text{CAr}_1\text{Ar}_2)(\text{N-N})]\text{BPh}_4$  (**5–8**) [ $\text{Ar}_1 = \text{Ar}_2 = \text{Ph}$  (**a**),  $\text{Ar}_1 = \text{Ph}$ ,  $\text{Ar}_2 = p\text{-tolyl}$  (**b**)]. Comparative DFT studies of the formation of metallocarbenes from aromatic diazoalkanes are reported. Phosphite complex  $[\text{Ru}(\eta^5\text{-C}_5\text{Me}_5)(\text{bpy})\{\text{P}(\text{OEt})_3\}]\text{BPh}_4$  (**9**) and acetylide derivatives  $[\text{Ru}(\eta^5\text{-C}_5\text{Me}_5)(\text{C}\equiv\text{CAr})(\text{bpy})]$  (**10, 11**) [ $\text{Ar} = \text{Ph}$  (**10**),  $p\text{-tolyl}$  (**11**)] were also obtained by substitution of the carbene in **5–8**. Unstable vinylidene cations  $[\text{Ru}(\eta^5\text{-C}_5\text{Me}_5)\{\text{C}=\text{C}(\text{H})\text{Ar}\}(\text{bpy})]^+$  (**12, 13**) were formed by protonation with  $\text{HBF}_4\cdot\text{Et}_2\text{O}$  of acetylide species **10, 11**. The complexes were characterized spectroscopically (IR and <sup>1</sup>H, <sup>13</sup>C, <sup>31</sup>P NMR) and by X-ray crystal structure determination of  $[\text{Ru}(\eta^5\text{-C}_5\text{Me}_5)\{\text{C}=\text{C}(\text{Ph})(p\text{-tolyl})\}(\text{bpy})]\text{BPh}_4$  (**5b**).

**Keywords:** Ruthenium; Half-sandwich; Diazoalkane; Carbene; Polypyridyl; Acetylide

## 1. Introduction

Diazoalkanes  $\text{Ar}_1\text{Ar}_2\text{CN}_2$  are a class of molecules which may behave as ligands in transition metal chemistry, giving rise to a number of stable and isolable complexes [1-4]. In some cases, diazoalkanes are also reported to react with metal complexes to give metallocarbene derivatives [5,6] after loss of  $\text{N}_2$ .

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