

CO₂-Driven N-Formylation/N-Methylation of Amines Using C-Scorpionate Metal Complexes

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C-scorpionate metal complexes, specifically, [NiCl₂(tpm)]·3H₂O, [CoCl₂(tpm)]·3H₂O and [PdCl₂(tpm)] [tpm = hydrotris(1H-pyrazol-1-yl)methane], were effective in the N-formylation and N-methylation of amines using carbon dioxide, as carbon source, in the presence of sodium borohydride. Various parameters were studied, including reaction time, temperature, solvent volume, presence of additives, and catalyst amount. These parameters were found to have a significant impact on the selectivity of the product. [NiCl₂(tpm)]·3H₂O exhibited good conversion at 80 °C, but its selectivity towards formamide decreased with prolonged reaction time. Increasing the amount of [NiCl₂(tpm)]·3H₂O, the selectivity changed. [PdCl₂(tpm)] showed different selectivity compared to [NiCl₂(tpm)]·3H₂O, while [CoCl₂(tpm)]·3H₂O presented poor results. Monitoring the reaction course by ¹H NMR revealed the presence of an intermediate species that influenced product formation. These results highlight the versatility and catalytic potential of C-scorpionate metal complexes in the N-formylation/N-methylation of amines in the catalytic system (NaBH₄/MeCN/CO₂).

Słowa kluczowe

CO₂, C-scorpionate, hydrotris(1H-pyrazol-1-yl)methane, catalyst, amine, N-formylation, N-methylation

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