



## Enzymatic and Model Carboxylation and Reduction Reactions for Carbon Dioxide Utilization (Nato Science Series C:) (Volume 314)

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The activation of carbon dioxide by transition metal complexes has been extensively studied, both experimentally and theoretically. 1 Central reactions in this chemistry are the insertion of CO<sub>2</sub> into M-X bonds, where X = H, C, O, and N. (eq. 1-4). We are presently investigating the mechanistic aspects of these reaction processes and will herein describe our current level of understanding. Comparisons of the pathway of the carbon-carbon bond forming process in transition metal chemistry with the well known analogous chemistry involving organolithium reagents will be presented. Furthermore, the role of these reaction types in both homogeneous and heterogeneous catalytic processes leading to useful chemicals will be elaborated.

1)  $LMt-H + \sim LMlopi \rightarrow LMt-OR + \sim [M]O$

2)  $[Mt-R + CO] \rightarrow [M]O + 2CR$

3)  $[Mt-OR + \sim [M]O] \rightarrow [M]O + 2COR$

4)  $[Mt-NR_2 + CO] \rightarrow [M]O + 2CNR_2$

Insertion of CO<sub>2</sub> into the Metal-Hydride Bond. The reaction of anionic group 6 (Cr, Mo, W) transition metal hydrides with carbon dioxide to afford metalloformates occurs readily at ambient temperature and 2 reduced pressures of carbon dioxide. This insertion process is referred to the normal pathway (Scheme 1). There are no documented cases of CO<sub>2</sub> insertion into the metal hydride bond to provide the alternative, metalcarboxylic acid, isomer (referred in Scheme 1 as abnormal).

3 Recent theoretical studies ascribe this preference to an unfavorable electrostatic interaction and poorer orbital overlap in the latter process. Nevertheless,

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OM> (1) M-H + ~ M-OPi OM> (2) [M-R + CO. 2 [M]O. 2CR OM> (3) [M-OR+ ~ [M]O. 2COR OM> (4) [M-NR<sub>2</sub> + CO. 2 [M]~CNR<sub>2</sub> Insertion of CO<sub>2</sub> into the Metal-Hydride Bond. The reaction of anionic group 6 (Cr, Mo, W) transition metal hydrides with carbon dioxide to afford metalloformates occurs readily at ambient temperature and 2 reduced pressures of carbon dioxide. This insertion process is referred to the normal pathway (Scheme 1). There are no documented cases of CO<sub>2</sub> insertion into the metal hydride bond to provide the alternative, metalcarboxylic acid, isomer (referred in Scheme 1 as abnormal). 3 Recent theoretical studies ascribe this preference to an unfavorable electrostatic interaction and poorer orbital overlap in the latter process. Nevertheless.

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