

Synthetic Access to Complex Metal Carbonates and Hydroxycarbonates via Sol-Gel Chemistry

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Abstract : Metal alkoxides are very versatile precursors for a broad array of complex functional materials. However, metal alkoxides, especially transition metal alkoxides, tend to form oligomeric structures due to the very strong M-O-M binding motif. This fact hinders their facile application in sol-gel-processes and complicates access to complex carbonate or oxidic compounds after hydrolysis of the precursors. Therefore, the development of a synthetic alternative with the aim to grant access to carbonates and hydroxycarbonates from simple metal alkoxide precursors via hydrolysis is key to this project. Our approach involves the reaction of metal alkoxides with unsaturated isoelectronic molecules, such as carbon dioxide. Subsequently, a stoichiometric insertion of the CO₂ into the alkoxide M-O bond takes place and leads to the formation of soluble metal alkyl carbonates. This strategy is a very elegant approach to solubilize metal alkoxide precursors to make them accessible for sol-gel chemistry. After hydrolysis of the metal alkyl carbonates, crystalline metal carbonates, and hydroxycarbonates can be obtained, which were then utilized for the synthesis of Cu/Zn based bulk catalysts for methanol synthesis. Using these catalysts, a comparable catalytic activity to commercially available MeOH catalysts could be reached. Based on these results, a complement for traditional precipitation techniques, which are usually utilized for the synthesis of bulk methanol catalysts, have been found based on an alternative solubilization strategy.

Keywords : metal alkoxides, metal carbonates, metal hydroxycarbonates, CO₂ insertion, solubilization

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