

Ni-based catalysts with controlled porosity for gas phase CO₂ conversion

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The huge scientific and technological interest towards the abatement of CO₂ emissions has brought about the carbon capture and utilization (CCU) technologies as more appealing strategies to the carbon capture and storage (CCS) ones. In this regard, CO₂ utilization through gas phase conversion to added-value products (synthetic methane, syngas) is a straightforward approach to its recycling [1]. However, the chemical inertness and thermodynamic stability of the CO₂ molecule is a great challenge, requiring highly active and selective catalysts for its efficient activation. Among the investigated catalysts for the conversion of CO₂, Ni-based ones are still the most appealing, owing to their lower cost compared to noble metal catalysts, besides their high activity. Besides, the nature, and properties of the catalytic support influence not only the dispersion of the active phase, and the metal-support interaction, but also the extent of CO₂ activation. In this respect, catalytic supports with high surface area, and controlled porosity can play a crucial role in fulfilling all these requirements.

Two classes of materials with controlled porosity were considered as catalytic supports for the conversion of CO₂: (1) metal-organic frameworks (MOFs), and (2) ordered mesoporous silica (OMS). Owing to their ultrahigh surface area and tunable porosity obtained by the connection of inorganic secondary building units (metal ions or clusters) with various organic linkers, leading to 1D, 2D or 3D highly crystalline porous structures, MOFs are very attractive candidates for gas-phase reactions. Our approach in using MOFs (MIL-101(Cr), MIL-53(Cr), MIL-53(Al), MIL-100(Al)) for the conversion of CO₂ was to obtain both MOF-based [2] and MOF-derived Ni catalysts [3]. In the case of MOF-based Ni catalysts the results obtained either by: (a) the encapsulation of Ni NPs in MOFs (Ni/MOF catalysts); or (b) the synthesis of MOF-Al₂O₃ composites followed by deposition of Ni NPs (Ni/MOF-Al₂O₃ catalysts) will be presented. On the other hand, MOF-derived Ni catalysts were obtained by (a) thermolysis of the MOF structure in air to give the corresponding metal-oxide which inherits to a large extent the porous structure of the parent MOF, followed by deposition of Ni (Ni/Oxide(MOF)); and (b) deposition of Ni in the parent MOF, followed by thermolysis in air (Ni@Oxide(Ni@MOF)). Among these strategies, the MOF-derived Ni catalysts revealed the best catalytic performance owing to the uniform distribution of small Ni nanoparticles, and the enhanced H₂ and CO₂ adsorption capacity. Another class of materials with designed and controlled porous structure used for CO₂ transformation were Ni/silica. The results obtained using Ni deposited on (a) multimodal pore structure SiO₂, promoted by MgO, CeO₂, or La₂O₃ (Ni/Oxide-SiO₂), and (b) SBA-15, with Ni deposited by 3 different strategies (impregnation, double-solvent method, or melt infiltration) will be presented. It was found that promotion by La₂O₃, as well as the MI method led to enhanced dispersion of Ni NPs, and consequently improved catalytic performance.

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References: