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Enhanced low-temperature CO₂ methanation over Ni-based catalyst: Effect of support and reaction mechanism

Abstract: The growing atmospheric concentration of CO₂ is considered one of the main causes of global warming. Power-to-gas (PtG) processes have the great potential to store the excess of H₂ generated from renewable energy, simultaneously reducing CO₂ emissions from the atmosphere. In the PtG technology concept, the renewable electricity is used to produce hydrogen by electrolysis of water, and subsequently hydrogen reacts with CO₂ to yield CH₄ via the Sabatier reaction. In this study, we focus on the catalytic behaviors of Ni catalysts supported on Y₂O₃, ZrO₂, CeO₂ and mixed oxides. A comprehensive understanding of the structure-activity correlations was provided to optimally design an effective catalyst for CO₂ hydrogenation. Ni-based catalysts were synthesized by the solution combustion synthesis (SCS) method and characterized by N₂-physisorption, XRD, CO-chemisorption, H₂-TPR, CO₂-TPD, XPS, and TEM. Methanation of CO₂ was investigated under atmospheric pressure, in the temperature range of 250-500°C, at constant stoichiometric reactants ratio of H₂/CO₂ = 4 and gas hourly space velocity (GHSV) of 30,000-120,000 NmL·g⁻¹·h⁻¹. Furthermore, the formation of intermediates was investigated by in-situ DRIFT tests. In general, the catalytic activity strongly depends on the nature of the Ni-oxide interactions. Based on DRIFT observations, the main reaction pathway involves the adsorption of CO₂ onto catalyst support as mono- or bi-dentate carbonate as well as the H₂ dissociation on metallic Ni sites. The adsorbed CO₂ was progressively hydrogenated to methane passing through the formation of formate species as active intermediate. However, the superior catalytic performance of Ni/CeO₂-ZrO₂ catalyst is mainly due to enhanced medium basicity as well as the relatively high Ni dispersion, as confirmed by CO₂-TPD and CO-chemisorption measurements. Finally, good anti-coking and anti-sintering ability after 100 h of lifetime test make Ni/CeO₂-ZrO₂ catalyst an effective catalyst to produce methane by CO₂ hydrogenation.

Biography: Cristina Italiano received her Ph.D. degree in “Chemical Technologies and Innovative Processes” at the University of Messina in 2013. She is a Researcher at National Research Council - Institute of Advanced Technologies for Energy (CNR-ITAE) since 2018. Author and co-author of 54 papers in reputed journals. The main research projects concern catalysts development for energetic applications, mainly i) reforming processes for hydrogen production; ii) methanation reactions for synthetic natural gas production; iii) ammonia synthesis and decomposition for hydrogen storage.