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Thermal and plasma-catalytic CO₂ methanation over nanostructured Ni/CeO₂

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PURPOSE OF THE ABSTRACT

CO₂ capture and utilization (CCU) is one of the main strategies to be developed in the direction of low-carbon budget and circular economy. With CCU, CO₂ is no longer considered as a waste molecule but as a reservoir of carbon to be exploited as building block for added-value compounds, for instance vector molecules for harvest of energy in the Power-to-gas technology [1]. Power-to-gas concept is beneficial for the development of electricity generation from renewable sources, which are seasonal and unpredictable, and therefore need an energy carrier or buffer, such as hydrogen. Due to the current limitations in the storage and utilization of hydrogen as a fuel, the reaction between H₂ and CO₂ to produce recycled hydrocarbons can be advantageous to obtain more stable and efficient carriers. In this scenario, Sabatier's reaction of CO₂ methanation is experiencing a renaissance for the development of Power-to-Methane and the production of synthetic fuels [2]. CO₂ methanation is exothermic but kinetically hindered, hence it requires a suitable catalyst for activation and selective conversion. In thermal heterogeneous catalysis the reaction typically operates between 200°C and 450°C, at pressure between atmospheric and 100 bar. The increase of temperature improves the CO₂ conversion but lowers the selectivity toward CH₄ and in addition causes catalyst deactivation. Recently, besides the efforts done in the conventional thermal catalysis field, novel methods for CO₂ hydrogenation have been investigated: electrocatalysis, photocatalysis, and plasma-assisted catalysis. It has been demonstrated that the use of a non-thermal plasma in combination with a Ni-containing catalysts results in an enhanced methane yield even at atmospheric pressure and mild reaction temperatures [3]. The study of synergy between plasma and catalytic materials represents a challenge but it is also very promising for the advancement of Power-to-gas technology.

In this study, Ni catalysts were prepared with nanostructured CeO₂ supports and tested for CO₂ methanation in a U-type tubular fixed bed reactor and in a packed dielectric barrier discharge (DBD) reactor in order to study the catalytic behaviour of Ni/CeO₂ materials in thermal and plasma-assisted processes. The nanoscale morphologies of CeO₂, namely nanoparticles (NP), nanorods (NR) and nanocubes (NC), were obtained through the modification of their hydrothermal synthesis conditions and the Ni phase (15% weight) was introduced via wet impregnation. The catalysts were characterized by means of XRD, XPS, SEM, H₂ TPR, CO₂ TPD, TGA, and N₂ physisorption before the reaction as well as post mortem to assess their physicochemical properties and possible modifications. We report that the synthesis conditions of the CeO₂ supports influence their catalytic performance and we observe that the key structural and chemical features for a good activity differ in thermal and plasma-assisted methanation. For instance, the support morphology has an impact on the properties of the catalysts and the yield in CO₂ methanation. Figure 1 and 2 show the different conversion obtained over the Ni/CeO₂ catalysts, ranking as NP>NR>NC in thermal and NR>NP>NC in plasma-assisted reaction.

FIGURES

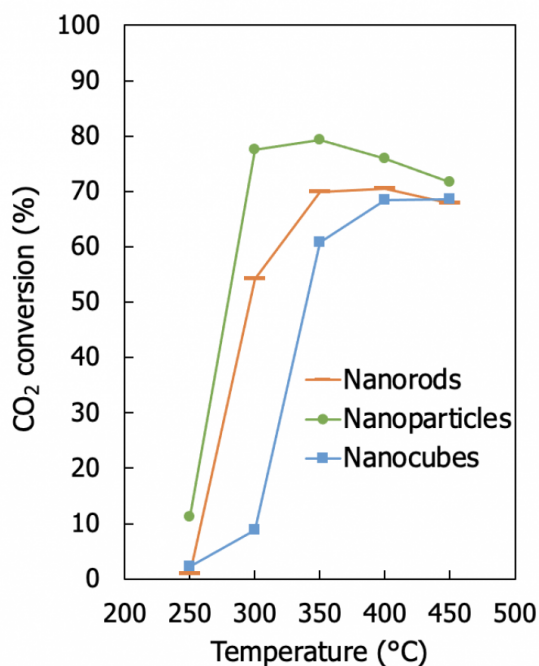


FIGURE 1

Catalytic performance in thermal CO₂ methanation of Ni/CeO₂ NC, Ni/CeO₂ NR, and Ni/CeO₂ NP.

CO₂/H₂/Ar 3/12/5

Total flow 100 mL/min

WHSV 30000 ml h⁻¹ g⁻¹

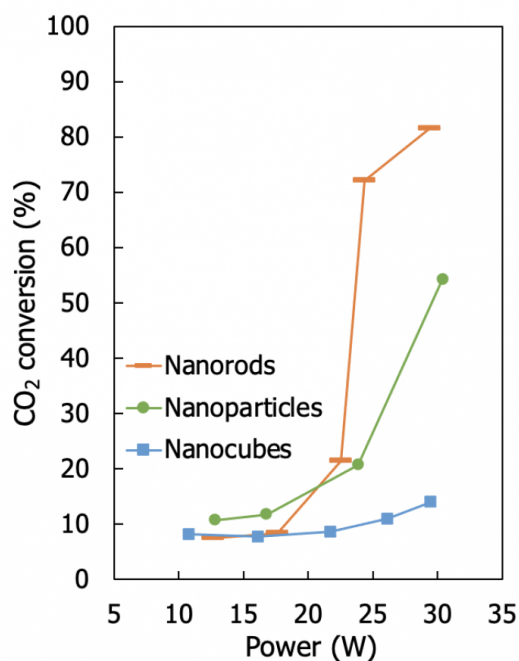


FIGURE 2

Catalytic performance in plasma-assisted CO₂ methanation of Ni/CeO₂ NC, Ni/CeO₂ NR, and Ni/CeO₂ NP.

21-24 kV, 12.3 kHz

CO₂/H₂ 1/4

Total flow 100 mL/min

WHSV 30000 ml h⁻¹ g⁻¹

KEYWORDS

Power-to-gas | Plasma catalysis | CO₂ capture and utilization | Cerium oxide

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