

Ni/CeO₂-catalyzed CO₂ Methanation: The effect of CeO₂ support nanoconfigurations.

P. Karga^{1, 2, #}, C. Drosou^{1,2}, E. Nikolaraki², M.A. Karakassides³, D. Gournis^{1,2}, Y.G. Deligiannakis^{1,4} and I.V. Yentekakis^{1,2,*}

¹Institute of Geoenergy, Foundation for Research and Technology-Hellas (IG/FORTH), Chania 73100, Greece

²Laboratory of Physical Chemistry and Chemical Processes, School of Chemical and Environmental Engineering, Technical University of Crete, Chania 73100, Greece.

³Department of Materials Science and Engineering, University of Ioannina, 45110 Ioannina, GR

⁴Department of Physics, University of Ioannina, 45110 Ioannina, GR

Presenting author: P. Karga, email: pkarga@tuc.gr

* Corresponding author: I.V. Yentekakis, email: i.yentekakis@ig.forth.gr

ABSTRACT

The catalytic hydrogenation of CO₂ to produce CH₄, also known as CO₂ methanation or the Sabatier reaction ($\text{CO}_2 + 4\text{H}_2 \rightleftharpoons \text{CH}_4 + 2\text{H}_2\text{O}$, $\Delta H_{298\text{K}} = -164 \text{ kJ/mol}$), is an exothermic reaction that is thermodynamically favored at low temperatures (approximately 200–400 °C). However, at lower temperature ranges, the reaction is subject to significant kinetic limitations, making the use of suitable catalytic systems necessary.

Sabatier reaction can be considered an alternative strategy for capturing and recycling emitted CO₂, offering significant potential for reducing fossil fuel dependence and consequently lowering atmospheric CO₂ concentration. Additionally, it can serve as a safe and efficient method for storing and transporting H₂, as part of the Power-to-Gas process. This approach overcomes the safety challenges and high cost associated with H₂ transport produced from renewable energy sources.

In the present study, the effect of the CeO₂ support synthesis method and the resulting nanostructures was investigated. Four different CeO₂ nano-configurations were studied: nanorods (CeO₂-NRs), irregular particles (CeO₂-IRRs), nanoflowers (CeO₂-NFs), and nanospheres (CeO₂-NSs). Specifically, CeO₂-NRs and CeO₂-NFs were synthesized via hydrothermal method, CeO₂-IRRs via precipitation, and CeO₂-NSs via a microwave-assisted hydrothermal method.

The active Ni phase (10 wt % Ni) was deposited onto the supports using the wet impregnation method. The catalytic performance and stability of the above materials were examined over CO₂ methanation reaction through kinetic experiments conducted over a temperature range of 150–600°C, as well as 12-hour stability tests at 380°C. The experimental conditions were as follows: H₂/CO₂ ratio = 4/1, balanced with Ar, with a total flow rate of $F_t = 19 \text{ cc/min}$ and a catalyst mass of $m_{\text{at}} = 60 \text{ mg}$. Additionally, the physicochemical and chemical properties of CeO₂ supports and counterpart catalysts were evaluated using various characterization techniques (BET, XRD, Chem-H₂, H₂-TPR, SEM, and TEM) to establish correlations between their activity and structural features. The supports synthesis method plays a crucial role in the performance of catalytic materials, as it affects the degree of nickel (Ni) dispersion and metal-support interactions. Notably, nanostructured supports like nanorods enhance nickel (Ni) catalysts, achieving up to 90% methane yield, at 355°C. These innovative materials, with specialized nanoconfigurations, offer both exceptional catalytic performance and improved stability, positioning them as promising materials in catalytic technology and methane production processes.

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