

Article

CO₂ Methanation: Solvent-Free Synthesis of Nickel-Containing Catalysts from Complexes with Ethylenediamine

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Abstract: CO₂ methanation was studied in the presence of nickel catalysts obtained by the solid-state combustion method. Complexes with a varying number of ethylenediamine molecules in the coordination sphere of nickel were chosen as the precursors of the active component of the catalysts. Their synthesis was carried out without the use of solvents, which made it possible to avoid the stages of their separation from the solution and the utilization of waste liquids. The composition and structure of the synthesized complexes were confirmed by elemental analysis, IR spectroscopy, powder XRD and XPS methods. It was determined that their thermal decomposition in the combustion wave proceeds in multiple stages with the formation of NiO and Ni(OH)₂, which are reduced to Ni⁰. Higher ethylenediamine content in the complex leads to a higher content of metal in the solid products of combustion. However, different ratios of oxidized and reduced forms of nickel do not affect the initial activation temperature of nickel catalysts in the presence of CO₂. It was noted that, after activation, the sample obtained from [Ni(C₂H₄N₂)₂](NO₃)₂ exhibited the highest activity in CO₂ methanation. Thus, this complex is a promising precursor for CO₂ methanation catalysts, and its synthesis requires only a small amount of ethylenediamine.

Keywords: CO₂ methanation; nickel; ethylenediamine; complex; solvent-free synthesis; solid-state combustion



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1. Introduction

In order to decrease the effect on the environment, new solvent-free methods need to be developed for the synthesis of catalysts, including CO₂ methanation (or CO₂ hydrogenation) catalysts. Currently, interest in this reaction is growing, since it allows a reduction in the carbon footprint of industry and energy while also solving the problem of the chemical storage of hydrogen by producing synthetic natural gas [1–3].



Traditionally, for this process, nickel catalysts are prepared using solvents. In this case, it is necessary to separate the catalyst from the solution and calcinate it at a high temperature to remove the solvent and form the active phase. These stages are common to most methods of CO₂ methanation catalyst synthesis: co-precipitation [4,5], the sol-gel method [6] and incipient wetness impregnation [7,8]. In the method of solid-state combustion (SSC), the use of solvents and the heat treatment of catalysts can be avoided. For this reason, stable nickel complexes containing high-energy ligands and oxidizing anions were used as precursors of active components [9,10]. It is preferable to use nitrogen-rich organic compounds as ligands since their thermal decomposition is accompanied by

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