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Kinetic exploration of CO_2 methanation over nickel loaded on fibrous mesoporous silica nanoparticles (CHE-SM)

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ABSTRACT

A novel series of nickel (Ni) loaded on Fibrous Mesoporous Silica Nanoparticles (CHE-SM) support with varying Ni contents (x=1–30 wt%) were synthesized, denoted as xNi/CHE-SM and then investigated for carbon dioxide (CO_2) methanation. The catalysts underwent comprehensive characterization using XRD, N_2 adsorption-desorption, FESEM, FTIR-KBr, H_2-TPR, and CO_2-TPD techniques. The XRD and FESEM analyses confirmed the structural integrity of CHE-SM, irrespective of the Ni loading. However, the size of the nanocrystalline NiO particles appeared to be influenced by the Ni loading. Notably, 20Ni/CHE-SM exhibited the highest CO_2 conversion of 92% at 350 °C, demonstrating its potential for low-temperature activation. H_2-TPR and CO_2-TPD results revealed favorable NiO reduction at lower temperatures, indicating medium-strength basicity that facilitated efficient CO_2 and H_2 adsorption and activation. Consequently, 20Ni/CHE-SM exhibited superior catalytic performance compared to other catalysts, with lower activation energy (61.5 kJ/mol). Kinetic studies focusing on 20Ni/CHE-SM indicated a molecular adsorption mechanism of CO_2 and H_2 on a single site after evaluation using four Langmuir-Hinshelwood models. This result was attributed to the high amount of medium strength basicity possessed by the 20Ni/CHE-SM catalyst which provided an abundance of adsorption sites, resulting in greater fractional coverage of reactants and enhancing the CH_4 formation rate.

1. Introduction

Our dependency on fossil fuels has brought about an environmental disaster due to rising carbon dioxide (CO_2) emissions, especially in industrial and transportation areas (Mazari et al., 2021; Tian et al., 2023). CO_2 is a major component of greenhouse gases (GHG), with properties that trap heat, causing a rise in temperature and leading to global warming (Tapia et al., 2016; Zhi et al., 2023). This issue has prompted researchers, engineers, and scientists to develop efficient mitigation methods for combating the increasing CO_2 emissions such as CO_2 capture and storage, as well as CO_2 utilization into high value-added products like methanol, methane, and higher hydrocarbons (Kilo, 1997; Omae, 2006; Wu and Chan, 2009; Yamasaki et al., 2006). However, in recent years, the utilization of CO_2 to produce substitute natural gas (SNG) has been witnessed as a lucrative strategy since this reaction is substantially faster than other approaches in thermodynamic aspects (Ocampo et al., 2009; Takht Ravanchi and Sahebdelfar, 2021).

This approach, known as CO_2 methanation, plays a pivotal role in power-to-gas (PtG) technologies, offering an innovative way to utilize fossil-carbon resources by producing SNG. Methane is a significant component of SNG that holds great promise as an energy source due to its high calorific value, leading to reduced CO_2 emissions and preserving

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