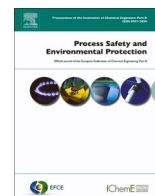




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Kinetic exploration of CO₂ methanation over nickel loaded on fibrous mesoporous silica nanoparticles (CHE-SM)

Muhammad Akmal Aziz^b, Aishah Abdul Jalil^{a,b,*}, Nurul Sahida Hassan^b, Mahadi Bin Bahari^c, Abdul Hakim Hatta^b, Tuan Amran Tuan Abdullah^{a,b}, Nurfatehah Wahyuni Che Jusoh^d, Herma Dina Setiabudi^e, Rajendran Saravanan^f

^a Centre of Hydrogen Energy, Institute of Future Energy, UTM Johor Bahru, Johor 81310, Malaysia

^b Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia, UTM Johor Bahru, Johor 81310, Malaysia

^c Faculty of Science, Universiti Teknologi Malaysia, UTM Johor Bahru, Johor 81310, Malaysia

^d Department of Chemical and Environmental Engineering, Malaysia-Japan International Institute of Technology, Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, Kuala Lumpur 54100, Malaysia

^e Faculty of Chemical and Process Engineering Technology, Universiti Malaysia Pahang, Lebuhr Persiaran Tun Khalil Yaakob, Gambang, Kuantan, Pahang 26300, Malaysia

^f Instituto de Alta Investigación, Universidad de Tarapacá, Arica 1000000, Chile

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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 (CO₂) methanation. The catalysts underwent comprehensive characterization using XRD, N₂ adsorption-desorption, FESEM, FTIR-KBr, H₂-TPR, and CO₂-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 CO₂ conversion of 92% at 350 °C, demonstrating its potential for low-temperature activation. H₂-TPR and CO₂-TPD results revealed favorable NiO reduction at lower temperatures, indicating medium-strength basicity that facilitated efficient CO₂ and H₂ 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 CO₂ and H₂ 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 CH₄ formation rate.

1. Introduction

Our dependency on fossil fuels has brought about an environmental disaster due to rising carbon dioxide (CO₂) emissions, especially in industrial and transportation areas (Mazari et al., 2021; Tian et al., 2023). CO₂ 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 CO₂ emissions such as CO₂ capture and storage, as well as CO₂ 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 CO₂ 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 Sahebdehfar, 2021).

This approach, known as CO₂ 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 CO₂ emissions and preserving

* Corresponding author at: Centre of Hydrogen Energy, Institute of Future Energy, UTM Johor Bahru, Johor 81310, Malaysia.

E-mail address: aishahaj@utm.my (A.A. Jalil).

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