



CO₂ methanation in a bench-scale bubbling fluidized bed reactor using Ni-based catalyst and its exothermic heat transfer analysis

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ARTICLE INFO

Article history:

Received 12 March 2020

Received in revised form

16 September 2020

Accepted 18 September 2020

Available online 22 September 2020

Keywords:

CO₂ methanation

Ni catalyst

Fluidized bed reactor

Heat transfer coefficient

RSM (response surface methodology)

ABSTRACT

CO₂ methanation, as a power-to-gas technology, is considered to be an important method to secure energy supply by utilizing CO₂ and H₂ gases. In this study, a 0.2 kW CH₄ bench-scale fluidized bed reactor was used for CO₂ methanation using approximately 13 kg nickel-based catalyst to investigate the effect of temperature, gas velocity, and H₂/CO₂ ratio on CO₂ conversion, CH₄ purity, and CH₄ selectivity. Response surface methodology (RSM) was employed to design the experimental conditions to statistically evaluate the effect of operating variables. Reduced quadratic model equations for CO₂ conversion and CH₄ purity were derived, which determined the optimal conditions within the experimental conditions. The suggested conditions for the highest CO₂ conversion were 297 °C, 4.66H₂/CO₂, and 4.0 U_g/U_{mf} (velocity ratio), whereas different conditions were determined for the highest CH₄ purity. Among the operating variables, temperature was the most influential factor, followed by the gas ratio. The highest CO₂ conversion and CH₄ purity were 98% and 81.6%, respectively. Additionally, the heat transfer coefficient (h_o) was found to be 115 W/m²·°C during a 10-h continuous CO₂ methanation experiment, which is an important design factor for the further scale-up of the process.

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1. Introduction

Power-to-gas (P2G) technology has received considerable attention for securing the energy supply by utilizing CO₂ gas in flue gas and H₂ from electrolysis of water. The initial report on CO and CO₂ methanation was prepared by Sabatier and Senderens in 1902, who produced natural gas from combustion flue gas [1]. The produced CH₄ gas is often called green methane because of the green methanation reaction [2] which uses a promising hydrogen carrier, produced from renewable solar and wind energy. The advantages of CH₄ for energy storage include 1) high storage capacity, 2) longer discharge time, and 3) safer transportation using existing natural gas infrastructures. Additionally, CO₂ gas from power plants can be utilized, which eventually reduces the severity of global warming.

The CO₂ methanation reaction is an exothermic reaction which

uses H₂ and CO₂ to produce CH₄ and H₂O. Fig. 1 shows the CO₂ conversion and CH₄ purity based on the gas concentrations at equilibrium, calculated using the Gibbs free energy minimization method. A thermodynamic analysis of CO₂ methanation under atmospheric conditions was performed at a temperature of 250 °C–600 °C and H₂/CO₂ ratios of 3.5–4.5. Fig. 1 (a) shows CO₂ conversion with increasing temperature and H₂/CO₂ ratio. A decrease in the temperature and an increase in the gas ratio enhanced CO₂ conversion, whereas a slightly different trend was observed for CH₄ purity under the same conditions. For each gas ratio, the CH₄ purity decreased with increasing temperature because of the lower CO₂ conversion rate as well as the production of H₂ gases over the water-gas shift reaction, as shown in Fig. 1 (c). However, the highest CH₄ purity was obtained with H₂/CO₂ = 4.0 at 250 °C because of the relatively high CO₂ conversion and low concentration of unreacted H₂. At H₂/CO₂ = 4.5, excess H₂ gas was found to reduce CH₄ purity, although CO₂ conversion was approximately 100%, whereas the lower CO₂ conversion efficiency at H₂/CO₂ = 3.5 reduced CH₄ purity. If carbon deposits are considered in the thermodynamic reaction, CO₂ conversion and CH₄ purity would

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