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CO_2 methanation in a bench-scale bubbling fluidized bed reactor using Ni-based catalyst and its exothermic heat transfer analysis

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CO_2 methanation, as a power-to-gas technology, is considered to be an important method to secure energy supply by utilizing CO_2 and H_2 gases. In this study, a 0.2 kW CH_4 bench-scale fluidized bed reactor was used for CO_2 methanation using approximately 13 kg nickel-based catalyst to investigate the effect of temperature, gas velocity, and H_2/CO_2 ratio on CO_2 conversion, CH_4 purity, and CH_4 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 CO_2 conversion and CH_4 purity were derived, which determined the optimal conditions within the experimental conditions. The suggested conditions for the highest CO_2 conversion were 297 °C, 4.66H_2/CO_2, and 4.0 U_g/U_{mf} (velocity ratio), whereas different conditions were determined for the highest CH_4 purity. Among the operating variables, temperature was the most influential factor, followed by the gas ratio. The highest CO_2 conversion and CH_4 purity were 98% and 81.6%, respectively. Additionally, the heat transfer coefficient (h_o) was found to be 115 W/m^2·°C during a 10-h continuous CO_2 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 CO_2 gas in flue gas and H_2 from electrolysis of water. The initial report on CO and CO_2 methanation was prepared by Sabatier and Senderens in 1902, who produced natural gas from combustion flue gas [1]. The produced CH_4 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 CH_4 for energy storage include 1) high storage capacity, 2) longer discharge time, and 3) safer transportation using existing natural gas infrastructures. Additionally, CO_2 gas from power plants can be utilized, which eventually reduces the severity of global warming.

The CO_2 methanation reaction is an exothermic reaction which uses H_2 and CO_2 to produce CH_4 and H_2O. Fig. 1 shows the CO_2 conversion and CH_4 purity based on the gas concentrations at equilibrium, calculated using the Gibbs free energy minimization method. A thermodynamic analysis of CO_2 methanation under atmospheric conditions was performed at a temperature of 250 °C–600 °C and H_2/CO_2 ratios of 3.5–4.5. Fig. 1 (a) shows CO_2 conversion with increasing temperature and H_2/CO_2 ratio. A decrease in the temperature and an increase in the gas ratio enhanced CO_2 conversion, whereas a slightly different trend was observed for CH_4 purity under the same conditions. For each gas ratio, the CH_4 purity decreased with increasing temperature because of the lower CO_2 conversion rate as well as the production of H_2 gases over the water-gas shift reaction, as shown in Fig. 1 (c). However, the highest CH_4 purity was obtained with $H_2/CO_2 = 4.0$ at 250 °C because of the relatively high CO_2 conversion and low concentration of unreacted H_2. At $H_2/CO_2 = 4.5$, excess H_2 gas was found to reduce CH_4 purity, although CO_2 conversion was approximately 100%, whereas the lower CO_2 conversion efficiency at $H_2/CO_2 = 3.5$ reduced CH_4 purity. If carbon deposits are considered in the thermodynamic reaction, CO_2 conversion and CH_4 purity would

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