Thermodynamic and experimental explorations of CO_2 methanation over highly active metal-free fibrous silica-beta zeolite (FS@SiO_2-BEA) of innovative morphology

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ABSTRACT

CO_2 methanation is a novel way for climate change mitigation by converting CO_2 into substitute natural gas. In this study, a highly active fibrous silica-beta zeolite (FS@SiO_2-BEA) catalyst was prepared for CO_2 methanation by a microemulsion process, and examined by N_2 adsorption–desorption, field emission scanning electron microscope (FESEM), transmission electron microscopy (TEM), and electron spin resonance (ESR) spectroscopy techniques. It was found that the FS@SiO_2-BEA catalyst possessed a fibrous silica morphology, leading to high surface area (609 m^2/g), oxygen vacancies, and basicity. A thermodynamic study was also carried out using Gibbs free energy minimization method, and it was found that low temperatures (25–350 °C) and high H_2: $CO_2 \geq 4$ ratios have enhanced the CO_2 methanation activity. The prepared FS@SiO_2-BEA catalyst exhibited high CO_2 conversion (65%), and CH_4 selectivity (61%) with a space–time yield of 3.30 g g_{cat}^{-1} h^{-1}. The obtained experimental results highly followed the thermodynamic calculations.

KEYWORDS

CO_2 methanation; Fibrous silica-beta zeolite; Gibbs free energy minimization; Microemulsion; Thermodynamics; Substitute natural gas

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