



High efficient nickel/vermiculite catalyst prepared via microwave irradiation-assisted synthesis for carbon monoxide methanation



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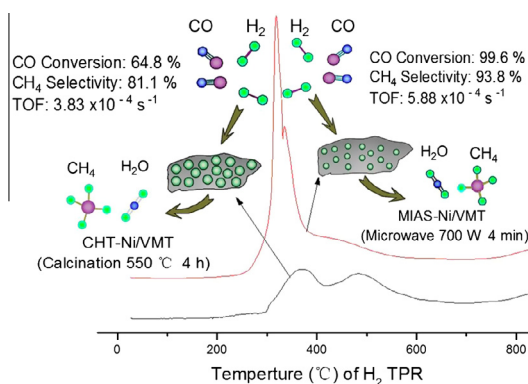
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HIGHLIGHTS

- Microwave irradiation assisted synthesis (MIAS) heats precursor rapidly.
- MIAS provides Ni/VMT catalyst with highly dispersed active points.
- Ni/VMT (MIAS) exhibits excellent CO conversion, CH₄ selectivity, and turn over frequency.

GRAPHICAL ABSTRACT

Compared with conventional heat treatment (CHT), microwave irradiation assisted synthesis (MIAS) heats the Ni/VMT precursor rapidly and endows the Ni/VMT catalyst with highly dispersed active points. Because of the well-dispersed active points, the as-obtained Ni/VMT (MIAS) exhibited excellent catalytic performance, including high CO conversion, CH₄ selectivity, and turn over frequency.



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ABSTRACT

Suitable supports for NiO species and NiO-support synthesis are important in CO methanation. We have used expanded multilayered vermiculite (VMT) as catalyst support and synthesized NiO/VMT composite by microwave irradiation-assisted synthesis (MIAS). Compared with conventional heat treatment (i.e., 550 °C for 4 h), the MIAS method provides rapid heating of the Ni/VMT precursor (i.e., 700 W for 4 min), which results in a Ni/VMT catalyst with highly dispersed active sites. The Ni/VMT(MIAS) exhibits excellent catalysis performance, including a high 99.6% CO conversion, 93.8% CH₄ selectivity, and turn over frequency of $5.88 \times 10^{-4} \text{ s}^{-1}$, at 400 °C, an gas hourly space velocity of 12,000 h⁻¹, a synthesis gas flow rate of 65 mL min⁻¹ and a pressure of 1.5 MPa. The design and fabrication of Ni/VMT(MIAS) may promote the synthesis of VMT-based catalysts with better CO methanation performance based on its outstanding structure and allow for its applications in other areas.

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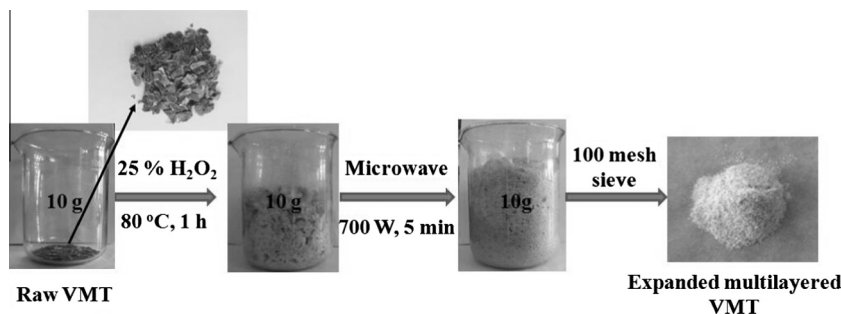


Fig. 1. Schematic illustration of processing steps to prepare expanded VMT and corresponding photos.

1. Introduction

Increasing energy needs and ecological concerns are great challenges of modern society [1,2]. Groundbreaking work by Sabatier and Senderens in 1902 [3] allowed for environmentally benign and economically efficient synthetic natural gas (SNG) to be obtained by the methanation of carbon oxides (CO or CO₂) with hydrogen [4]. Advanced and efficient catalysts are required to extend the usage of SNG energy [5,6]. In general, SiO₂-supported Ni is used as a catalyst in CO methanation (i.e., syngas methanation). Although Ni/SiO₂ exhibits good methanation activities for CO, it also exhibits a weak metal-support interaction, which results in aggregate formation of the active components Ni, carbon deposition, and eventual catalyst deactivation [7]. Although active component Ni nanoparticles can minimize these effects, they may be unstable and become deactivated, which is problematic from an industrial and fundamental points of view [8]. A study on the preparation of desired high-stability and high-mechanical-strength catalyst supports that can be integrated with efficient reactive species to reduce the aggregation and enhance CO methanation is still required [9–11].

Recently, other catalyst supports (such as Al₂O₃, MgO, TiO₂, ZrO₂ and CeO₂) have been introduced as Ni supports in CO methanation [12]. Al₂O₃-supported Ni catalysts show good activity and excellent resistance to carbon deposition [13,14]. However, high calcination temperatures have led to an increase in Ni particle size and a weakened interaction between Ni nanoparticles and Al₂O₃ supports [12]. Hu et al. [15] found that Ni/Al₂O₃ with adding a small amount of MgO (e.g., 2.0 wt.%) could yield a high catalytic activity, catalyst stability and strong resistance to carbon deposition. Compared with Ni/Al₂O₃, Mg–Al oxides-supported Ni catalysts have excellent thermal stability and an improved metal-support interaction [16].

Vermiculite (VMT), is a natural layer-structured clay mineral [17] that consists mainly of SiO₂, MgO and Al₂O₃, and has been used recently as a catalyst support, such as VMT-supported TiO₂ as photocatalyst for methylene blue photocatalytic degradation [18] and water decontamination [19]. In 2013, Liu et al. [20] used modified-VMT supported Ni catalyst for CO₂ reforming of methane to syngas and found that it exhibited potential as a Ni component carrier because of its thermal stability. NiO supported on VMT can be reduced more easily than Al₂O₃-supported NiO catalyst. To date, Ni/VMT has not been reported as a viable catalyst in the reaction of CO methanation. Ni/VMT prepared by conventional heat treatment (CHT, i.e., 550 °C for 4 h) exhibits a low CO conversion and CH₄ selectivity for CO methanation. This occurs because of the large NiO material crystallite size that results from the high temperature and long sintering time used in CHT method. Compared with CHT, advantages of microwave irradiation-assisted synthesis (MIAS) include energy- and time-efficient microwave irradiation, rapid precursor heating and ideal heat treatment for an efficient and eco-

nomous synthesis route [21–23]. MIAS can also help to optimize particle size and improve active component dispersion on catalyst carrier materials [24,25].

Herein, we have synthesized a highly efficient VMT-supported Ni catalyst via MIAS at 700 W for 4 min for CO methanation. The as-obtained MIAS-Ni/VMT exhibited excellent CO conversion and CH₄ selectivity because of excellent Ni nanoparticle size [26–28]. We hope that the MIAS will provide an additional strategy for rapid heat treatment and show potential for the preparation of other catalysts.

2. Experimental

2.1. Preparation of expanded multilayered VMT

Raw VMT (Xinjiang Yuli Xinlong Vermiculite Co., Ltd., China) is composed mainly of 38.76% SiO₂, 24.05% MgO, 20.04% Al₂O₃, 6.22% K₂O, 4.93% Fe₂O₃, 3.03% CaO, 2.02% Na₂O, and 0.94% TiO₂. VMT (10 g) and H₂O₂ (25% concentration, 100 mL) was placed in a 500 mL beaker (see Fig. 1). The mixture of VMT and H₂O₂ was heated at 80 °C for 1 h using a constant temperature water bath, and then dried in a microwave oven at 700 W for 5 min. The dried expanded multilayered VMT was ground and sieved through a 100 mesh sieve. The as-obtained VMT was collected and placed in a glass dryer prior to use.

2.2. Ni/VMT catalyst preparation

The as-obtained VMT was impregnated with an aqueous solution of Ni(NO₃)₂·6H₂O to achieve 10 wt.% Ni loading. The impregnated sample was stirred and soaked at 80 °C for 12 h. The precursor was obtained after the impregnated sample was dried at 110 °C for 12 h and crushed in an agate mortar. The as-prepared precursor was divided into two equal parts. One part was calcined at 550 °C for 4 h using CHT and was termed CHT-NiO/VMT. The other part was treated by microwave irradiation at 700 W for 4 min via MIAS and was termed MIAS-NiO/VMT. Both samples were reduced in H₂ atmosphere at 500 °C for 2 h and were termed CHT-Ni/VMT and MIAS-Ni/VMT, respectively.

2.3. Catalyst characterization

X-ray diffraction (XRD) analysis was carried out on a Bruker D8 Advance X-ray diffractometer with Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$). Scanning electron microscopy (SEM) images were obtained using a Hitachi S-4300 microscope. Transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) images were obtained with a Tecnai F30 field emission transmission electron microscope. The Brunauer–Emmett–Teller (BET) specific surface area and Barrett–Joyner–Halenda (BJH) pore structure were determined at

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