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High-performance Fe_5C_2@CMK-3 nanocatalyst for selective and high-yield production of gasoline-range hydrocarbons

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ABSTRACT

Highly-loaded and well-dispersed Fe_5C_2 nanoparticles within ordered mesoporous carbon CMK-3 (Fe_5C_2@CMK-3) were prepared via a simple melt infiltration method. They were successfully applied to high-temperature Fischer-Tropsch synthesis, and showed high CO conversion (91%) and activity ($5.1 \times 10^{-4}\ mol_{CO}\ g_{Fe}^{-1}\ s^{-1}$) as well as good selectivity (38 wt%) for gasoline-range hydrocarbons (C_5–C_{12}). The catalytic property of Fe_5C_2@CMK-3 was newly interpreted, based on theoretical data obtained by computational simulations.

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1. Introduction

Fischer–Tropsch synthesis (FTS) has been a key technology that can produce a high quality petroleum substitute by coupling CO and H_2 gases derived from fossil resources such as coal, natural gas, and biomass [1,2]. Using this reaction for selective, high-yield production of synthetic fuel has been a significant challenge [3]. Typically, gasoline and lower-olefin products are effectively obtained by high-temperature FTS (HT-FTS) operated at temperatures of 300–350 °C using an iron-based catalyst [4]. Recently, some research has been reported for ways to provide more selective production of lower-olefins (C_2–C_4) [5–7]. However, profound studies and efficient new ways to produce gasoline-range hydrocarbons (C_5–C_{12}), without additional use of zeolite materials as a cracking catalyst, are rare [8].

Until now, iron-based nanocatalysts with carbon support (e.g. activated carbon, charcoal, carbon nanotubes, carbon nanofibers, and graphene) have been used for HT-FTS due to their high specific surface areas and good thermal stability [9–14]. The recent use of ordered mesoporous carbon materials with high surface area, large pore volume and uniform mesopores enabled the uniform dispersion of tiny nanoparticles and efficient mass transfer [15–17].

Generally, alkali metals serving as a base promoter (e.g. Na, K, Cs) have been exploited in order to improve the catalytic activity of the catalysts [18–21]. However, when using a large amount of base promoter, the selectivity and productivity for gasoline-range hydrocarbons tend to decrease, because of the high basicity of the active surfaces formed by the promoters [22]. In the present work, we report Fe_5C_2 nanoparticles (4.6 nm) encapsulated within amorphous carbon CMK-3 (Fe_5C_2@CMK-3) as an efficient catalyst for selective production of gasoline-range hydrocarbons. The catalyst, which was prepared via melt infiltration of hydrated iron nitrate salt and a subsequent thermal activation process pre-optimized by screening various temperatures and gas conditions, showed higher activity and better selectivity for C_5–C_{12} hydrocarbons than those in graphene. Furthermore, the catalytic property of Fe_5C_2@CMK-3 was newly interpreted, based on theoretical data obtained by computational simulations.

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