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Deep eutectic solvent-derived carbon-based FeNi bimetallic catalysts for selective hydrogenation of furfural

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Keywords:
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ABSTRACT

Cheap Ni catalysts have excellent catalytic hydrogenation activity, but also result in low selectivity. Ni-based alloy catalysts can improve performance, but controlling their preparation is a challenge. This paper reports an alloying strategy by using a deep eutectic solvent (DES) as the catalyst precursor, and carbon-based catalysts ($FeNi_4$-NC-D) containing $FeNi_3$ alloys with high specific surface area were prepared by a simple one-step calcination. The necessity of DES for the formation of $FeNi_3$ alloys was demonstrated. In comparison to the inactive of $FeNi_4$-NC and Fe-NC-D, as well as the 60.3 % of $FeNi_2$-NC-D and 25.3 % of Ni-NC-D, the $FeNi_4$-NC-D with dual active sites in the selective hydrogenation of furfural (FF) to furfuryl alcohol (FAL) achieved a yield of 99.9 % at a mild condition of 110 °C. According to serial characterization, the explanation for this difference is that DES not only promotes a substantial increase in specific surface area and pore volume, but also effectively modulates the electronic environment of the carbon substrate in $FeNi_4$-NC-D. More significantly, DES forms a composite structure of Ni and $FeNi_3$ alloy phase in $FeNi_4$-NC-D. The $FeNi_3$ and Ni double sites in this structure markedly enhance the dissociation of H_2. Moreover, the formation and enhancement of Lewis acid-base sites by $FeNi_3$ promotes the activation and adsorption of FF. Meanwhile, DFT calculations further demonstrated that $FeNi_3$ plays an important modulating role as an active site in enhancing the activity, especially in promoting the FAL selectivity. Furthermore, the carbon coated structure resulted from DES could alleviate metal nanoparticle agglomeration and metal leaching, which improved the stability of the catalyst. This work is of great significance for the preparation of low-cost and efficient bimetallic catalysts by DES.

1. Introduction

The use of renewable biomass as feedstock for the production of biofuels and refined chemicals can significantly reduce the use of fossil resources [1–3]. Biomass-derived furfural (FF) has great potential as a material for the production of oils and chemicals [4]. About 65 % of the global FF is used to produce furfuryl alcohol (FAL) [5,6]. FAL performs essential role in the chemical industry as an intermediate for upgrading products [7,8]. Currently, a commercial process for the hydrogenation of FF to FAL has been achieved using Cu-Cr catalysts [9,10]. However, the exploration of high-performance chromium-free catalysts has become a priority due to the serious hazards of Cr to humans and the environment [8,11]. Generally, noble metal catalysts exhibit better catalytic performance in this reaction than non-precious metals [12]. Considering the economic cost [13] and environmental issues [14], non-noble metal catalysts such as Ni, Fe, Co, and Cu may be a better choice even though they have limited catalytic performance [15,16]. Therefore, the development of inexpensive and high-efficiency porous carbon-based non-noble metal catalysts has become an urgent need for the selective hydrogenation of FF.

The hydrogenation activity of inexpensive Ni-based catalysts is particularly remarkable [17–19]. However, because of adsorption modes and thermodynamics, monometallic Ni catalysts are more inclined to C = C hydrogenation than C = O [20,21]. This leads to excessive hydrogenation of FF and reduced the selectivity of the target product [22]. It was proven that the introduction of a second metal to

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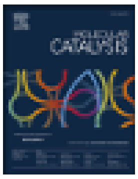

Cu-Doped CoZn@NC promotes catalytic activity of furfural hydrogenation at low temperature: Regulation of copper

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ABSTRACT

The low-cost Cu-based catalysts have excellent selectivity in biomass hydrogenation, but it is challenging to improve their catalytic activity at low temperature. Herein, the trimetallic $Cu_xCoZn@NC$ (where $x = 0.1, 0.3, 0.5$ mmol is the molar mass of Cu) were prepared by in suit pyrolysis using trimesic acid with stronger coordination ability. When it was applied to the furfural (FF) selective hydrogenation process, the catalytic yield of the optimal $Cu_{0.3}CoZn@NC$ reached >99 % under mild condition of 90 °C, which was 29 times superior to that of the CoZn@NC without Cu doping. The results demonstrate that the doping of Cu reduces the activation energy of $Cu_xCoZn@NC$ and increases the catalytic activity at low temperature relative to CoZn@NC. The characterizations and density functional theory (DFT) calculations show that the incorporation of suitable Cu content enhances the interaction between Cu and Co. The amount of surface-active Co^0 and the electron density are significantly enhanced, which effectively stimulates the dissociation of hydrogen. Moreover, the Lewis acid content is significantly increased to facilitate the adsorption and activation of FF. Meanwhile, the impact of Cu content variation on the performance of the multi-metal catalyst is discussed. This work is promising to establish a foundation for the accurate design of polymetallic catalysts through adjusting the Cu content to regulate the reaction activity and selectivity.

1. Introduction

Abundant biomass and its derivatives are renewable resources for the manufacture of high value-added compounds [1]. Furfural (FF) is an essential biomass platform molecule can be derived from the degradation of hemicellulose [2]. Numerous high-value compounds like furoic acid, furfuryl alcohol (FAL), cyclopentanone, furan, etc. can be generated through varying degrees of hydrogenation [3], of which about 65 % of FF is used to produce FAL [4]. In thermodynamics, FF exhibits that C=C addition is easier than C=O [5]. This leads to the generation of tetrahydrofurfuryl alcohol (THFAL), 2-methylfuran (2-MF) and other co-products besides the target product FAL [6]. Therefore, it is difficult to regulate the catalytic selectivity of FF to FAL. Noble-metal catalysts exhibited superior catalytic activity at moderate conditions, but the selectivity is difficult to regulate [7]. In the meantime, noble-metal catalysts with low reserves also have the problem of high cost [8]. Born et al. research reported that when Pd-based catalysts were used for the selective conversion of Furfural, in addition to the target product THFAL, by-products such as FAL and 2-MF were generated [9]. At present, low-cost non-noble metal catalysts including Co, Cu, Fe and Ni were developed [10]. However, non-noble metals have low activity and require harsh conditions like high temperature and reaction pressure to achieve selective hydrogenation of FF [11,12]. Although the improvement of reaction conditions will increase the activity, it will also lead to the decrease of selectivity. Consequently, designing non-noble metal catalysts that can upgrade furfural to furfuryl alcohol with highly selective hydrogenation at low temperatures is a challenge.

Metal doping as a way to further boost the catalytic performance of hydrogenation [13–15]. Through introduced one or more metal components, the catalytic activity and selectivity could be enhanced by adjusting the active sites [16] and the electronic structure through the electronic interactions between different metals [17,18]. For example,

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