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Full Length Article

Steam reforming of acetic acid over Ni/Al_2O_3 catalyst: Correlation of calcination temperature with the interaction of nickel and alumina

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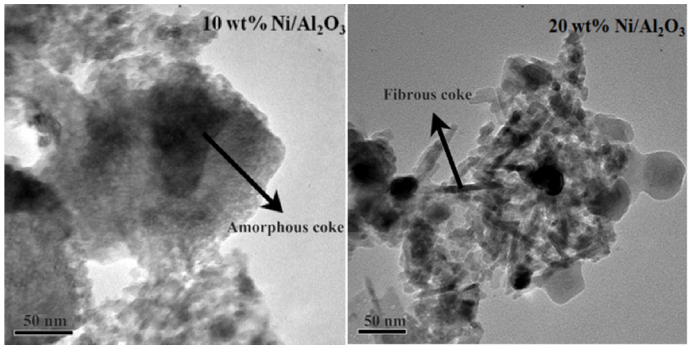
GRAPHICAL ABSTRACT



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ABSTRACT

This study investigated the interaction of nickel species with alumina versus calcination temperature and nickel loading. A total of 22 Ni/Al_2O_3 catalysts, calcined at the temperature from 500 to 1000 °C at a 50 °C increment, were employed for the study. High calcination temperature led to the formation of nickel–alumina spinel via the solid phase reaction, which shifted the reduction temperature to higher ranges. The catalyst, however, still achieved good activity after full reduction of the nickel–alumina spinel. Nevertheless, the high calcination temperature led to the collapse of the small pores and the formation of the big one, resulting in the significant decrease of specific surface area. At higher nickel loadings, more nickel species weakly interacted with alumina formed as the reactive center of alumina was saturated. With the increase of nickel loading, the catalytic activities were not varied much but the catalytic stability and the resistivity towards coking enhanced. The coke produced over the catalyst at low nickel loading tended to be amorphous, while the coke produced at the high nickel loading was more fibrous. Furthermore, the coke produced at the low loading contained more small aromatic rings and more oxygen-containing functional groups. The higher nickel loading possibly promoted the catalytic cracking reactions to form more catalytic coke while the low nickel loading probably favored the polymerization reactions to form the polymeric coke. In addition, the calcination at the higher temperature could enhance the stability of the catalysts, which might be related to the enlarged pore sizes.

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1. Introduction

Nickel–based catalysts have many applications in various catalytic reactions such as steam reforming, hydrogenation, methanation and etc. [1–5]. In some cases unsupported nickel such as Raney Ni is used as the catalyst, while more frequently nickel is supported on a support for the use as a catalyst [6,7]. The use of a support instead of using pure nickel is more advantageous. For example, a support with a high surface area could enhance the dispersion of nickel which consequently decreases the usage of nickel and might reduce the cost of catalyst manufacturing. In addition, some supports are not inert materials, which could involve and play a role in the catalytic reactions [8–12]. One example is Pt/ZrO_2, a bifunctional catalyst used for steam reforming of acetic acid [13]. Pt mainly activate acetic acid while ZrO_2 helps to activate steam for the reforming reactions. The support interacts with nickel catalyst, which significantly affect the catalytic properties [14–16].

One typical example is nickel supported on alumina. The surface of alumina (the gamma form) has some acidity and the functionality such as hydroxyl group, which could have strong metal–support interactions [17–19] At elevated temperatures such as above 800 °C, alumina and nickel could react, forming nickel aluminum spinel ($NiAl_2O_4$) [20]. The formation of $NiAl_2O_4$ significantly affect the behaviors of nickel species as $NiAl_2O_4$ is much more difficult to be reduced than nickel oxides. The interaction of nickel with alumina is a well–known phenomenon [21]. However, how does the essential parameters for preparation of the Ni/Al_2O_3 catalyst affect the extent of the interaction between nickel and alumina needs further investigation. Understanding this could help to optimize the parameters for preparation of the Ni/Al_2O_3 catalyst for maximizing its performances for catalytic reactions, which could also provide useful information for preparation of other heterogeneous supported catalysts.

In this study, the effects of nickel loading (10 wt% and 20 wt%) and the calcination temperatures (from 500 to 1000 °C with an increment of 50 °C) on the interaction of nickel species with the alumina carrier were investigated. The catalysts prepared were characterized in detail and evaluated in steam reforming of acetic acid. The selection of steam reforming of acetic acid as a probe reaction is due to the fact that acetic

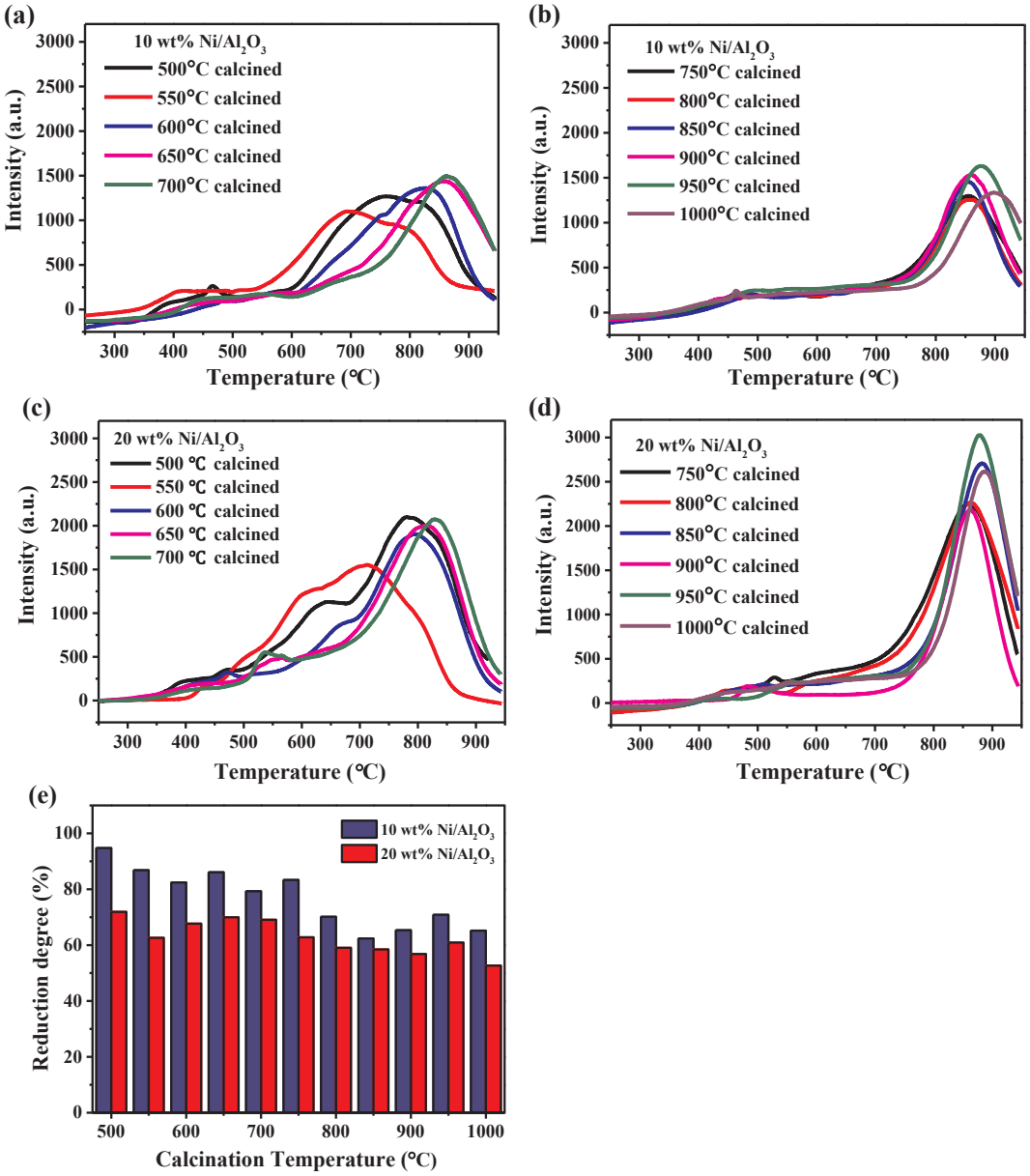

Fig. 1. H_2–TPR and reduction degree for the Ni/Al_2O_3 catalysts with different calcination temperature and nickel loadings.