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Selective hydrogenation of acetylene in excess ethylene over SiO_2 supported Au–Ag bimetallic catalyst

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ABSTRACT

Supported gold nanocatalysts have been reported to be active in selective hydrogenation of acetylene. In this work, SiO_2 supported Au–Ag bimetallic catalyst is studied in the selective hydrogenation of acetylene in excess ethylene. Au and Ag were reductively deposited on a silica surface functionalized by APTES (3-aminopropyltriethoxysilane). They form Au–Ag alloy nanoparticles of very small size. The catalytic activity of Au–Ag bimetallic system showed better catalytic activity at high temperature than that of monometallic gold catalyst. According to the TEM and XRD results, Ag stabilized the nanoparticles against sintering during high temperature calcinations. Non-thermal O_2 plasma was applied to remove the APTES under mild conditions instead of high temperature calcination. The results showed that the conversion of acetylene was much higher over Au–Ag/SiO_2 catalyst pretreated by O_2 plasma than that of pretreated by calcination at 500 °C, although the latter catalyst had similar particle size.

1. Introduction

During the production of ethylene by naphtha cracking, it is always accompanied by impurity of acetylene which is detrimental to the catalytic ethylene polymerization processes. Selective removal of the acetylene in the ethylene stream is therefore highly desirable for the industrial process [1,2]. Pd-based catalysts are typically applied to the selective hydrogenation. However, they suffer from poor selectivity of acetylene conversion to ethylene which leads to production of a large amount of ethane and causes waste of the ethylene [3,4]. To modulate the selectivity and activity of Pd-based catalysts, recent trend has been to find a second metal to form alloy or core-shell structure with Pd for increasing the selectivity [5–7]. Nonetheless, high selectivity together with high conversion for hydrogenation of acetylene is still a challenge.

Supported gold nanaocatalyts have attracted tremendous interests, after the pioneering work of Haruta et al. [8] and Hutchings [9], in a variety of redox reactions. Among these, gold-based nanocatalysts have been applied to selective hydrogenation reactions [10–12]. The selectivity of acetylene over gold catalysts is very high due to the unique properties of gold in having much stronger adsorption toward acetylene than olefin molecules [13,14]. But the conversion is relatively low at low temperatures [15,16]. It has been reported that the catalytic performance of gold catalyst is sensitive to the particle size and the optimum size is about 3 nm [17]. Therefore, it would be interesting to increase the conversion of acetylene over supported gold catalysts; one way is to prepare bimetallic systems and the other way is to synthesize highly dispersed gold nanoparticles. It has been reported that Au–Pd [18] and Au–Ni [19] bimetallic catalysts exhibited higher acetylene conversion than the monometallic gold system. However, Au–Ag system has not been investigated in selective hydrogenation of acetylene before, especially in excess ethylene which is close to the industrial conditions. Silver has been added as a promoter for Pd-based catalysts to increase the selectivity of acetylene hydrogenation at the expense of some conversion [7,11,20]. Silver was not recognized as a good candidate for improving the conversion of acetylene. Nevertheless, it is shown in our previous work that there was a strong synergetic effect between gold and silver in the CO oxidation or preferential CO oxidation in rich H_2 [21]. Silver was found to prevent sintering of gold-based nanoparticles under high temperature calcinations, so it is anticipated that it may help to maintain the optimum particle sizes of Au particles at about 3 nm for acetylene hydrogenation. Although unsupported silver is not known for interacting with hydrogen, it was reported that silica-supported silver can activate H_2 at moderate temperature [22] and to catalyze the acetylene hydrogenation [23] and hydrogenation of chloronitrobenzenes [24]. Silver supported on alumina was also known to

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