Facile synthesis of palladium nanoparticles decorated nitrogen doped graphene and their catalytic study for acetylene hydrogenation in excess ethylene

Firozeh mansourkhani[a], Alireza Badiei[a], Ali Morad Rashidi[b], Saeid Khajehmandali[c]

a school of chemistry, college of science, University of Tehran, Enghelab Ave, Tehran, Iran
b Research Institute of Petroleum Industry, Tehran, Iran.
c Research and Development Department, Shazand Petrochemical Company, Arak, Iran

f.mansourkhani@ut.ac.ir

Abstract

In this research, a palladium nanocatalyst was synthesized over a nitrogen doped graphene support and then applied for selective hydrogenation of acetylene in an ethylene-rich flow stream. The N-doped graphene domain is synthesized through co-growth of naphtalene and urea by chemical vapor deposition (CVD). The nitrogen content of N-doped graphene was further investigated by CHN analysis. The total N content is 4.8 %. Nitrogen doping of graphene can influence the growth kinetics of metal nanoparticles that lead to their small particle size, uniform dispersion, and different morphology. Also, nitrogen doping of the support materials can give better chemical binding between support and metal nanoparticles resulting enhanced stability [1].

Pd nanoparticles are decorated over nitrogen doped graphene (Pd/N-G) by kinetically controlling the polyol reduction process [2-3]. The kinetic control of the growth of the nanoparticles and nitrogen doping of the supporting material leads to the formation of highly dispersed anisotropic nanoparticles over the graphene support [4]. TEM images of Pd/N-G shows that Pd nanoparticles are uniformly dispersed over nitrogen doped graphene with high dispersion and narrow size distribution (6-8 nm).

Performance tests of Pd/NG nanocatalyst were conducted for selective hydrogenation of acetylene from the ethylene rich stream(contains 88, 10, 1.4, 0.8 Vol % for ethylene, ethane, hydrogen and acetylene, respectively) at 60–200 °C. Pd/FG nanocatalyst showed excellent ethylene conversion and selectivity and also, its selectivity increased with both temperature and acetylene conversion. Figure 1 shows the acetylene conversion and ethylene selectivity versus temperature during the reaction. The maximum selectivity to ethylene (89.5%) and yield (83.5%) were observed at 120 °C. These excellent catalytic properties may be attributed to the formation of highly dispersed Pd nanoparticles on the support, increasing electron density on supported palladium and modifying metal/support interactions, which could enhance the ethylene desorption and also decrease the consecutive hydrogenation to ethane and green oil formation. While alumina is common commercial support for Pd catalysts, remarkable improvements in selectivity could be made by the use of Nitrogen doped graphene supports and the superior catalytic properties of the Pd/NG nanocatalyst are promising for the development of new catalytic technologies for the selective hydrogenation processes.

References

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Figures

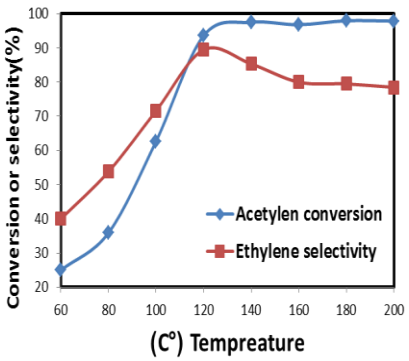

Fig. 1. Acetylene conversion and ethylene selectivity for Pd/NG catalyst