Tuning the Particle size in Supported Au-Pd Alloy Nanoparticles for Catalytic evaluation

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Introduction

The development of methods capable of synthesizing highly monodispersed alloy nanoparticles of controlled size is of great importance and challenging in the field of nanoscience [1]. Particles of the sub 10 nm regime shows potential catalytic activity in the oxidation reactions namely benzyl alcohol oxidation [2], toluene oxidation [3]. In this present study, we demonstrate the effect of calcination using TiO_2 supported Au-Pd alloy nanoparticles as it affects the particle size within 10 nm range.

Experimental

To an aqueous $PdCl_2$ and $HAuCl_4$ solution of the desired concentration, the required amount of a PVA solution (1 wt %) was added (PVA/ (Au+Pd) (w/w) = 1.2); a freshly prepared solution of NaBH4 (0.1 M, $NaBH_4$/ (Au +Pd)(mol/mol)=5) was then added to form a dark-brown sol. After 30 min of sol generation, the colloid was immobilized by adding TiO_2 (acidified at pH 1 by sulphuric acid) under vigorous stirring conditions. The amount of support material required was calculated to have a total final metal loading of 1 wt%. After 2 h, the slurry was filtered and the catalyst was washed thoroughly with distilled water (neutral mother liquors) and dried at 120 °C overnight. Monometallic gold catalysts were also prepared using a similar methodology. The calcined catalysts were pre-treated at 200-400 ^oC under static air for 3h using a heating rate 5 °C/min.These catalysts were tested for benzyl alcohol and glycerol oxidation in a stirred reactor (100 mL, Parr reactor) and 50 mL glass/autoclave reactor respectively. For the analysis of the products a GC (a Varian star 3400 cx with a 30 m CP-Wax 52 CB column) and HPLC (Metacarb 67H column) were employed respectively.

Results/Discussion

The catalytic results show that the dried catalyst is more active than the calcined catalysts. We attributed this catalytic performance due to increase of particle size with the temperature increment during the heat treament. The catalytic activity drops gradually with the increase in particle size whereas selectivity to the oxygenated products is affected. The TEM images reveal that the particle size increases progressively with the calcination temperature. As a result of this, we see decrease in activity with increase in particle size. Furthermore, characterization using XPS has shown Pd enrichment with the increase of calcination temperature. The proper tuning of particle size via calcination can improve the selectivity to the desired product

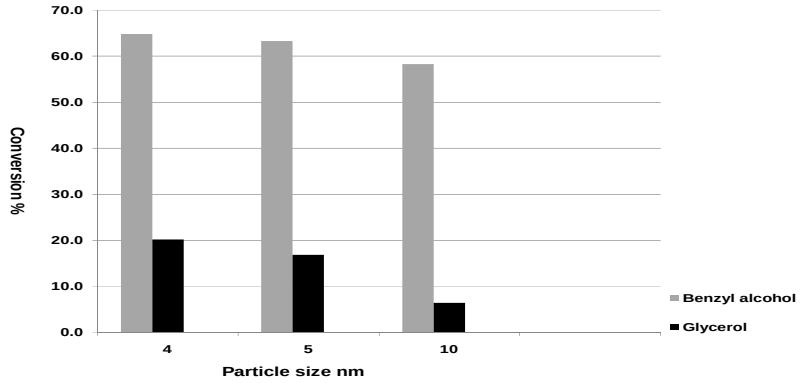

Figure 1: Activity plot against particle size

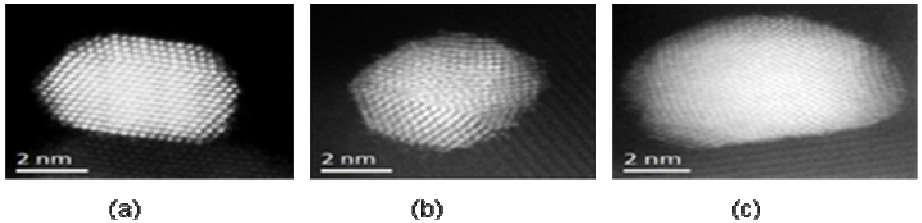

Figure 2: HAADF STEM images of 1%Au Pd/TiO_2 catalyst (a) Dried at 120 °C (b) Calcined at 200 °C (c) Calcined at 400 °C

Reference:

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