Investigating the effect of gold-palladium bimetallic nanoparticles on TiO_2 and the catalytic activity in CO oxidation.

By

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DECLARATION

I declare that the study titled "Investigating the effect of gold-palladium bimetallic nanoparticles on TiO_2 and the catalytic activity in CO oxidation is my own, unaided work submitted for the degree of Master of Science in the University of the Witwatersrand, Johannesburg. It has not been submitted for any degree or examination in any other University, and all sources used or quoted here have been indicated and acknowledged by means of references.

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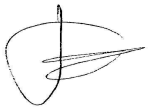
Signature: 

Abstract

In recent years, studies have shown that supported Au catalysts have high activity for CO oxidation at low or ambient temperatures. However, the activity of these catalysts is dependent on a lot of synthesis conditions as reproducibility of small sized gold particles is hard. In this study supported Au catalysts were prepared via deposition precipitation-method (DP). The small sized Au particles were supported on TiO_2 (P25). The suitable synthesis conditions such as pH, aging, metal loading and catalyst pre-treatment were investigated in order to obtain optimum synthesis conditions. The catalysts were characterized with TEM, XRD, and HRTEM. It was found that 3.7 nm Au particles were best synthesized when Au metal loading is 3% at pH 8 and aged for 72 h. The suitable calcination temperature was 200 °C. It was found that the Au particle size was 4.5 nm when Au was supported on SiO_2 thus making TiO_2 a suitable support.

Bimetallic catalyst was synthesized via DP where Pd metal was incorporated as the second metal. It was found that the type of bimetallic formed was heterostructed where both metals where separately attached on the support. The interatomic distance measured from HRTEM results confirmed that both metal were individually attached on the support. XRD results showed that there was no Au-Pd alloy phase or PdO confirming that the Pd metal on the support was indeed in metallic form.

Carbon monoxide (CO) oxidation reactions were undertaken in a tubular glass flow reactor. The monometallic Au catalyst showed superior activity at 200 °C with almost 100% CO conversion. It was also observed that the activity of these catalyst decreased as temperature increased. The CO-TPD studies showed that as temperature increased there was a low CO adsorption due to a decrease in adsorption sites. Varying Pd composition in the bimetallic catalyst did not enhance catalytic activity. However, 25Au75Pd catalyst showed a better conversion as compared to other Au-Pd catalysts. Temperatures studies on bimetallic catalysts showed that as temperature increased there was a decrease in activity. The observed decrease could be attributed to catalyst formation of large particle aggregates. It was also

assumed that the low activity was due to how these catalysts were prepared as there was no surfactant utilized during preparation.

DEDICATION

TO MY LATE MOTHER THAKANE SHEILA NTHOLENG

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ABBREVIATIONS

UNFCCC	united nations framework convention on climate change
GHG	greenhouse gases
TWC	three-way catalyst
DOC	diesel oxidation catalyst
CO	carbon monoxide
CO_2	carbon dioxide
PGM	platinum group metal
TCD	thermal conductivity detector
TPD	temperature programmed desorption
TEM	transmission electron microscopy
MTPs	multiply twinned particles
STM	scanning tunnelling miscrosopy
LBH	local barrier height
ER	eley rideal
DFT	density functional theory
HOMO	high occupied molecular orbital
XPS	x-ray photoelectron spectroscopy
TPR	temperature programmed reduction
IEP	isoeletric point
ALD	atomic layer deposition
$HAuCl_4$	chloroauric acid; hydrogen tetrachloroaurate
WGS	water gas shift
VA	vinyl acaetate
DP	deposition precipitation method
HRTEM	high resolution electron microscopy
ICP-OES	inductively coupled plasma-optical emission spectrometer
XRD	x-ray diffraction
CVD	chemical vapour deposition
Wt %	weight percentage

GC	gas chromatography