Supporting Information

Multifunctional supported bimetallic catalysts for a cascade reaction with hydrogen auto transfer: synthesis of 4-phenylbutan-2-ones from 4-methoxybenzyl alcohols

Moataz Morad,[a,b] Ewa Nowicka,[a] Mark Douthwaite,[a] Sarwat Iqbal,[a] Peter Miedziak,[a] Jennifer K. Edwards,[a] Gemma L. Brett,[a] Qian He,[a] David Morgan,[a] Hamed Alshammari,[a,c] Donald Bethell,[a] David W. Knight,[a] Meenakshisundaram Sankar[a]* and Graham J. Hutchings[a]**

[a]*Cardiff Catalysis Institute, School of Chemistry, Cardiff University, CF10 3AT Cardiff, United Kingdom.*

[b]*Chemistry Department Faculty of Science, Umm al-qura University, PO Box 21955, 9264 Makkah, Saudi Arabia.*

[c]*Chemistry Department, Faculty of Science, Ha'il University, P. Box 2440, 81451 Ha'il, Saudi Arabia.*

** Dr. Meenakshisundaram Sankar, Cardiff Catalysis Institute, School of Chemistry, Cardiff University, Cardiff, CF10 3AT (UK), Fax: (+44) 2920-874-030, E-mail: sankar@cardiff.ac.uk.*

*** Prof Graham J. Hutchings, Cardiff Catalysis Institute, School of Chemistry, Cardiff University, Cardiff, CF10 3AT (UK), Fax: (+44) 2920-874-030, E-mail: hutch@cardiff.ac.uk*

Table S1. *Synthesis of 4-(4-methoxyphenyl)but-3-ene-2-one (**3**) from 4-methoxybenzaldehyde (**2**) and acetone using different metal oxide catalysts.*

Catalyst	**Conversion (%)**	**Product Selectivity (%)[£]**	
		3	**1**
No Catalyst	-	-	-
MgO Fresh	<1	-	99
MgO Calcined	<1	-	99
ZnO Fresh	<1	-	99
ZnO Calcined	<1	-	99
La_2O_3 – Fresh	<1	16	-
La_2O_3 – Calcined	<1	18	43
Al_2O_3 - Fresh	23	84	-
Al_2O_3 - Calcined	13	80	-
TiO_2 - Fresh	20	98	-
TiO_2 - Calcined	33	98	-

Reaction conditions: *4-methoxybenzaldehyde: 1.3g; acetone: 6.3g; mesitylene (internal standard): 1g; catalyst: 0.5g, T: 348K; time: 1h. [a]: Selectivity is calculated based known and identified products and we have used a normalisation method based on these products. The C balance for these reactions were calculated to be between 70-80%. Remaining products are unidentified (20-30%).*

Table S2. *Synthesis of 4-(4-methoxyphenyl)butan-2one* ***(4)*** *from 4-methoxybenzaldeyde* ***(2)*** *and acetone using supported metal bi-functional catalysts*

Catalyst	**Conv**	**Selectivity (%)** [a]				
	(%)	**1**	**3**	**4**	**5**	**6**
1%Pd/TiO_2	16	9	21	68	1	0
5%Pd/TiO_2	100	1	0	0	0	99
1%AuPd/TiO_2	63	62	1	16	4	16
1%AuPd/Al_2O_3	7	67	3	27	0	2
1%AuPd/MgO	100	40	0	58	2	0
0.5%AuPd/MgO	100	31	0	61	7	0
0.5%Au/MgO	90	1	98	1	0	0
0.5%Pd/MgO	100	20	0	74	5	0

Reaction conditions: *4-methoxybenzaldehyde: 1.3g; acetone: 6.3g; mesitylene (internal standard): 1g; catalyst: 0.5g, pH_2: 5 bar T: 348K; time: 1h. [a]: Selectivity is calculated based known and identified products and we have used a normalisation method based on these products. The C balance for these reactions were calculated to be between 70-80%. Remaining products are unidentified (20-30%).*

Table S3. *MP-AES analysis: the total metal weight loading of the fresh 1% AuPd/MgO catalyst.*

Total Metal Loading (wt. %)	Metal Loading (wt. %)		Molar Ratio (%)	
	Au	Pd	Au	Pd
1.12	0.69	0.43	46.4	53.6

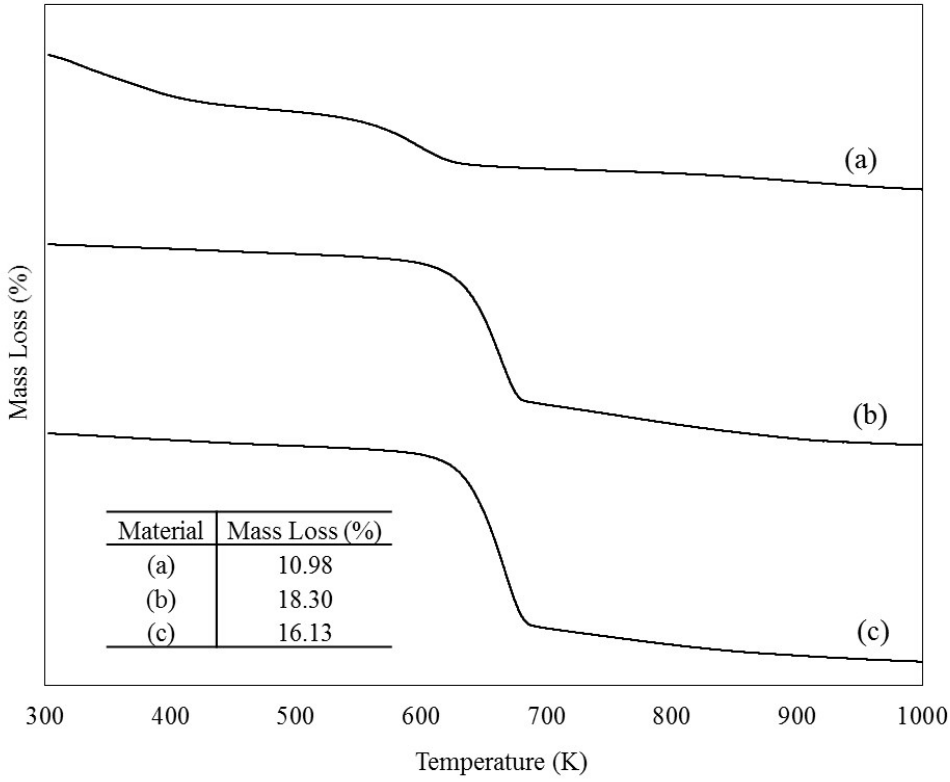

Figure S1. *Thermogravimetric analysis traces corresponding to (a) AuPd/MgO catalyst prior to reaction, (b) the used AuPd/MgO catalyst after the reaction with 4-methoxybenzyl alcohol under N_2 and (c) the used catalyst after the reaction with 4-methoxybenzyl alcohol under H_2.*

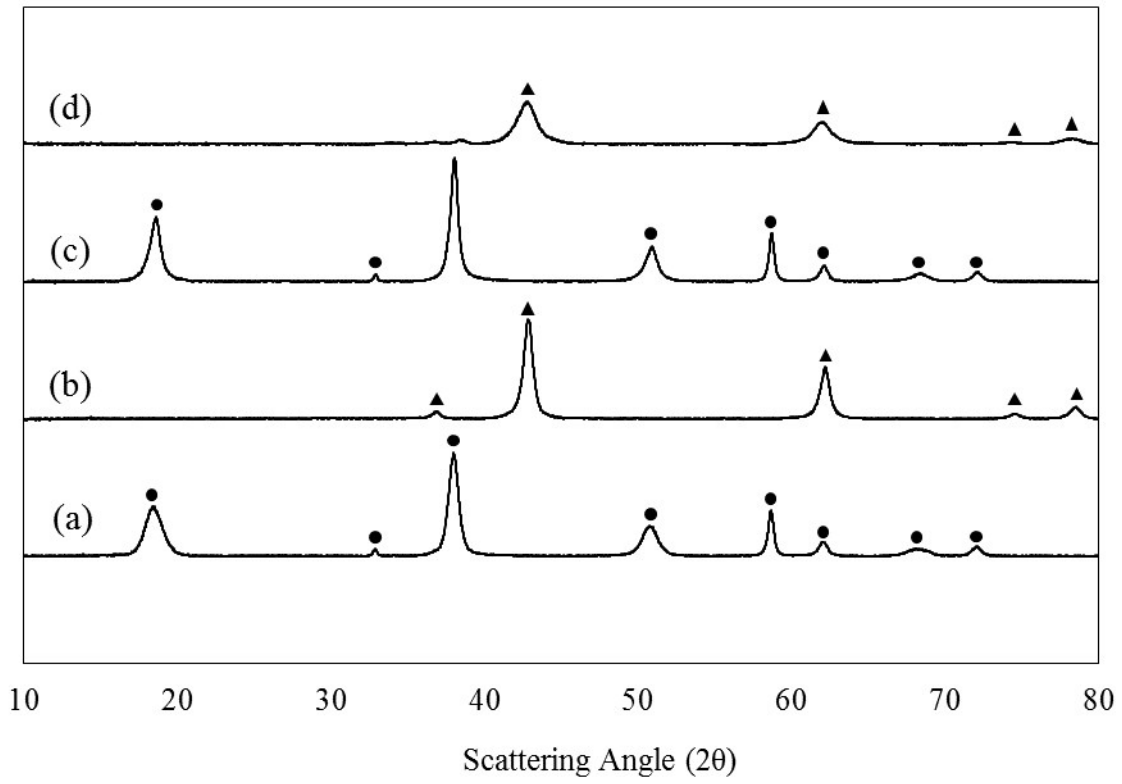

Figure S2. *XRD data corresponding to (a) the $Mg(OH)_2$ support, (b) the MgO support formed from the $Mg(OH)_2$ after the first heat treatment, (c) sol-immobilised 1% $AuPd/Mg(OH)_2$, (d) sol immobilised 1% AuPd/MgO after heat treatment under N_2. ●: $Mg(OH)_2$, ▲: MgO phase.*

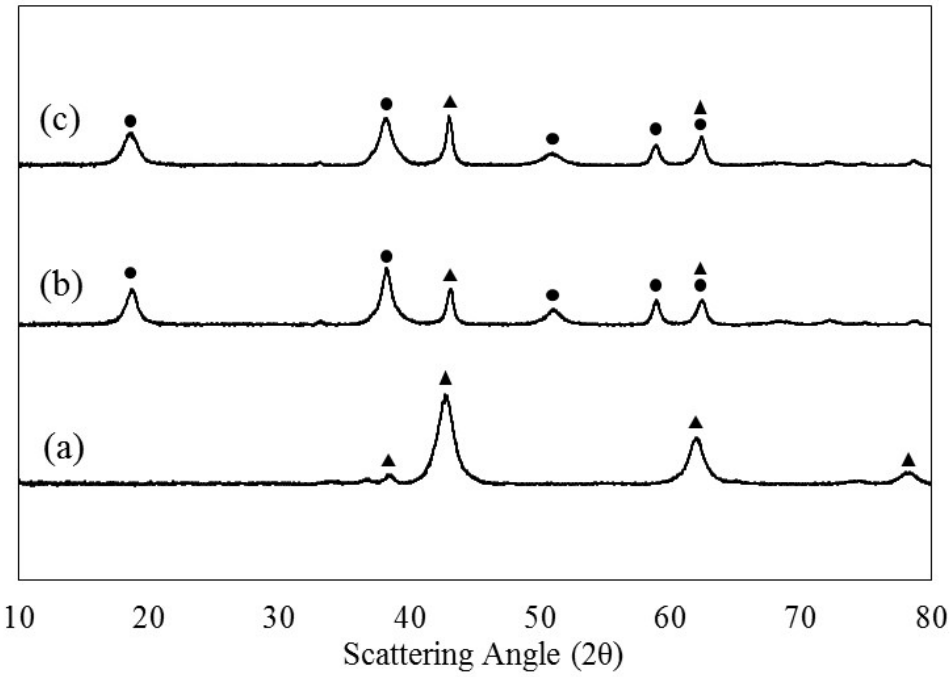

Figure S3. *XRD data corresponding to (a) AuPd/MgO catalyst prior to reaction, (b) the used AuPd/MgO catalyst after the reaction with 4-methoxybenzyl alcohol under H_2 and (c) the used catalyst after the reaction with 4-methoxybenzyl alcohol under N_2. ●: $Mg(OH)_2$ phase; ▲: MgO phase*