

Table S1. Activity and selectivity data for POM reaction over CeO₂-ZrO₂ support, Au/ZrO₂, Au/CeO₂-ZrO₂ and Au/CeO₂-TiO₂-ZrO₂ catalysts at different temperatures ranges for O₂ : CH₃OH molar feed ratio 0.5 and GHSV =30000 h⁻¹ with methanol flow rate 0.52 cm³ hr⁻¹.

Catalysts	Conversion& selectivity	Temperature (°C)							
		275	300	325	350	375	400	425	450
CeO ₂ -ZrO ₂	O ₂ Conversion	1.79	7.14	14.29	32.14	1.43	91.07	99.86	100
	CH ₃ OH Conversion	2.75	3.89	7.89	15.91	45.08	51.37	61.4	64.87
	H ₂ Selectivity	0	0	0	0	4.59	10.52	16.46	21.69
	CO Selectivity	0	0	0	31.79	35.49	48.22	58.29	66.19
Au/ZrO ₂	O ₂ Conversion	7.22	17.59	32.41	59.6	75.93	85.56	91.85	96.3
	CH ₃ OH Conversion	12.77	18.5	33.41	44.88	48.8	49.86	57.04	65.99
	H ₂ Selectivity	0	0	0	0	0	4.17	8.66	15.95
	CO Selectivity	0	0	0	9.52	24.84	41.62	57.08	68.24
Au/CeO ₂ -ZrO ₂	O ₂ Conversion	99.8	99.8	100	100	100	100	100	100
	CH ₃ OH Conversion	49.1	58.9	70	81.4	91.6	94.3	97	98.79
	H ₂ Selectivity	31.6	44.8	57.1	68.7	75.5	68.1	63.1	57.8
	CO Selectivity	7.5	9.9	16.3	19.5	22.3	42.7	58.5	62.17
	Carbon balance	94.7	99.5	97.5	99.5	98.7	96.5	97.8	96.3
Au/CeO ₂ -ZrO ₂ -TiO ₂	O ₂ Conversion	99.8	99.8	99.8	100	100	100	100	100
	CH ₃ OH Conversion	44.7	51.4	57.4	69.4	82.2	93.6	99.2	99.44
	H ₂ Selectivity	6	14.5	37.6	46.2	59.8	67.8	70.6	66.6
	CO Selectivity	1.3	3.2	7.7	13.4	21	32.3	45	57.13
	Carbon balance	94.7	92.9	97.5	94.2	92.3	93.0	93.7	96.9

Supplementary calculation for mole balance

Carbon balance (For Au/CeZrO_x catalyst at 375 °C)

CH₃OH moles IN = 0.00021485 moles/min

O₂ moles IN = 0.000112303 moles/min

Feed ratio O₂:CH₃OH = 0.491

Reactor outlet mole balance:

Exit flow rate 50 cc/min measured by bubble flow meter.

The product mole flow rate were calculated using GC response factor to evaluate the mole fraction for each element then multiplied by the total exit flow rate. Ideal gas rate law was assumed in all the calculations.

CH₃OH moles out = 0.000018009 moles/min

O₂ moles out = 0.0 moles/min

H₂ moles out = 0.00029727 moles/min

CO moles out = 0.0000479 moles/min

CO₂ moles out = 0.000146257 moles/min

$$\text{Conversion of CH}_3\text{OH} = \frac{\text{Reacted moles of CH}_3\text{OH}}{\text{inlet moles of CH}_3\text{OH}} \times 100\% = 91.62\%$$

Selectivity of H₂

$$= \frac{\text{moles of H}_2\text{ produced}}{\text{moles of CH}_3\text{OH reacted} \times 2} \times 100\% = \frac{0.00029727}{(0.00021485 - 0.000018009) \times 2} \times 100\% = 75.5\%$$

Carbon balance:

Moles of carbon IN = 0.00021485 moles/min

Moles of carbon out = 0.000018009 + 0.0000479 + 0.000146257
= 0.000212166 moles/min

$$\text{Carbon balance} = \frac{\text{outlet moles of carbon}}{\text{Inlet moles of carbon}} \times 100\% = \frac{0.000212166}{0.00021485} \times 100\% = 98.7\%$$

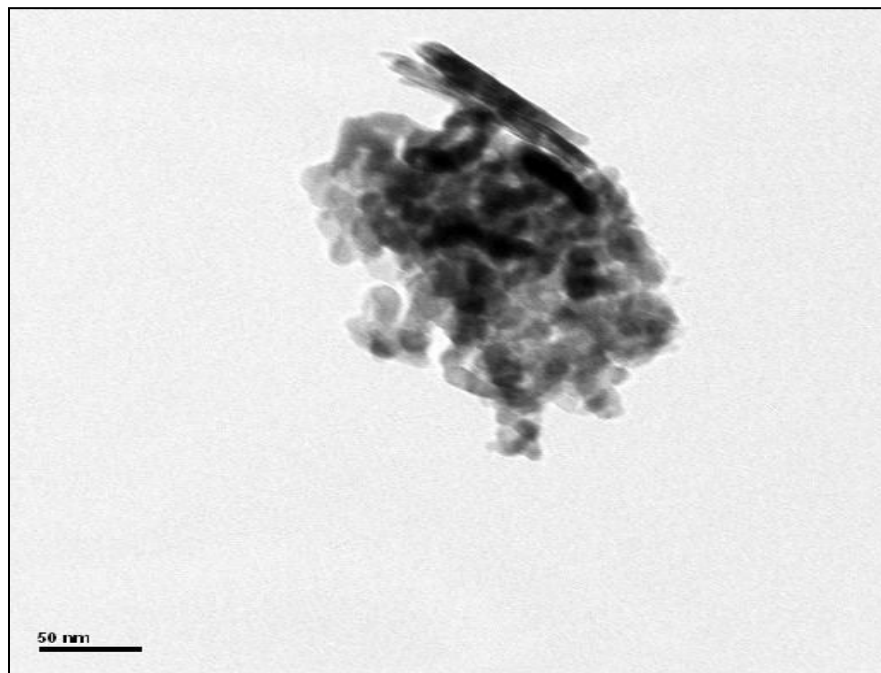
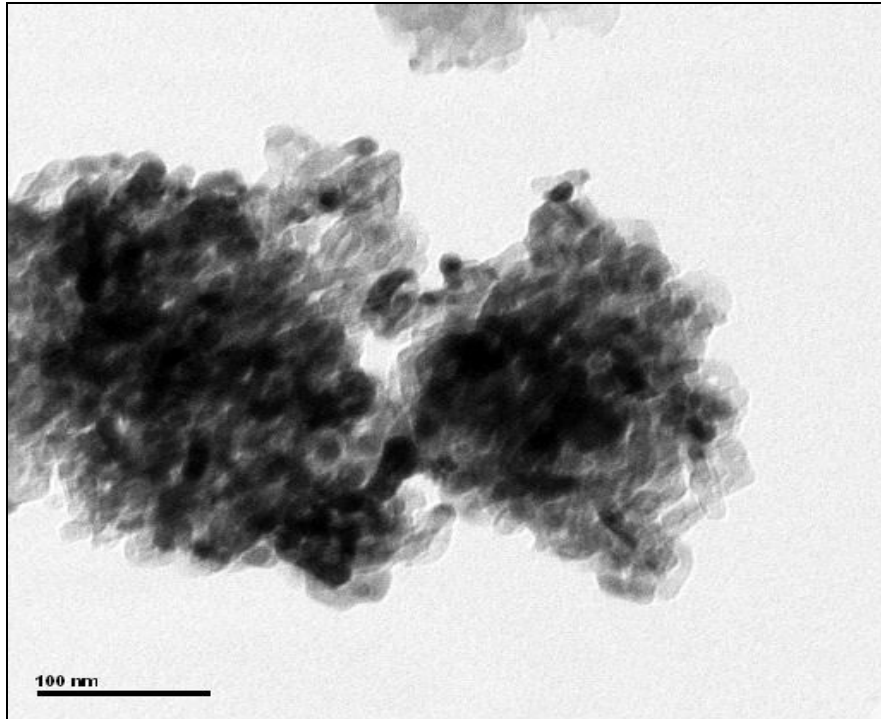


Figure S1. TEM (235KX magnification) show that the powders essentially comprised of finely divided material in the form of clusters. Each cluster consists of entangled tape-like structures, each strand being 5-10nm in width. Hence the overall material is very porous.