

Supporting Information

**Hierarchically nanostructured  $\text{MnCo}_2\text{O}_4$  as active catalysts for synthesis of *N*-benzylideneaniline from benzyl alcohol and aniline**

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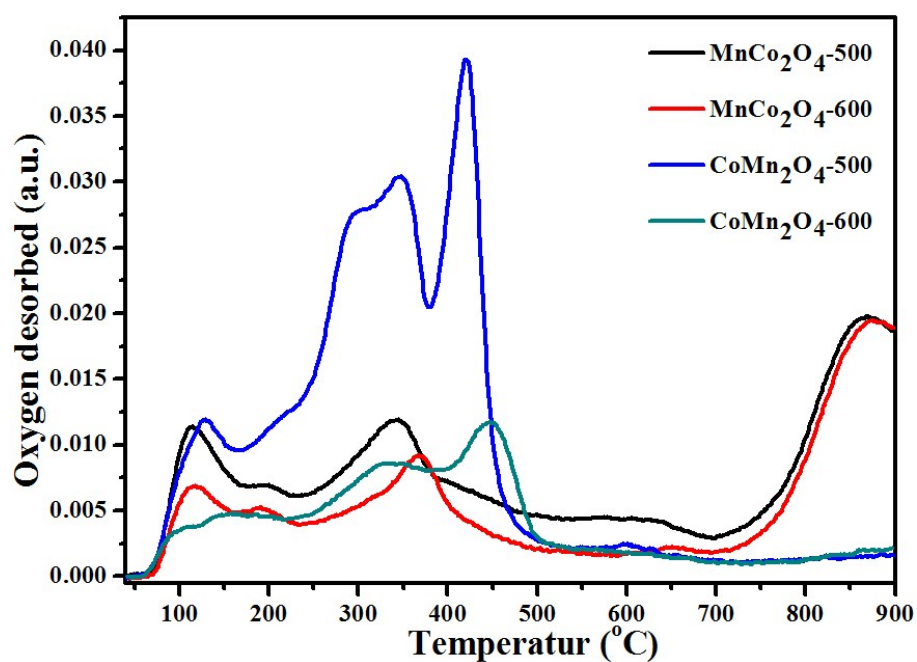
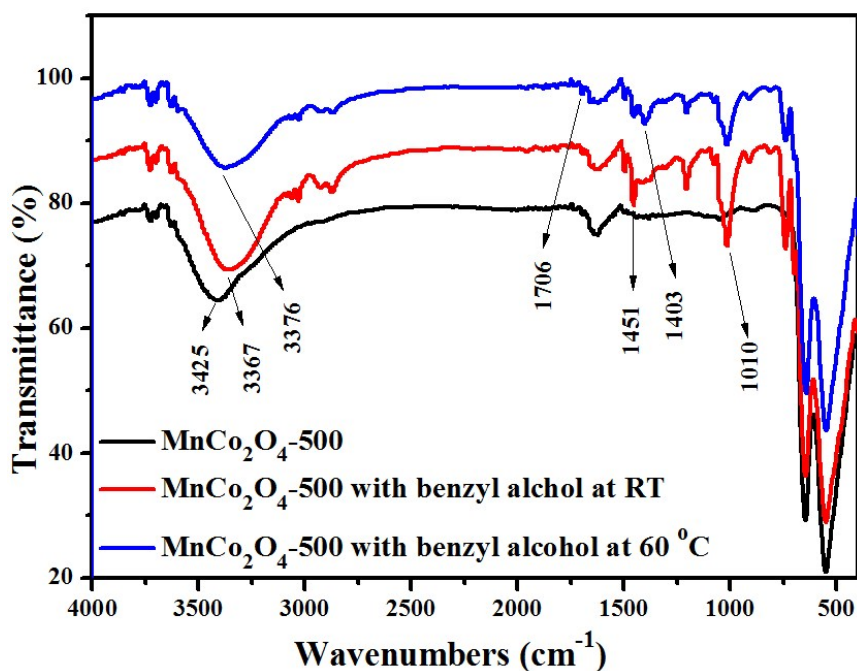
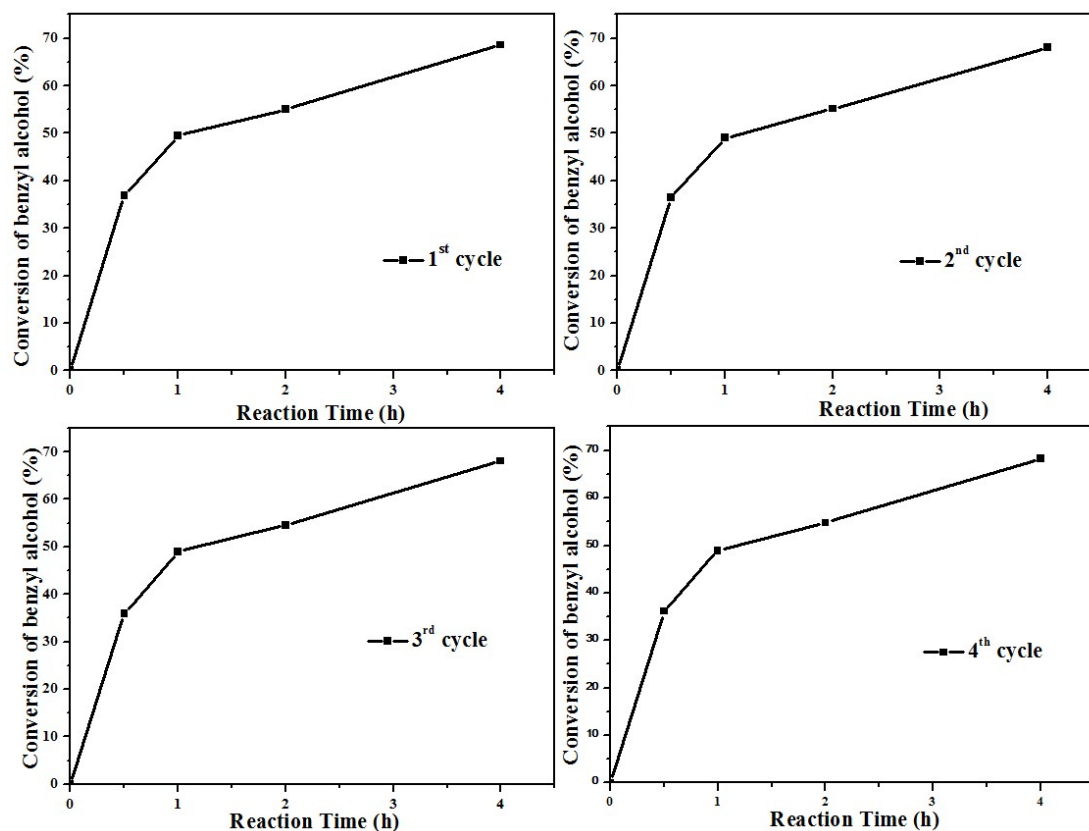


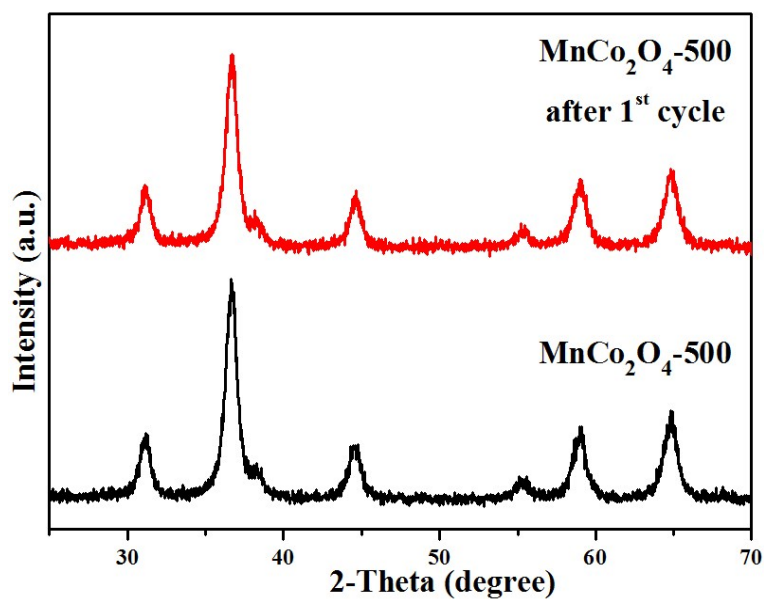
Figure S1. O<sub>2</sub>-TPD profiles of as-prepared catalysts.



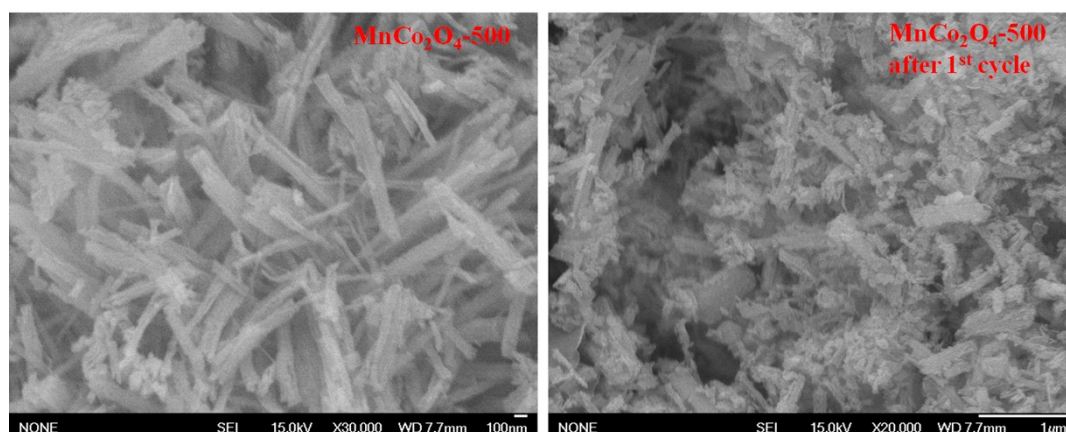
**Figure S2.** FT-IR spectrum of MnCo<sub>2</sub>O<sub>4</sub>-500, MnCo<sub>2</sub>O<sub>4</sub>-500 with benzyl alcohol at room temperature, MnCo<sub>2</sub>O<sub>4</sub>-500 with benzyl alcohol after heat treatment at 60 °C under air for 1 h.



**Figure S3.** Recycle test of MnCo<sub>2</sub>O<sub>4</sub>-500 at 60 °C under 1 atm O<sub>2</sub> for 4 h. The catalyst after each cycle was collected by centrifugation and washing with ethanol three times to remove the products, and dry at 80 °C for 12 h, followed by calcination at 400 °C in air for 4 h. After this treatment, the obtained catalyst was tested for the second time reaction.



**Figure S4.** XRD patterns of fresh  $\text{MnCo}_2\text{O}_4\text{-500}$  and after 1<sup>st</sup> catalytic cycle.



**Figure S5.** SEM images of fresh  $\text{MnCo}_2\text{O}_4\text{-500}$  and after 1<sup>st</sup> catalytic cycle.

**Table S1.** Comparison of catalytic activity of  $\text{MnCo}_2\text{O}_4\text{-500}$  and reported catalysts in the synthesis of *N*-benzylideneaniline from benzyl alcohol and aniline.

catalyst	Amount (mg)	BA (mmol)	Aniline (mmol)	gas	T (°C)	Time (h)	Con. (%)	Sel. (%)	Ref.
$\text{MnCo}_2\text{O}_4\text{-500}$	35	0.5	1.0	air	80	12	93.9	92.8	this work
$\text{CeO}_2$	50	1.0	2.0	air	60	24	99	97	Ref. 1
K-OSM-2	50	1.0	2.0	air	110	24	100	99	Ref. 2

#### References

- Masazumi Tamura, Keiichi Tomishige, Redox Properties of  $\text{CeO}_2$  at Low Temperature: The Direct Synthesis of Imines from Alcohol and Amine, *Angew. Chem. Int. Ed.* **2015**, 54, 864 – 867.
- Shanthakumar Sithambaram, Ranjit Kumar, Young-Chan Son, Steven L. Suib, Tandem catalysis: Direct catalytic synthesis of imines from alcohols using manganese octahedral molecular sieves, *Journal of Catalysis*, 2008, 253, 269–277.