# Supporting Information

## Selective oxidation of alcohols by graphene-like carbon with electrophilic oxygen and integrated pyridinic nitrogen active sites

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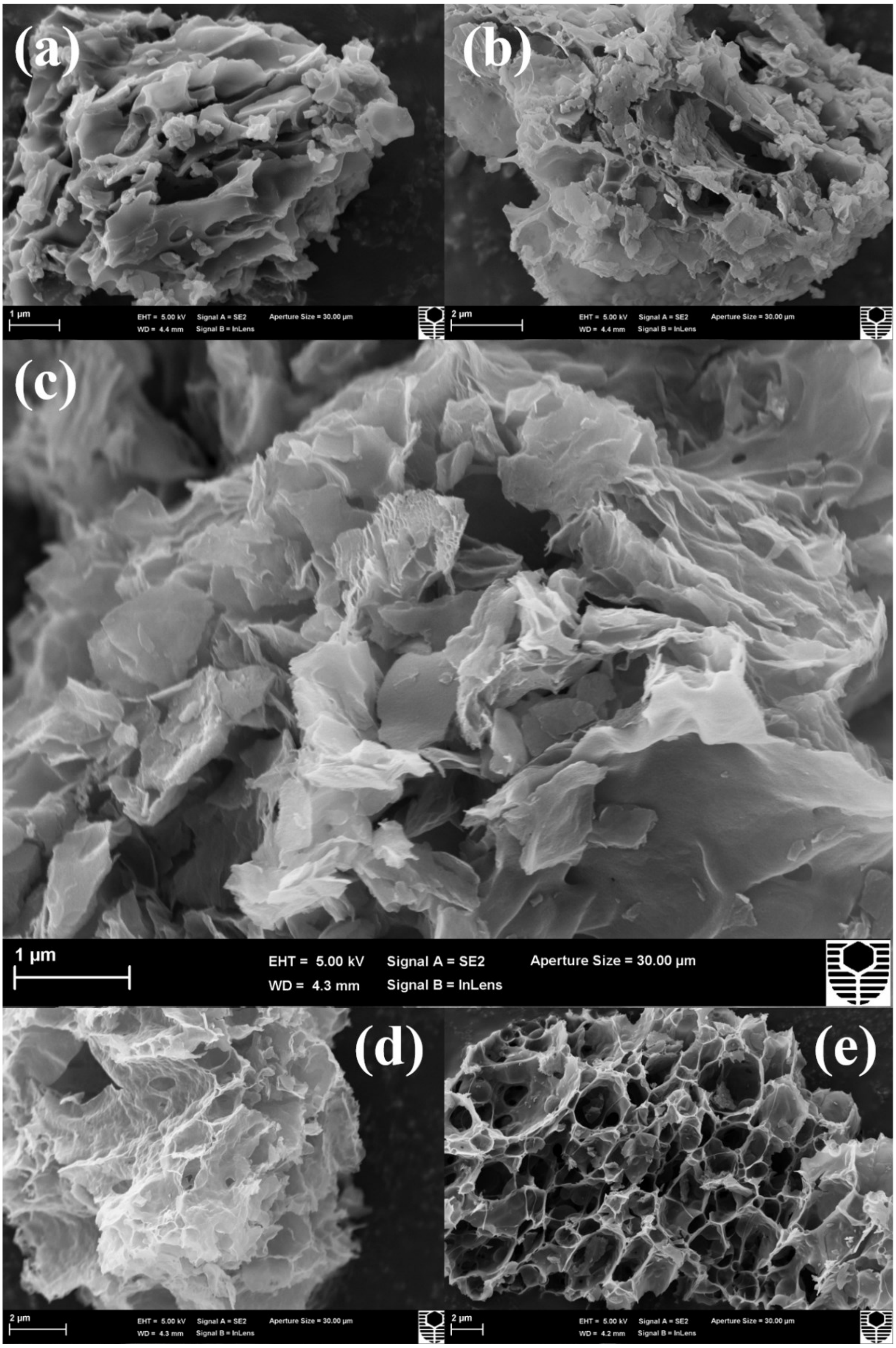

**Fig. S1.** SEM images of (a) NG-600, (b) NG-700, (c) NG-800, (d) NG-900 and (e) NG-1000.

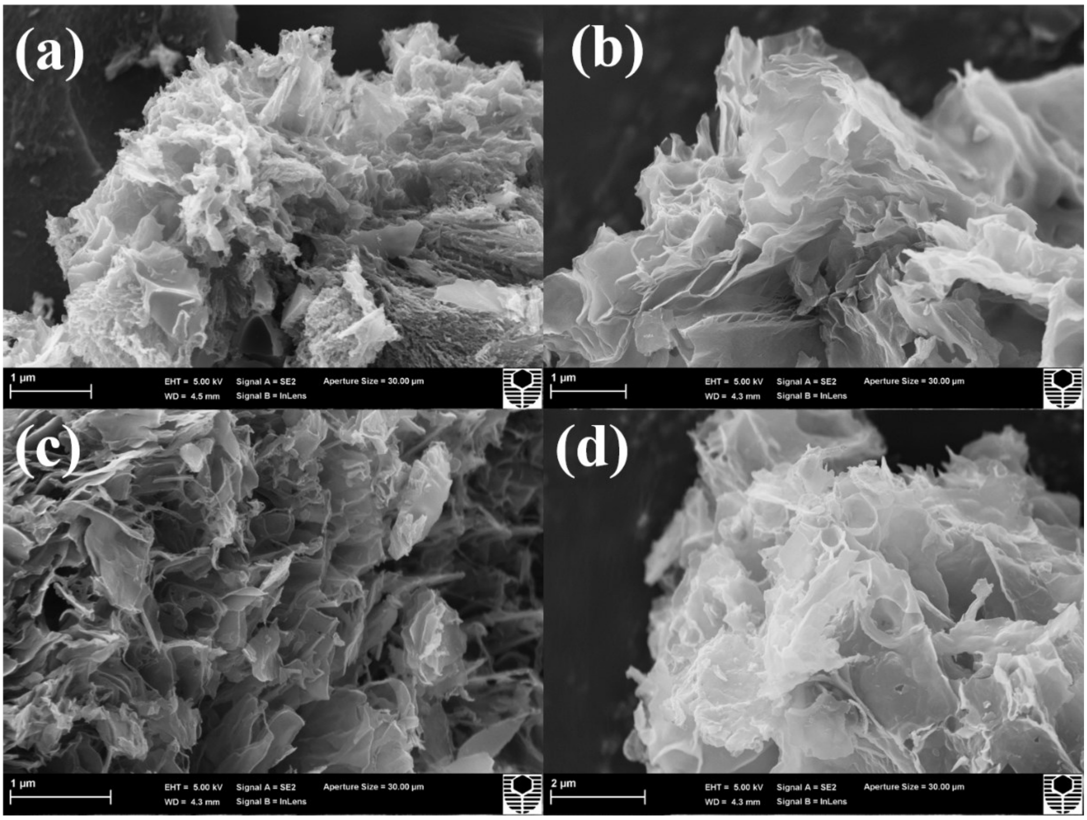


**Fig. S2.** SEM images of (a) NG(blank), (b) NG(AN), (c) NG(ACl) and (d) NG(MgN)

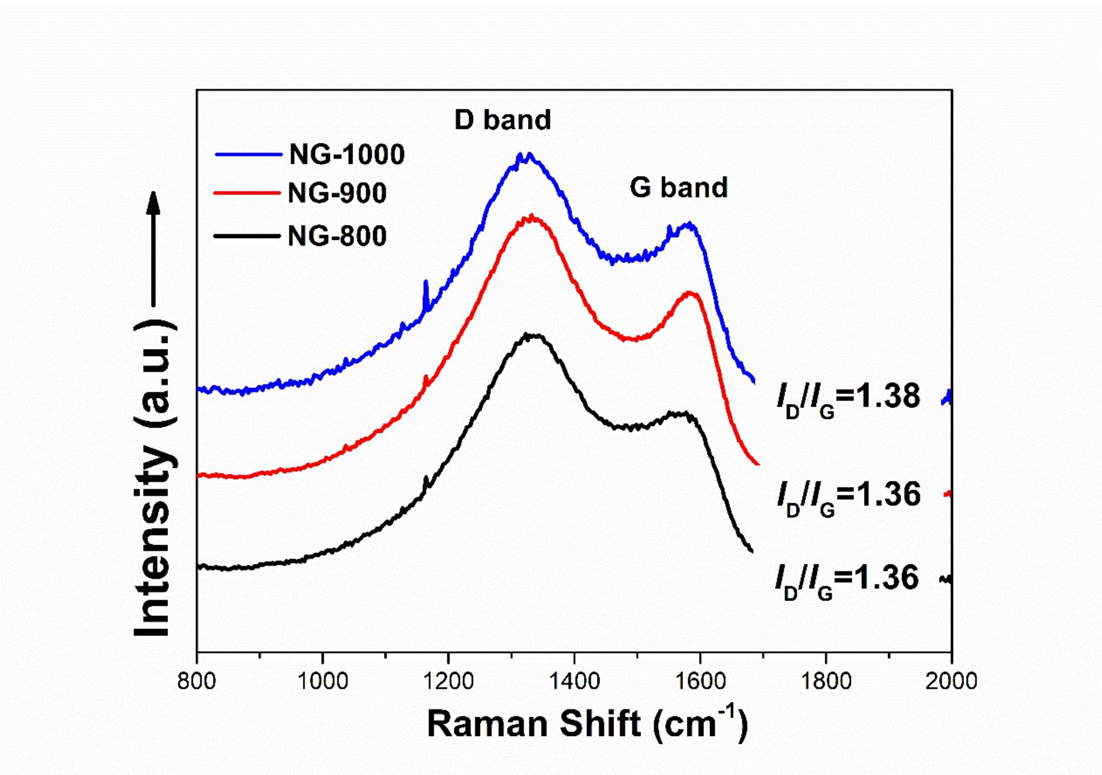


**Fig. S3.** Raman spectra of NG samples.

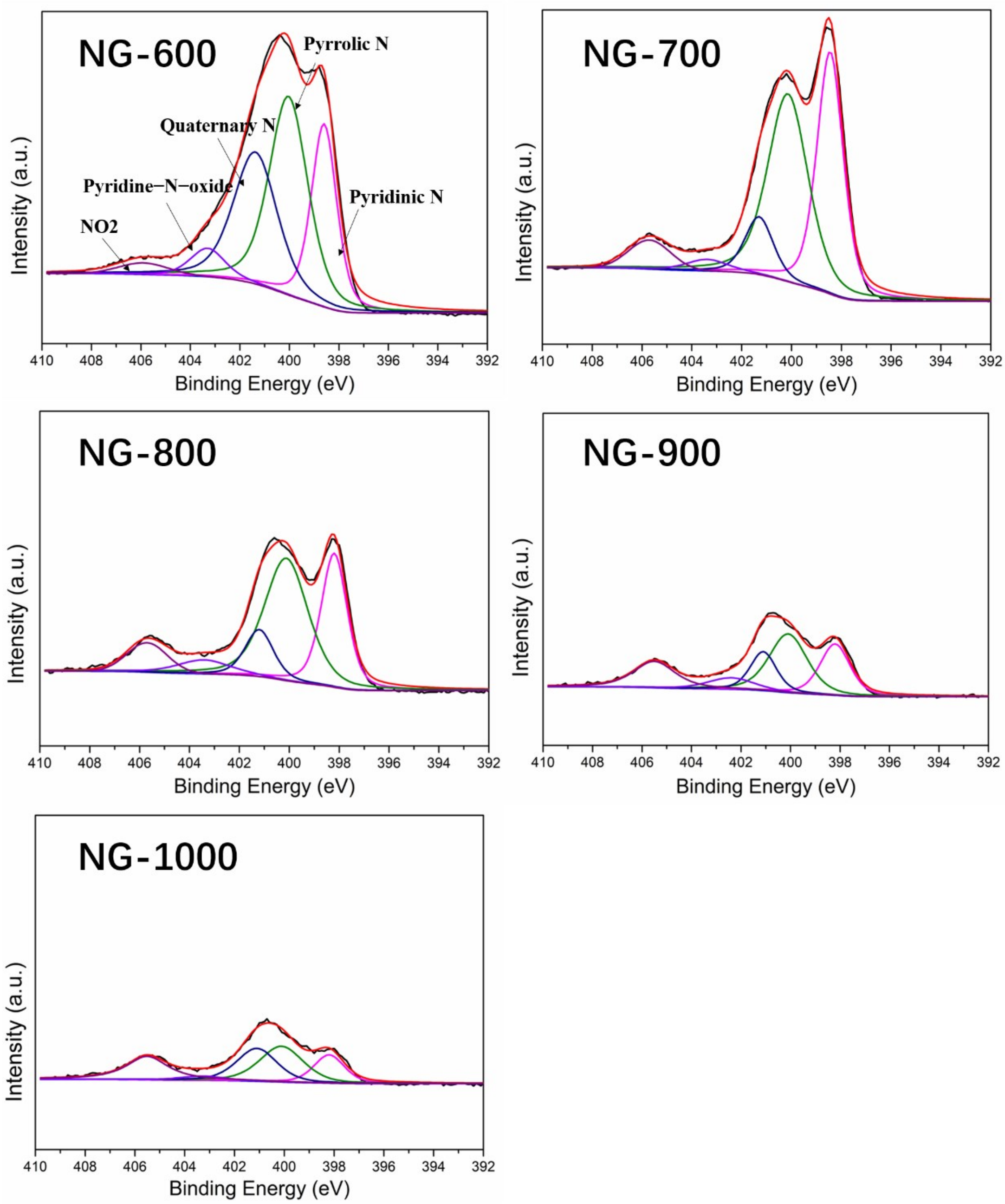


**Fig. S4.** Deconvolution of N1s XPS spectra of NG-600, NG-700, NG-800, NG-900 and NG-1000.

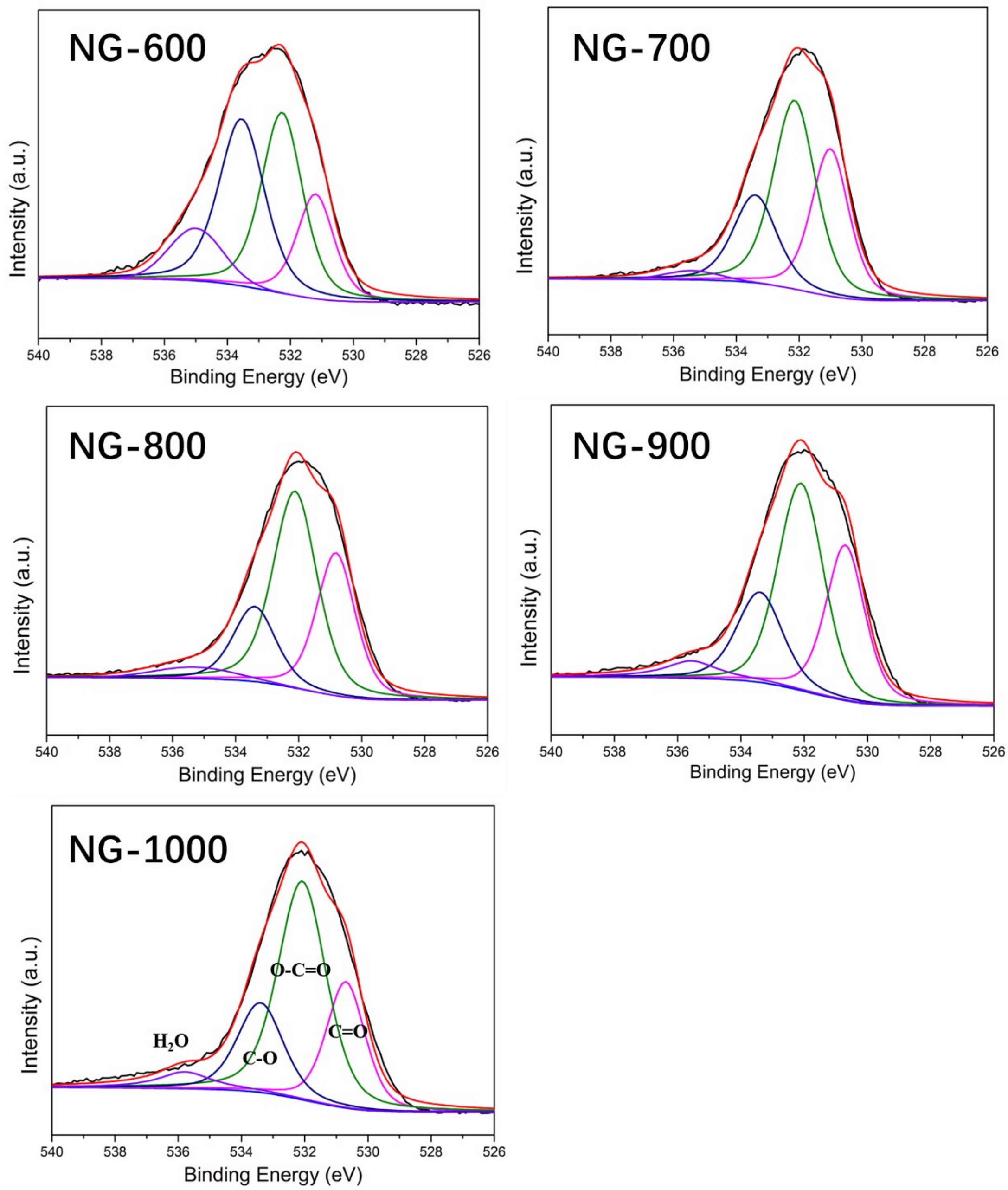


**Fig. S5.** Deconvolution of O1s XPS spectra of NG-600, NG-700, NG-800, NG-900 and NG-1000.

**Table S1.** Composition of O and N species on NG-600, NG-700, NG-800, NG-900 and NG-1000 derived from deconvolution of XPS spectra.[a]

| Samples | Pyridinic N | Pyrrolic N | Quaternary N | Pyridine−N−oxide | $NO_2$ | C=O | O=C−O | C−O | $H_2O$ |
|---|---|---|---|---|---|---|---|---|---|
| NG-600 | 5.5 | 9.1 | 6.9 | 1.2 | 0.6 | 1.7 | 3.2 | 3.3 | 1.0 |
| NG-700 | 6.7 | 7.9 | 1.7 | 0.5 | 1.1 | 3.1 | 4.6 | 2.2 | 0.2 |
| NG-800 | 3.6 | 5.6 | 1.4 | 0.8 | 1.0 | 2.8 | 4.4 | 1.6 | 0.4 |
| NG-900 | 1.7 | 2.5 | 1.1 | 0.6 | 1.3 | 2.9 | 4.3 | 2.0 | 0.6 |
| NG-1000 | 0.8 | 1.5 | 1.2 | 0.2 | 1.1 | 2.1 | 4.5 | 1.7 | 0.5 |

[a] The content of each N or O component is provided in atomic percentage.

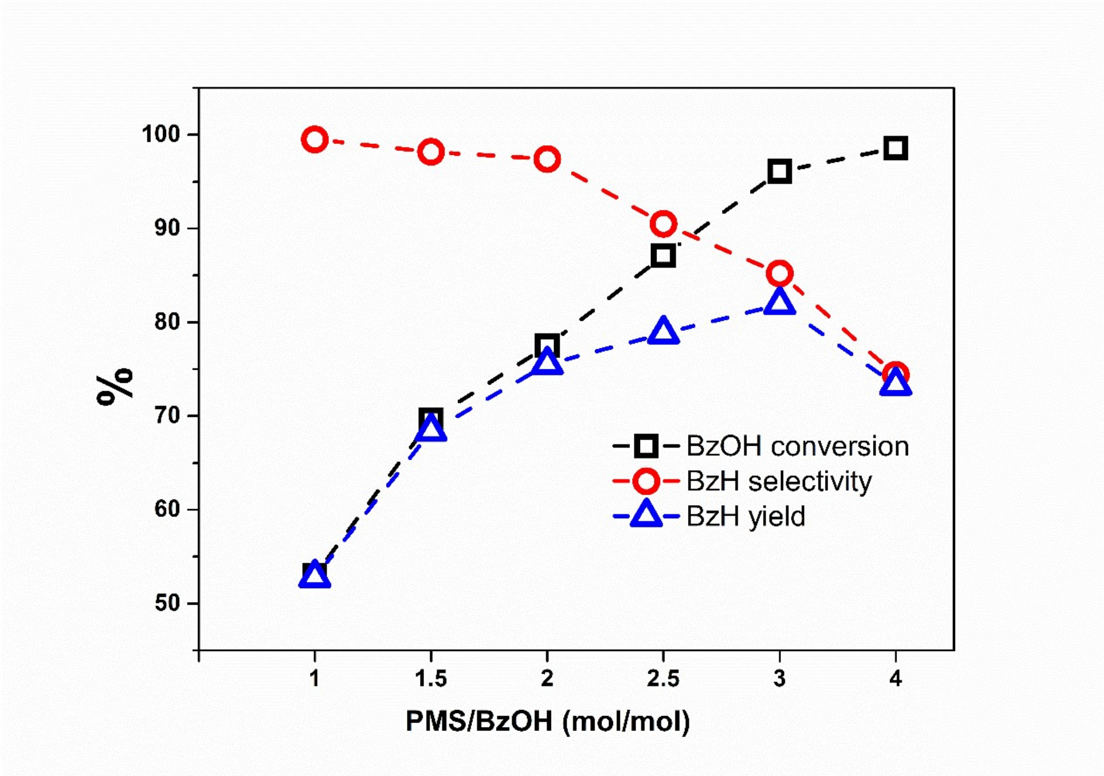


**Fig. S6.** The effect of PMS concentration on the catalytic efficiency of BzOH oxidation with NG-800. Reaction conditions: 5 mg catalyst, 0.1 mmol BzOH, 5 mL acetonitrile/water (1:1, volume ratio), 50 ºC, 5 h.

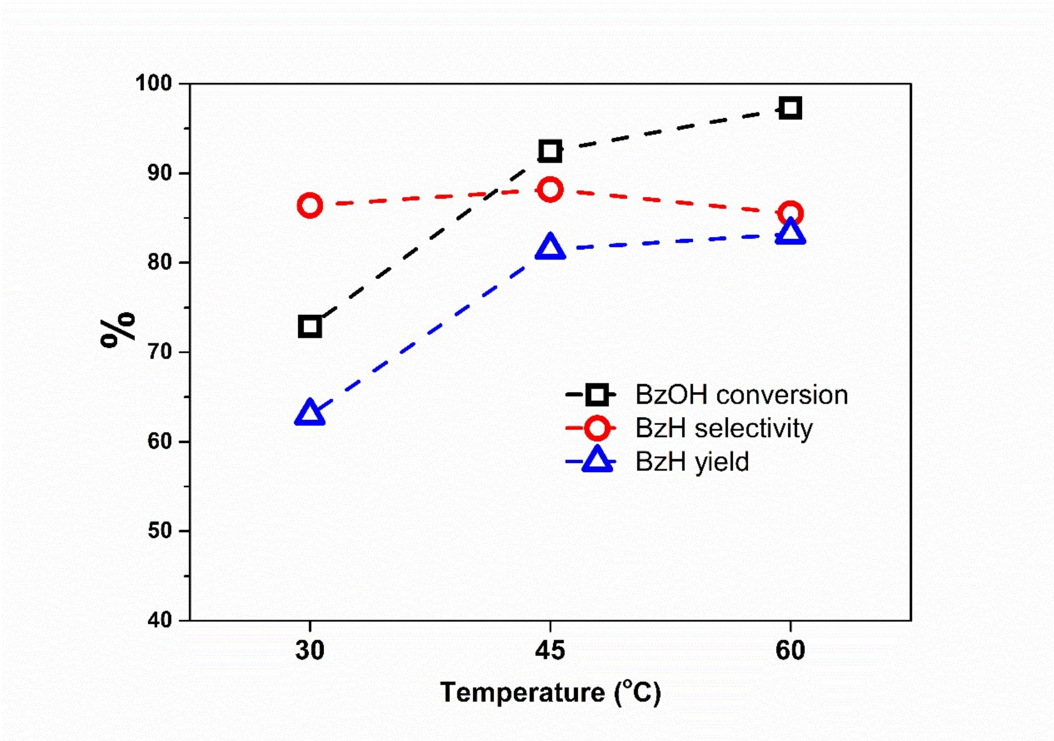


**Fig. S7.** The effect of reaction temperature on the catalytic efficiency of BzOH oxidation with NG-800. Reaction conditions: 5 mg catalyst, 0.1 mmol BzOH, 0.3 mmol PMS, 5 mL acetonitrile/water (1:1, volume ratio), 5 h.

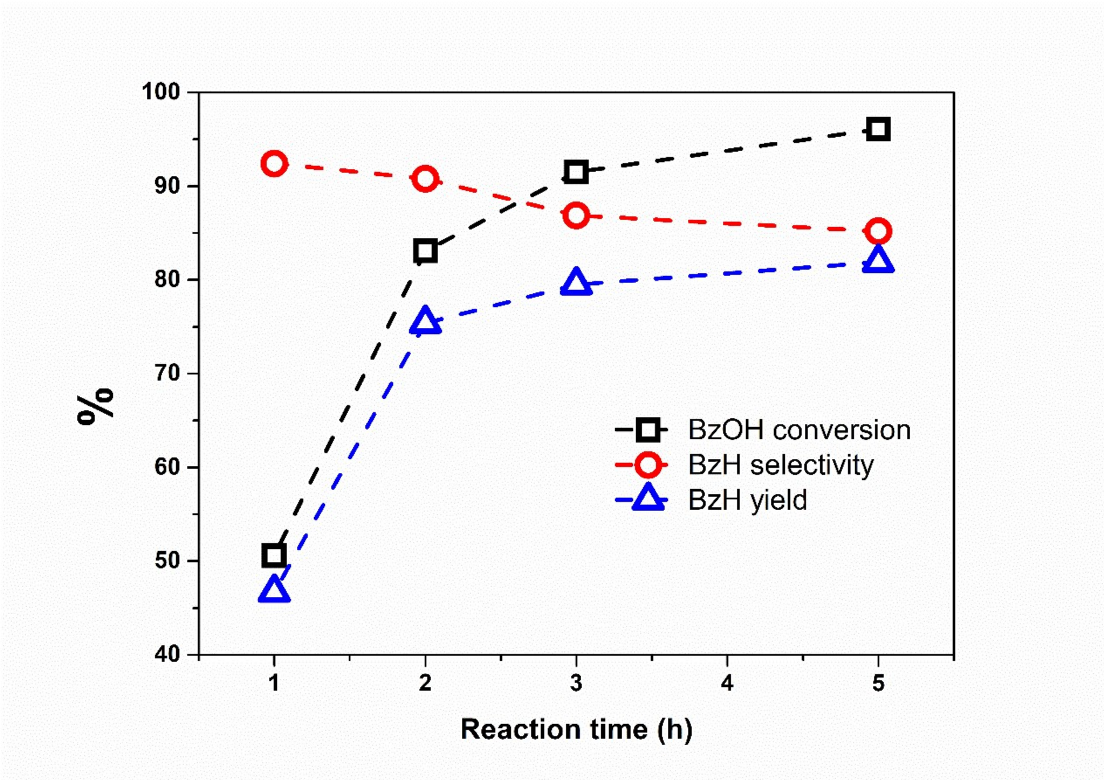


**Fig. S8.** The effect of reaction time on the catalytic efficiency of BzOH oxidation with NG-800. Reaction conditions: 5 mg catalyst, 0.1 mmol BzOH, 0.3 mmol PMS, 5 mL acetonitrile/water (1:1, volume ratio), 50 ºC.

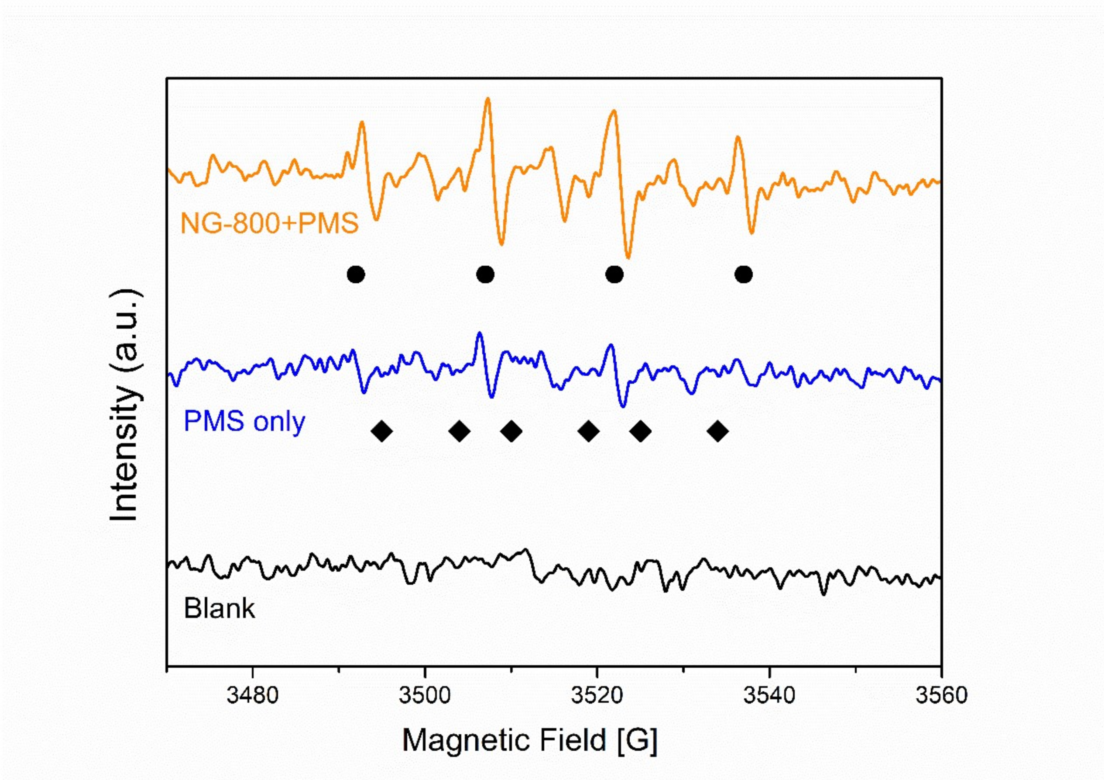


**Fig. S9.** EPR spectra in the presence of DMPO (DMPO-•OH-●, DMPO- $SO_4^{\bullet -}$-♦).

**Table S2.** Comparison of the oxidation efficiency of benzyl alcohol in this work with the reported literatures.

| Entry | Catalyst loading (relative to the mass of BzOH) | Oxidant | T/ °C | t/h | Additive | BzOH conversion/ % | BzH selectivity /% | BzH yield/% | Ref |
|---|---|---|---|---|---|---|---|---|---|
| 1 | Graphene oxide (200%) | $O_2$/1 atm | 100 | 24 | – | – | – | 90 | [1] |
| 2 | NCNT (2%) | $O_2$/15 atm | 130 | 8 | – | 44.7 | 94.1 | – | [2] |
| 3 | N-graphene (300%) | $O_2$/1 atm | 70 | 10 | – | 12.8 | 100 | – | [3] |
| 4 | P-doped carbon (80%) | Air/1 atm | 120 | 5 | – | – | – | 99.7 | [4] |
| 5 | AuPd bimetal (0.5%) | $O_2$/8 atm | 110 | 1 | – | 83 | 70 | – | [5] |
| 6 | $NiFe_2O_4$ (10%) | TBHP | 60 | 3 | – | – | – | 85 | [6] |
| 7 | $Au/Al_2O_3$ (2.5%) | TBHP | 125 | 5 | – | 89.9 | 89.2 | – | [7] |
| 8 | N-nanodiamond (5%) | TBHP | 70 | 24 | – | 88.5 | 99 | – | [8] |
| 9 | – | PMS | – | 3 | NaBr | – | – | 87 | [9] |
| 10 | Carbon nanotubes (50%) | PMS | 50 | 5 | – | 57.1 | 84.3 | 48.1 | [10] |
| 11 | NG (50%) | PMS | 50 | 5 | – | 96.1 | 85.2 | 81.9 | Herein |

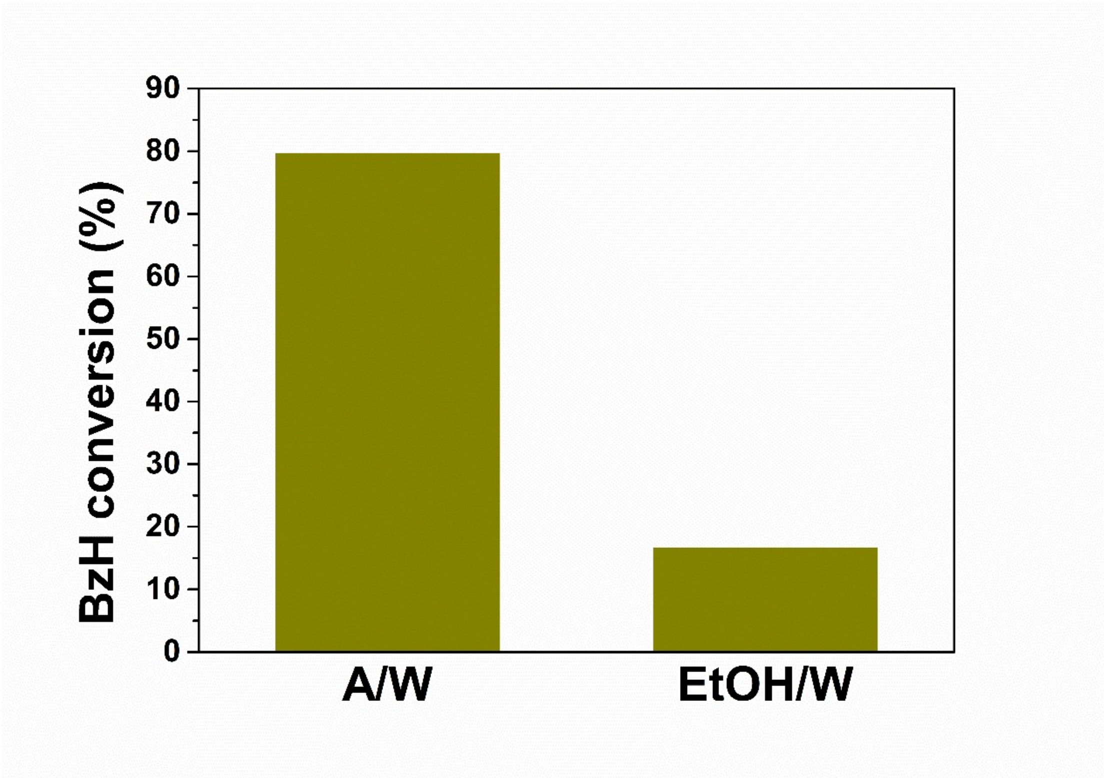


**Fig. S10.** The oxidation of BzH with NG-800 in acetonitrile/water 1:1 solvent (A/W) and ethanol/water 1:1 solvent (EtOH/W). Reaction conditions: 5 mg catalyst, 0.1 mmol BzH, 0.2 mmol PMS, 5 mL acetonitrile/water (1:1, volume ratio), 50 ºC, 5 h.

## References

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