Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supplementary material

Role of different coordinated Cu and reactive oxygen species on the highly active Cu-Ce-Zr mixed oxides in NH_3-SCO: A combined *in situ* EPR and O_2-TPD approach

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Catalyst Preparation

1. Citric Acid Sol-gel Method (SOL)

The Cu-Ce-Zr mixed oxide was first prepared by citric acid sol-gel method (SOL). The cerium (III) nitrate ($Ce(NO_3)_3$•$6H_2O$), zirconium nitrate ($Zr(NO_3)_4$•$5H_2O$) and copper nitrate ($Cu(NO_3)_2$•$3H_2O$) were mixed in deionized water according to the desired molar ratio. Citric acid was added as the complexing agent with a 1.3:1 ratio of the acid to metal ions including Ce^{3+}, Zr^{4+} and Cu^{2+}. Appropriate polyglycol with the weight of 50% citric acid was added. The blended solution was continuously stirred in a magnetic stirrer and heated at 90°C till transparent gel was formed. The resulting gel was dried at 100°C overnight, and the obtained sample was labeled as Cu-Ce-Zr (SOL).

2. Homogeneous Precipitation Method (HP)

The Cu-Ce-Zr mixed oxide was then prepared by a homogeneous precipitation method using urea as precipitator (HP). The aqueous solutions of $Cu(NO_3)_2$•$3H_2O$, $Ce(NO_3)_3$•$6H_2O$ and $Zr(NO_3)_4$•$5H_2O$ were mixed. Excessive urea aqueous solution was then added into the mixed solution, with a urea/(Cu +Ce + Zr) molar ratio of 10:1. The mixed solution was then heated to 90°C and held there for 24 h with vigorous stirring. After filtration and washing with deionized water, the resulting precipitant was dried at 100°C overnight, and the obtained sample was labeled as Cu-Ce-Zr (HP).

3. Incipient wetness impregnation method (IW)

First, CeO_2, ZrO_2 and Ce-Zr mixed oxide with Ce/Zr molar ratio of 4 were prepared by the surfactant-templated method[1]. Then, the catalyst was prepared by the incipient wetness impregnation method. The Ce-Zr mixed oxide support was added to $Cu(NO_3)_2$•$3H_2O$ aqueous solution, and the obtained sample was denoted as Cu/Ce-Zr (IW); In addition, appropriate quantities of CeO_2 and ZrO_2 power were mixed in an agate mortar to prepare CeO_2-ZrO_2 oxide by mechanical mixing. Subsequently, the CeO_2-ZrO_2 support was added to the $Cu(NO_3)_2{\bullet}3H_2O$ aqueous solution, and the obtained sample was denoted as Cu/CeO_2-ZrO_2 (IW). Finally, all obtained slurries were stored at room temperature overnight and then dried at 100°C for 12 h.

Figure Captions

Fig. S1 The Madon-Boudart test of NH_3 oxidation at 180°C over Cu-Ce-Zr (SOL) catalyst with different particle size

Fig. S2 The Madon-Boudart test of NH_3 oxidation at 180°C and 200°C over Cu-Ce-Zr (SOL) catalyst with the same particle size (20-40 mesh).

Fig. S3 The TOF of NH_3 oxidation in the presence of H_2O, SO_2 and CO_2.

Fig. S4 Comparison of H_2-TPR patterns of a: fresh Cu-Ce-Zr (SOL), b: after treatment of NH_3 at 230°C over fresh Cu-Ce-Zr (SOL) and c: after treatment of O_2 at 230°C following by step b.

Figure S1

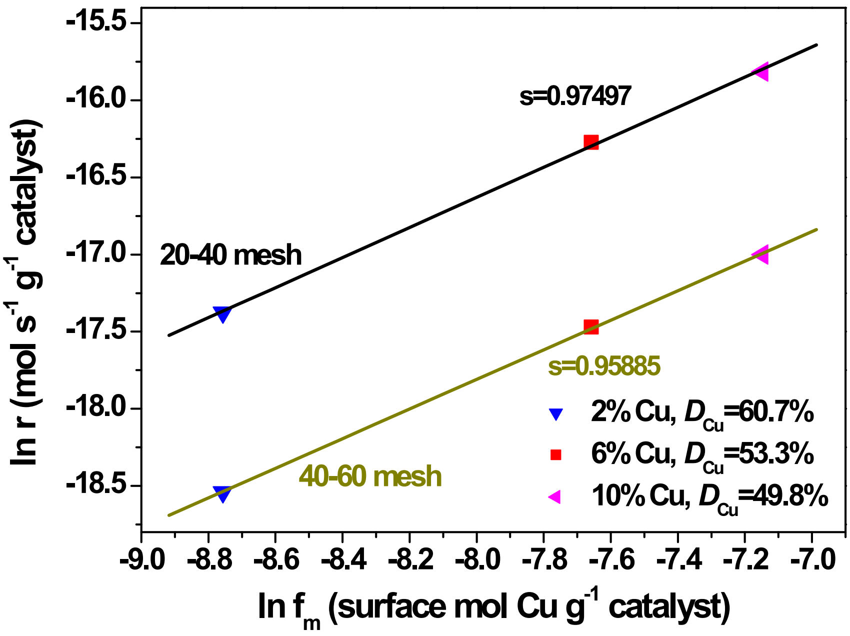

Figure S2

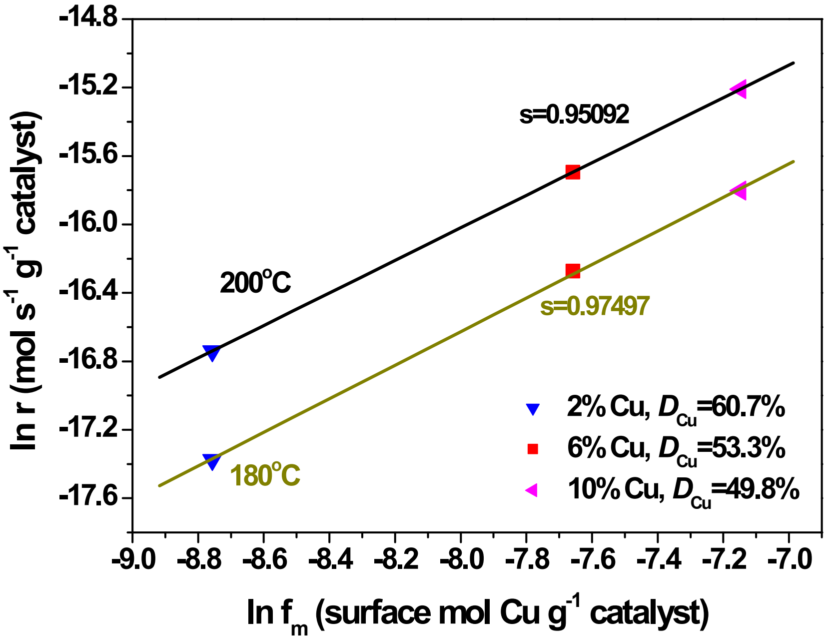

Figure S3

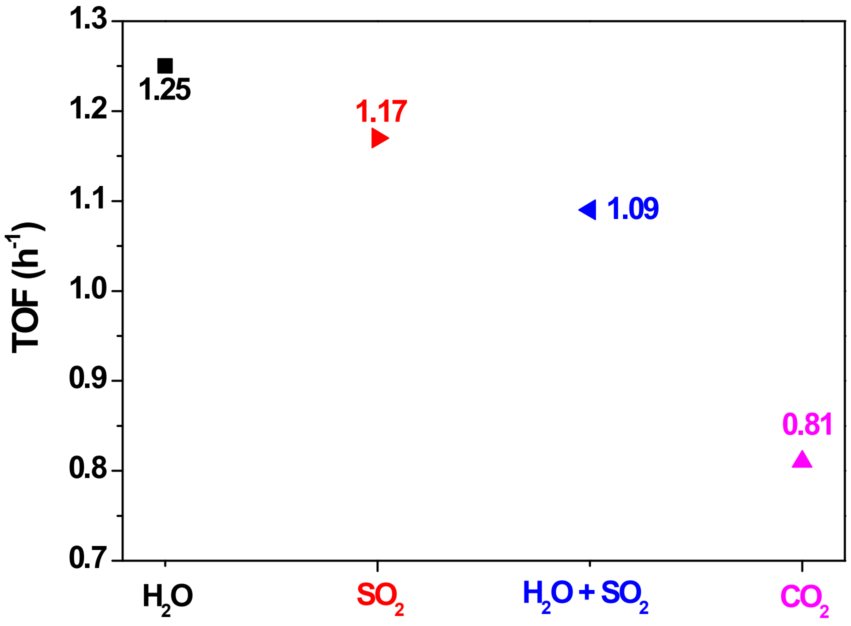

Figure S4