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Effect of preparation methods on VO_x/CeO_2 catalysts for the selective catalytic reduction of NO_x with NH_3

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Figure S1 exhibits the NH₃-SCR activity over VO_x/CeO₂ with different loadings prepared by the homogeneous precipitation method. With the increase of VO_x loading, NO_x conversion increased obviously, especially in the temperature window of 150-300 °C. 3% wt.% VO_x/CeO₂ showed higher catalytic activity than 1%, 0.5%, and 0.1%. Due to the toxicity of vanadium to the human body, a vanadium-based catalyst with too high loading was not preferred. Therefore, we chose the 3% VO_x/CeO₂ catalyst to investigate rather than catalysts with higher loading.

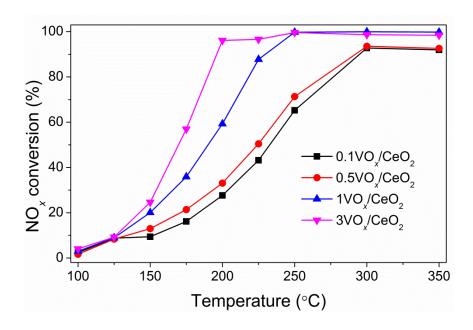


Figure S1 NH₃-SCR activity over VO_x/CeO₂ with different loadings.

Reaction conditions: [NO] = [NH₃] = 500 ppm, [O₂] = 5 vol. %, N₂ balance, total flow rate 500 ml/min and GHSV = 50 000 h⁻¹.

The N_2 selectivity of VO_x/CeO_2 catalysts prepared by different methods is shown in Figure S2. All the catalysts presented higher than 90% N_2 selectivity and only a little N_2O was produced at the temperatures we investigated.

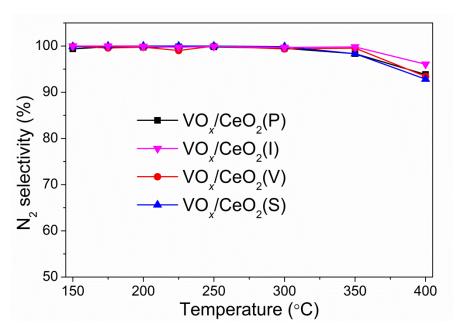


Figure S2. The N₂ selectivity in NH₃-SCR reaction over VO_x/CeO₂ catalysts prepared by different methods.

The NH₃-SCR performance of VO_x/CeO₂ catalysts after SO₂ poisoning for 24 h is shown in Figure S3. After SO₂ poisoning, the activity of all the catalysts decreased noticeably. However, the activity of VO_x/CeO₂(P) was still higher than that of other catalysts. 100% NO_x conversion could be obtained on the VO_x/CeO₂(P) catalyst at 250 °C and 70% on VO_x/CeO₂(S). This proved again that the VO_x/CeO₂(P) catalyst showed the strongest SO₂ resistance.

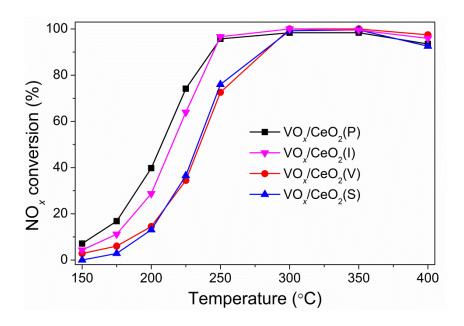


Figure S3. NH₃-SCR activity of VO_x/CeO₂(P) and VO_x/CeO₂(S) catalysts after 100 ppm SO₂ poisoning for 24 h.

Reaction conditions: [NO] = [NH₃] = 500 ppm, [O₂] = 5 vol. %, N₂ balance, total flow rate 500 ml/min and GHSV = 50 000 h⁻¹.

The separate oxidation activities of NO and NH₃ on VO_x/CeO₂ catalysts are shown in Figure S4. The NO oxidation activity of VO_x/CeO₂(P) was a little higher than that of VO_x/CeO₂(S). It has been reported that the enhancement of NO oxidation to NO₂ could significantly promote the low temperature SCR activity due to the occurrence of "fast SCR": NO and NO₂ react with NH₃ to form N₂ and H₂O.¹⁻² Higher catalytic activity over the VO_x/CeO₂(P) catalyst could be related to its slightly higher NO oxidation activity.

The $VO_x/CeO_2(P)$ catalyst showed a bit higher NH_3 oxidation activity than $VO_x/CeO_2(S)$ in the temperature range investigated. This indicates that the $VO_x/CeO_2(P)$ catalyst adsorbed and activated NH_3 more easily and therefore achieved higher NH_3 -SCR activity than the $VO_x/CeO_2(S)$ catalyst.

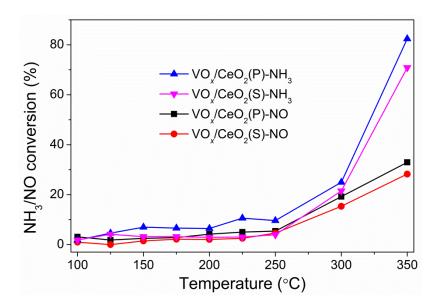


Figure S4. NH₃/NO conversion in separate NH₃ or NO oxidation reactions over $VO_x/CeO_2(P)$ and $VO_x/CeO_2(S)$ catalysts.

Reaction conditions: [NO] = 500 ppm or [NH₃] = 500 ppm, [O₂] = 5 vol. %, N₂ balance, total flow rate 500 ml/min and GHSV = 50 000 h⁻¹.

The changes in band intensities of nitrate species on NO_x pre-adsorbed catalysts during the introduction of NH_3 are shown in Figure S5. When NH_3 was introduced, the intensity of the bands attributed to monodentate nitrate and bridging nitrate species decreased slightly. The amount of bidentate nitrate species increased markedly, which may be due to the transformation of monodentate and bridging nitrate to bidentate nitrate.

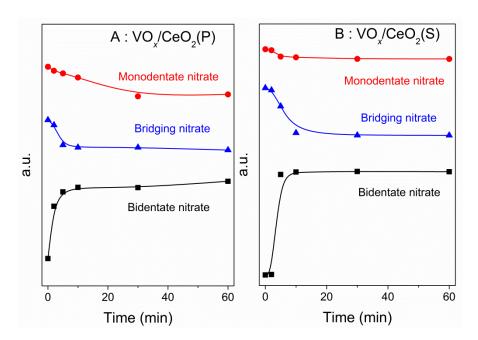


Figure S5.The band intensities of nitrate species calculated from DRIFT spectra (Figure 9) centered at 1542 cm⁻¹ (monodentate nitrate), 1203 cm⁻¹ (bridging nitrate) and 1245 cm⁻¹ (bidentate nitrate) of VO_x/CeO₂(P) (A) and VO_x/CeO₂(S) (B).

References

- (1) G. Qi, R. T. Yang, Appl Catal B-environ 2003, 44, 217-225.
- (2) R. Q. Long, R. T. Yang, J Catal 2001, 198, 20-28.