Support shape effect on the catalytic performance of VO_x/CeO_2 catalysts for the selective catalytic reduction of NO_x with NH_3

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Abstract: VO_x/CeO_2 catalysts, employing CeO_2 nanocubes (NCs), nanorods (NRs), and nanopolyhedrons (NPs) with predominately oxposed {100}, {110}, and {111} facets as the supports, were prepared by an incipient wetness imprenation method. The catalysts were characterized in detail and applied to the selective catalytic reduction (SCR) of NO_x with NH_3. The catalytic tests showed that V-CeO_2-NPs could achieve significantly higher NO conversion than V-CeO_2-NCs and V-CeO_2-NRs across the entire temperature range. The characterization results confirmed that the redox and acidic properities of VO_x/CeO_2 catalysts were closely related to the exposed facets of corresponding CeO_2 supports.

Keywords: CeO_2 support, facets, VO_x species.

1. Introduction (11-point boldface)

Currently, nitrogen oxides (NO_x, including NO and NO_2) from stationary and mobile sources have been regarded as major air pollutants.1 Ceria (CeO_2) has been attracted much attention due to its high oxygen storage capacity and nice redox properity. Recently, CeO_2 nanocrystals with well-defined morphologies were succesfully synthesized and used as catalysts or supports for various catalytic reactions, in which the shape-denpendent catalytic performances were observed clearly.3,4 Here, VO_x/CeO_2 catalysts employing CeO_2 nanocubes (NCs), nanorods (NRs) and nanopolyhedrons (NPs) as the supports were prepared (denoted as V-CeO_2-NCs, V-CeO_2-NRs， V-CeO_2-NPs) and applied to the NH_3-SCR reaction. The catalytic perdormances of these catalysts were found to be quite different. To elucidate this difference, the surface structure, redox and acidic properities of VO_x/CeO_2 catalysts were investigated using various characterization techniques. The results indicated that the catalysts displayed significant support-morphology-denpendent SCR activity.

2. Experimental (or Theoretical)

CeO_2 supports with various morphologies were prepared by a hydrothermal method according to the previous report.8 The V-CeO_2-NCs catalyst with a vanadia (VO_x) loading of 5 wt% was prepared by an incipient wetness imprenation method. The CeO_2-CNs support was impregnated with an aqueous solution of NH_4VO_3 in oxalic acid (n_{NH4VO3}:$n_{oxalic\ acid}$ = 1:2). Then, the sample was dried at 100 °C overnight and calcined at 500 °C for 4 h. The V-CeO_2-NRs and V-CeO_2-NPs catalysts were prepared by a similar method.

3. Results and discussion

The TEM and HRTEM images of CeO_2 supports are collected in Figure 1. Furthermore, the schematic illustrations of the idealized support shapes are also shown in Figure 1. As shown in the Figure 1, we could see that CeO_2-NCs only exposes {100} facets, while CeO_2-NRs and CeO_2-NPs mainly expose {110} and {111} facets, respectively.

Figure 2 shows the NH_3-SCR performance of VO_x/CeO_2 catalysts at temperature ranging from 150 to 400 °C under a GHSV of 120, 000 mL g^{-1} h^{-1}. A significant difference on the catalytic performance is observed over the three catalysts. V-CeO_2-NPs displays the best SCR activity, followed by V-CeO_2-NCs, and V-CeO_2-NRs shows negligible NO conversion. Furthermore, more than 90% N_2 selectivity is obtained over V-CeO_2-CPs in the whole temperature range investigated (Figure 2b).

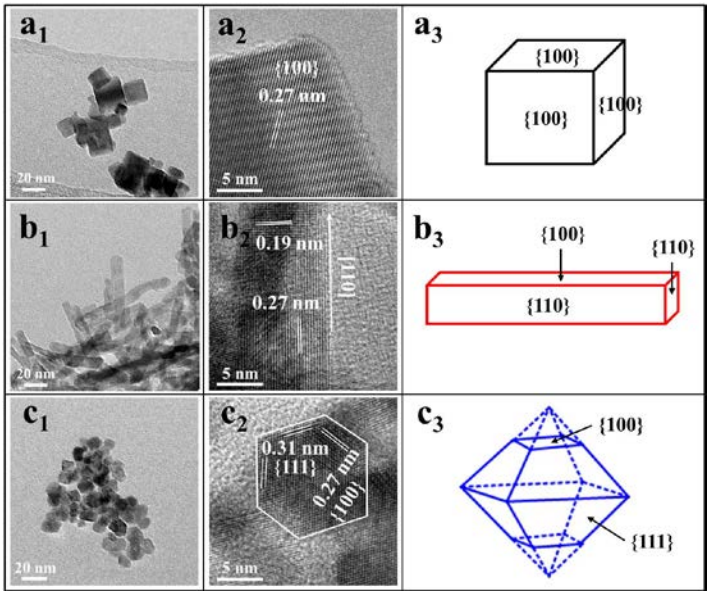

Figure 1. TEM images, HRTEM images, and schematic illustrations for CeO_2-NCs (a_1, a_2, a_3), CeO_2-NRs (b_1, b_2, b_3), CeO_2-NPs (c_1, c_2, c_3).

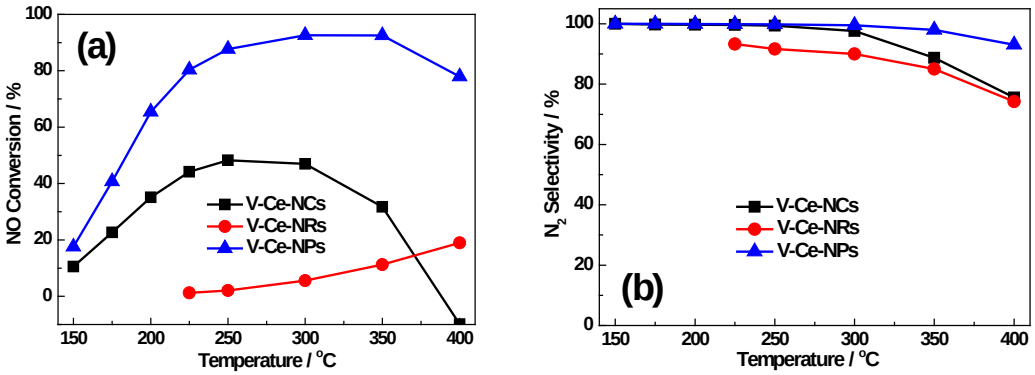

Figure 2. NO conversion (a) and N_2 selectivity (b) as a function of temperature in the NH_3-SCR reaction. Reaction conditions: [NO] = [NH_3] = 500 ppm, [O_2] = 5%, [H_2O]= 5.5% (when used), [SO_2]=110 ppm (when used), N_2 balance, total flow rate 200 mL min-1 and GHSV = 120,000 mL g^{-1} h^{-1}.

4. Conclusions

In summary, VO_x/CeO_2 catalysts with same VO_x loading with tunable CeO_2 morphology/facet were prepared, and then the catalytic properities of such catalysts were further investigated in the NH_3-SCR resction. The combination of characterization techniques and activity test demonstrated that VO_x/CeO_2 catalysts displayed sinificantly support-morphology-dependent catalytic activity. This study not only provides a facile method for the preparation of highly efficient CeO_2-based SCR catalysis, but also enables a better understanding of the relationship between the support morphology and catalytic performance of supported vanadium catalysts.

References

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