

Diffusion Limitations and Effectiveness Factor of Mesoporous and Hierarchically Structured Catalysts for SCR-DeNO_x

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Due to stricter environmental regulations for minimizing NO_x emissions, the urge to apply efficient catalysts is a worldwide concern. Among the technologies for controlling NO_x-emissions, the selective catalytic reduction of NO_x (SCR-DeNO_x) is the main industrial application. Vanadia supported on mesoporous titania is the most common catalyst wash-coated on SCR monoliths [1]. Such wash-coats lack a well-defined pore architecture, which leads to diffusion limitations and compromised material use. Theoretical studies have shown that by introducing macropores in a conventional SCR catalyst, its efficiency can be increased by up to 180% [2]. However, a direct proof of the effect of pore hierarchy on the catalytic activity of SCR-DeNO_x reaction is lacking.

In order to experimentally investigate the effect of additional macropores in a SCR catalysts, mesoporous and hierarchically structured (meso-/macroporous) V₂O₅/TiO₂ mixed oxides with different V₂O₅-contents (1, 3.5 and 5 wt.%) were prepared via a modified sol-gel method [3]. Characterization results show that the catalysts have similar content and nature of V₂O₅-species,

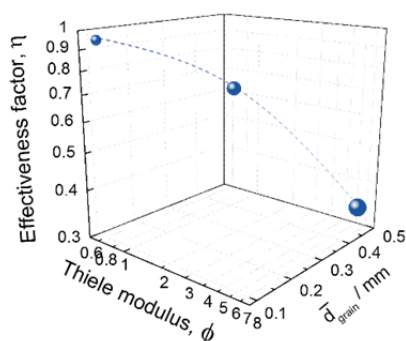


Figure 1: Effectiveness factor and Thiele modulus for different grain sizes of the 5 wt.% V₂O₅/TiO₂ mesoporous catalyst.

independent of their pore architecture. The diffusion effect on the catalytic activity was measured for a mesoporous catalyst by varying its grain size. By using the Weisz-Prater criterion to estimate the Thiele modulus and the effectiveness factor from the observed reaction rates, diffusion limitations were indeed found for the DeNO_x reaction ($\phi \sim 3$ and $\eta \sim 0.7$) for $\bar{d}_{\text{cat.grain}} = 0.25$ mm (Figure 1). The results from SCR of NO with NH₃ ($m_{\text{cat.}} = 0.2$ g, $\bar{d}_{\text{cat.grain}} = 0.25$ mm, $T = 423\text{--}773$ K, GHSV = 21,300 h⁻¹, $n(\text{NO})/n(\text{NH}_3) = 2/1$) over mesoporous and hierarchical V₂O₅/TiO₂ catalysts show that NO conversion increases with V₂O₅-content. However, the hierarchical catalysts exhibit a higher overall reaction rate over the whole temperature range ($T_R = 423\text{--}773$ K) when compared to their mesoporous counterparts with same V₂O₅-content. The NO consumption rate (r_{NO}) increases by up to 200%, and the light-off temperature ($T_{\text{Light-off}}$) decreases by 50 K when additional macropores are present (Figure 2). Considering the surface properties of the materials and the fact that DeNO_x is limited by intraparticle diffusion, the higher activity of the meso-/macroporous catalysts (for the same V₂O₅ content) can be attributed to diffusion facilitation within the catalyst particle. These results provide the first experimental proof of the advantage of pore hierarchy in SCR-DeNO_x as predicted earlier from theoretical calculations [2].

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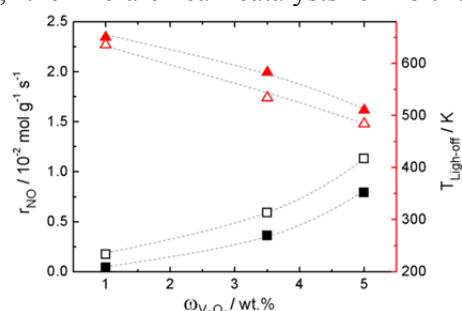


Figure 2: Reaction rate (r_{NO}) at 523 K and light-off temperature ($T_{\text{Light-off}}$) during SCR-DeNO_x over mesoporous and hierarchical V₂O₅/TiO₂ catalysts with different V₂O₅-content.

References

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