Diffusion Limitations and Effectiveness Factor of Mesoporous and Hierarchically Structured Catalysts for SCR-DeNO\textsubscript{x}

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Due to stricter environmental regulations for minimizing NO\textsubscript{x} emissions, the urge to apply efficient catalysts is a worldwide concern. Among the technologies for controlling NO\textsubscript{x}-emissions, the selective catalytic reduction of NO\textsubscript{x} (SCR-DeNO\textsubscript{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\textsubscript{x} reaction is lacking.

In order to experimentally investigate the effect of additional macropores in a SCR catalyst, mesoporous and hierarchically structured (meso-/macroporous) V\textsubscript{2}O\textsubscript{5}/TiO\textsubscript{2} mixed oxides with different V\textsubscript{2}O\textsubscript{5}-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 V\textsubscript{2}O\textsubscript{5}-species, 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\textsubscript{x} reaction (\(\phi\approx 3\) and \(\eta\approx 0.7\)) for \(d_{\text{cat,grain}} = 0.25\) mm (Figure 1). The results from SCR of NO with NH\textsubscript{3} (\(m_{\text{cat}} = 0.2\) g, \(d_{\text{cat,grain}} = 0.25\) mm, \(T = 423\)–\(773\) K, GHSV = 21.300 h\(^{-1}\), \(n(\text{NO})/n(\text{NH}_3) = 2/1\)) over mesoporous and hierarchical V\textsubscript{2}O\textsubscript{5}/TiO\textsubscript{2} catalysts show that NO conversion increases with V\textsubscript{2}O\textsubscript{5}-content. However, the hierarchical catalysts exhibit a higher overall reaction rate over the whole temperature range (\(T_R\) = 423–773 K) when compared to their mesoporous counterparts with same V\textsubscript{2}O\textsubscript{5}-content. The NO consumption rate (\(r_{\text{NO}}\)) 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\textsubscript{x} is limited by intraparticle diffusion, the higher activity of the meso-/macroporous catalysts (for the same V\textsubscript{2}O\textsubscript{5} 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\textsubscript{x} as predicted earlier from theoretical calculations [2].

References