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Regeneration of Pt-catalysts deactivated in municipal waste flue gas with H₂/N₂ and the effect of regeneration step on the SCR catalyst



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1. Introduction

MUNICIPAL waste flue gas was previously^[1] found to deactivate the Pt-based CO oxidation catalyst severely. In the specific case studied, siloxanes were found to cause the deactivation. An on-site method for complete regeneration of the catalyst activity was found without shutdown of the flue gas stream, i.e. by *in situ* treatment of the Pt-catalyst by reductive H₂-gas, outlined in Fig. 1.

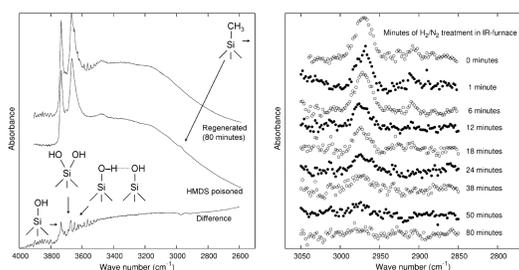


Figure 1: (Right) High temperature-FTIR (250°C) spectra of hexamethyldisiloxane-poisoned Pt/TiO₂ catalyst. (Left) Poisoned catalyst before and after treatment with 10% H₂/N₂^[1]

However, introduction of H₂ gas in the gas stream could also affect other units in the tail pipe gas cleaning system. Of special interest here, is the effect of hydrogen gas on the performance of the DeNO_x + SCR catalytic process, i.e. the reaction $4\text{NO} + 4\text{NH}_3 + \text{O}_2 \xrightarrow{\text{V}_2\text{O}_5-\text{WO}_3/\text{TiO}_2} 4\text{N}_2 + 6\text{H}_2\text{O}$.

2. NO SCR Activity

THE impact of hydrogen on the catalytic activity of the 3%V₂O₅-7%WO₃/TiO₂ catalyst at 250°C was investigated in two steps.

Exposure of the SCR catalyst to 10% H₂/N₂ reveals a dual-influence on the activity (Fig. 2, right): Up until ≈30 min in H₂ atmosphere, only the surface vanadate groups (V⁵⁺ and V⁴⁺) are reduced to V³⁺, decreasing the catalytic performance. At longer times of exposure, the slope has decreased, indicating that WO₃ or bulk TiO₂ is being reduced.

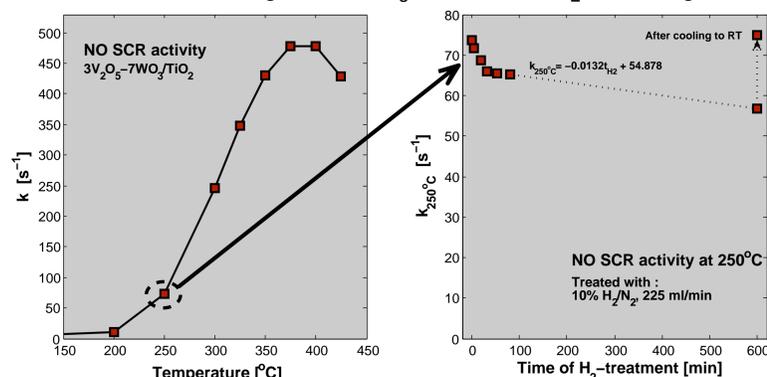


Figure 2: NO SCR activity vs. temperature for the standard SCR-catalyst, 3V₂O₅-7WO₃/TiO₂ (left). NO SCR activity at 250°C of the catalyst vs. time of preliminary 10% H₂/N₂ exposure of the catalyst (right)

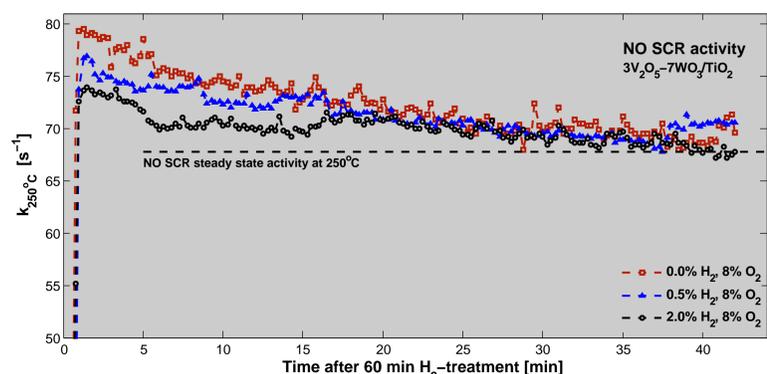


Figure 3: Time of regeneration in SCR gas of 3V₂O₅-7WO₃/TiO₂ at 250°C after 60 min exposure to different H₂-conc. in 8% O₂. Notice that initial steady-state activity is regained within about 30 min at these conditions

3. EPR spectroscopy

FROM the spectra it is evident that vanadium in the catalyst is reduced upon exposure to the hydrogen/oxygen mixture, observed as a reduction in signal from the V⁴⁺ species – the only EPR active vanadium species. It is possible that both the V⁵⁺ and V⁴⁺ species are reduced to V³⁺.

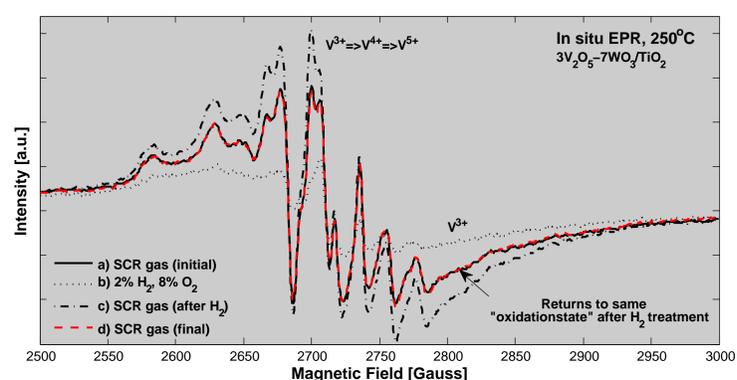


Figure 4: In situ EPR at 250°C at four key-points, a) start of NO SCR gas treatment, b) 2% H₂, 8% O₂ gas at steady state, c) immediately after switch back to SCR gas, detected increase in concentration of V⁴⁺, due to formation of V⁴⁺ and V⁵⁺ from V³⁺, d) steady state SCR conditions, notice that the system is completely reversible

The observed variation in the continuous spectrum (Fig. 5) is due to the oxidation of V(III) firstly to V(IV) and then to V(V): V³⁺ = V⁴⁺ = V⁵⁺, creating the maximum in the spectra vs. time.

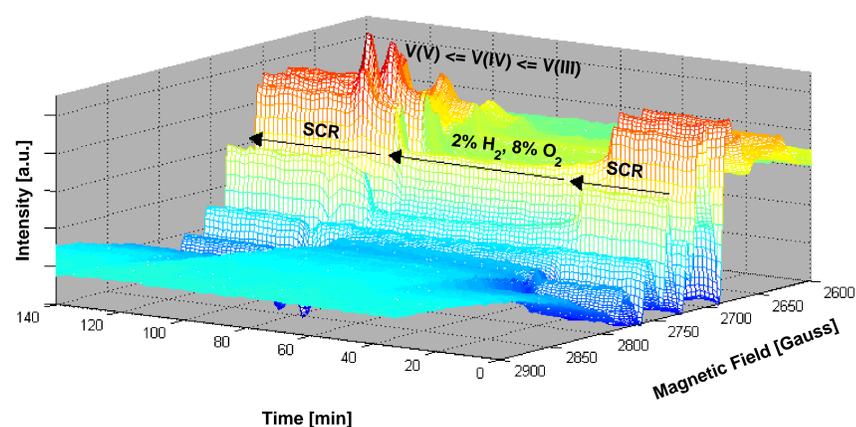


Figure 5: EPR spectra of V⁴⁺ in NO SCR gas, shift to 2% H₂, 8% O₂ for 60 min and shift back to SCR conditions

However, the 'excess' of the formed V(IV) is only intermediate, and the system quickly returns to the same distribution of vanadium oxidation states as before the hydrogen treatment.

4. Conclusion

TRADITIONAL titania-based SCR catalysts is practically not influenced by temporarily exposure to reductive gases. Thus, no permanent deactivation of the industrial SCR catalyst would be expected by exposure of such catalysts to diluted hydrogen/oxygen gas.

References

[1] Rasmussen, S.B., Kustov, A.L., Due-Hansen, J., Siret, B., Tabaries, F., Fehrmann, R., *Appl. Catal. B*, 69 (2006) 10–16