



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

Due-Hansen, Johannes; Rasmussen, Søren Birk; Kustov, Arkadii; Siret, Bernard; Tabaries, Frank; Fehrmann, Rasmus

Publication date:
2007

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Due-Hansen, J., Rasmussen, S. B., Kustov, A., Siret, B., Tabaries, F., & Fehrmann, R. (2007). *Regeneration of Pt-catalysts deactivated in municipal waste flue gas with H₂/N₂ and the effect of regeneration step on the SCR catalyst*. Poster session presented at 8th European Congress on Catalysis, Turku, Finland.

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

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



Johannes Due-Hansen^{a,*}, Søren B. Rasmussen^a,
Arkady L. Kustov^a, Bernard Siret^b, Frank Tabaries^b, Rasmus Fehrmann^a

^a Department of Chemistry and Center for Sustainable and Green Chemistry, Technical University of Denmark

^b LAB S.A., 25 rue Bossuet, 69455 Lyon Cédex 06, France

*jdh@kemi.dtu.dk

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.

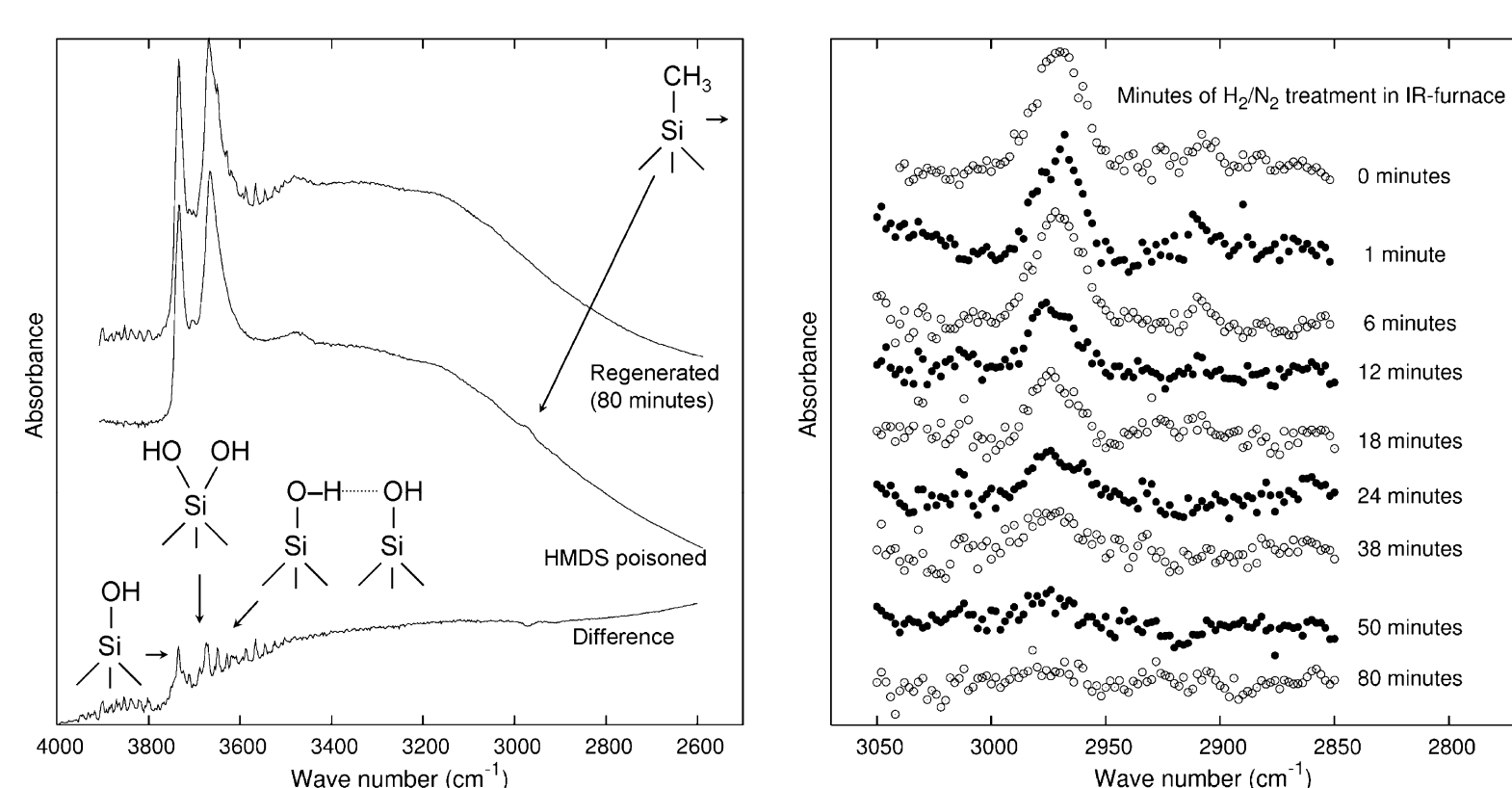


Figure 1: (Right) High temperature-FTIR (250°C) spectra of hexamethyldisiloxane-poisoned Pt/TiO₂ catalyst. (Left) Poisoned catalyst before and after treatment with 10% H₂/N₂^[1]

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.

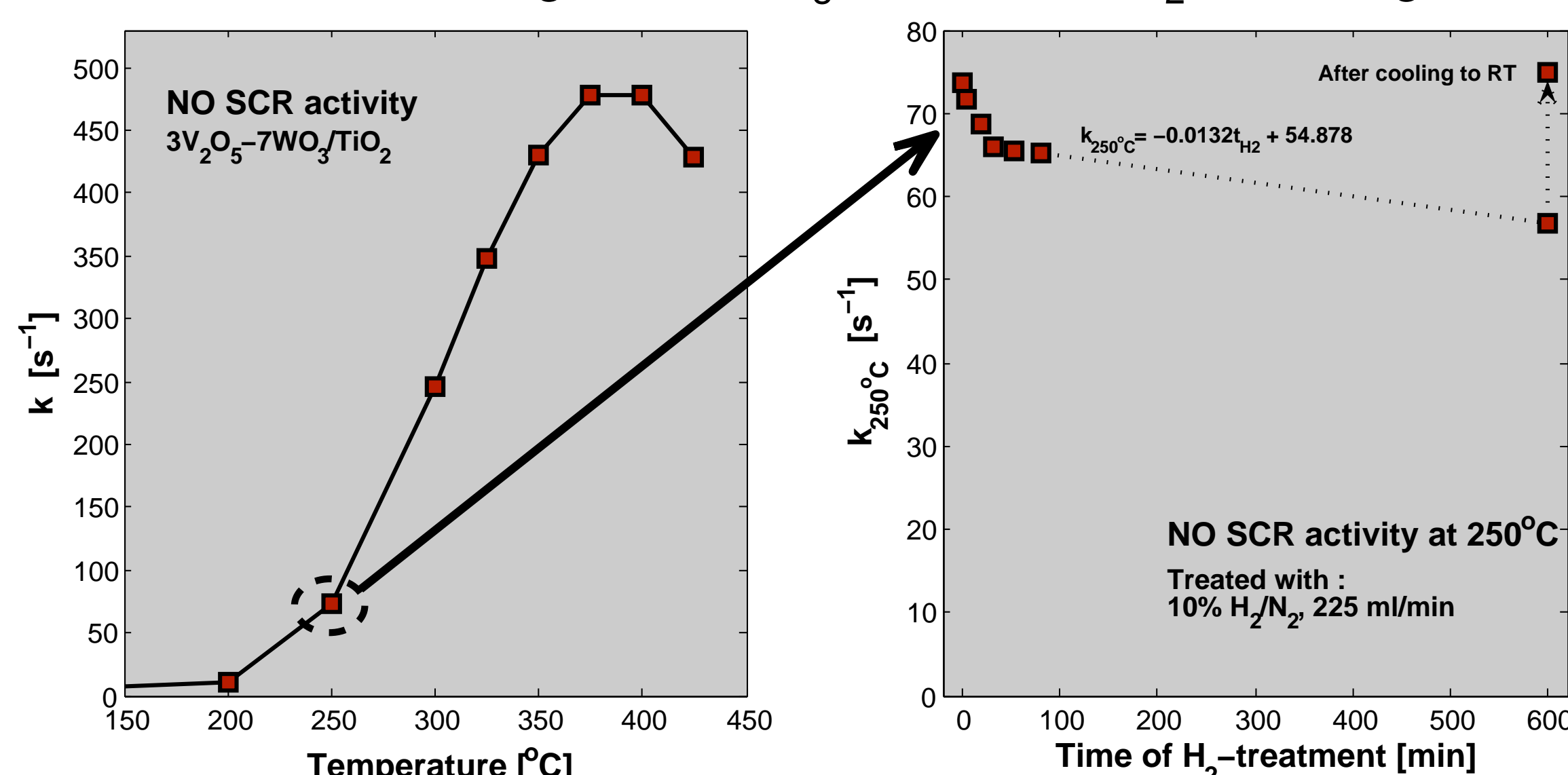


Figure 2: NO SCR activity vs. temperature for the standard SCR-catalyst, 3V₂O₅-7WO₃/TiO₂ (left). NO SCR activity at 250°C of the catalyst vs. time of preliminary 10% H₂/N₂ exposure of the catalyst (right)

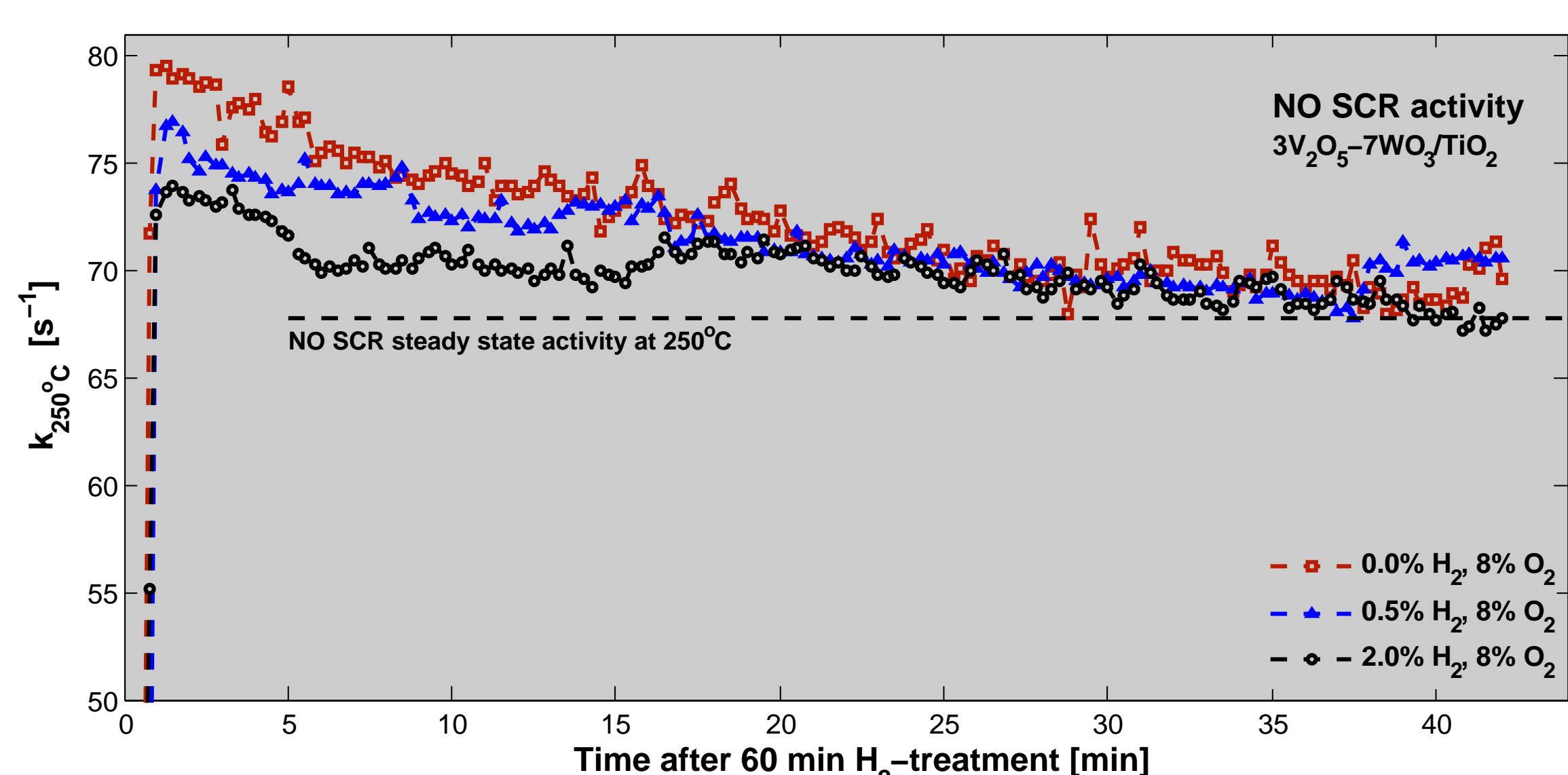


Figure 3: Time of regeneration in SCR gas of 3V₂O₅-7WO₃/TiO₂ at 250°C after 60 min exposure to different H₂-conc. in 8% O₂. Notice that initial steady-state activity is regained within about 30 min at these conditions

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⁺.

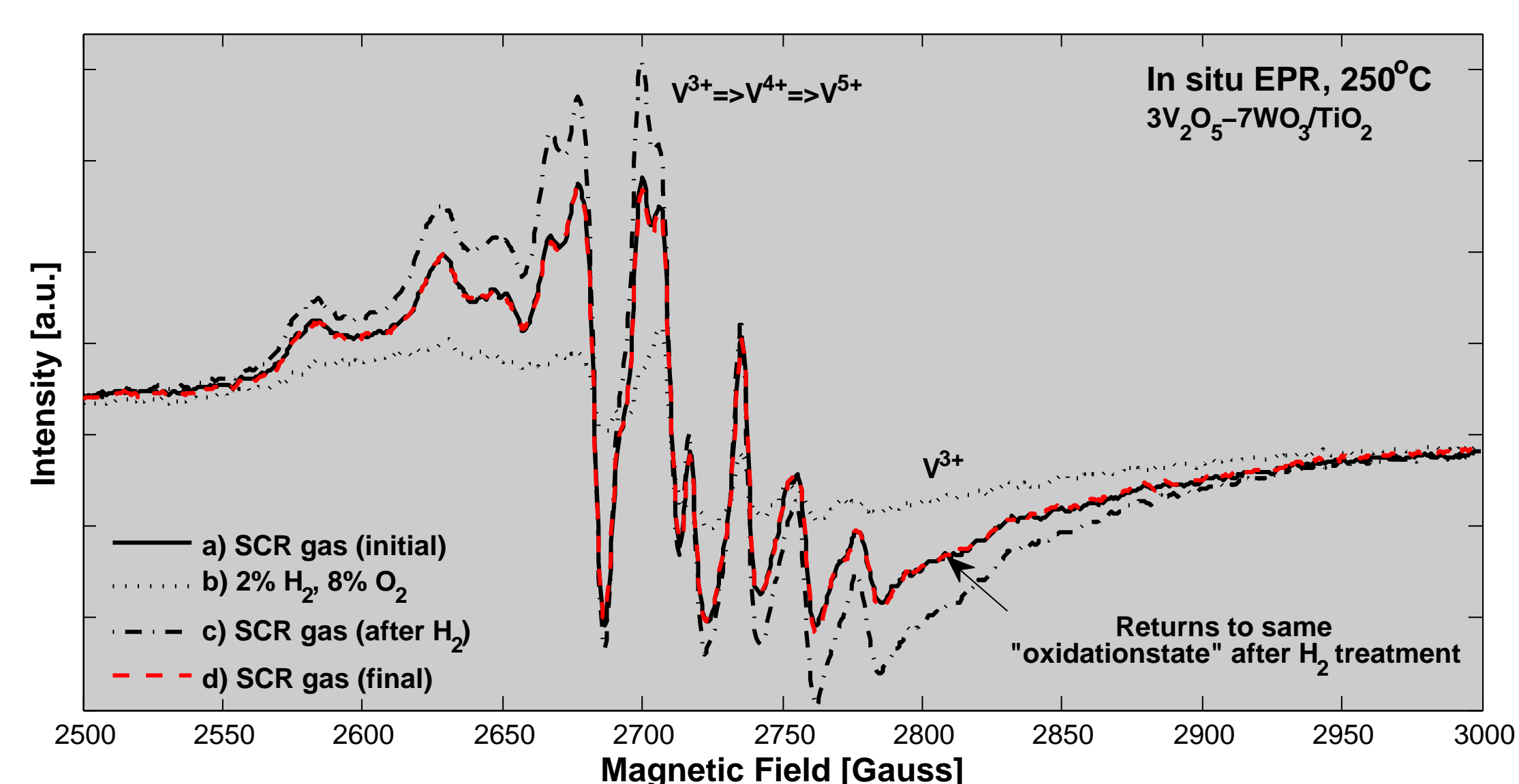


Figure 4: In situ EPR at 250°C at four key-points, a) start of NO SCR gas treatment, b) 2% H₂, 8% O₂ gas at steady state, c) immediately after switch back to SCR gas, detected increase in concentration of V⁴⁺, due to formation of V⁴⁺ and V⁵⁺ from V³⁺, d) steady state SCR conditions, notice that the system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

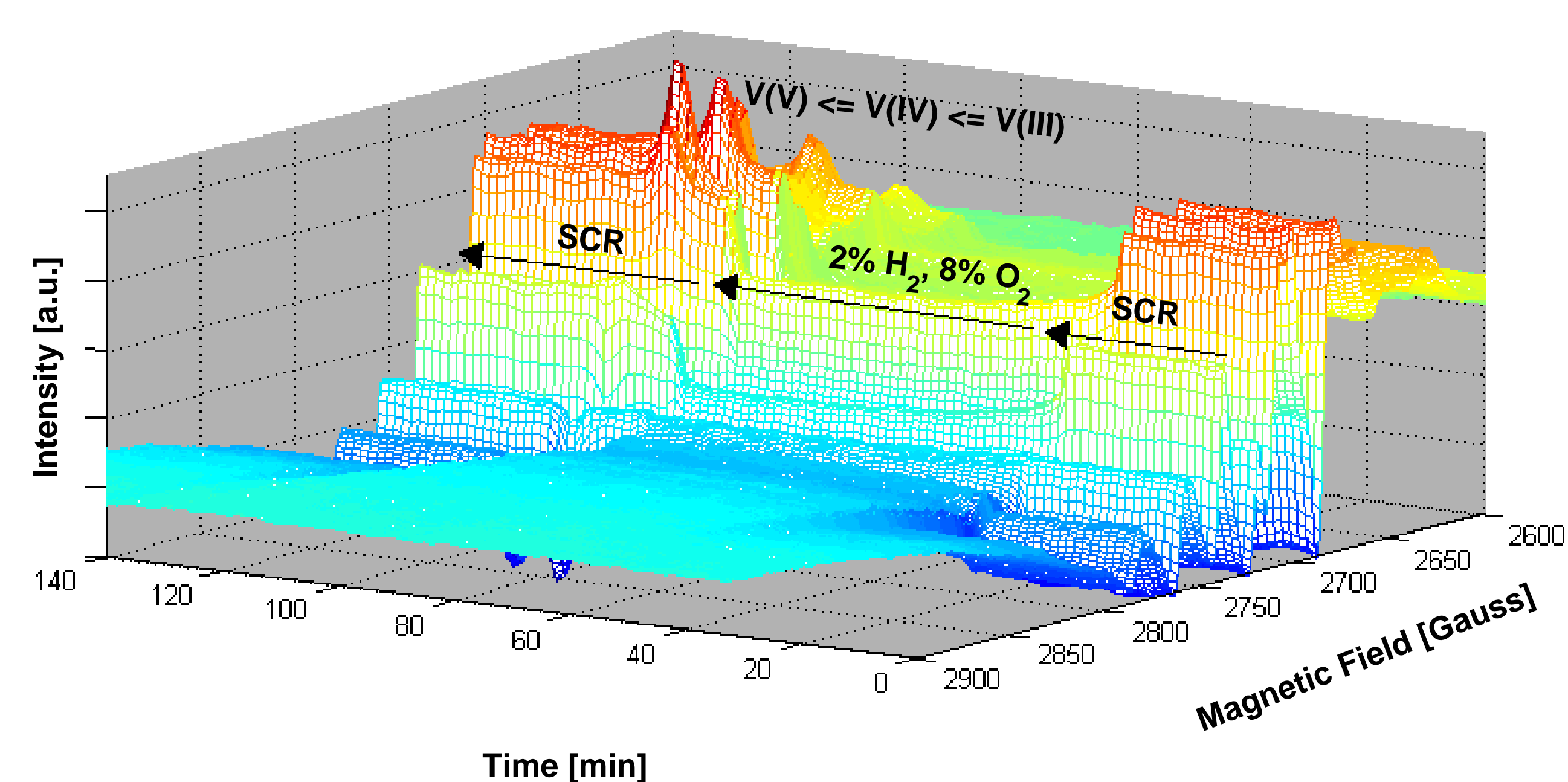


Figure 5: EPR spectra of V⁴⁺ in NO SCR gas, shift to 2% H₂, 8% O₂ for 60 min and shift back to SCR conditions

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