Kinetic modeling of H_2-assisted C_3H_6 Selective Catalytic Reduction of NO over Silver Alumina catalyst

Muhammad Mufti Azis[a,b], Hanna Härelind[b] and Derek Creaser[a]

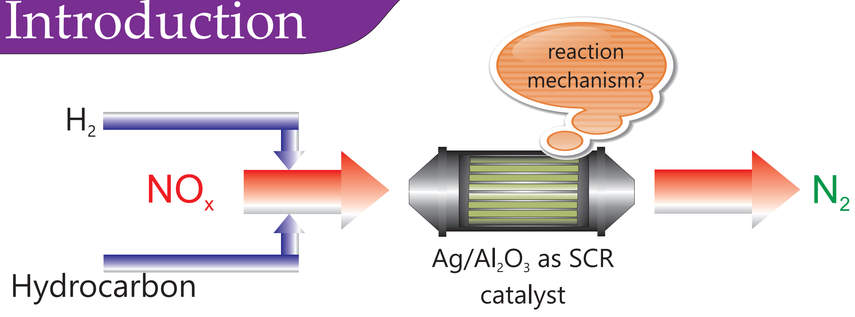
[a]Division of Chemical Engineering, Department of Chemical and Biological Engineering

[b]Competence Centre for Catalysis (KCK)

Chalmers University of Technology, SE-41296, Gothenburg, Sweden

Email: muhammad.azis@chalmers.se

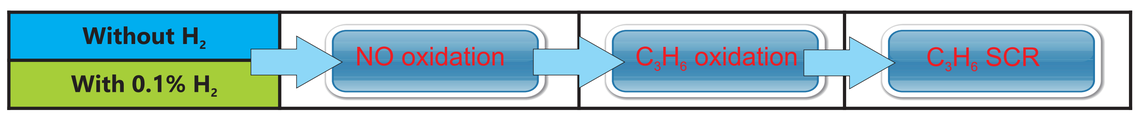
Introduction



- Ag/Al_2O_3 catalyst is a potential catalyst for NO_x abatement with HC-SCR technology. Further, addition of H_2 has been shown to give better HC-SCR performance on Ag/Al_2O_3 [1].
- The present work aims to set up a global kinetic model to describe mechanistic role of H_2 to promote C_3H_6 SCR over Ag/Al_2O_3 catalyst.

Experimental methods

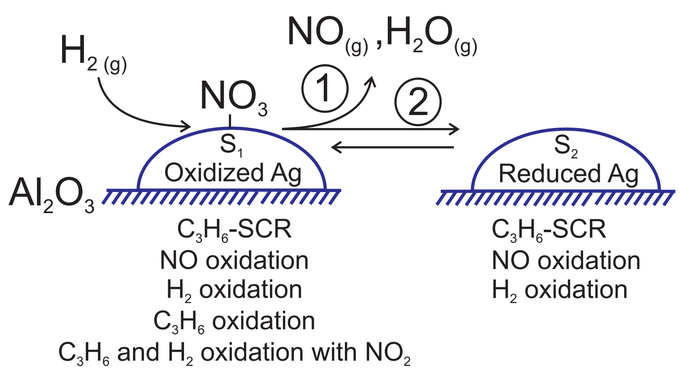
- Ag/Al_2O_3 with 2 wt.% Ag loading was prepared by sol-gel freeze dried method. The catalyst powder was washcoated on cordierite monolith.
- Activity measurement was conducted in a flow reactor equipped with gas FTIR and MS.
- Temperature-programmed reaction and transient experiments were used.
- The model aimed to simulate wide range of temperature and inlet feed concentration.



Modeling methods

Development of reaction mechanisms:

- Experimental data shows NO oxidation and C_3H_6 SCR are always higher in the presence of H_2 even for high temperature range.
- Initial screening of mechanisms suggested that single role of H_2 to remove inhibiting nitrate was insufficient to reproduce wide ranging experimental conditions.
- Key point in reaction mechanism: H_2 is proposed to have dual role to remove inhibiting nitrate and simultaneously form more active reduced sites, S_2.



- Reactor model: single channel model was used to describe mass transfer and reaction kinetic. The model also includes mass transfer of gas components inside washcoat [2].

References

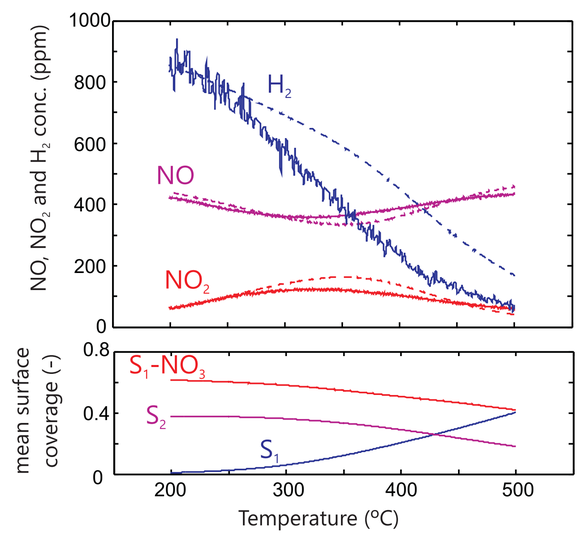
1. S. Satokawa, J. Shibata,K.-i. Shimizu, A. Satsuma and T. Hattori, Appl. Catal. B, 42(2003) 179-186.
2. M.M. Azis, H. Härelind and D. Creaser, Chem. Eng. J., 221 (2013) 382-397.

Acknowledgements

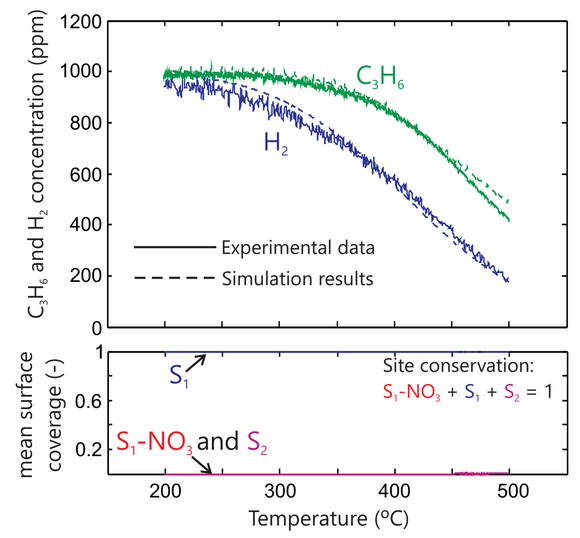
The financial support by the Swedish Research Council is gratefully acknowledged. This work was performed within the Competence Centre for Catalysis, which is hosted by Chalmers University of Technology and financially supported by the Swedish Energy Agency and the member companies AB Volvo, ECAPS AB, Haldor Topsøe A/S, Scania CV AB, Volvo Car Corporation AB and Wärtsilä Finland Oy.

Results and discussion

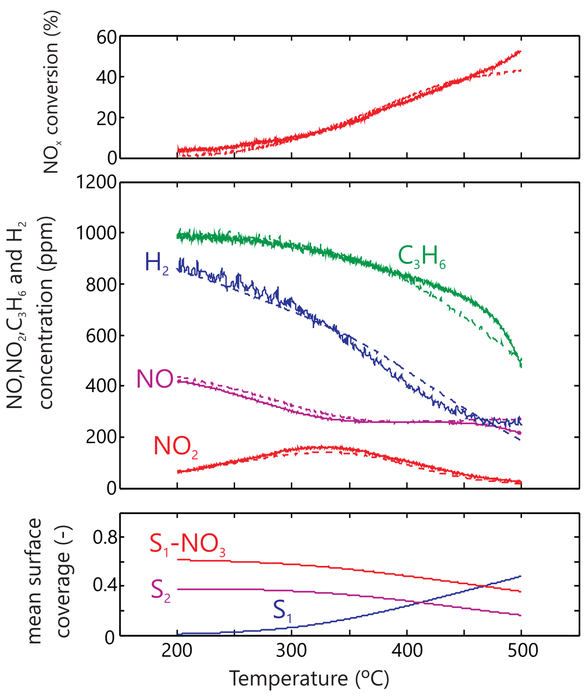
A sample of experimental and simulation results:



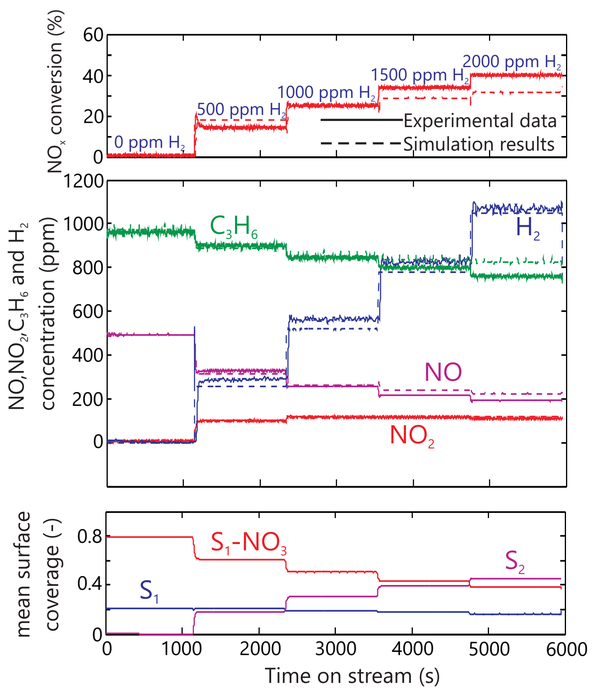
Temperature-programmed reaction of H_2-assisted NO oxidation. Feed: 500 ppm NO, 8% O_2, 0.1% H_2 and 5% H_2O.



Temperature-programmed reaction of H_2+C_3H_6 oxidation. Feed: 1000 ppm C_3H_6, 8% O_2, 0.1% H_2 and 5% H_2O.



Temperature-programmed reaction of H_2-assisted C_3H_6 SCR. Feed: 500 ppm NO, 8% O_2, 1000 ppm C_3H_6, 0.1% H_2 and 5% H_2O.



Transient experiment by varying H_2 concentration in C_3H_6 SCR at 375°C. Feed: 500 ppm NO, 8% O_2, 1000 ppm C_3H_6, 0 to 2000 ppm H_2 and 5% H_2O.

- The proposed H_2 role reproduced experimental data well for both temperature-programmed and transient experiments. Similarly, model validation under NO, C_3H_6 oxidation and C_3H_6 SCR with/without H_2 at 250 and 400°C gave good agreement between simulations and transient experiments.
- All reactions on reduced Ag sites (S_2) are more rapid than on oxidized Ag sites (S_1) which is indicated by lower activation energies on S_2 than on S_1 for the same reactions.
- Weisz modulie evaluations were made for H_2-assisted C_3H_6 SCR and C_3H_6 SCR indicated that larger fraction of experimental data was free from mass transport resistance.

Conclusions

- A global kinetic model for H_2-assisted C_3H_6 SCR has been developed to effectivey capture a wide range of feed concentrations and temperature.
- The model proposes dual role of H_2 to eliminate nitrate as well as to form a reduced sites, S_2. NO oxidation, H_2 oxidation as well as C_3H_6 SCR reactions are more rapid on the reduced sites, S_2.
- A larger fraction of experimental data was free from the influence of mass transport resistance.

CHALMERS