NH_3-Selective Catalytic Reduction over Ag/Al_2O_3 Catalysts

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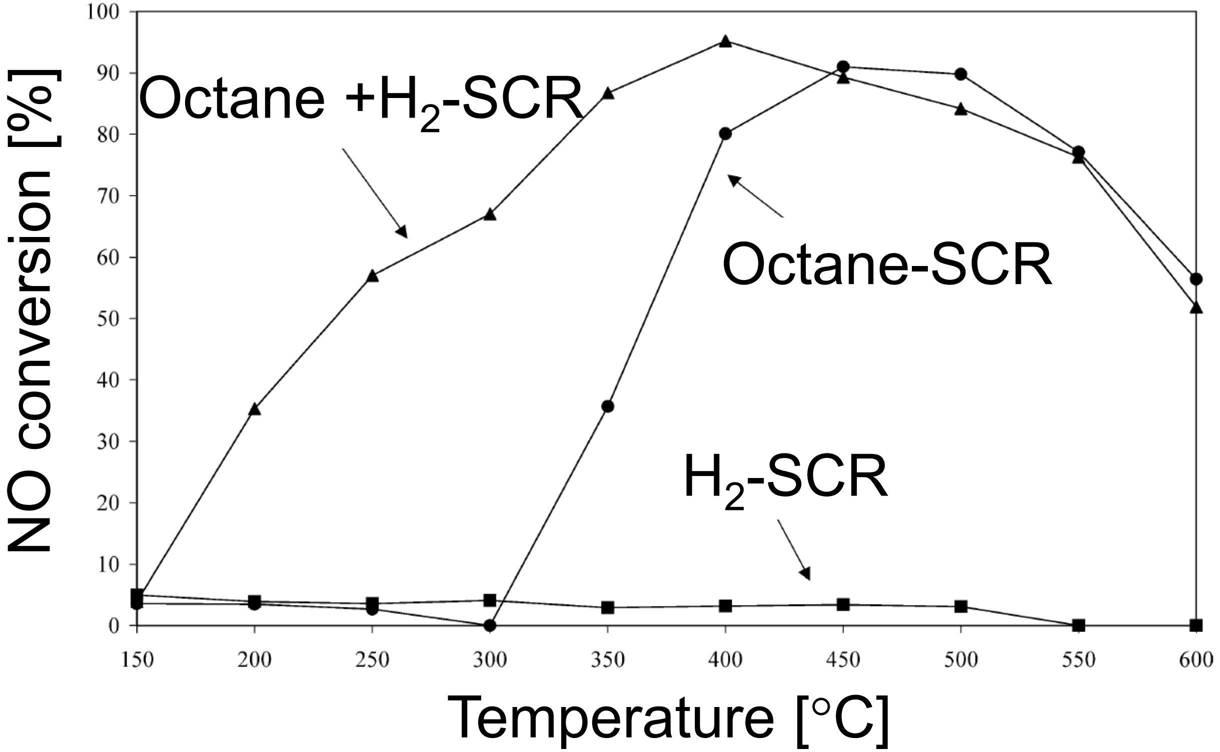
HC-SCR over Ag/Al_2O_3

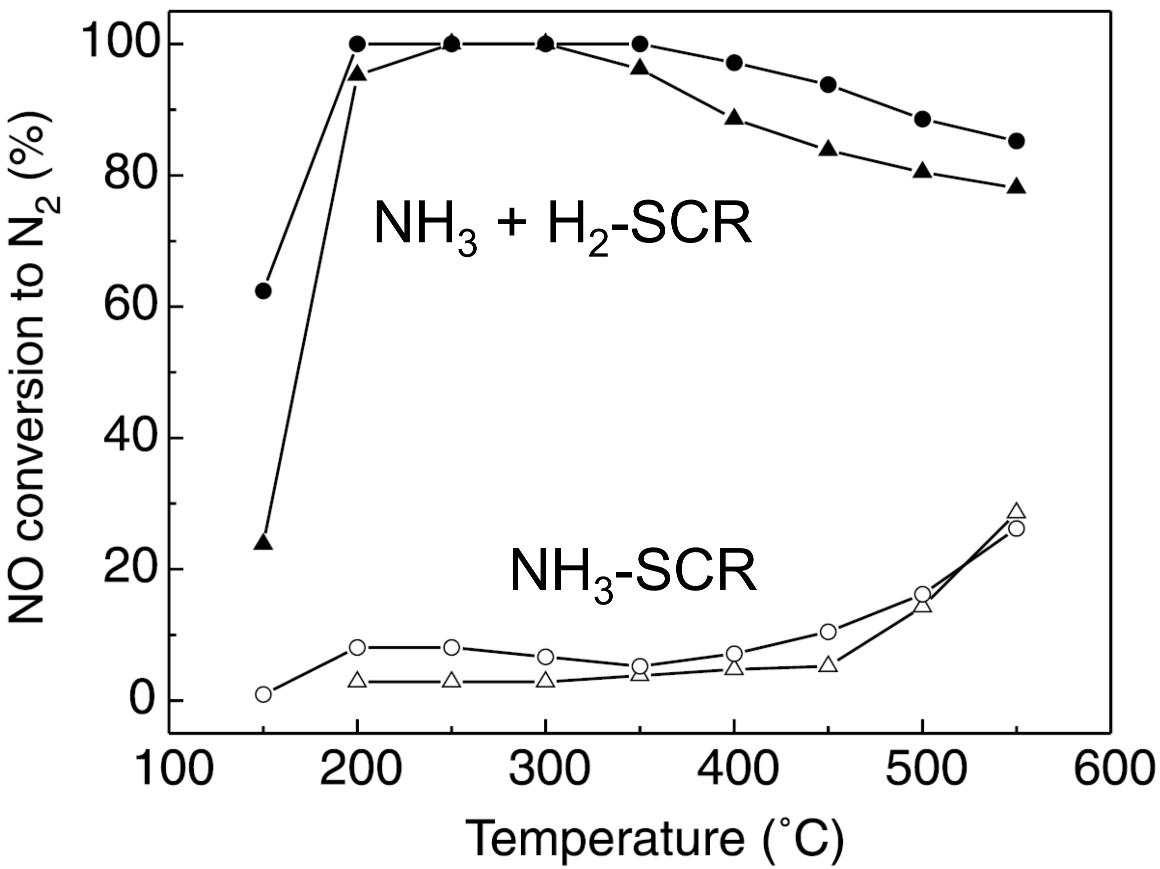


- H_2 lowers the temperature, where Ag/Al_2O_3 is active for HC-SCR
- Ag/Al_2O_3 is not active for H_2-SCR
- H_2 is a co-reductant

- Concitions: 500 ppm NO, 375 ppm C_8H_{18}, 1 vol% H_2, 6 vol% O_2, 10 vol% CO_2, 350 ppm CO, 12 vol% H_2O in He.
- K. Eränen et al., Journal of Catalysis 227 (2004) 328-343

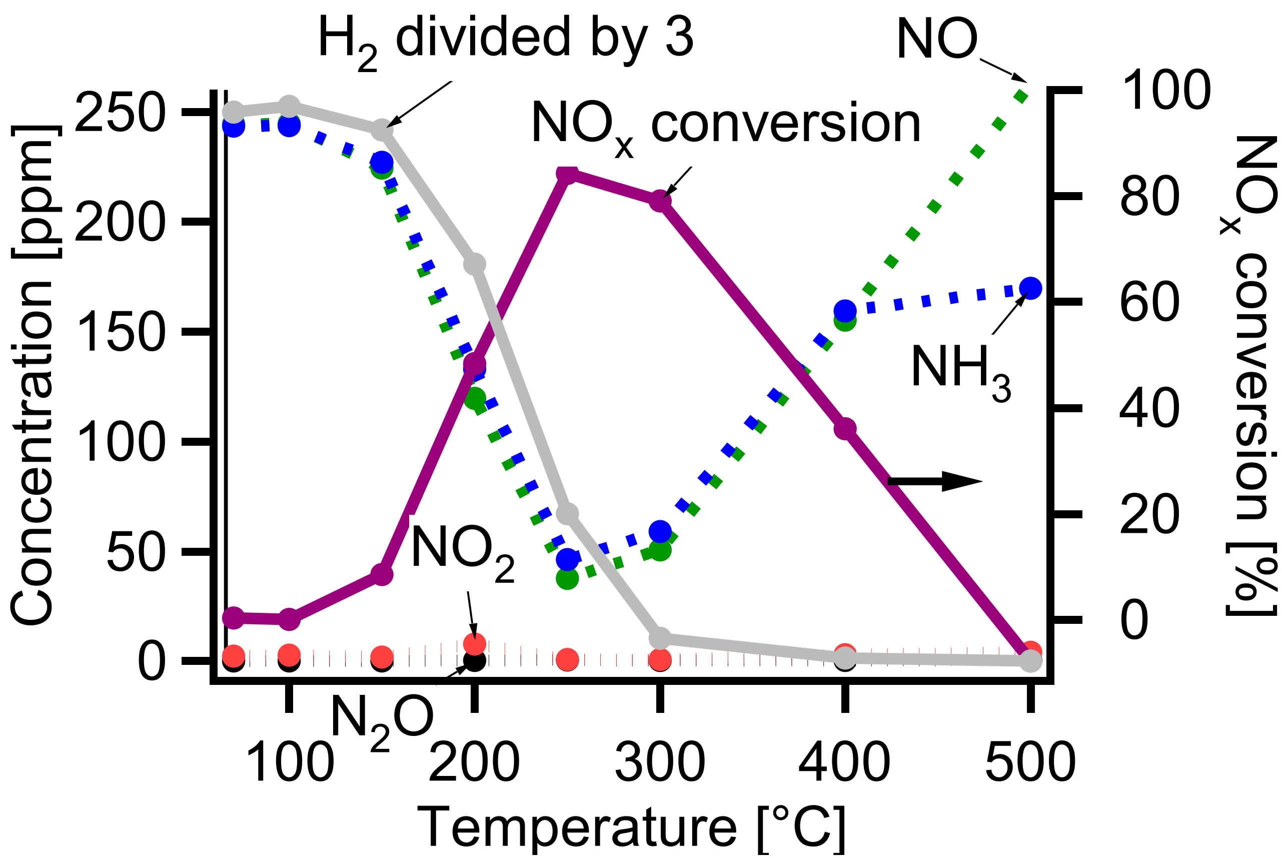
NH_3-SCR over Ag/Al_2O_3

- High activity at low temperature
- H_2 is needed for NH_3-SCR



- Concitions: 1000 ppm NO, 1000 ppm NH_3, 1 vol%H_2, 6 vol% O_2, 7 vol% H_2O. GHSV = 30000 h^{-1}
- M. Richter, R. Fricke, R. Eckelt, Catalysis Letters 94 (2004)115-118.

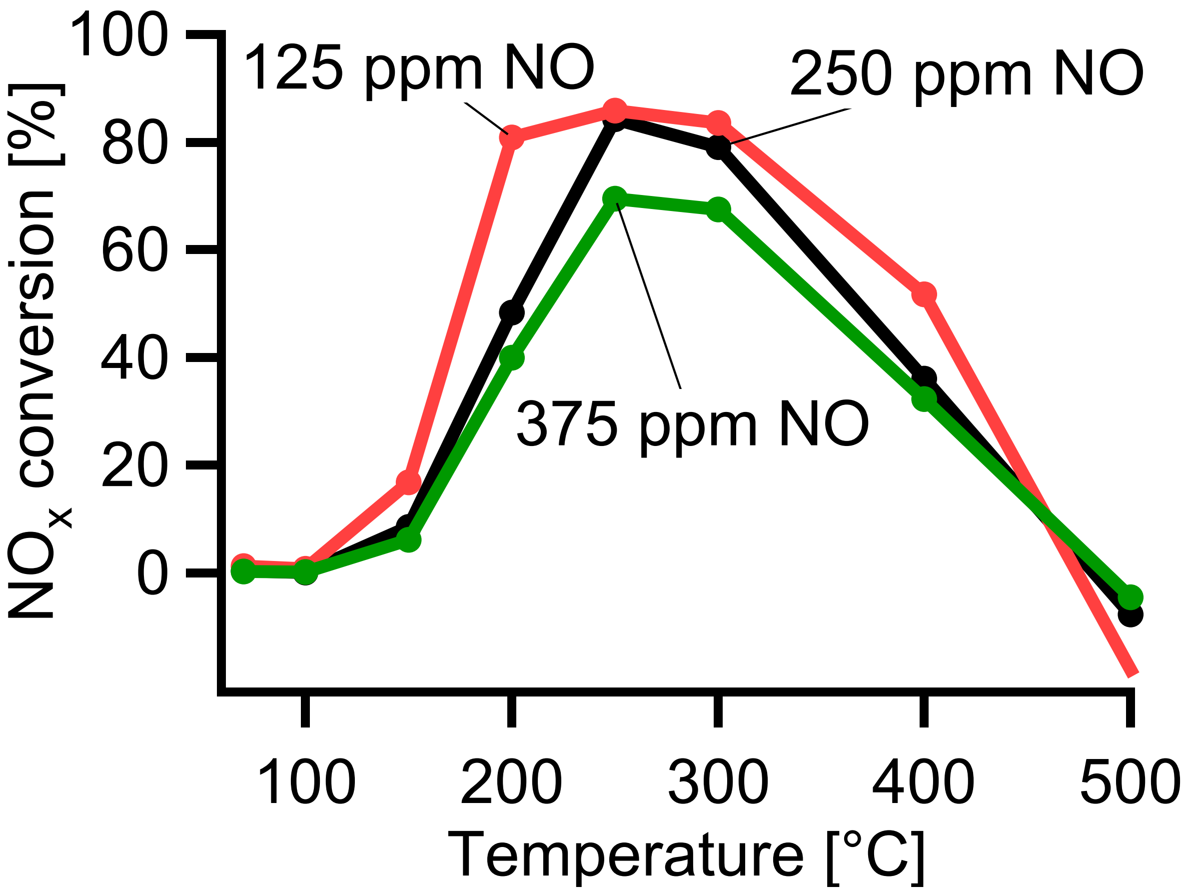
H_2-assisted NH_3-SCR - own results



- 6 wt% Ag on Al_2O_3
- Presulfated monolith catalyst
- 250 ppm NO
- 250 ppm NH_3
- 750 ppm H_2
- 10% O_2
- 5% H_2O
- in Ar
- GHSV =33 100 h^{-1}

- The hydrogen concentration is divided by 3.

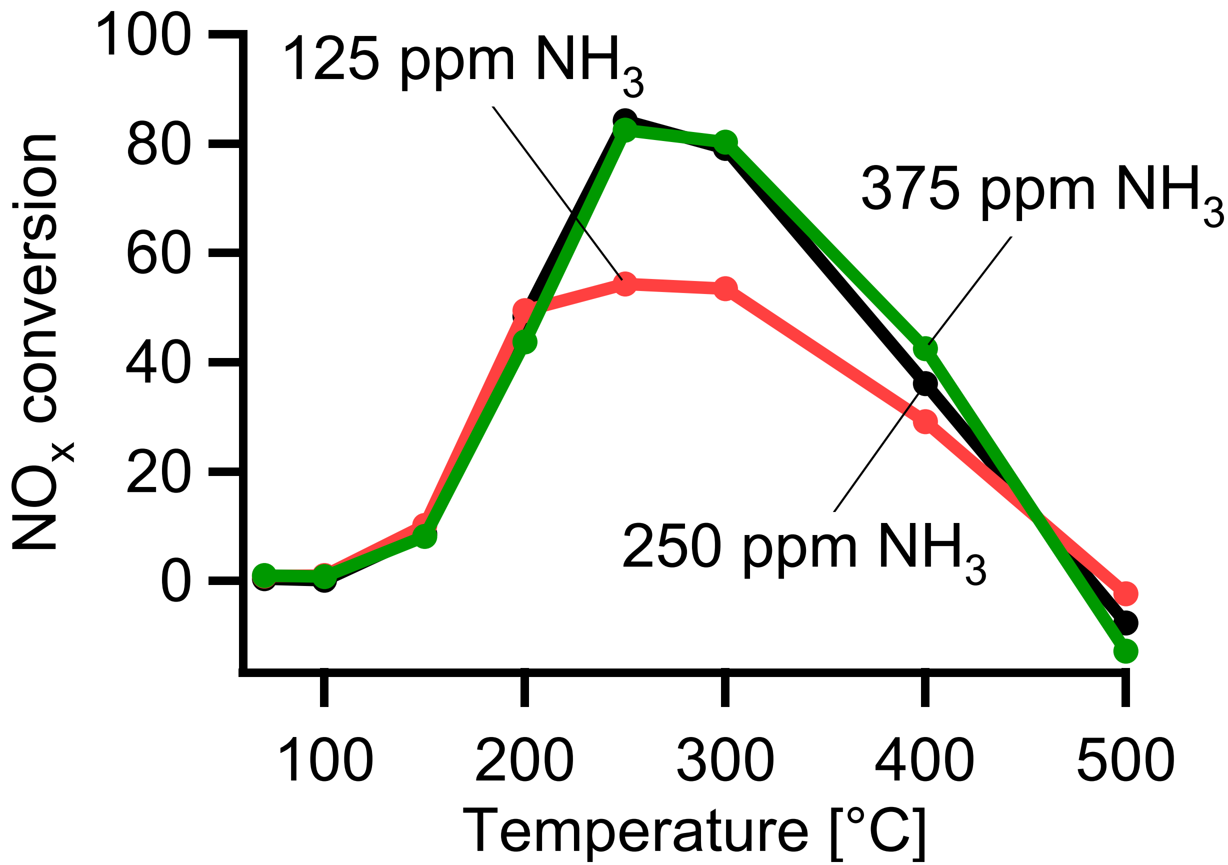
NO variation



- 125 – 375 ppm NO
- 250 ppm NH_3
- 750 ppm H_2
- 10% O_2
- 5% H_2O
- in Ar
- GHSV =33 100 h^{-1}

- NO_x conversion decreases with increasing NO concentration.

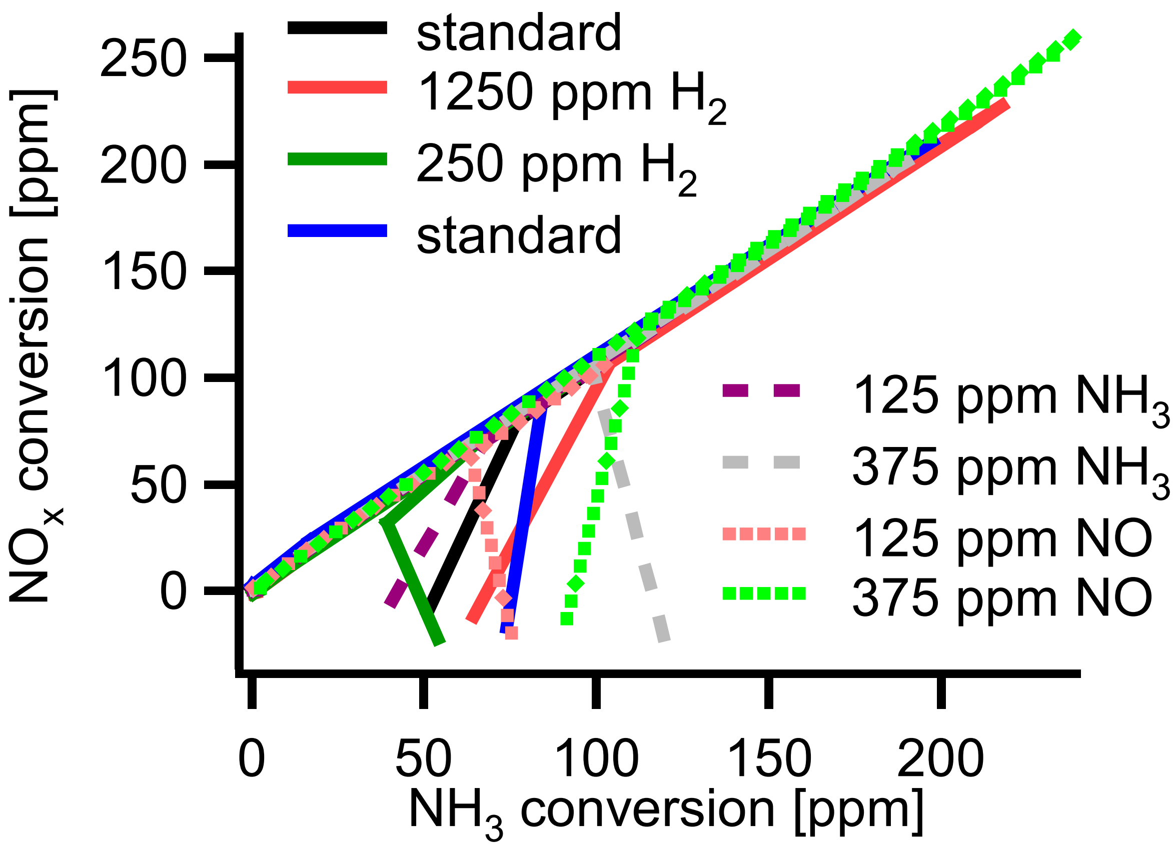
NH_3 variation



- 250 ppm NO
- 125 – 375 ppm NH_3
- 750 ppm H_2
- 10% O_2
- 5% H_2O
- in Ar
- GHSV =33 100 h^{-1}

- NO_x conversion increases with increasing NH_3 concentration to a ratio of 1:1.

NO:NH_3 ratio



Standard

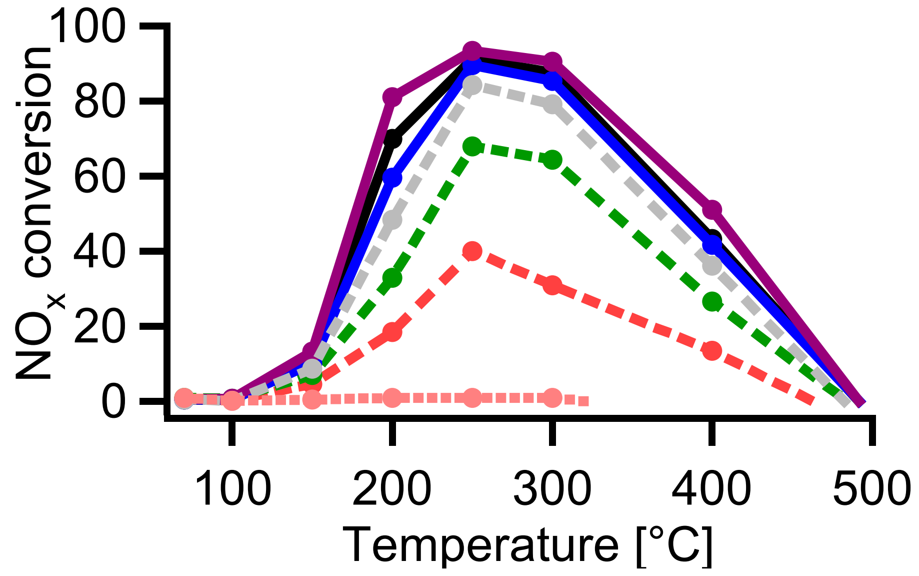
- 250 ppm NO
- 250 ppm NH_3
- 750 ppm H_2
- 10% O_2
- 5% H_2O
- in Ar
- GHSV =33 100 h^{-1}

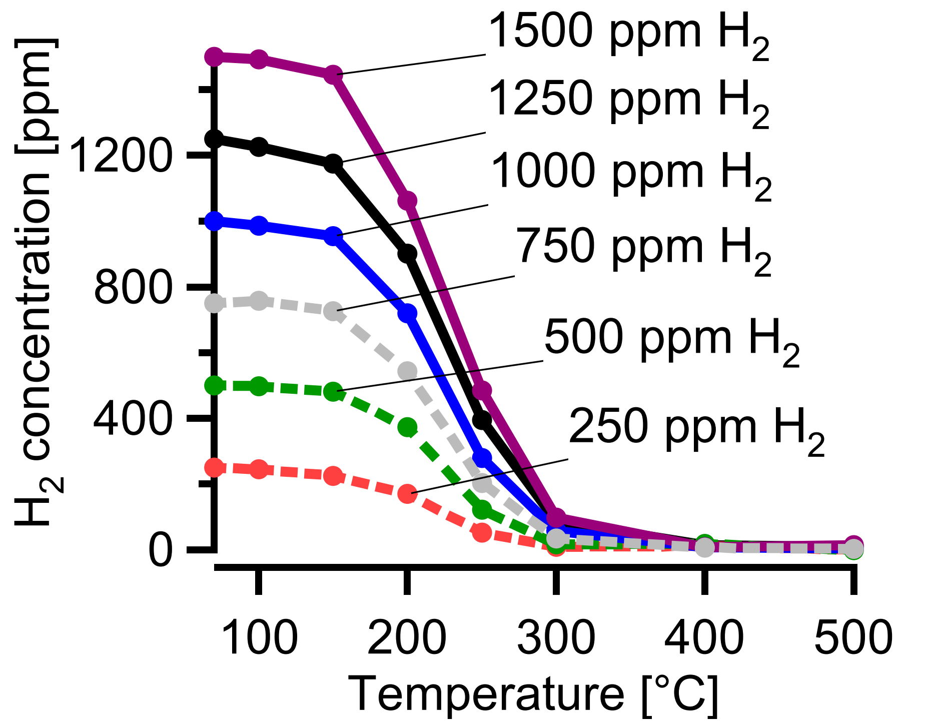
- The ratio between NO and NH_3 conversion was in all experiments1:1.

Variation of H_2

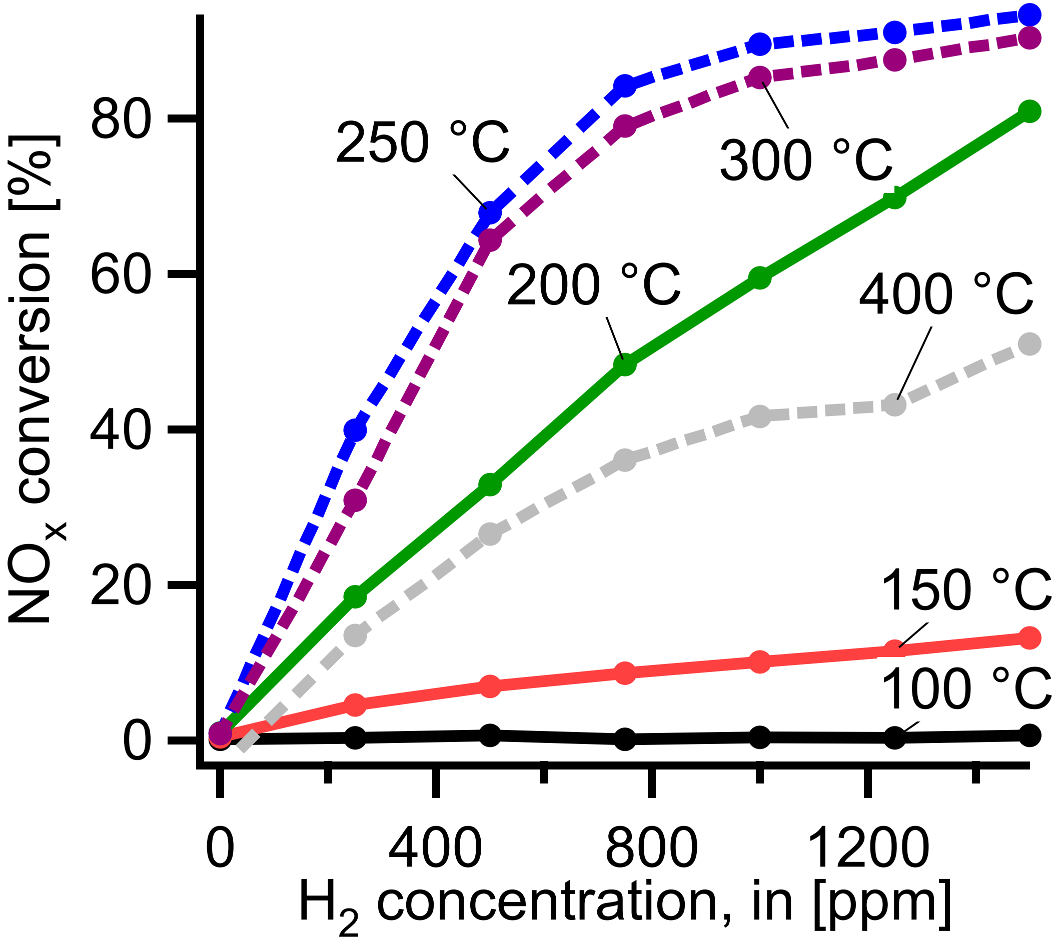
- Higher NO_x conversion with higher H_2 concentration
- Above 300ºC all H_2 is converted
- H_2 conversion is independent on NO : H_2 ratio

- 250 ppm NO
- 250 ppm NH_3
- 0-1500 ppm H_2
- 10% O_2
- 5% H_2O
- in Ar
- GHSV = 33 100 h^{-1}





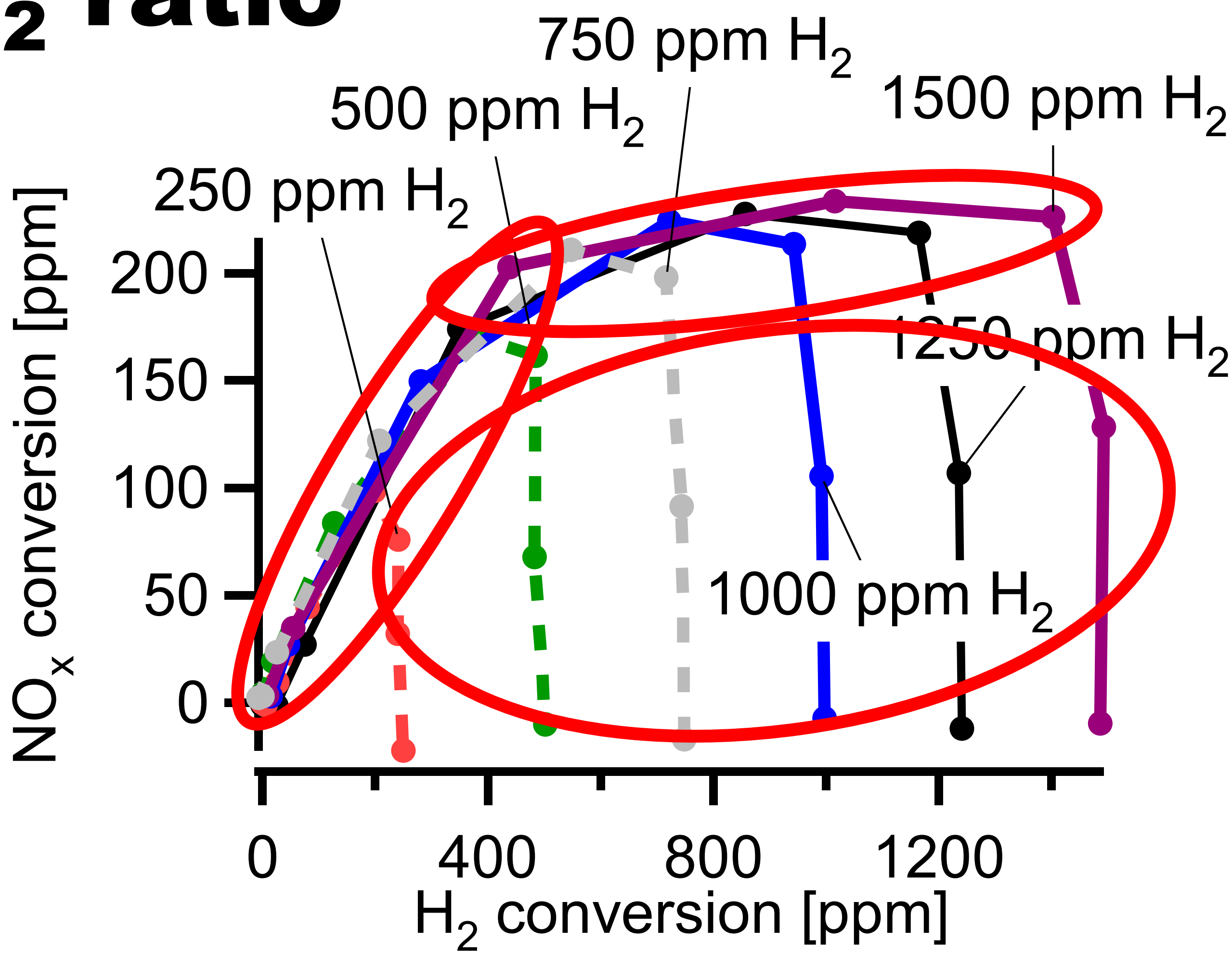
Variation of H_2



- 250 ppm NO
- 250 ppm NH_3
- 0 - 1500 ppm H_2
- 10% O_2
- 5% H_2O
- in Ar
- GHSV = 33 100 h^{-1}

- NO_x conversion levels out at 250 and 300 ºC but increases constantly at 150, 200 and 400 ºC.

NO : H_2 ratio



- The ratio between NO and H_2 conversion was in all experiments1:2 without unselective H_2 oxidation.

Role of H_2

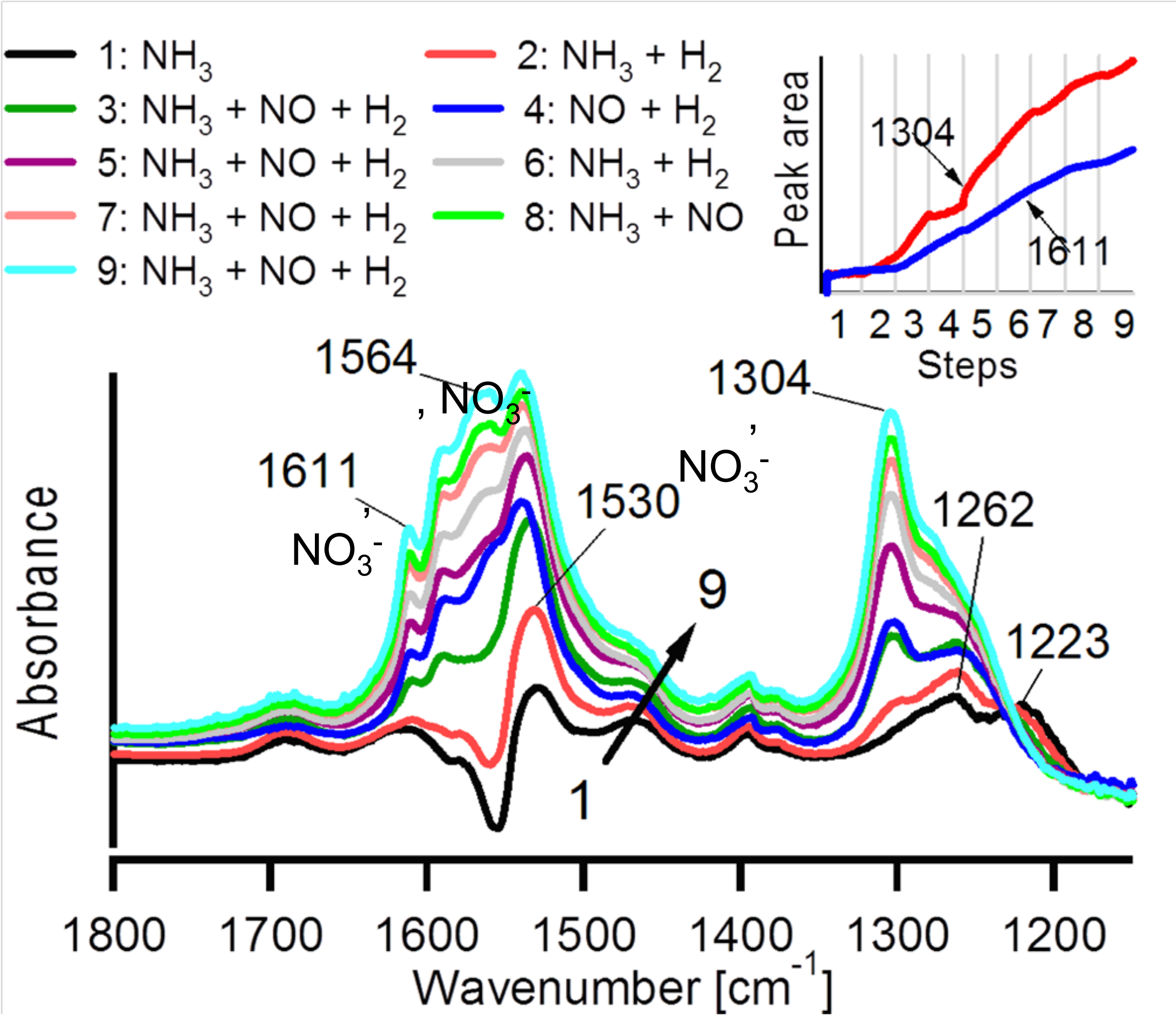
From own experiments

- Limit in NO_x conversion due to H_2
- Defined ratio of $NO:H_2 = 1:2$

Proposals in the literature

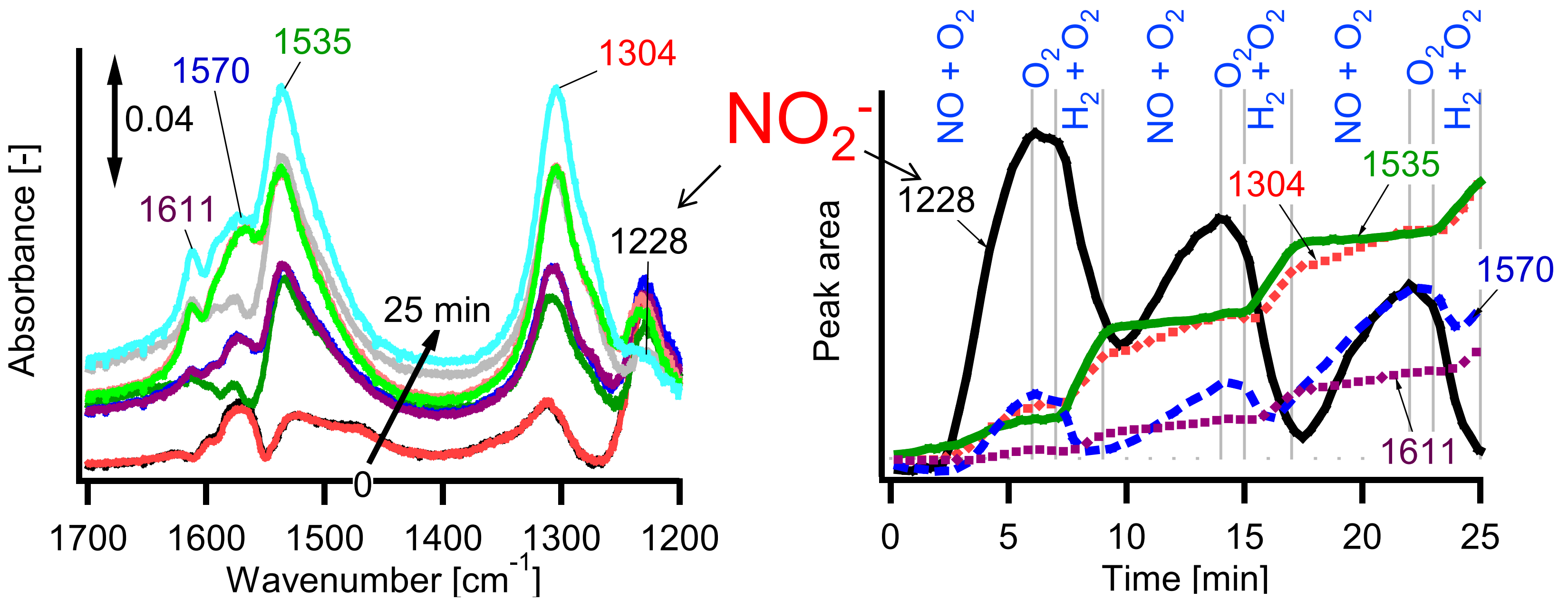
- increase of the number of small silver clusters
- reduction of silver species
- increase of the amount of surface nitrates
- hydrogen reduces or removes nitrates from the catalyst

H_2 reduces nitrates?



- Step – response DRIFT experiment at 250ºC
- NH_3, NO and H_2 are switched in and out
- Step length: 1 h
- **Nitrates increase from step 3**

H_2 assists nitrite conversion



- Step – response DRIFT experiment at 250ºC
- 5 min nitrate/nitrite formation from NO + O_2 → 1 min 10% O_2 → 2 min 1000 ppm H_2 + 10% O_2

Summary

- Ratio between NO : NH_3 : H_2 = 1 : 1 : 2
- Increasing the H_2 concentration => NO_x conversion levels out at 250 and 300ºC but increases constantly at 150, 200 and 400ºC.
- H_2 concentration can limit the NO_x conversion although NH_3 is available in excess.
- H_2 assists the conversion of nitrites to nitrates

Acknowledements

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