Deactivation of Cu/SAPO-34 During Low-Temperature NH_3-SCR

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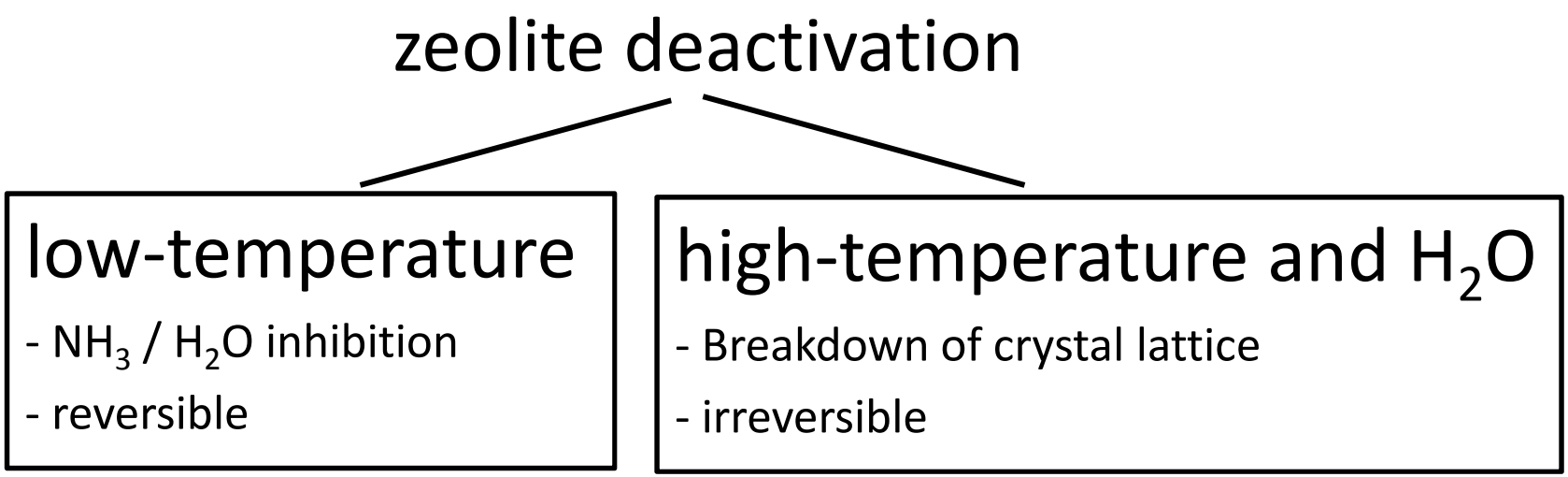
Introduction



Small-pore zeolites less prone to HT collapse of lattice.

Is the Cu/SAPO-34 structure stable at low T?

Experimental Methods

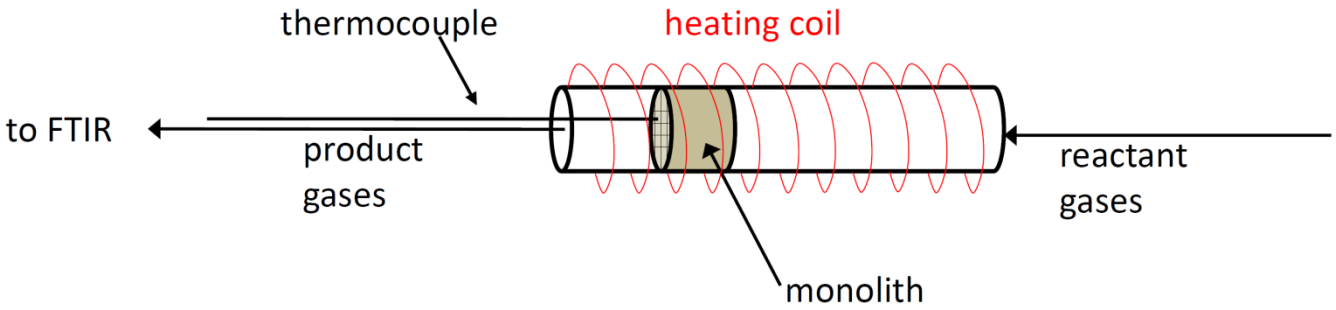
SAPO-34: hydrothermal synthesis

Cu/SAPO-34
1.27 wt.% Cu

Aqueous ion exchange

Cu/SAPO-34
2.60 wt.% Cu

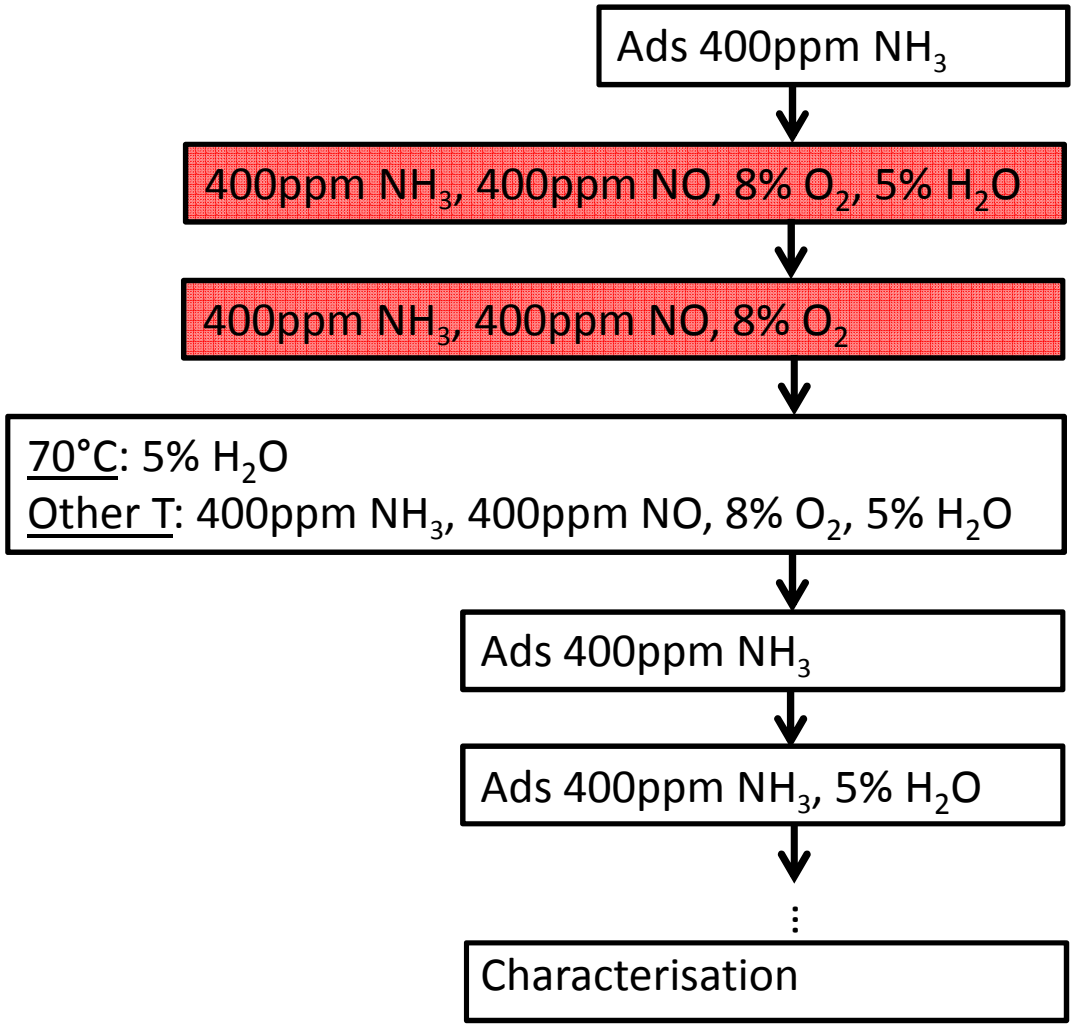
Exposure to SCR conditions and water vapour



Degreening: 700 °C
3500 ml/min
400 ppm NH_3 / NO
5% H_2O (φ=16%)
8% O_2
T: 70-200 °C

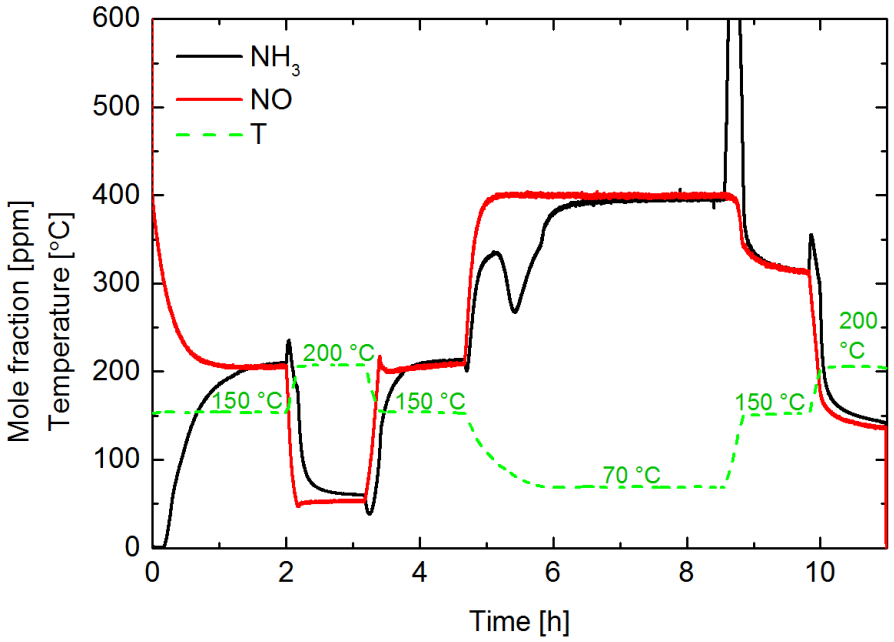
Characterisation of Cu/SAPO-34 before (powder) and after (scraped off monolith) experiments

Experiments Performed in Order

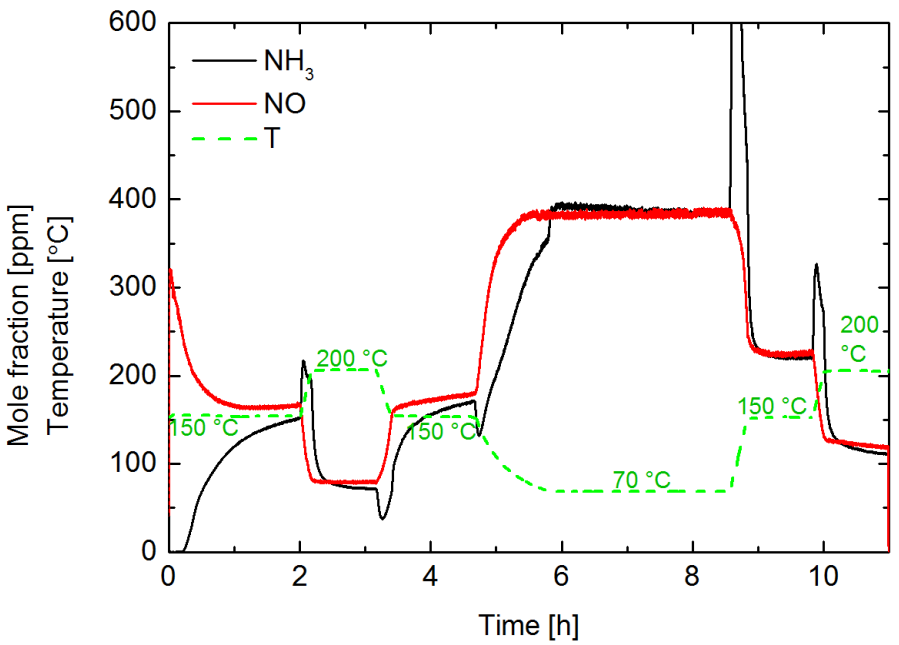


SCR over Cu/SAPO-34 (2.60 wt.%Cu)

SCR+H_2O



SCR



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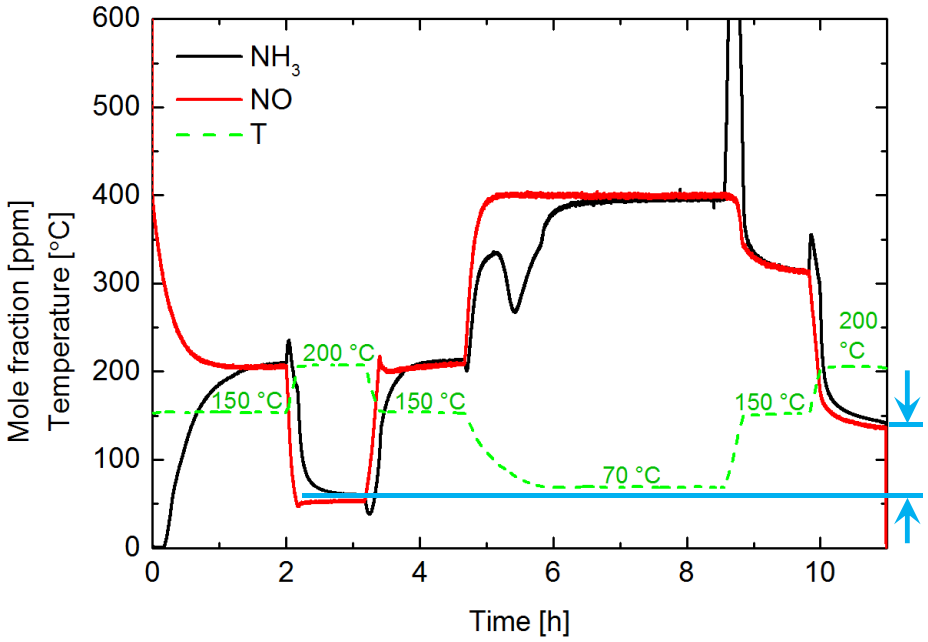
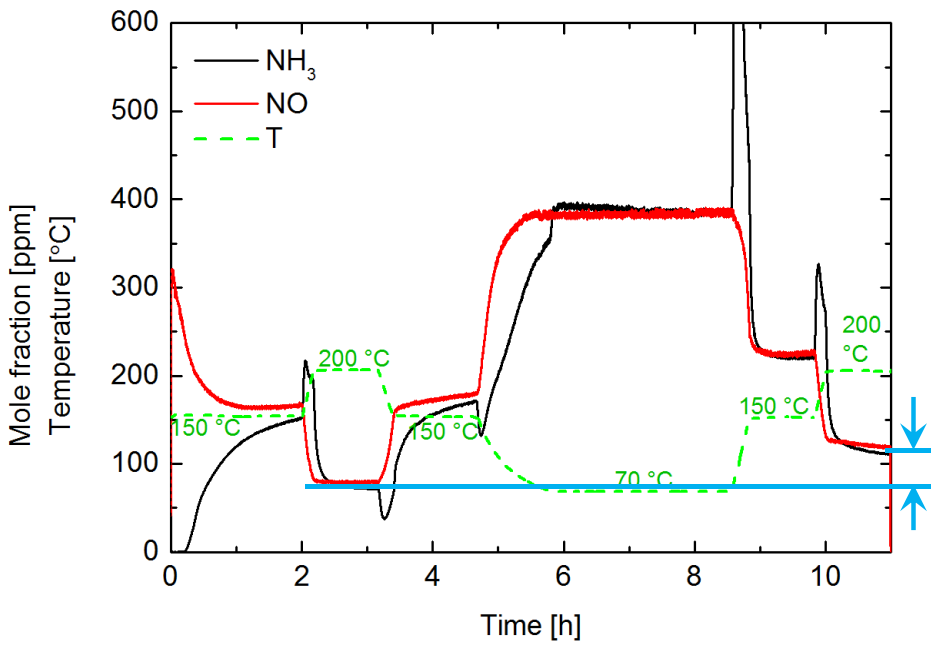


Loss of activity: from 87 to 66%

SCR



No H_2O: Loss of activity smaller

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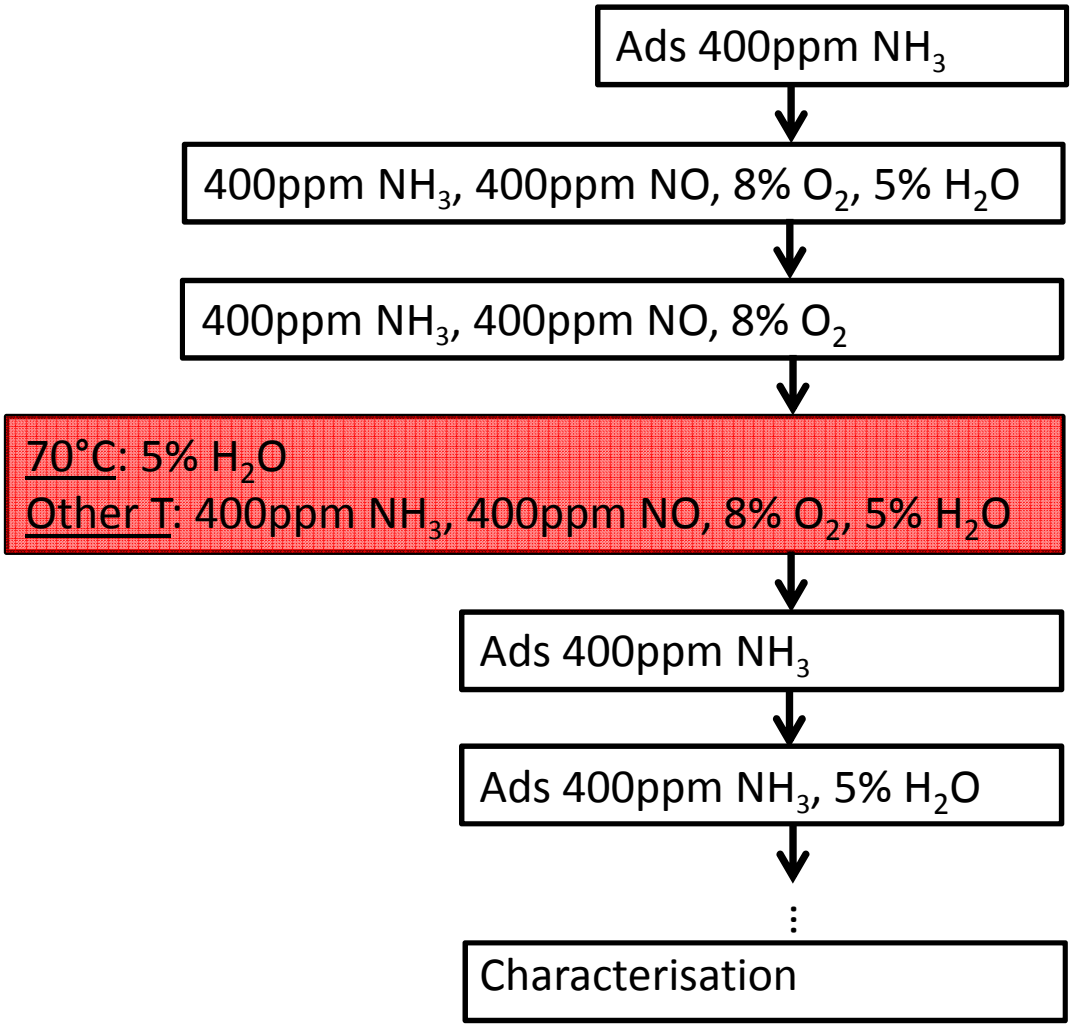
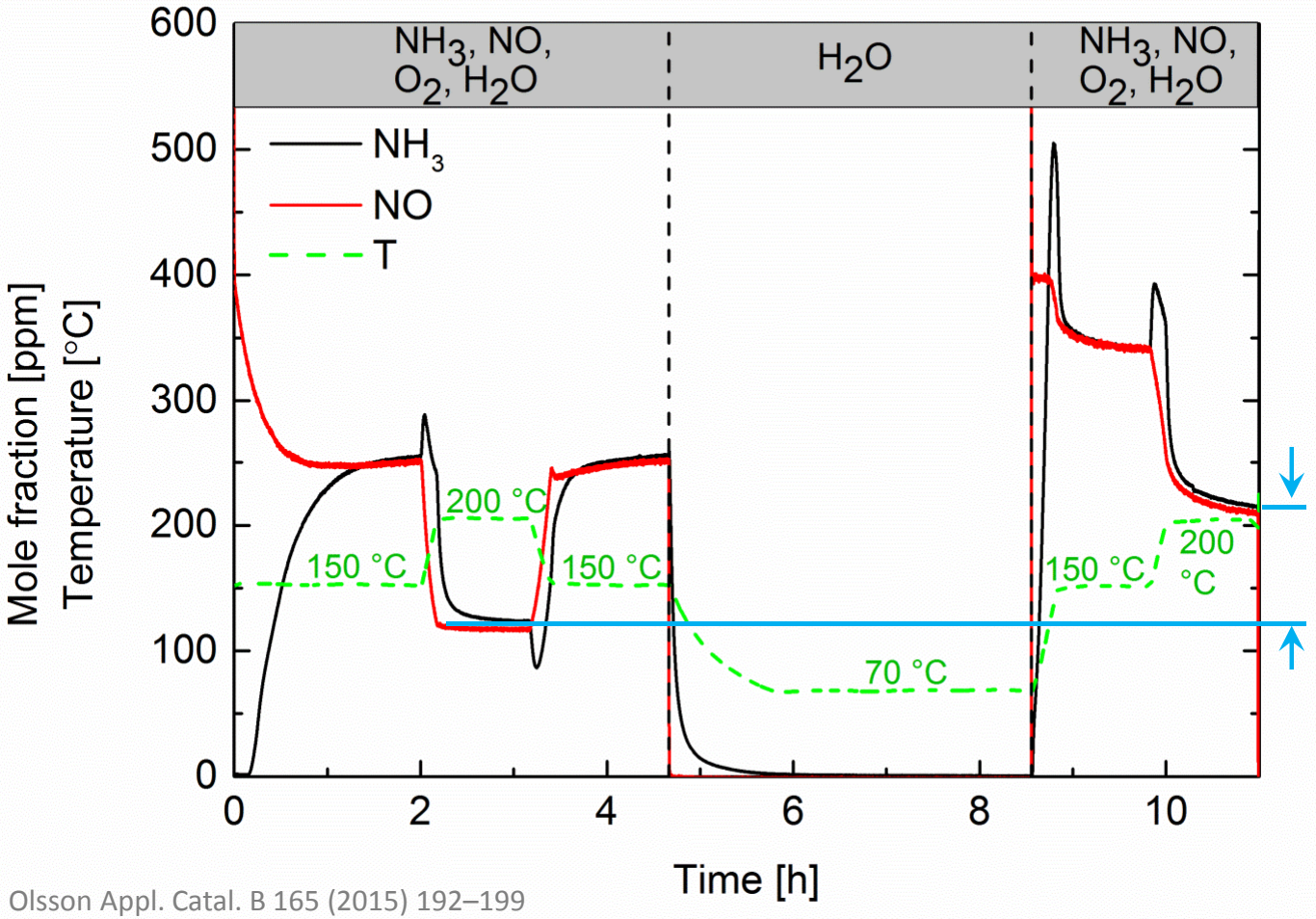
Experiments Performed in Order



SCR over Cu/SAPO-34 (2.60 wt.%Cu)
Only water vapour at 70 °C



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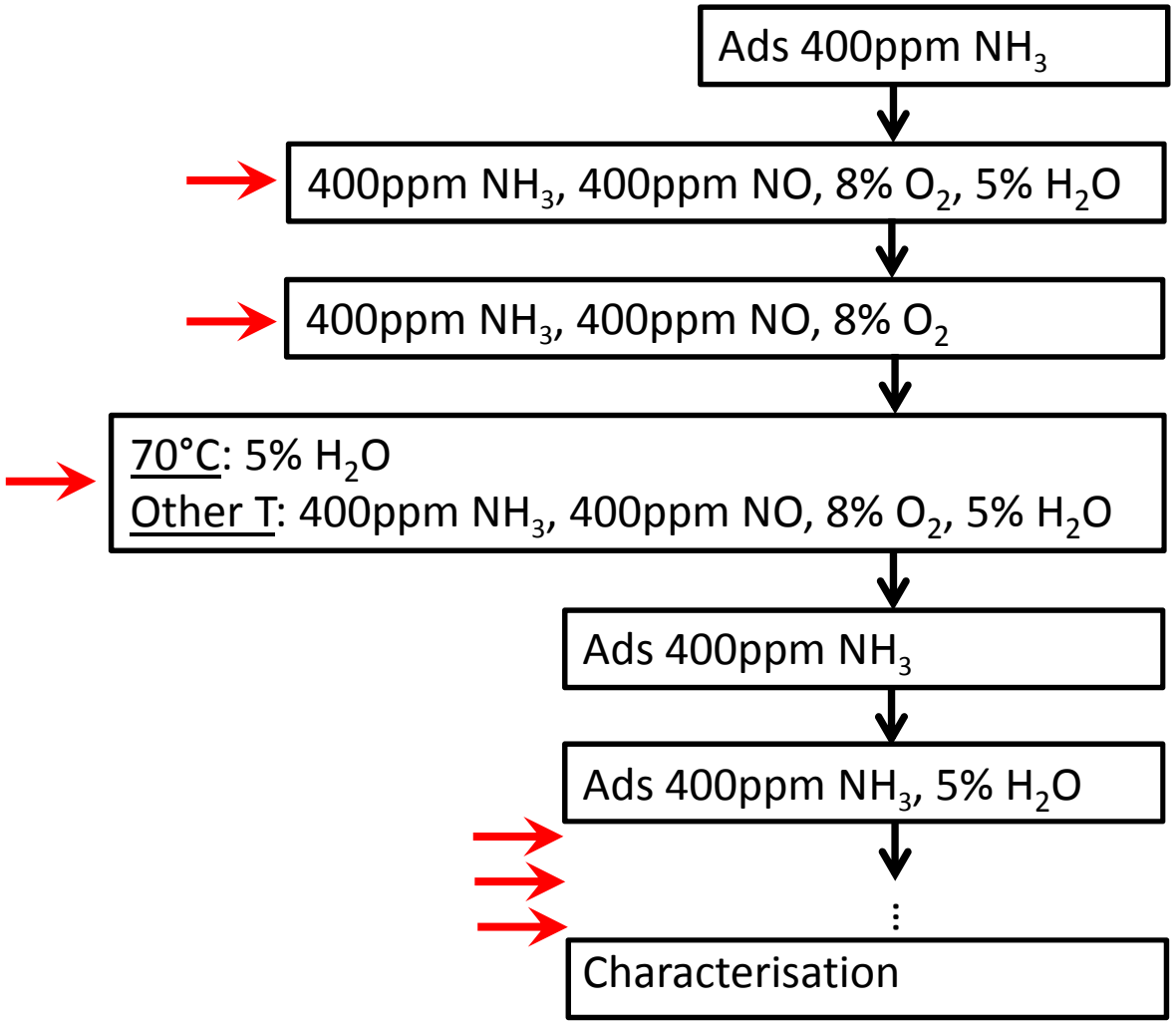
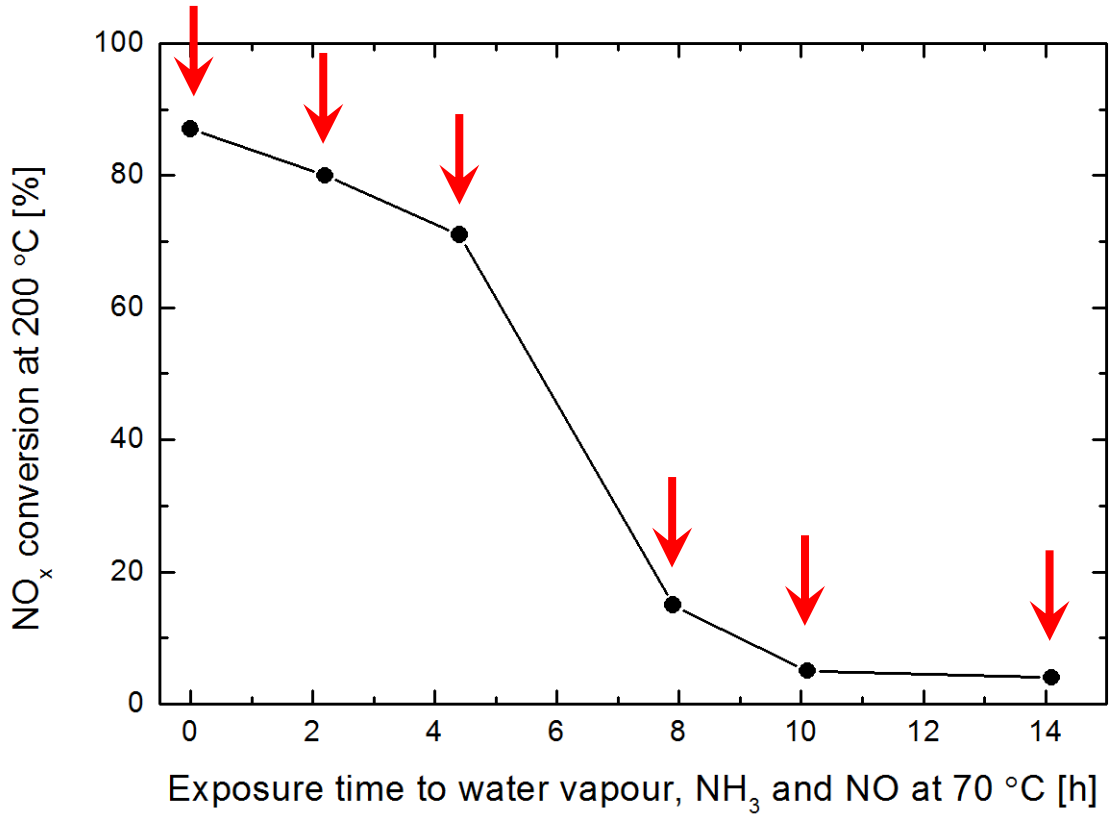
Experiments Performed in Order



Loss of Activity Over Time (2.60 wt.%Cu)



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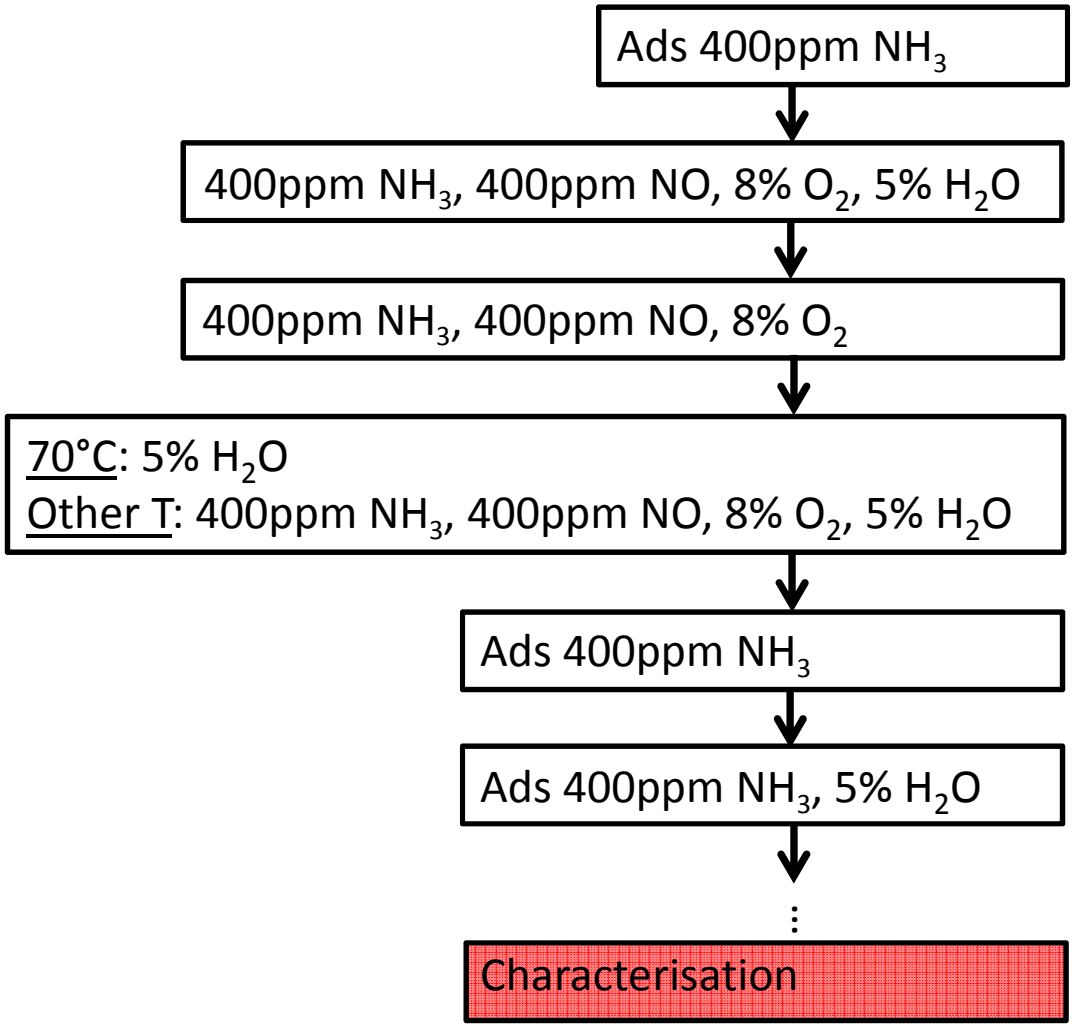
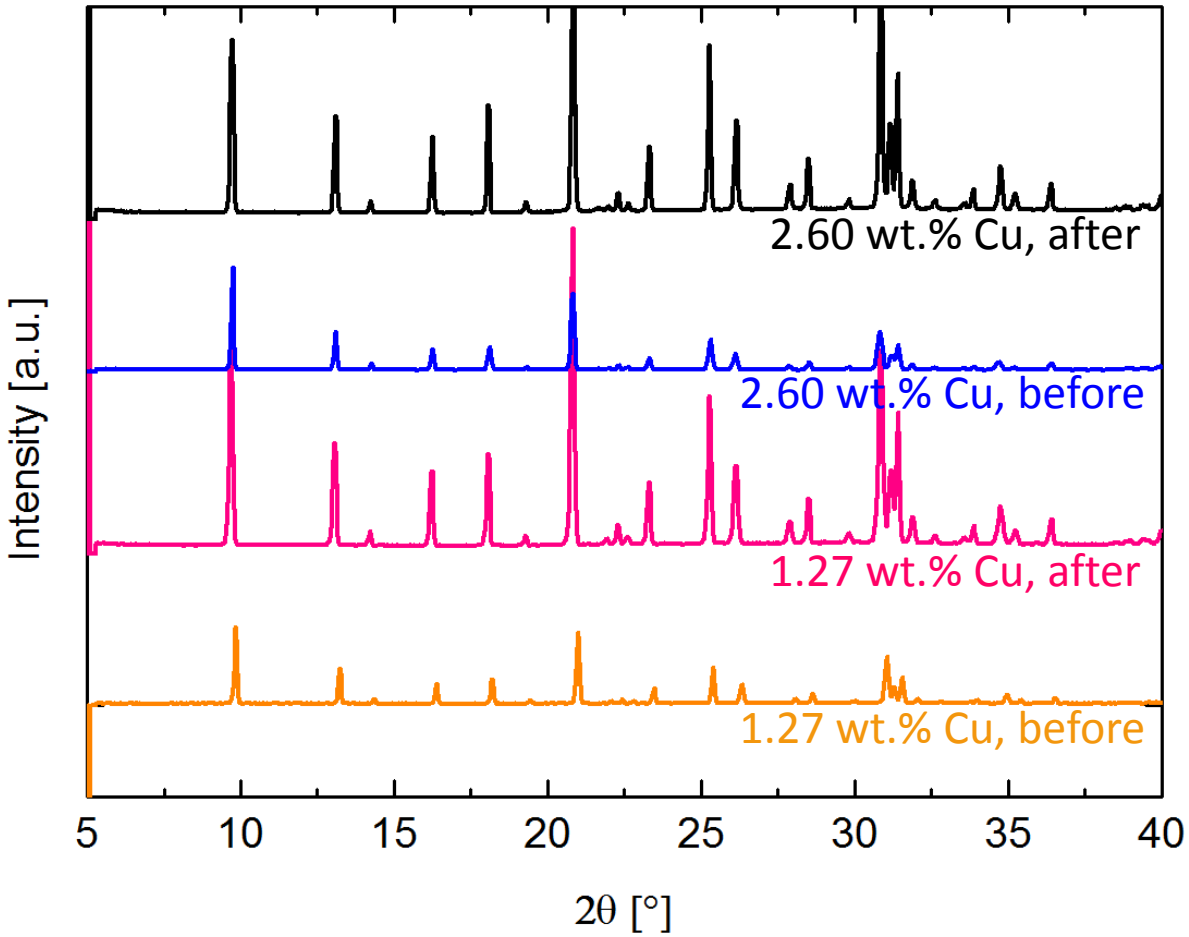
Experiments Performed in Order



Loss of Crystallinity?

XRD



BET

544 m^2/g

↓

473 m^2/g

582 m^2/g

↓

420 m^2/g

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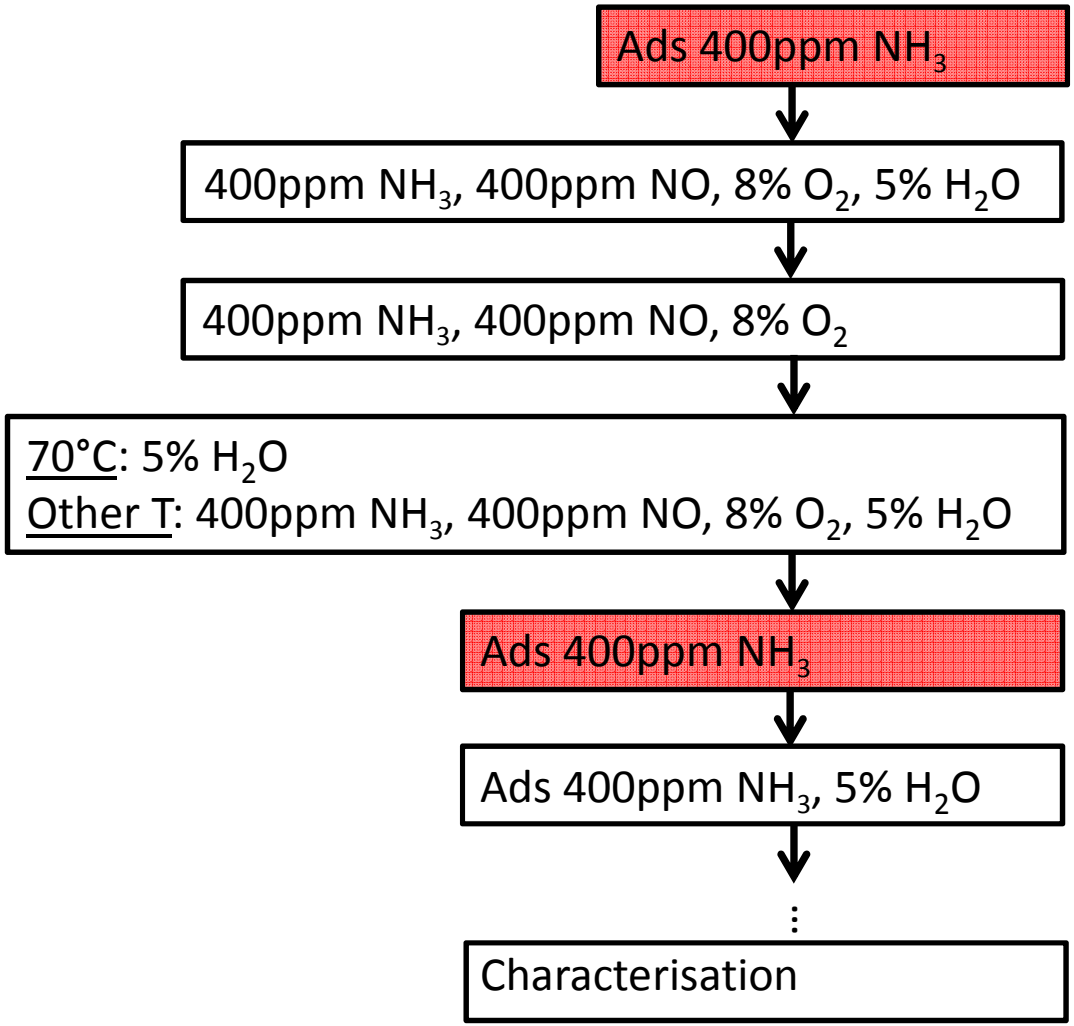
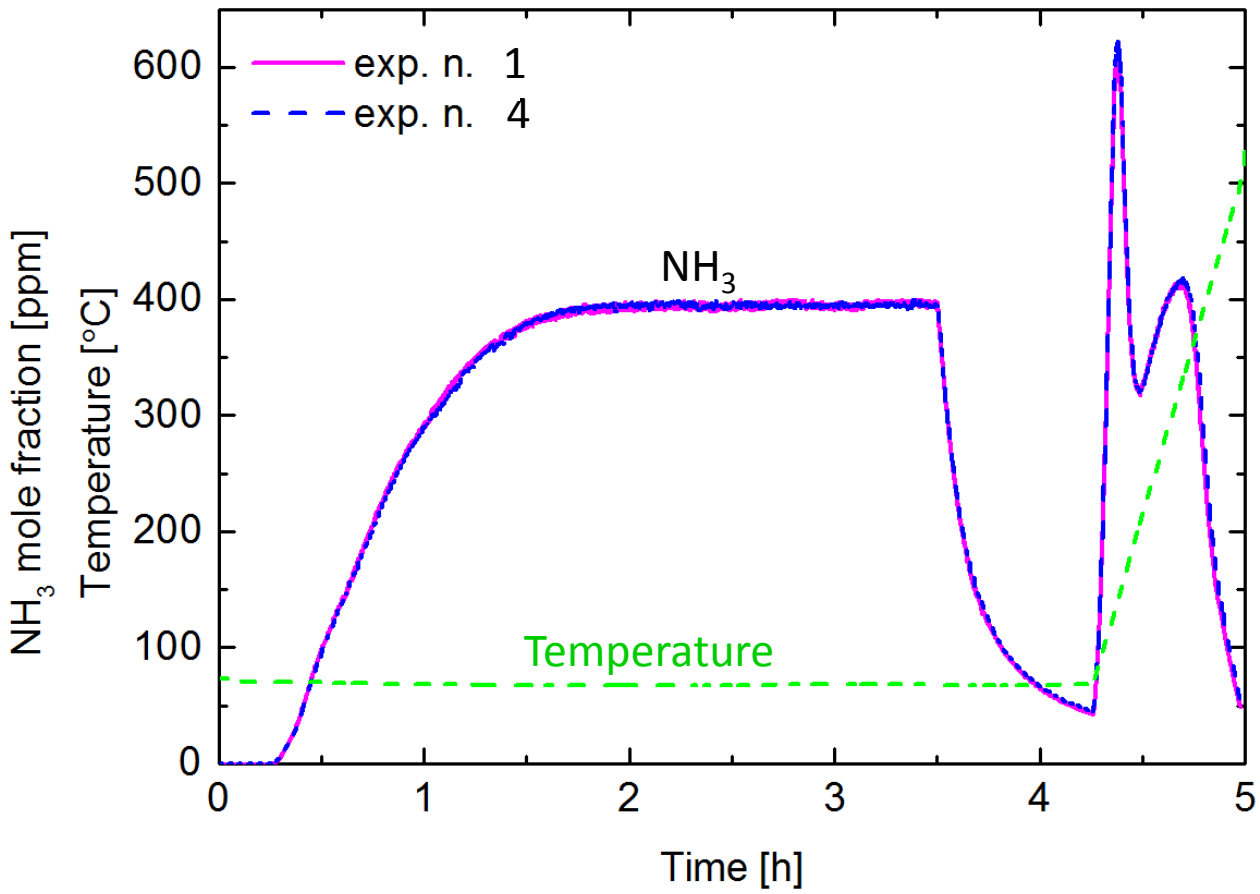
Experiments Performed in Order



NH_3 Adsorption over Cu/SAPO-34 (2.60 wt.%Cu)



NH_3 storage identical after ca. 4.5h exposure to water vapour at 70 °C

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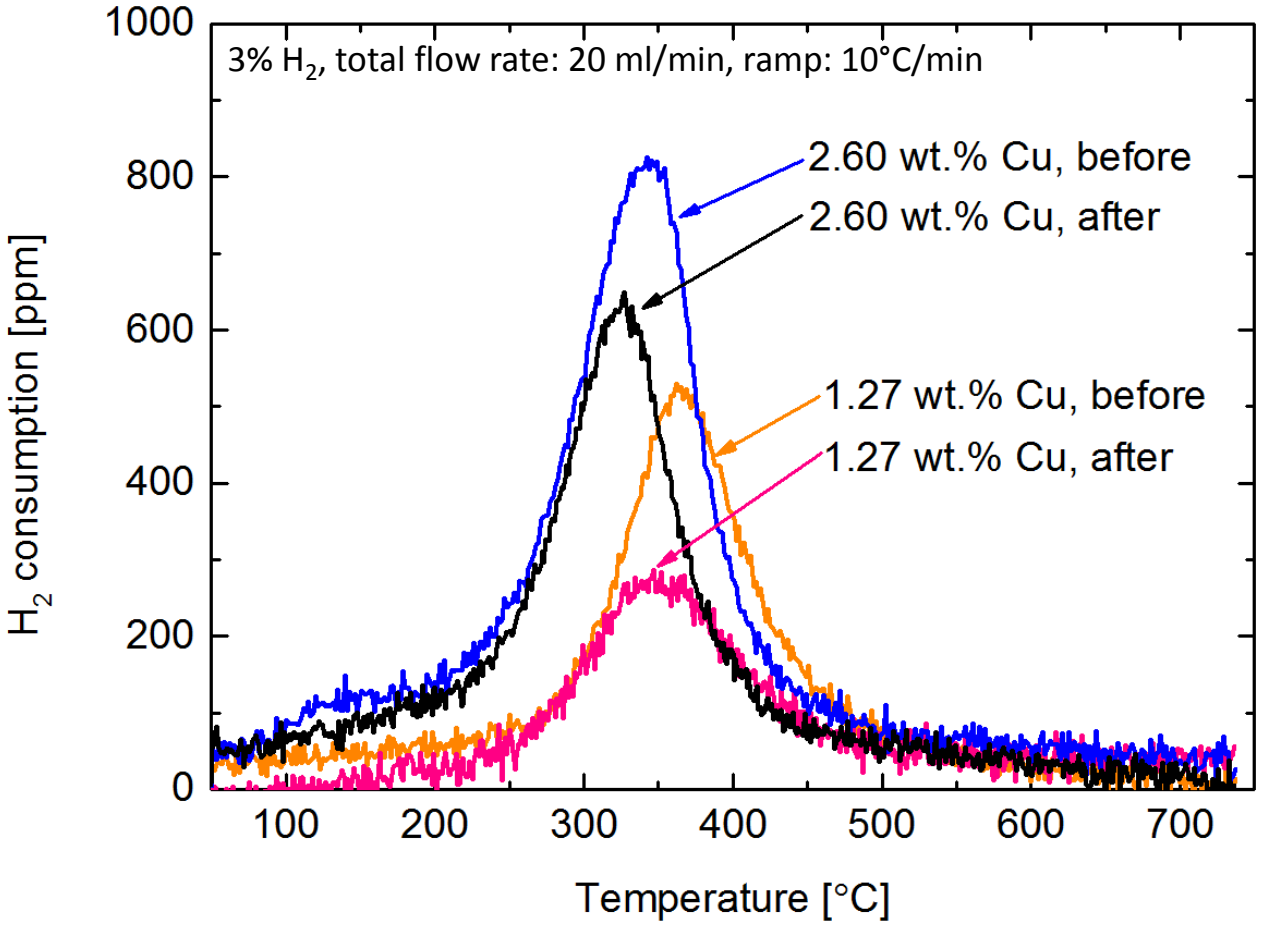
H_2-TPR



Hydrogen consumption decreased by 26 and 38% (or less: up to 13% binder in "after" samples) - small compared to loss of activity

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Conclusions

- Synthesis of 1.27 wt.% Cu and 2.60 wt.% Cu Cu/SAPO-34
- 14 h enough for complete deactivation
- 600 °C treatment does not reverse deact.
- Plugging of pores and blocking of sites ruled out
- Breakage of crystal framework not dominating

→ transformation of copper sites

Acknowledgements

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