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Marie-Laure Tarot, Fabien Can, Daniel Duprez, Vincent Lauga, Eduard Emil Iojoiu, et al.. Influence of sodium loading on the deactivation of Cu/FER exchanged zeolites for SCR of NO_x with NH₃. Effect of copper loading. EuropaCat 2017, Aug 2017, Florence (IT), Italy. hal-04595905

HAL Id: hal-04595905

<https://hal.science/hal-04595905>

Submitted on 3 Jun 2024

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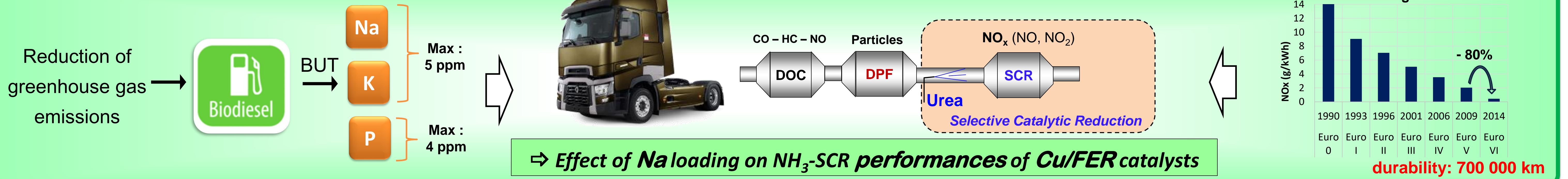
Influence of sodium loading on the deactivation of Cu/FeR exchanged zeolites for SCR of NO_x with NH₃.

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CONTEXT



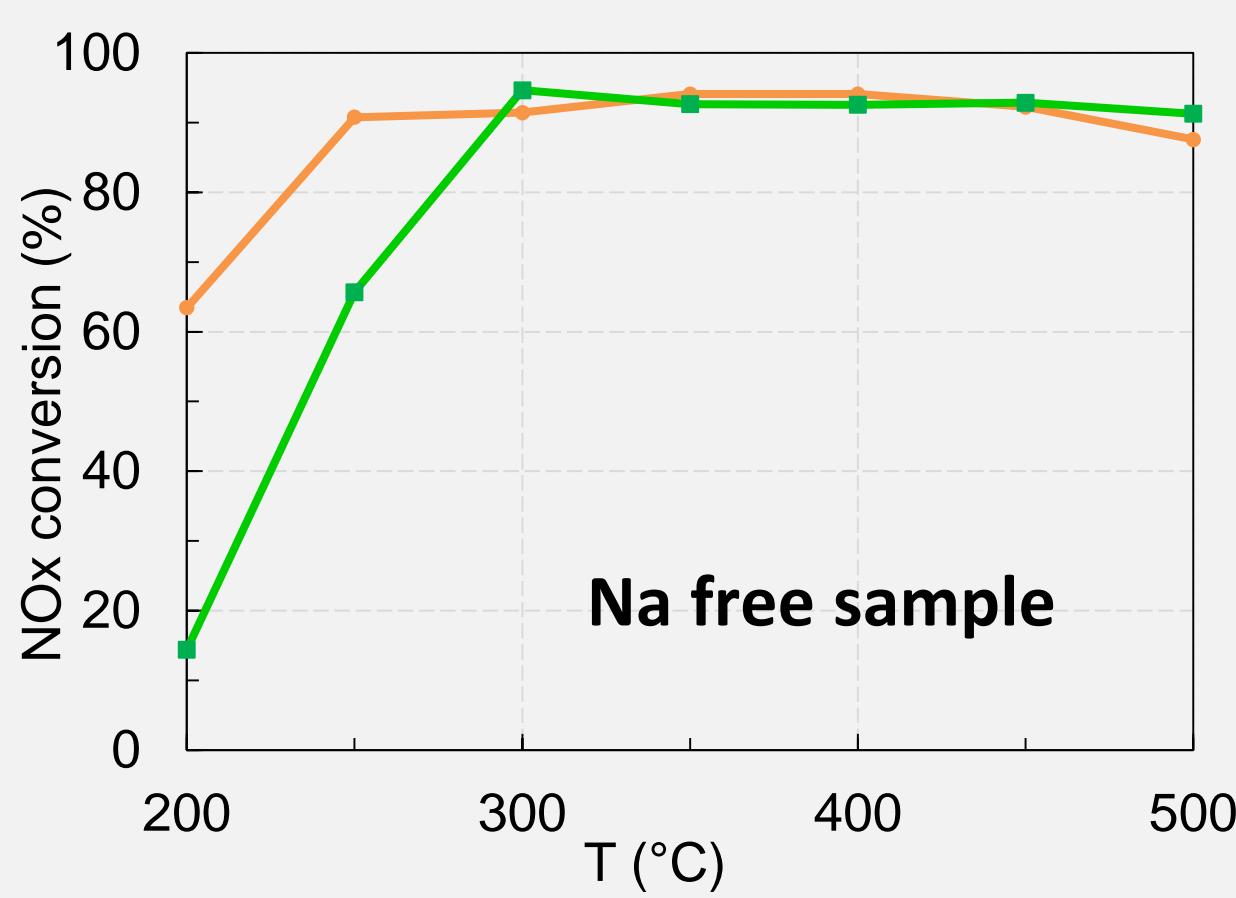
Catalyst, Na addition & reference tests

Catalyst: Cu(2.8%)/FeR (zeolite with small channels) prepared by ion-exchange. Hydrotreatment 16 hours at 600 °C with 10 % O₂, 10% H₂O in N₂.

Na addition: impregnation of Na (precursor NaNO₃), evaluation of two solvents:

Ethanol: limiting interactions between the zeolite and the solvent
Water: Simulate condensation/evaporation of water (cold start)

Experimental conditions: 500 ppm NO (= standard SCR) or 250 ppm NO, 250 ppm NO₂ (= fast SCR)
500 ppm NH₃, 10% O₂, 10% CO₂, 10% H₂O balanced with N₂
(Flow rate: 250 ml.min⁻¹)



fast SCR: w_{cata} = 15 mg
 $2 \text{ NH}_3 + \text{NO} + \text{NO}_2 + \frac{1}{2} \text{ O}_2 \rightarrow 2 \text{ N}_2 + 3 \text{ H}_2\text{O}$

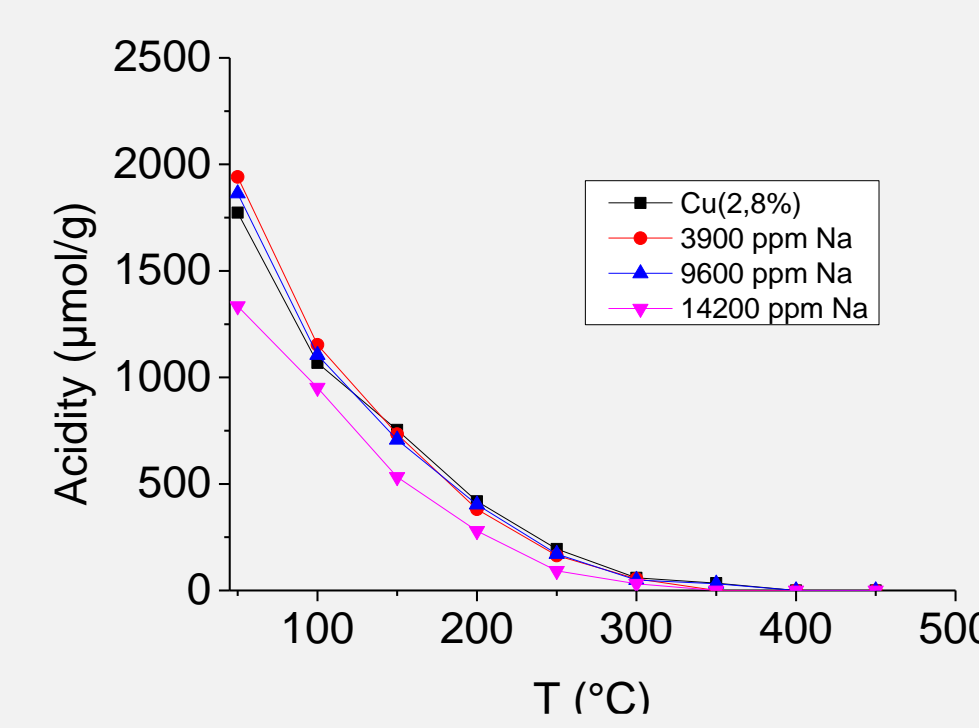
standard SCR: w_{cata} = 50 mg
 $2 \text{ NH}_3 + 2 \text{ NO} \rightarrow 2 \text{ N}_2 + 3 \text{ H}_2\text{O}$

➤ Higher activity in fast-SCR compared to standard-SCR

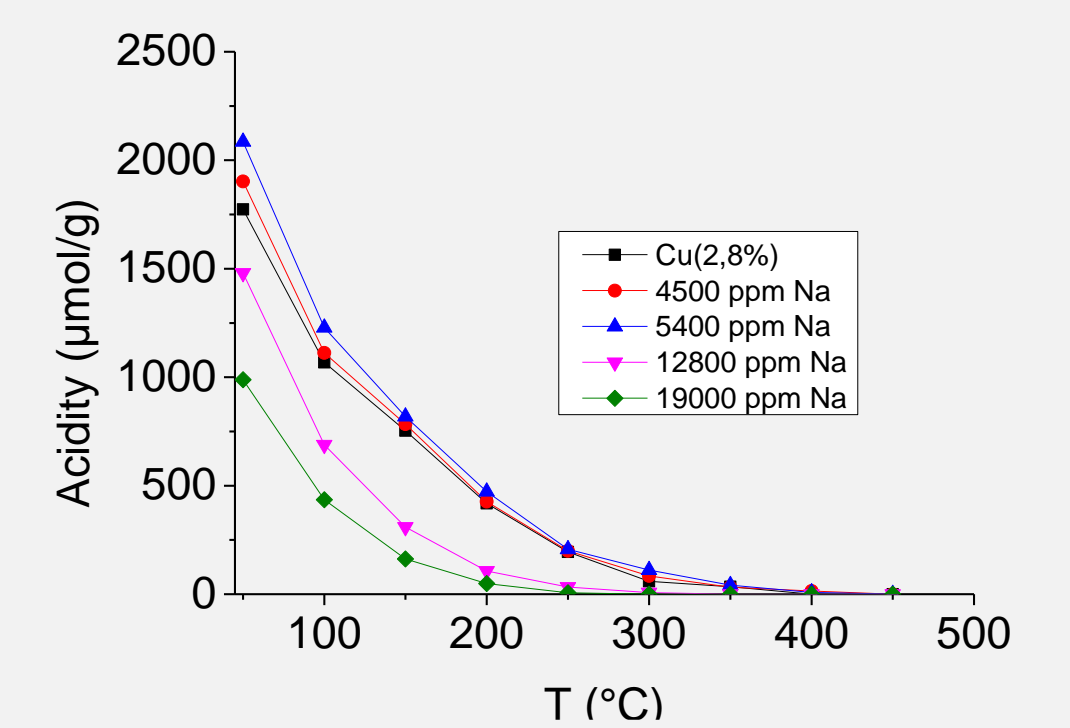
Acidic properties (NH₃ / FT-IR)

NH₃ as a probe for acidity: pyridine do not enter in all the zeolite channels

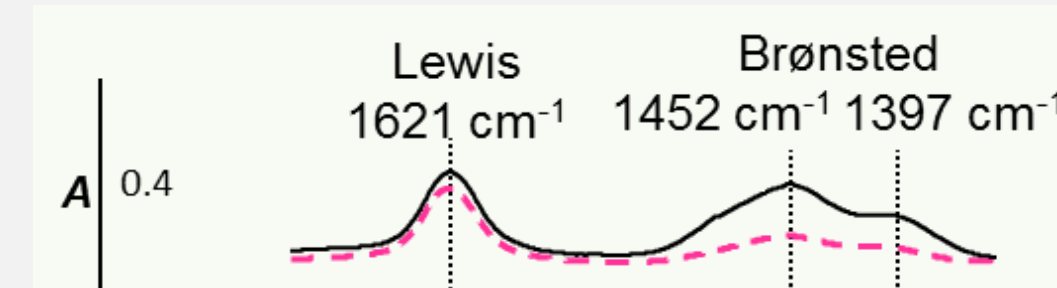
Na imp. in EtOH



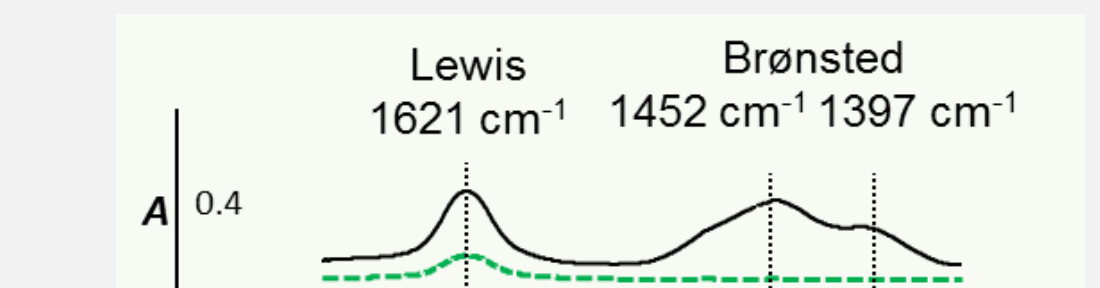
Na imp. in H₂O



H₂O ≈ EtOH; acidity ↓ for [Na] > 13 000 – 14 000 ppm:



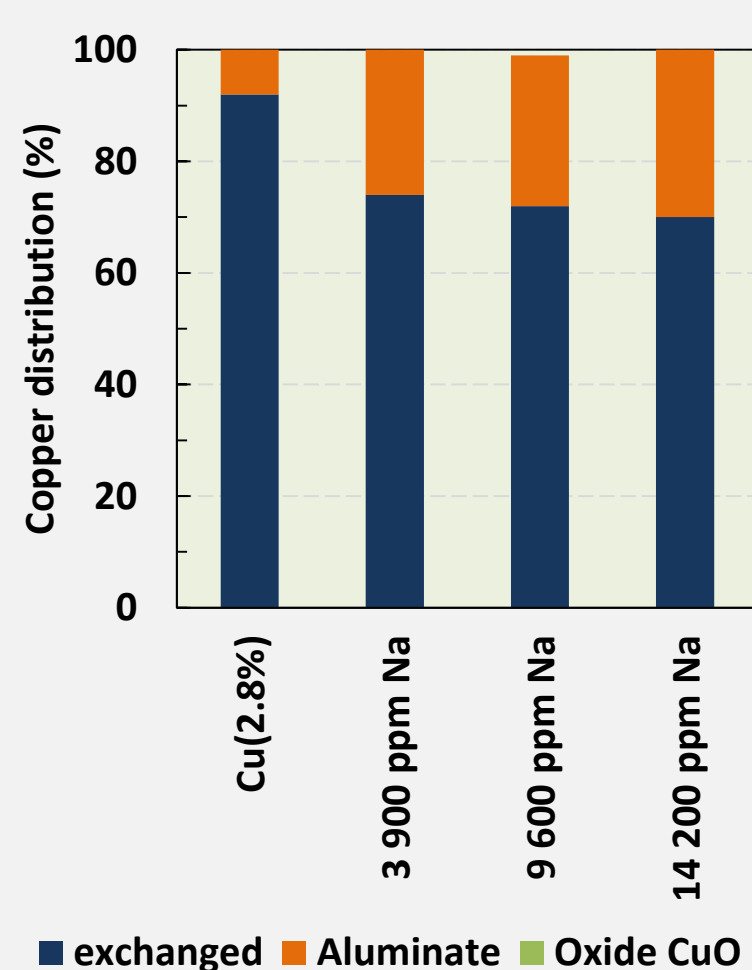
➤ Still Brønsted acidity with 14 200 ppm of Na



➤ Brønsted acidity ↓ preferentially

H₂-TPR - copper state

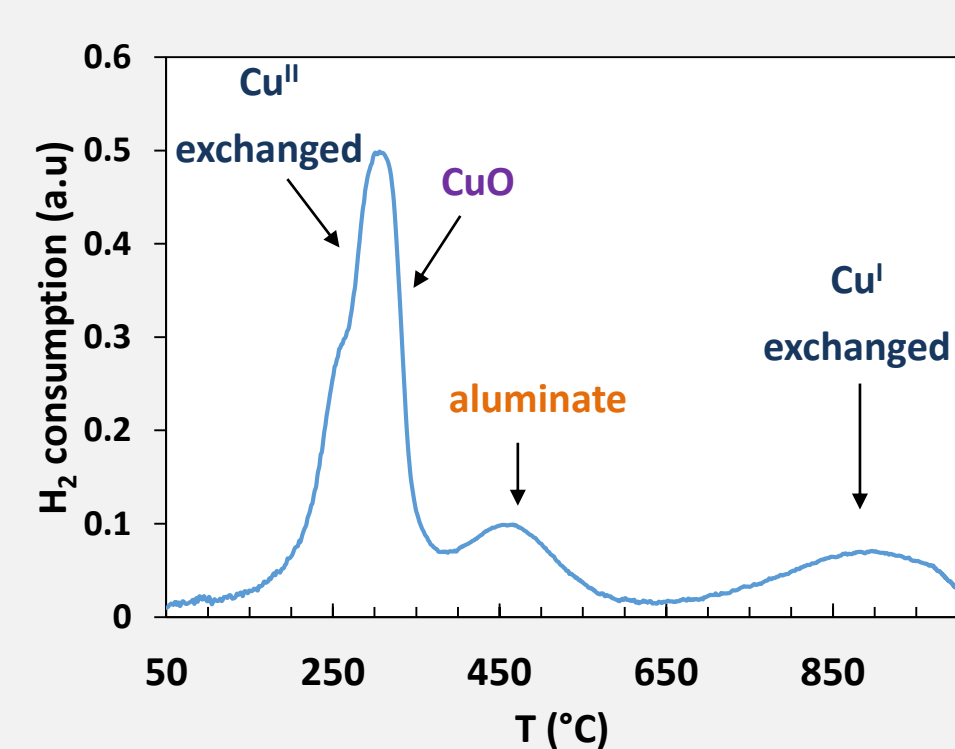
Na imp. in EtOH



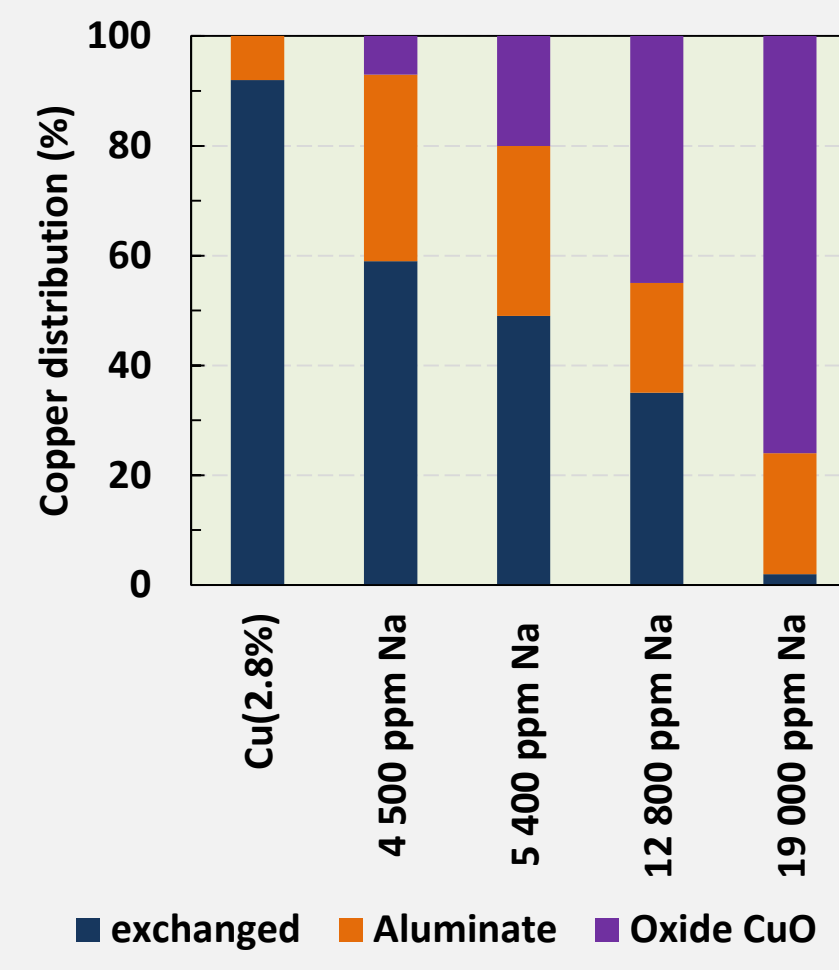
➤ Few change in copper distribution, only ↑ copper aluminate

initial state of copper: only Cu^{II}
 $\frac{\text{Cu}^{\text{loading}}}{\text{H}_2 \text{ consumption}} = 1$

Copper distribution (Cu^I exchanged = Cu^{II} exchanged)



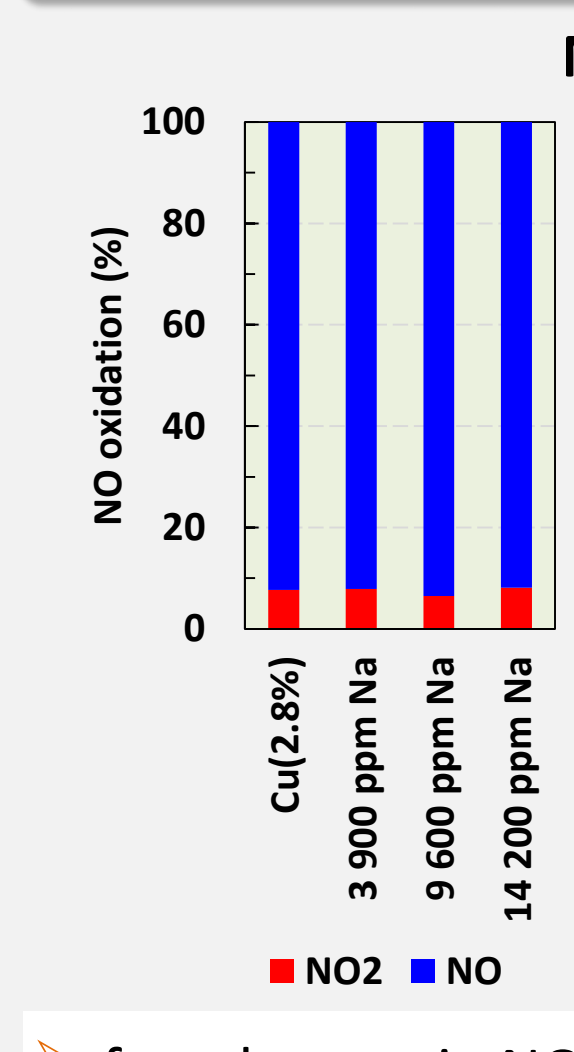
Na imp. in H₂O



➤ ↓ exchanged copper
➤ ↑ CuO (extra framework)

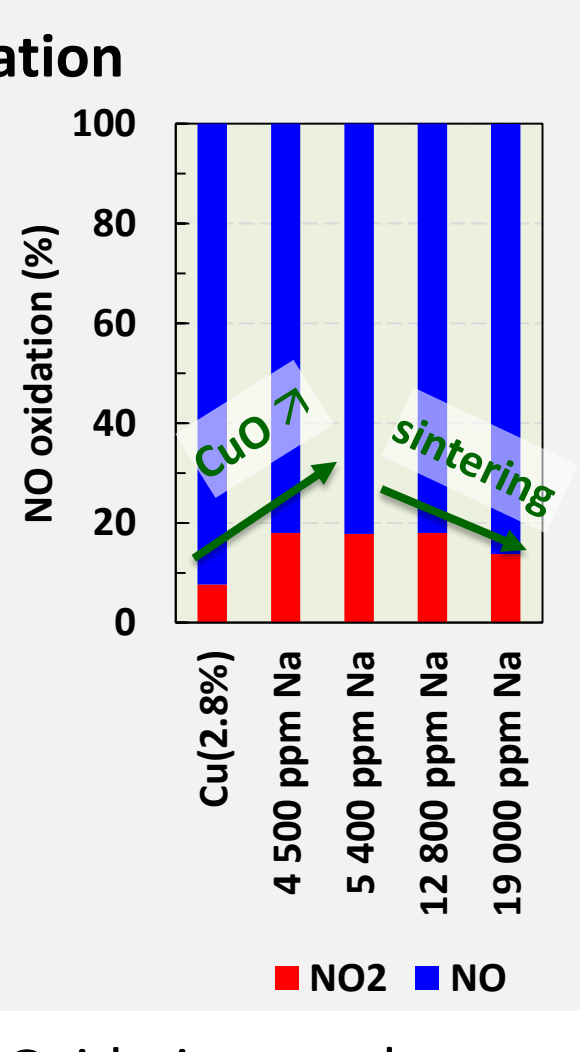
Catalytic oxidation properties

Na imp. in EtOH



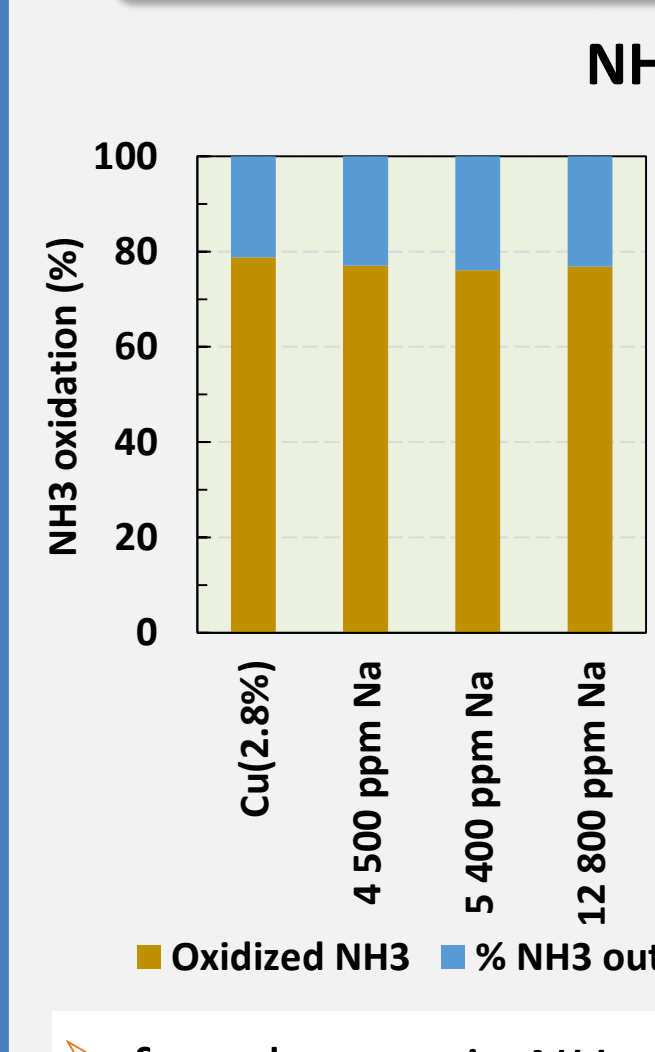
➤ few changes in NO oxidation, correlated with few changes in Cu distribution

Na imp. in H₂O



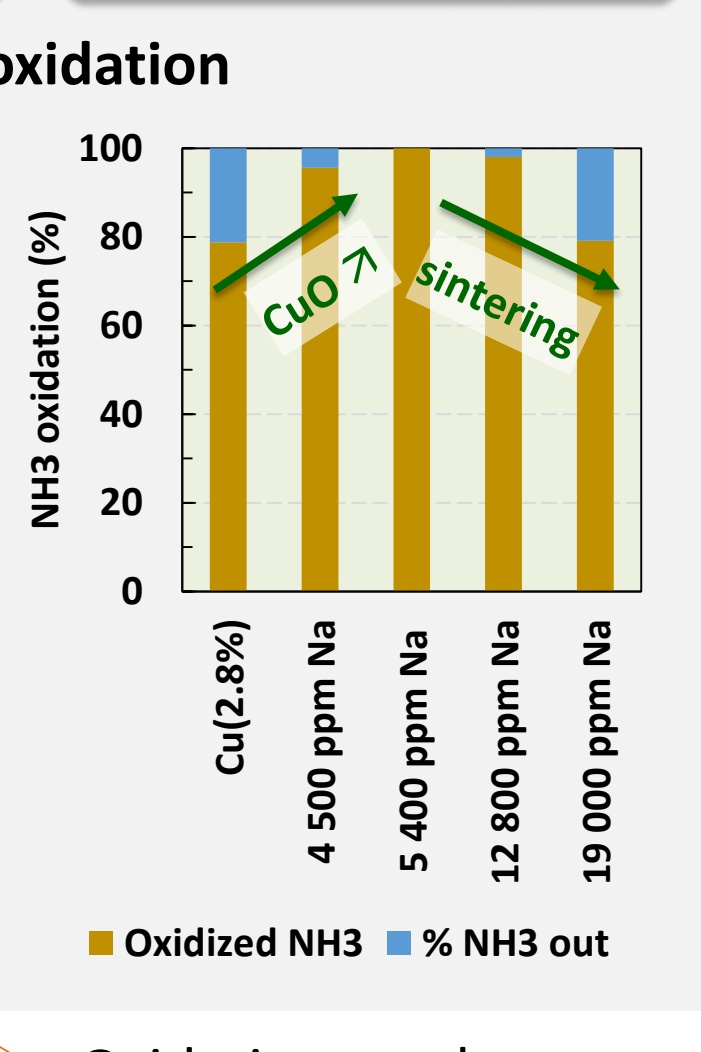
➤ Oxidation reaches a maximum: CuO formation → NO ox. ↑
CuO sintering → NO ox. ↓

Na imp. in EtOH



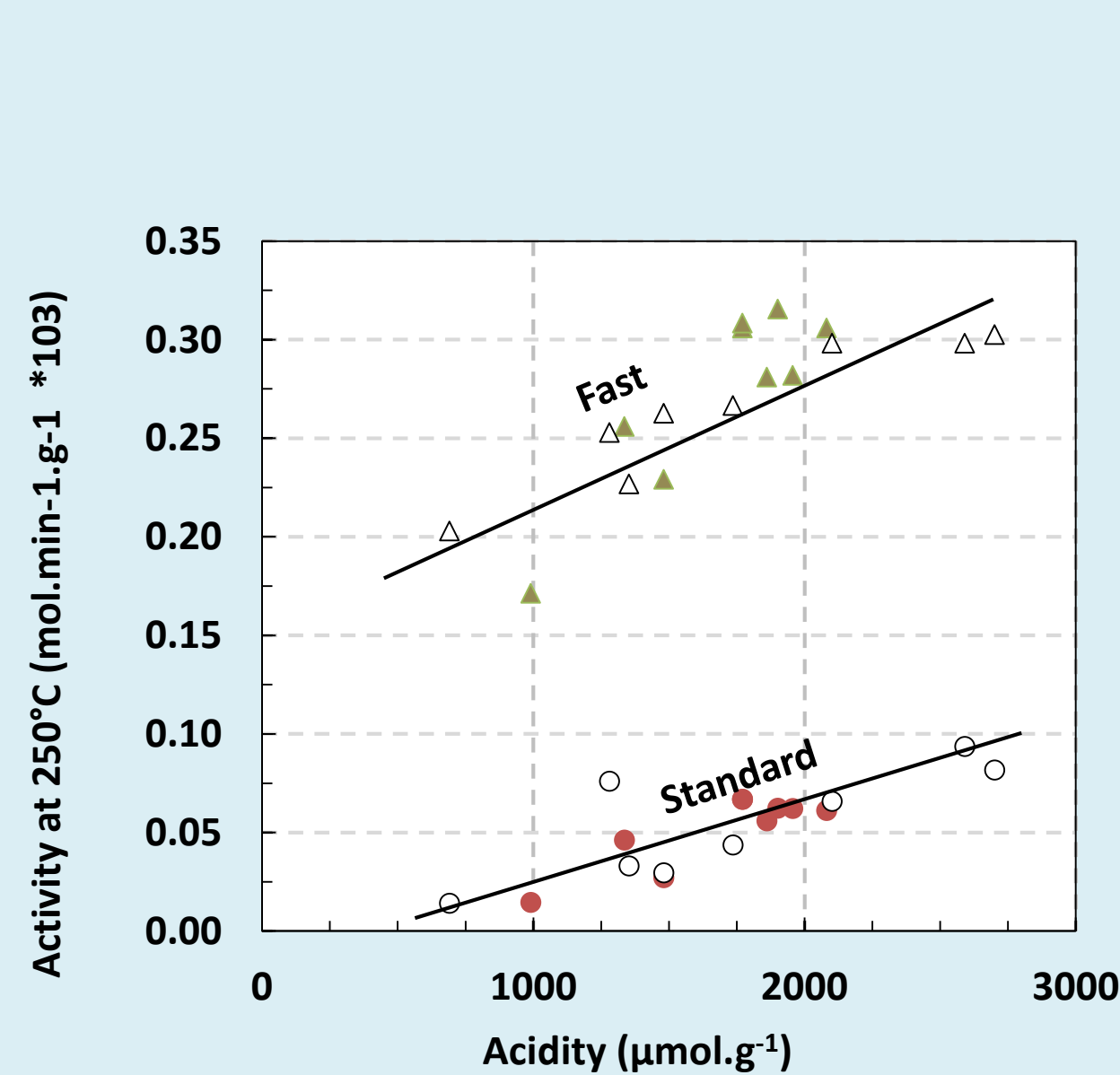
➤ few changes in NH₃ oxidation, correlated with few changes in Cu distribution

Na imp. in H₂O

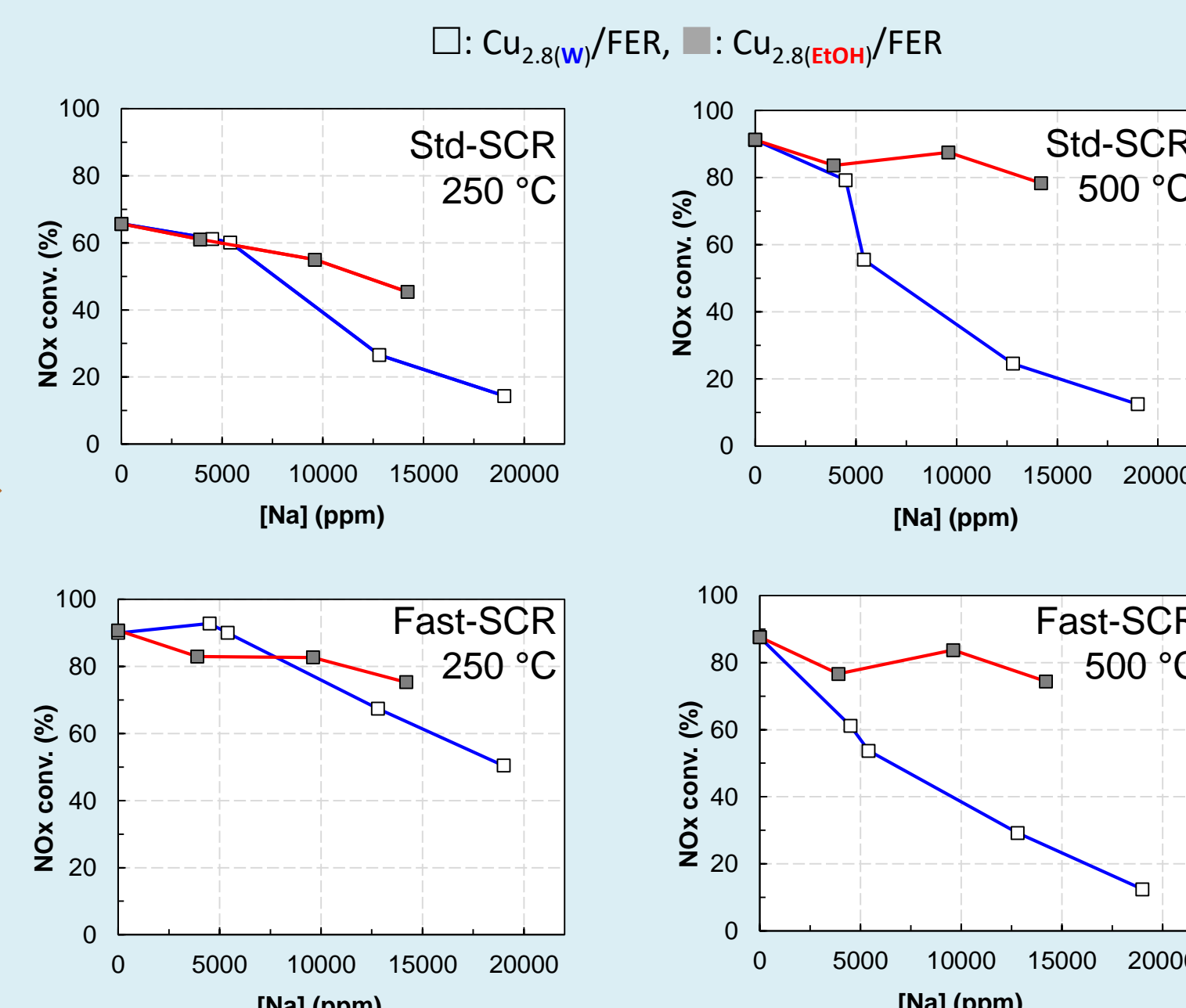


➤ Oxidation reaches a maximum: CuO formation → NH₃ ox. ↑
CuO sintering → NH₃ ox. ↓

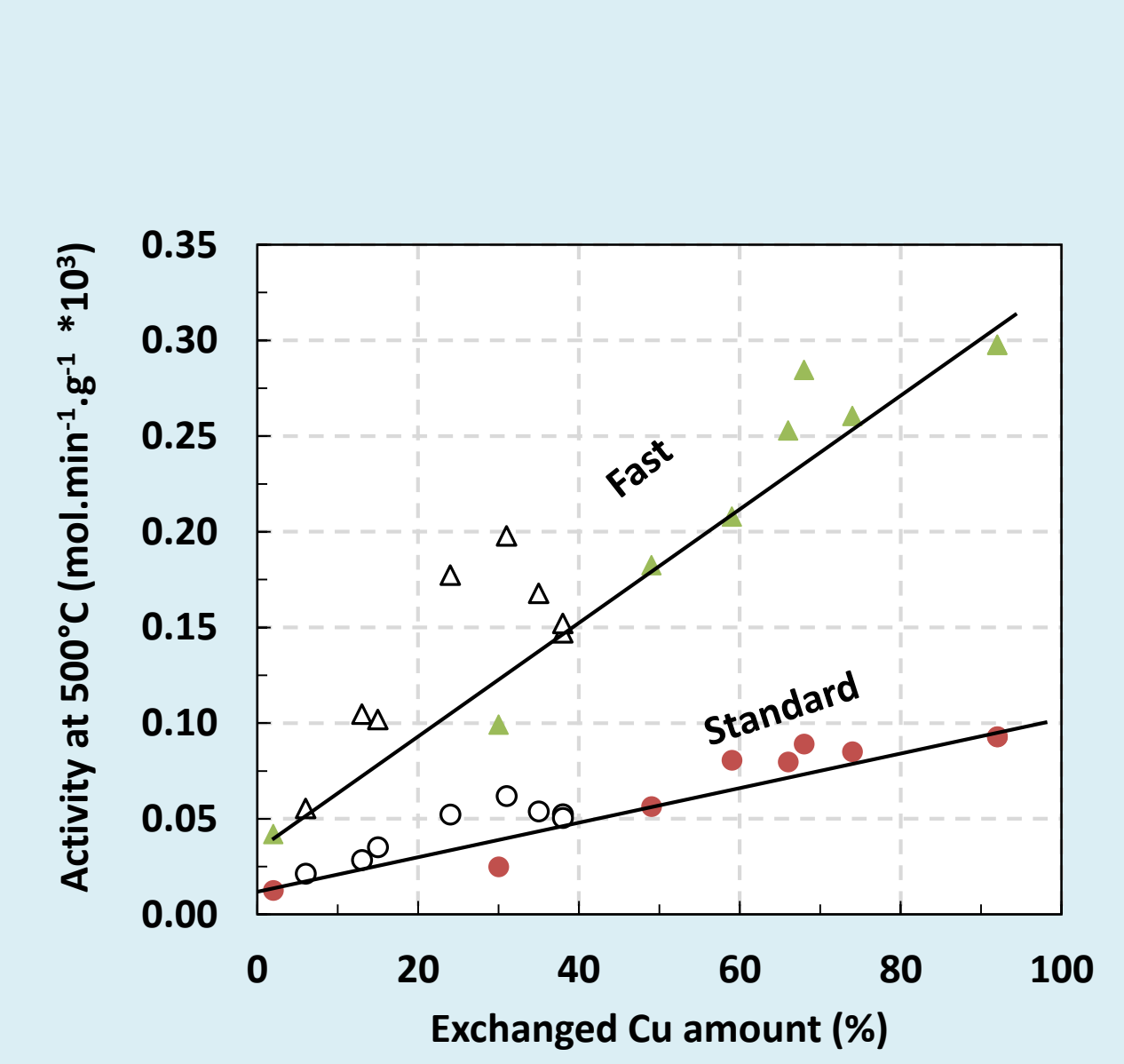
Correlations



↘ NO_x conversion at low temperature (250°C)
→ ↘ acidity



Less deactivation by EtOH compare to H₂O, in particular at high temperature



NO_x conversion at high temperature (500°C)
→ ratio of exchanged copper

CONCLUSION

Impregnation in ethanol leads mainly to a decrease of acidity, which affects the DeNO_x efficiency at low temperature.

Impregnation of Na in water decreases acidity and causes a migration of copper species from the exchange sites to extra framework CuO.

For Cu(2.8%) whatever the reaction mixture (Fast or Standard SCR), the NO_x conversion decrease with Na addition appears linked to the decrease of acidity at low temperature (250°C), whereas the activity at high temperature (500°C) mainly depends on the ratio of exchanged copper.