



Formation and reactivity of nitrates on Cu(II) sites in copper substituted CHA zeolite

Møller, Nicklas ; Isaksen, Oliver ; Godiksen, Anita; Rasmussen, Søren Birk; Vennestrøm, Peter N. R.; Mossin, Susanne

Publication date:
2016

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

[Link back to DTU Orbit](#)

Citation (APA):
Møller, N., Isaksen, O., Godiksen, A., Rasmussen, S. B., Vennestrøm, P. N. R., & Mossin, S. (2016). *Formation and reactivity of nitrates on Cu(II) sites in copper substituted CHA zeolite*. Poster session presented at 17th Nordic Symposium on Catalysis 2016, Lund, Sweden.

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.

Formation and reactivity of nitrates on Cu(II) sites in copper substituted CHA zeolite

Nicklas Møller¹, Oliver Isaksen¹, Anita Godiksen¹, Søren Birk Rasmussen², Peter N. R. Vennestrøm², Susanne Mossin^{1*}

¹ – Centre for Catalysis and Sustainable Chemistry, Department of Chemistry, Technical University of Denmark, Kemitorvet 207, 2800 Lyngby, Denmark

² – Haldor Topsøe A/S, Haldor Topsøe alle 1, 2800 Lyngby, Denmark

* slmo@kemi.dtu.dk

Copper exchanged zeolites are active for the selective catalytic reduction (SCR) of nitrogen oxides with ammonia.[1] The reactivity of small-pore zeolites and zeotypes with the CHA framework topology for removal of NO_x in diesel exhaust is due to favorable properties such as high stability towards hydrothermal ageing and high resistance towards the presence of hydrocarbons.

One of the key intermediates in the SCR reaction with ammonia is the formation of a nitrate. EPR spectroscopy is very sensitive towards Cu²⁺ and offers the possibility for both quantification and speciation with unrivaled sensitivity.[2] Nitrate coordinated to ion exchanged Cu²⁺ has a distinctive EPR signal, that makes it possible to follow the formation of this species.[3]

Here Cu-CHA with different Si/Al and Cu/Al ratios was first reduced in NO and NH₃ and then exposed to NO and O₂ gas mixtures under *in-situ* conditions. The formation of [CuNO₃]⁺ was observed on 1Al Cu sites in the CHA structure but not on 2Al Cu sites even though both Cu sites are observed to be oxidized from Cu⁺ to Cu²⁺.

Conclusions for the NH₃-SCR reaction on these materials will be presented.

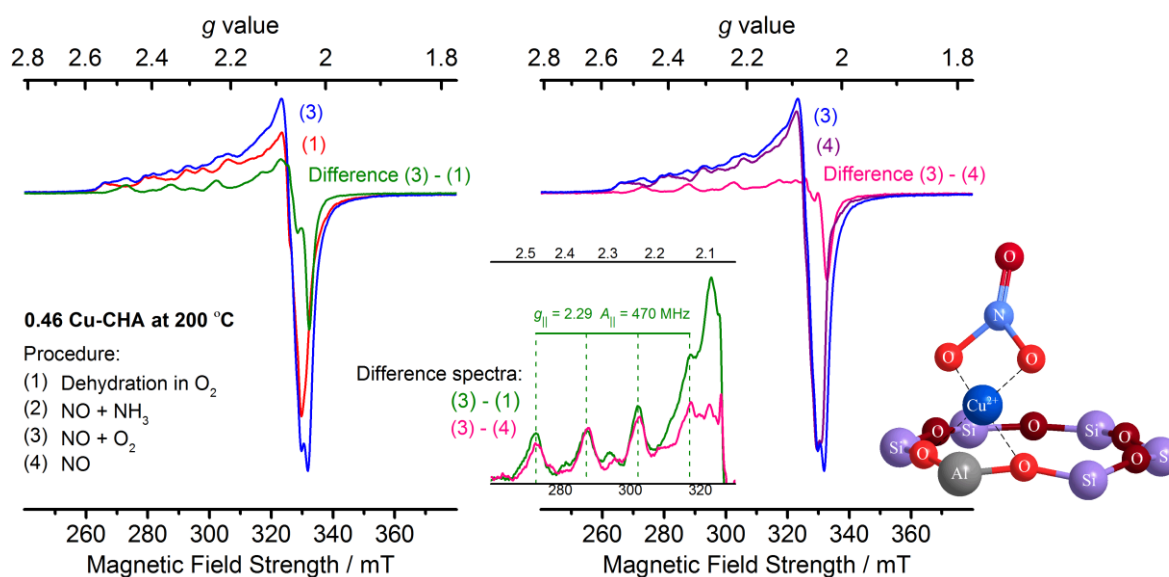


Figure 1. EPR spectra for Cu-CHA with Si/Al = 15 and Cu/Al = 0.46 during the steps of an *in-situ* procedure with NH₃ (1200 ppm), NO (1000 ppm) and O₂ (10 %) at 200 °C and GHSV = 400,000 h⁻¹. The difference spectra show that the EPR active nitrate species is first formed in NO + O₂ (step 1 to 3) and then is consumed completely after reaction with NO (step 3 to 4). The nitrate species [CuNO₃]⁺ above the 6mr is shown to the right.

[1] U. Deka et. al. *ACS Catal.* **2013**, 3, 413.

[2] A. Godiksen et. al. *J. Phys. Chem. C.* **2014**, 118, 23126.

[3] T. V. W. Janssens et. al. *ACS Catal.* **2015**, 5, 2832.