



A consistent reaction mechanism for the selective catalytic reduction of NO with NH₃

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ABSTRACT BODY:

Abstract: In the NH₃ selective catalytic reduction (SCR) harmful NO and NO₂ is removed from exhaust gases by using NH₃ as the reducing agent. The advantages demonstrated by copper substituted chabazite, Cu-CHA, include a widespread thermal stability range and high activity under lean combustion e.g. in fuel-efficient diesel engines. A full understanding of the structural activity and mechanism can help to improve the future design of SCR catalysts. By example of copper substituted chabazite zeolite, a mechanism for the SCR reaction has therefore been formulated on a single redox active copper center cycling through the oxidation steps +1 and +2. Only spectroscopically characterized copper species and stable charge-neutral gas phase molecules are suggested, avoiding the use of e.g. ½ O₂ or migration of charged species in order to match the stoichiometry of the reaction. Thus the suggested mechanism is relevant for the low end of the temperature interval relevant for the SCR reaction on these materials (250 °C - 300 °C) whereas other reactions may become important under more harsh conditions. The steps of the suggested cycle has been followed spectroscopically by in-situ EPR spectroscopy, in-situ FTIR spectroscopy and in-situ XAS spectroscopy and selected results will be presented. The suggested mechanism is also in accord with data already in the literature, of which some comparisons will be presented. The mechanism can be translated to other transition metal-based SCR catalysts and will be exemplified by vanadia (V₂O₅) on anatase (TiO₂) relevant for removal of NO and NO₂ from stationary sources. [1] F. Giordanino et. al. Dalton Trans., 2013, 42, pp 12741-12761 [2] A. Godiksen et. al. J. Phys. Chem. C, 2014, 118, 40, pp 23126–23138 [3] F. Giordanino et. al. Phys. Chem. Lett. 2014, 5, pp 1552-1559 [4] E. Borfecchia et. al. Chem. Sci. 2014, accepted [5] C. Paolucci et. al. Angew. Chem. Int. Ed. 2014, in press

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