Selective Catalytic Reduction of NOx by ammonia: Adsorption of NO on Cu-SSZ-13 using Ab initio simulations

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The goal of the selective catalytic reduction is to limit the emission of nitroxides (NOx) originating from combustion processes. Due to the upcoming tightening of the European legislation concerning emission from mobile sources, the importance of this process is rapidly increasing. In this process, fume gasses enriched with ammonia or urea, are brought into contact with a catalyst, effectively converting NOx into harmless gasses, i.e. nitrogen and water. Recently, experiments indicated that metal exchanged zeolites are particularly suited for this job, due to their high selectivity and hydrothermal stability [1]. This is especially true for copper-exchanged zeolites with a chabazite topology,i.e. Cu-SSZ-13, due to the relatively small pores. Moreover, the (almost) unique location of the active site (Cu^{2+} ion), makes this particular catalyst very interesting from a research point of view [1,2].

Despite the importance of the selective catalytic reduction, many questions concerning the reaction mechanism and the precise role of the catalyst remain unanswered. Therefore, Ab initio simulations have been performed on Cu-SSZ-13. In a first stage, the adsorption of NO is investigated using periodic DFT-based simulations. The adsorption energy and the optimal Cu-NO configuration within the zeolite framework are determined. In a second stage, a smaller cluster is isolated from the zeolite, in order to focus more on the charge distribution and to calculate IR-spectra [3], as experimental validation. The results of this study will serve as stepping stone towards unraveling the reaction mechanism.

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