Overview of ZSM-5, CLI, and FAU catalysts with metallic dimers (Cu, Fe, Zn) and their sensitivity to poisoning by sulfur dioxide (SO_2) in the DeNOx process.

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Significance and Relevance

The study analyzes the impact of SO_2 present in the reaction mixture during the industrial deNOx process. Results indicate that SO_2 molecules adsorb onto the metal sites in dimeric structures within all investigated zeolites. This interaction demonstrates that SO_2 acts as a catalyst poison. Furthermore, the sensitivity to poisoning is strongly dependent on the type of catalyst. Among the tested zeolites, clinoptilolite exhibits the highest resistance to SO_2-induced deactivation.

Preferred and 2nd choice for the topic: Automotive and stationary emission control, Fundamental advances in understanding catalysis
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Introduction and Motivations

The emission of nitrogen oxides NOx has become the main problem among air pollutants emitted from various sources, such as automotive, energy and heavy industry. In addition, these oxides are a very serious problem both in terms of impact on human health and the environment cause for eg. acid rains and photochemical smog.[1] Currently, regulations require a significant reduction of these harmful oxides and the existing thresholds may be lowered again in the near future. The most effective way to dispose of nitrogen oxides to meet the required limits is the SCR deNOx reaction. Transition metal modified zeolites have recently gained the greatest interest among scientists.[2-4] The resistance of catalysts to sulfur oxide poisoning is also a critical factor in.[5] Actual exhaust gases typically contain 50–300 ppm SO_2. Selecting an appropriate support material is the most effective strategy for enhancing low-temperature catalytic activity and improving resistance to SO_2-induced deactivation.

Materials and Methods

The ab initio density functional theory (DFT) method was used to calculate the electron structure of the presented clusters with StoBe software. The exchange and correlation functional was approximated with a Perdew-Burke-Ernzerhof (PBE) functional. It was used for the electron exchange and correlation. Milliken populations and Mayer's bond order factors were used to precisely analyse the electron structure of the clusters.

Results and Discussion

The analysis presents an overview of the three zeolites: ZSM-5, FAU and clinoptilolite. Different variants of metallic dimers (CuOCu, CuOFe, CuOZn) were also selected. In addition, each dimer was presented in a partially hydrated form - an OH group on the bridging oxygen and an OH group on one of the metals (two variants for each dimer). This representation of the dimers is due to the presence of H_2O in the reaction mixture and the ability of the dimers to hydrate easily. The study showed that sulphur dioxide adsorbs onto the surface of the metal dimer in each of the three selected zeolites. The zeolite with the highest resistance to sulphur dioxide poisoning was found to be the clinoptilolite. This natural zeolite showed high efficiency in deNOx and deN_2O processes, and its resistance to poisoning confirms its validity for use in these processes.

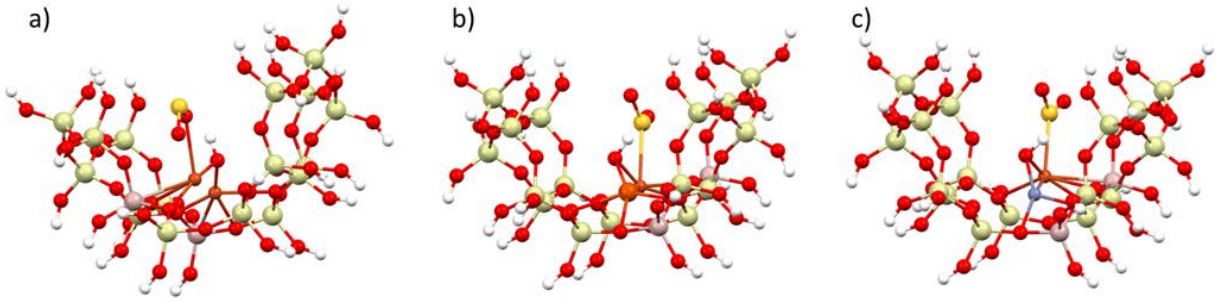

Figure 1. Structure of zeolite clinoptilolite ($Al_2Si_{16}O_{50}H_{28}$) with bimetallic dimers (a) Cu-OH-Cu (b) Cu-OH-Fe and (c) Cu-OH-Zn with adsorbed SO_2.

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