Impact of SO2-poisoning over the lifetime of a Cu-CHA catalyst for NH3-SCR

Cu-CHA catalysts for NH3-SCR in exhaust after treatment systems of heavy-duty vehicles, are constantly exposed to SO2 during their lifetime of about 10000h. In order to study the development of deactivation by SO2, a Cu-CHA catalyst was exposed to SO2 at 200, 300, 400 and 500°C for different durations up to 120h, resulting in total SO2 exposures that are comparable to that of the lifetime of a Cu-CHA catalyst in an after treatment system. The measured deactivation increases very fast to a steady level in the range 0.85-0.95, dependent on the exposure temperature, which shows the need for frequent regeneration of the catalyst. Regeneration at 550°C can restore the activity of the catalyst to 80 % of its fresh activity level even after 120h exposure, suggesting frequent regeneration as a feasible method for overcoming SO2-poisoning. ICP analyses showed that SO2 exposure led to S/Cu ratios in the range 0.5-1, indicating that sulfur is associated with Cu. After regeneration the S/Cu ratios did not exceed 0.2, suggesting that only certain Cu sites are able to form Cu,S species that are thermally stable above 550°C. This together with the observations that the deactivation before and after regeneration impact differently on the activation energy of the SCR reaction, and that the deactivation never exceeded 0.95, suggests that SO2-poisoning of Cu-CHA depends on the structural properties of this material. TGA measurements of the mass uptake during SO2 exposure was consistent with a process where SO2 is initially adsorbed on Cu, and then slowly oxidized to SO3 at 200°C, whereas the mass uptake at 500°C was consistent with an immediate adsorption of SO3, which is accredited to a faster oxidation rate at higher temperature.

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