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# Lifetime impact of $SO_2$-poisoning of a Cu-CHA catalyst for $NH_3$-SCR

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**Abstract:** $NO_x$ ($NO+NO_2$) emissions from diesel vehicles are reduced by selective catalytic reduction with $NH_3$ ($NH_3$-SCR) over $V_2O_5$- or metal-zeolite-based catalysts. Cu-CHA zeolites are particularly relevant due to their superior low-temperature activity [1]. However, the inevitable presence of $SO_2$ in diesel exhaust causes deactivation of Cu-CHA catalysts [1-3]. As the required lifetime of an SCR catalyst is ca. 10000 h, it is important to know how the activity of the catalyst develops with long-term $SO_2$ exposure, and if catalyst regeneration is an option. It has been shown that the deactivation by $SO_2$ is partially reversible [3]. In this contribution, we focus on the $SO_2$ uptake by the Cu-CHA catalyst, and how the reversible and irreversible deactivation are related to the amount of sulfur in the catalyst.

The long-term effects of $SO_2$ exposure are studied here by measurement of the deactivation of 24 Cu-CHA catalysts (Si/Al = 14.6, 2.4 wt% Cu) obtained by exposure to 50 ppm $SO_2$ at 200, 300, 400 and 500 °C for 1, 5, 15, 30, 60, and 120 h. The total amount of $SO_2$ passing the catalyst in 120 h is comparable to that in the lifetime of a heavy-duty vehicle. The regeneration of the catalysts is determined by measurement of the activity after heating each of the 24 samples to 550 °C in an $SO_2$-free gas for 6 h.

The most important effect of exposure to $SO_2$ is a decrease of the $NO_x$ conversion in the low-temperature range (200-300 °C). Most of the activity can be regenerated by heating to 550 °C. Fig. 1 shows that deactivation of the Cu-CHA catalysts occurs fast, and already at a total $SO_2$ exposure of 750 ppm·h, the final activity levels for both the $SO_2$ exposed and regenerated catalysts has been reached. $SO_2$ exposure leads to a saturated deactivation level in the range 85-95 %, and about 20 % for the regenerated catalysts. Full deactivation of the Cu-CHA has not been observed, suggesting that some of the Cu remains active. The data in Fig. 1 also imply that regeneration at 550 °C in $SO_2$-free gas restores the activity to about 80 % of the fresh catalyst even after 6000 ppm·h $SO_2$ exposure, showing that the predominant part of the deactivation by $SO_2$ can be reversed by an active regeneration. The irreversible deactivation can be dealt with by proper dimensioning of the catalyst.

Chemical analysis by ICP-OES of the 24 catalyst samples shows a fast initial increase in S/Cu ratios with $SO_2$ exposure, which coincides with the fast initial deactivation. The S/Cu ratio is never significantly higher than 1, indicating that all S atoms in the deactivated catalysts are associated with the Cu ions. The S/Cu ratios after regeneration at 550 °C are limited to 0.2, and does not depend on the exposure temperature. The S/Cu ratios of the $SO_2$ exposed catalysts reach different levels depending on the exposure temperature. The highest S uptake occurs at 300 °C, which means that the S uptake is actually close to its maximum at typical SCR operating temperatures.

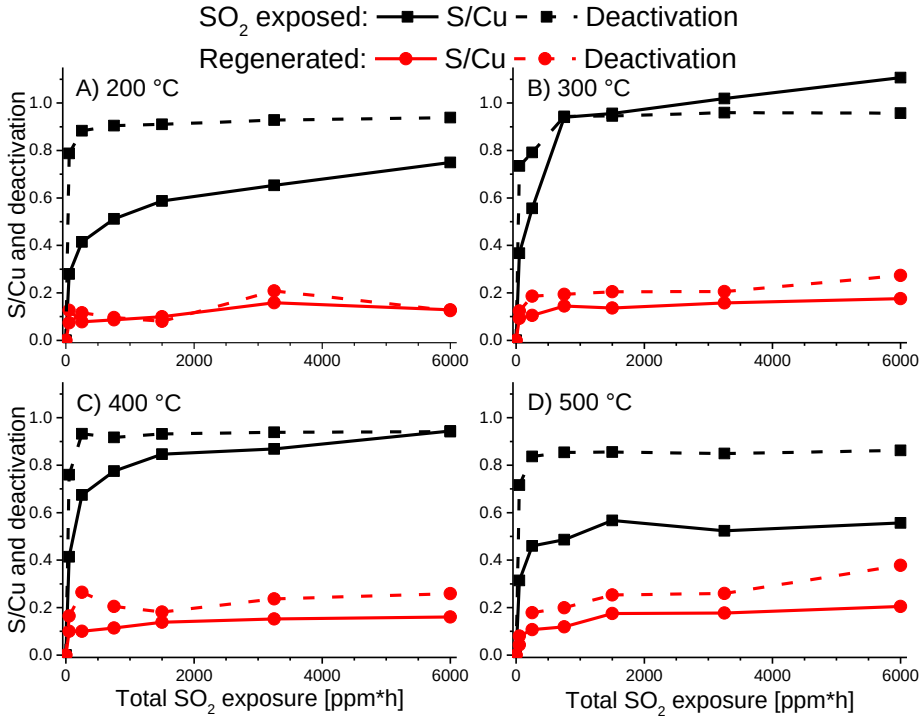


*Figure 1 – S/Cu ratio and deactivation as functions of total $SO_2$ exposure, for $SO_2$ exposed and regenerated states of a Cu-CHA catalyst. The $SO_2$ exposures were at A) 200 °C, B) 300 °C, C) 400 °C, and D) 500 °C, and the regeneration always at 550 °C. The level of deactivation was measured at 220 °C.*

In summary, the deactivation of Cu-CHA builds up fast, and regeneration at 550 °C restores the activity to about 80 % of the original level. Furthermore, the S uptake also increases fast initially to a saturation S/Cu level between 0.5-1, dependent on the exposure temperature. Some Cu remains active, which indicates that certain Cu sites appear to be inert to $SO_2$-poisoning.

**Key words:** $NH_3$-SCR, Cu-CHA, $SO_2$ poisoning, deactivation, regeneration

**References**

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