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Lifetime impact of SO₂-poisoning of a Cu-CHA catalyst for NH₃-SCR

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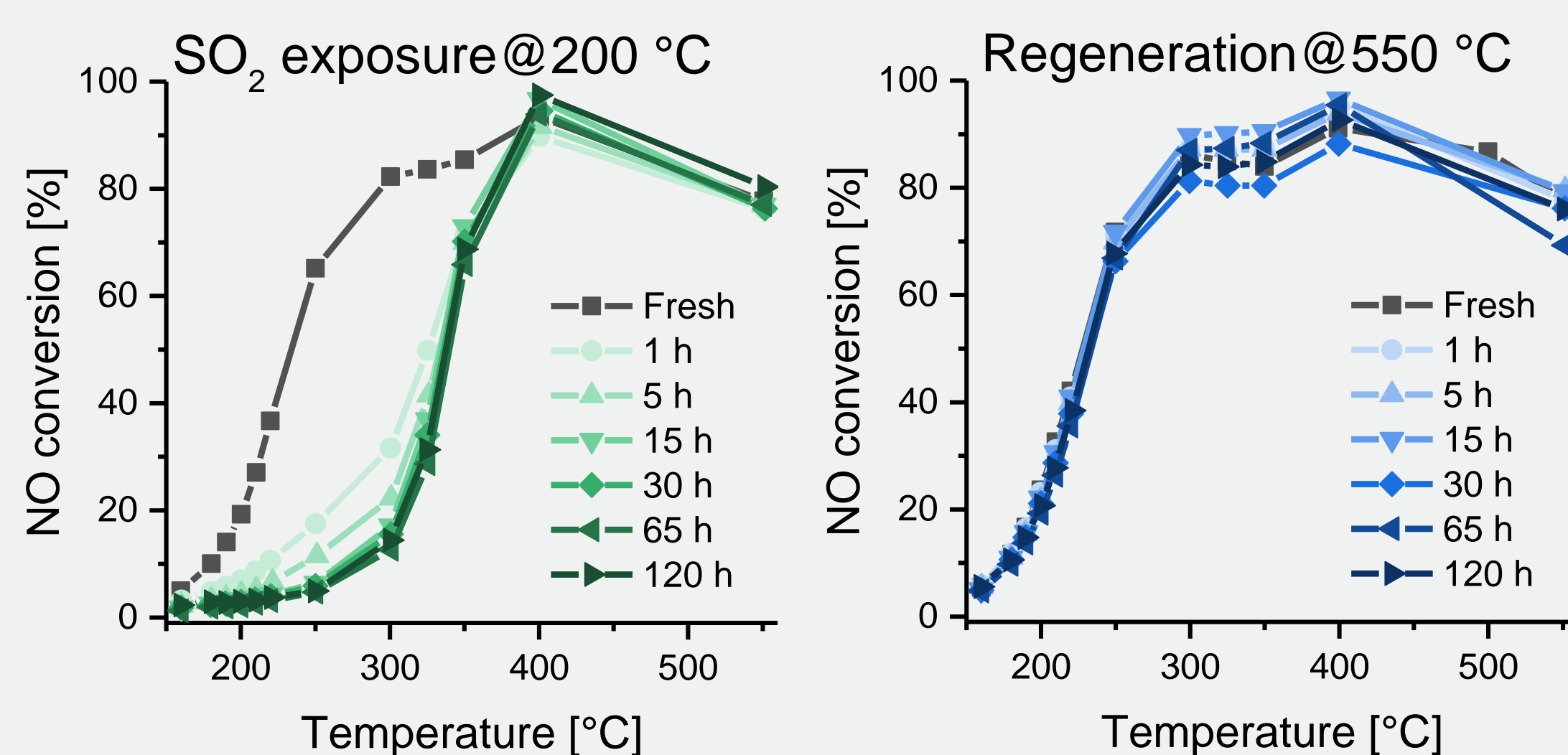
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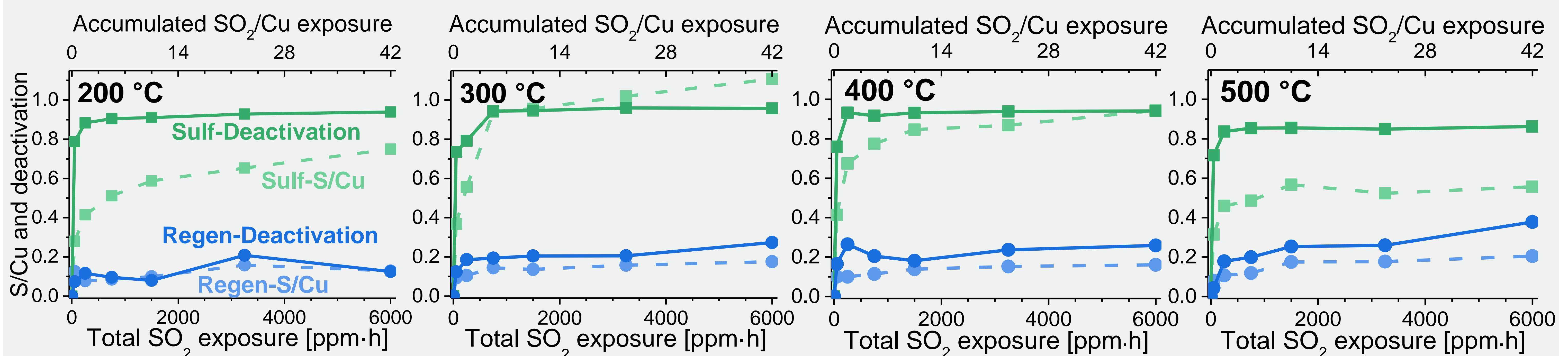
Background

The highest activity for removal of NO_x (NO and NO₂) in the temperature range 180-250 °C, by selective catalytic reduction (SCR) with NH₃ in the after treatment system of heavy-duty vehicles, is obtained with Cu-zeolite catalysts. Improved fuel efficiency that is necessary for lower CO₂ emissions, has led to colder exhausts, which has increased the importance of efficient NO_x removal in that temperature range. However, Cu-zeolites deactivate by the inevitable presence of SO₂ in diesel exhaust, and therefore, we have investigated if the lifetime impact of SO₂ on the activity of a Cu-CHA zeolite catalyst is prohibitive for practical application.



Typical SO₂-poisoning behavior

- Exposure to SO₂ results in significantly lower NO conversions in the temperature range 180-350 °C
- Regeneration at 550 °C in SO₂-free gas can restore the predominant part of the drop in NO conversion inferred by SO₂



Experimental

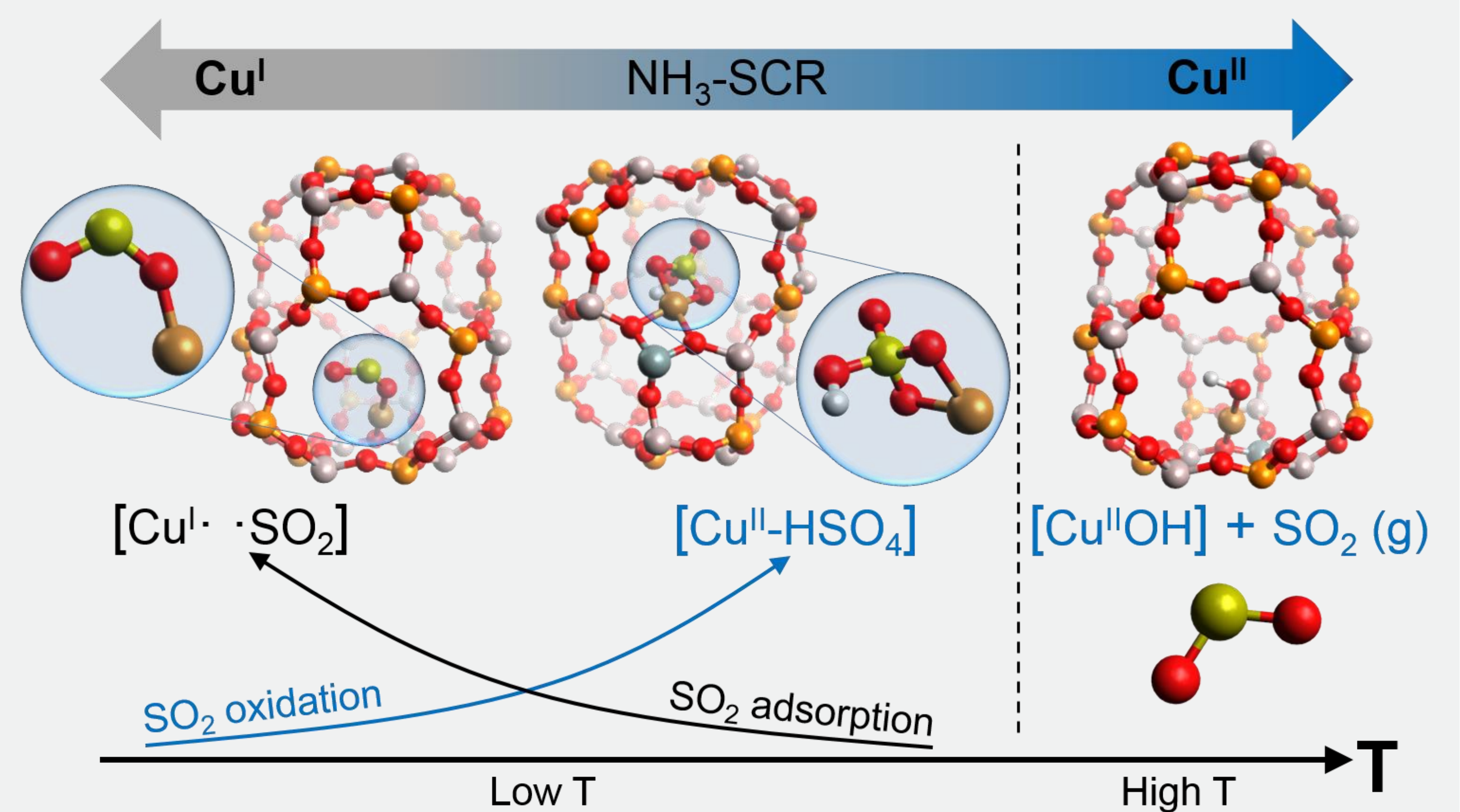
Catalyst: Cu-CHA powder with Si/Al of 14.6 and Cu/Al of 0.42 (2.76 wt% Cu on dry matter basis), and of sieve fraction 150-300 μm.

SCR activity measurements: 5 mg catalyst, 500 ppmv NO, 533 ppmv NH₃, 10 % O₂, 5 % H₂O, with N₂ at a total flow rate of 225 NmL/min.

SO₂ exposure: 50 ppmv SO₂, 10 % O₂ and 5 % H₂O with N₂ to 1.67 NL/min at 200, 300, 400 or 500 °C for 1, 5, 15, 30, 65 or 120 h.

Regeneration: 6 h at 550 °C in 10 % O₂ and 5 % H₂O gas.

ICP-OES: S/Cu ratios of the sulfated and regenerated catalysts.



Potential lifetime SO₂/Cu exposure

Assumptions for the lifetime of a heavy-duty vehicle:

- Distance: 800 000 km
- Average mileage: 30 L diesel/ 100 km
- Diesel: 5 wt ppm S (Ultra-low sulfur diesel in: EU: <10 wt ppm S, USA: <15 wt ppm S)
- Cu-CHA: 1.5 kg with 3 wt% Cu (45 g Cu)
- Potential accumulated lifetime SO₂/Cu exposure: **44**

Conclusions

- Deactivation occurs fast regardless of exposure temperature
 - Maximum deactivation levels reached already at low S/Cu ratios
- Regeneration at 550 °C can limit deactivation to 20 % throughout the catalyst lifetime
- Practical application of Cu-CHA as SCR catalyst is contingent on:
 - Efficient regeneration
 - Use of ultra-low sulfur diesel