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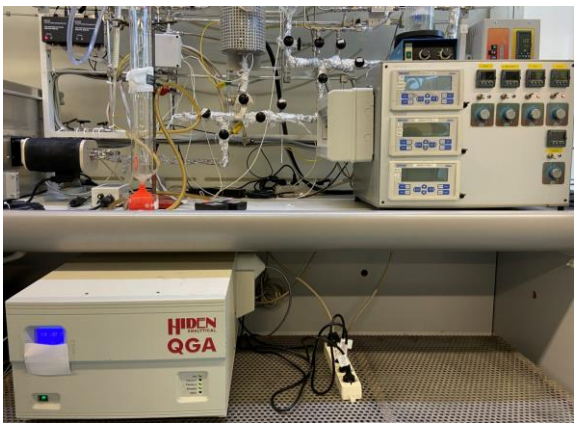
Unexpected Low-Temperature DeNOx Activity of AdSCR Systems for Cold start NOx Abatement

Urea-SCR aftertreatment systems have been implemented in lean burn vehicles to reduce NOx emissions in order to meet government regulations. Current commercial catalysts (Cu- and Fe- promoted zeolites) are able to achieve very high NOx removal efficiencies once they reach operating temperatures above 200°C. However, their performances are relatively poor at low temperatures, e.g. during the engine cold start. Indeed, in this temperature region: (i) the inability of decomposing urea prevents fully exploiting the SCR technology; (ii) the metal-promoted zeolite catalysts have a limited deNOx activity.

In the present work we extend the investigation of a novel catalytic device designed to improve the abatement of NOx during the engine warm up phase, the so called AdSCR system (AdSCR = Adsorption + Selective Catalytic Reduction). The formulation of AdSCR systems is based on the combination of a Urea-SCR catalyst (e.g. Cu-CHA) and of a NOx storage material (e.g. BaO/Al_2O_3) in the form of physical mixtures. They are characterized by a dual functionality: they can operate both as NOx adsorbers, trapping NOx during the low temperature transient, and as SCR catalysts, reducing the previously stored NOx with NH_3 at higher temperatures.

A set of experiments was carried out over a physical mixture of SCR catalysts (Cu-CHA) and NO_x storage materials (BaO/Al_2O_3) to study their deNOx efficiency. The typical protocol consisted of cold start mimicking runs, aimed at simulating the aftertreatment system warm-up: NO (300 ppm) was fed in a stream of 8% of O_2 at r.t. followed by a temperature (T)-ramp (15 °C/min). At 170 °C, i.e. the T-threshold for urea injection, NH_3 (800 ppm) was fed to start the SCR reactions. The same protocol was repeated in the presence of NH_3 pre-adsorbed at 320 °C to investigate the reactivity of stored NO_x and NH_3. In these experiments the dynamics of NO, NH_3, and NO_2 at the reactor outlet were continuously measured both by a UV analyzer (ABB LIMAS 11HW) and by a quadrupole mass spectrometer (QGA Hiden Analytical) which could, furthermore, detect N_2 and N_2O.

Our results from dry cold-start runs show that: i) after a NO step feed, NOx were completely trapped for up to 420 s before NO breakthrough, with a NO storage efficiency of 93% before NH_3 injection; ii) after NH_3 injection, a drop in the NO outlet concentration and a release of N_2 (850 ppm) were observed due to the onset of the SCR reactions. Moreover, in the presence of pre-adsorbed ammonia the formation of N_2 was unexpectedly observed already at 40 °C. These remarkable results identify AdSCR as promising systems and may warrant further investigation.



The Hiden QGA in the Laboratory

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