Performance analysis of isostructural Cu-CHA-zeolites in NSR-SCR Hybrid DeNO$_x$ Technology for Diesel Engines

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Introduction.

Road-transpor sector is still the major air pollution contributor of harmful NO$_x$ emissions in Europe. The new Euro 7 regulation declared the importance of the development of DeNO$_x$ technologies, based on the combination of the current technologies and knowledge of all relevant pollutants control systems. Two catalytic technologies are widely used for this purpose are NO$_x$ Storage and Reduction (NSR) and Selective Catalytic Reduction (SCR) in diesel engines. The coupling between a NO$_x$-Trap followed by a NH$_3$-SCR catalyst leads to an improved N$_2$-selectivity, by the storage and reaction of the undesired NH$_3$ produced in the NSR catalyst in the downstream SCR system. This contribution is focused on the synthesis of two isostructural CHA-zeolites with Cu integrated in their structure, SAPO-34 and SSZ-13, and the combination and analysis of the behavior in NO$_x$ abatement activity in the NSR-SCR hybrid system.

Experimental/methodology.

For the NSR system, a model catalyst (Pt-Ba-K/Al$_2$O$_3$) was used [1]. As SCR catalysts, Cu-CHA-zeolites precursor gel phase was synthesized containing 2 wt.% of Cu by an ultrasound-assisted hydrothermal method and the calcined materials were identified as 2Cu-SAPO-34 and 2Cu-SSZ-13. Cu-zeolites were well-characterized. DeNO$_x$ activity of the NSR+SCR system was evaluated in a fixed-bed tubular reactor in isothermal and transient regime using a MultiGas MKS 2030 FTIR analyzer and the QMS200 Mass Spectrometer for online monitoring the outlet product distribution. Cyclic alternating lean (3%O$_2$) and rich (1%H$_2$) fuel conditions were used maintaining the concentration of NO (600 ppm), CO$_2$ (0.3%) and H$_2$O (1.5%) He-balanced, GHSV=30000h^{-1} and the T-range of 200-350ºC.

Results and discussion.

In NH$_3$-TPD profiles (Fig. 1) a low-temperature (185ºC) signal related to Brønsted acid sites was observed, a medium-temperature signal (286ºC) due to NH$_3$-adsorbed on strong Lewis acidic sites, associated with isolated Cu^{2+} sites, was identified for both Cu-zeolites but more evident for Cu-SAPO-34. The existence of different Cu-species (confirmed by FTIR analysis with probe molecules) causes the shift to the T-max desorption signal of the Brønsted sites and influences the deNO$_x$ activity. .

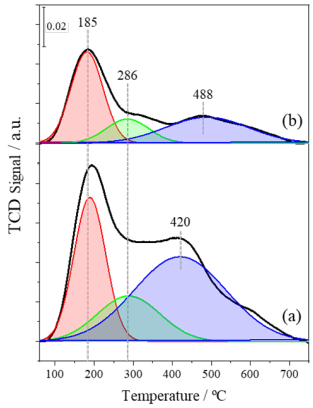

Figure 1. NH$_3$-TPD profiles for Cu-CHA-zeolites: (a)2Cu-SAPO-34 and (b) 2Cu-SSZ-13

Table 1. DeNO$_x$ activity results at 250 ºC

Configuration	X_{NO_x} (%)	S_{N_2} (%)	S_{NH_3} (%)	S_{N_2O} (%)
NSR	52.3	69.6	21.6	8.8
NSR+2Cu-SAPO-34	63.7	89.4	0.02	9.6
NSR+2Cu-SSZ-13	58.8	94.1	0.00	5.8

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References

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