Challenges and performance of Cu-SCR catalysts under H2-ICE conditions

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Significance and Relevance

This work shows the challenges which Cu-SCR will have to face if used as technology for NOx removal in H2-ICE applications. Activity testing, Operando XAS Spectroscopy and reactive characterisation were combined to collect preliminary evidence of the SCR performance and durability of state-of-the-art Cu-CHA catalyst under H2-ICE conditions, characterised by high water concentration (20-30%) and H_2 (up to 2%).

Preferred and 2nd choice for the topic: automotive and stationary emission control; sustainable and clean energy production and transport.
Preferred presentation: (Oral only / Oral preferred or Short Oral / Poster) Oral and Short oral

Introduction and Motivations

Internal combustion engines operated using H_2 as fuel (H2-ICE) will have a key role in the transition towards a zero-carbon transport society and they will expect to contribute to the global HD powertrain from 2030 onwards. Despite a history dating back to the 19th century, H2-ICEs are not yet competitive to Diesel and Gasoline engines and need to be developed. H2 ICEs will still require an after-treatment system: despite the use of a completely carbon free energy source and therefore the absence of CO and CO_2 emissions, NOx and high levels of water are still produced as by-products of the hydrogen combustion process. Due to the nature of the pollutants, Pt/Pd zeolites could be considered as a suitable solution for reducing NOx to N_2 using H_2 itself as reductant (H_2 SCR – Selective Catalytic Reduction) [1,2,]. However, it is well known the poor activity and selectivity to N_2 of these materials. NOx reduction by NH_3-SCR, combined with an ASC (Ammonia Slip Catalyst) to deal with the unconverted ammonia, could represent a better choice because of their typical outstanding performance under Diesel conditions[3]. H2-ICE conditions could be more challenging than Diesel though: this work will provide an overview on the challenges which state-of-the-art Cu-SCR catalyst will face when exposed to H2-ICE exhausts and how its SCR performance is impacted by combining activity testing, Operando Spectroscopy and reactive characterisation under relevant conditions.

Results and Discussion

H2-ICE exhausts are characterised by high water concentration (20-30%), potentially high levels of NOx (3000 ppm) and H_2 spikes (up to 2%) and S (depending on the H_2 source - i.e. grey H_2 - and from lubricant oil). The feasibility of the use of Cu-zeolite catalysts for the removal of NOx in presence of high-water and H_2 was preliminary investigated by comparing NH_3-SCR performance of a state-of-the-art Cu-CHA catalyst under Diesel conditions (7%H_2O, no H_2) and H2-ICE conditions (20% H_2O, 2%H_2). Data showed a negative impact on the NOx conversion below 250°C given by the presence of high-water concentration, with an increase in the N_2O production. However, when H_2 is added, while the activity is still affected, the selectivity improves. This effect seems to suggest an important role of H_2 on the oxidation state of Cu sites: indeed, formation of N_2O has been associated with Cu^{2+} dimers[4] and addition of H_2 could modify their population, lowering the average oxidation state of Cu sites and therefore the formation of this by-product. Dedicated Operando Spectroscopy Experiments were also performed at Diamond Light Source which confirmed an increasing fraction of Cu^{+} species on increasing H_2 concentration in the SCR base feed (Figure 1).

The durability of Cu-SCR technology in presence of higher water content, H_2 and S was also investigated. Activity data and Operando XAS data were collected on sulfated Cu catalysts under Diesel

conditions and H2-ICE conditions, showing a harsher impact of high-water content on the extent of S poisoning of Cu sites, which are less available to be involved in the SCR redox cycle with negative consequence on the SCR performance.

To summarise, preliminary data have shown that Cu-CHA are still effective catalysts for dealing with the NOx produced in H2-ICEs. However, the different environment in which Cu-CHAs are exposed could have several impacts on the SCR mechanism and the durability of these catalysts which will need to be addressed for selecting the best formulation for H2 ICE after-treatment systems.

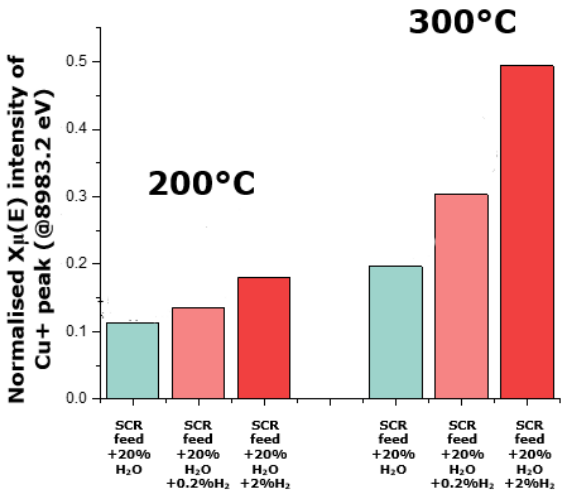

Figure 1 Normalised Xμ(E) intensity of Cu^+ peak (@ 8983.2 eV) of XAS Spectra collected during in situ experiment over a Cu-CHA catalyst at steady state conditions at 200 and 300°C. Green: SCR base feed (500 ppm NO, 500 ppm NH_3, 10% O_2) +20% H_2O; Pink: SCR base feed +20% H_2O + 0.2%H_2; Red: SCR base feed +20% H_2O + 2%H_2.

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