



Effect of promoters on the performance of Pt/TiO₂ catalyst for NO_x removal by selective catalytic reduction (SCR) with H₂

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Significance and Relevance

The influence of basic (Mg and Ba) and acidic (W and Nb) promoters on the activity of a 0.5 wt.%Pt/TiO₂ catalyst for the selective catalytic reduction (SCR) of NO_x using H₂ as a reductant has been systematically studied. Pronounced effects on the NO_x conversion profile were identified for the different promoters and especially on the catalyst selectivity towards nitrogen and ammonia formation. Additionally, NH₃ emissions were detected for the catalysts doped with basic promoters while NH₃ seems to be converted to N₂ via NH₃-SCR in the presence of acidic promoters.

Preferred and 2nd choice for the topic: Automotive and stationary emission control, Air cleaning and combustion

Preferred presentation: Oral preferred

Introduction and Motivations

To reduce the CO₂ emissions, the use of carbon free fuels has gained major attention during recent years. Due to its potential to improve the efficiency of internal combustion engines while reducing the emissions of harmful pollutants, hydrogen is regarded as an interesting alternative fuel for the automotive sector¹. Herein, the use of the on-board available H₂ for NO_x removal instead of the ammonia/urea solution is an economically relevant approach² but very challenging. Since the direct oxidation of hydrogen at higher temperatures and the unwanted reaction pathways forming either N₂O or NH₃ instead of the desired N₂ can occur.³ For this process, noble metal-based catalysts in multicomponent systems have demonstrated a promising performance^{4,5}. However, to comply with the current legislation significant improvement in their activity and selectivity is still necessary. In this context, this study reports a systematic investigation of the effect of catalyst promoters on the Pt/TiO₂ catalyst activity and selectivity.

Materials and Methods

The different catalysts were prepared by a nitrate-based precursor incipient wetness impregnation of commercial titania support to obtain the aimed promoter loadings. After drying and calcination of the doped support, the noble metal precursor was added by incipient wetness impregnation to achieve a 0.5 wt % loading of Pt. In addition, a reference catalyst with the same Pt loading without the addition of a promoter was produced. The obtained catalysts were characterized by N₂ physisorption, X-ray diffraction (XRD) and elemental analysis using inductively coupled plasma - optical emission spectroscopy (ICP-OES). Additionally, *operando* X-ray absorption spectroscopy (XAS)⁶ investigations of selected samples were conducted to identify the noble metal state under reaction conditions as a function of catalyst composition. Catalytic tests for NO_x reduction were performed for the as prepared and reductively treated samples in a plug-flow reactor while analyzing the evolved gases by MS and FT-IR spectroscopy.

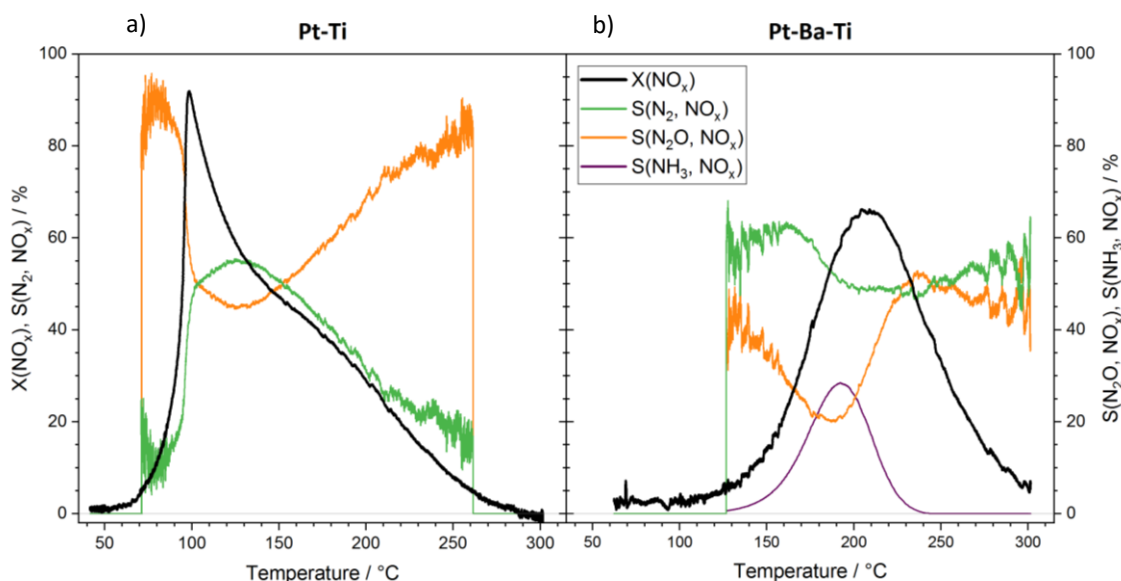


Figure 1: Activity profiles of the plug-flow reactor tests in a gas mixture containing: 1000 ppm NO, 5000 ppm H₂, 10 % O₂, 5 % H₂O in N₂ between 50 – 300 °C (WHSV of 90.000 L*h⁻¹*gPt⁻¹), after an oxidative treatment at 350 °C in 20 % O₂ for 1 h. The conversion of NO_x (black) and selectivity towards nitrogen (green), nitrous oxide (orange) and ammonia (purple) of NO_x are shown line in their respective colors.

Results and Discussion

X-ray diffraction patterns of the produced supports did not show any platinum reflexes, indicating a good distribution of small Pt particles on the titania support irrespective of the promoter type. Plug-flow reactor activity tests show a significant increase of the low temperature activity after catalyst pre-reduction, as illustrated in Figure 1 for Pt/TiO₂.

The catalysts containing basic promoters (Mg and Ba) display increased selectivity for NH₃ formation. In contrast, no NH₃ trace could be detected for the undoped catalysts and the catalysts containing acidic promoters (W and Nb). While the production of NH₃ is limited to a narrow temperature window for the Ba-promoted catalyst, nitrogen formation does not vary significantly over the whole activity range (Fig 1b). In line with the observed H₂-SCR activity profile, *operando* XAS studies and following linear combination analysis revealed a faster reduction of Pt in the tungsten promoted catalyst, while slower redox response was observed for the noble metal in the barium promoted catalysts indicating the influence of the promoters on the noble metal-support interactions.

In summary, a better understanding of the catalyst composition effects and enhancement on the H₂-SCR activity and selectivity of Pt-Ti catalysts was obtained, which enables a knowledge driven catalysts design and efficient noble metal use.

References

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