NOx emission control by H_2-SCR over Pt-ZSM5 catalyst in mobile applications

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Significance and Relevance

The development of H_2-fueled vehicles has been proposed to contribute to the limitation of the large fraction of CO_2 emissions associated to the transports sector because hydrogen is a carbon-free fuel. However, there are operating points of the engine promoting the formation of thermal NO_x. The use of H_2, already present on board as a fuel, for the Selective Catalytic Reduction (SCR) of NO_x, replacing ammonia, would avoid the additional device for storage and injection of urea.
ZSM5 supported Pt catalysts have been investigated for SCR of NO_x and the effect of additives considered to inhibit the undesired NO oxidation.

Preferred and 2nd choice for the topic: Automotive and stationary emission control
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Introduction and Motivations

In order to mitigate the global warming limiting CO_2 emissions, the development of electric cars has been proposed as primary solution for the transports sector. Nevertheless, approaches still based on Internal Combustion Engine (ICE) have been also considered, such as the use of hydrogen as fuel. Since a completely NO_x free combustion cannot be ensured at all operating points[1] of the H_2-fueled engine which works under lean condition, a catalyzed exhaust after-treatment system is required. H_2 as a reducing agent of NO_x would have the advantage that only one fuel must be carried on board both for propulsion and NO_x reduction, thus avoiding the device for storage and injection of urea, as for the more common NH_3-SCR.
The NO_x reduction by H_2 produces only elemental nitrogen and water:

$$2\ NO + 4\ H_2 + O_2 \rightarrow N_2 + 4\ H_2O \qquad (1)$$

MFI zeolite supported Pt catalysts exhibited higher N_2 selectivity than metal oxide supported catalysts, however, in the presence of excess O_2, Pt can easily catalyse NO oxidation to NO_2[2,3]. In this work, the effect of Pt load at very low levels (0.1-1%) on the activity and selectivity and that of the addition of sodium to depress the oxidation activity of platinum have been investigated.

Materials and Methods

A commercial ZSM5 powder with a SiO_2/Al_2O_3 ratio=23 was impregnated with platinum using an aqueous solution of Tertraammineplatinum(II) chloride hydrate with a platinum/zeolite concentration ratio to obtain 0.1%Pt, 0.5 or 1wt% nominal load. Portions of 0.1%Pt/ZSM5 sample were impregnated with a sodium nitrate solution at different concentration to obtain 5 and 10% Na.
H_2-SCR experiments were carried out in a fixed bed reactor in the temperature range 50-450°C and were also simulated in a DRIFT environmental chamber to investigate the adsorbed NO_x species involved in the reaction.

Results and Discussion

Catalytic tests performed on Pt/Z catalysts with different platinum loads showed that the addition of an amount as low as 0.1%Pt promoted NO conversion to N_2 in the temperature range 80-200°C. The temperature corresponding to the maximum NO conversion decreased with Pt load. Nevertheless,

even with a so low Pt load, at T>200°C NO is mostly oxidized to NO_2. The addition of sodium strongly inhibited the undesired NO_2 formation and, unexpectedly, the N_2O production occurring simultaneously with the main SCR reaction (Fig.1). Catalysts were fully characterized (elemental analysis, XRD, porosimetric analysis, NH_3-TPD) to determine the role of Pt dispersion and of the acid properties on the catalytic performance.

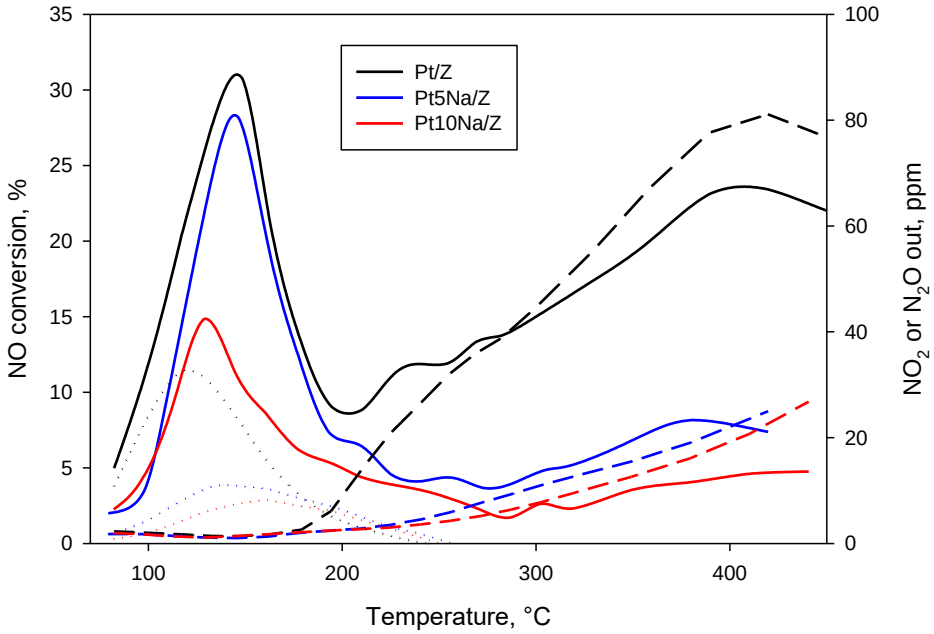

Fig. 1. NO conversion (solid line) and outlet concentration of NO_2 (dashed line) and N_2O (dotted line) of Pt/Z, Pt5Na/Z and Pt10Na/Z.

An Operando-DRIFT study was done to determine the role of adsorbed NOx species involved in the SCR reaction and to identify the possible reaction intermediates.

References

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