Abstract

Abatement technologies for soot and NOX have become a topic of relevant research over the years due to the stricter legislations. Oxidation-based technologies may be a useful solution to reduce soot by converting it to CO2. However, high temperatures are needed. As a result, catalysts that promote soot combustion at lower temperatures are required. Since soot oxidation by O2 requires temperatures above 600°C to start uncatalysed soot oxidation, the possible solution is to use NO2, which is a better oxidant.

Soot can be collected by placing a filter device in the exhaust line. But the problem is that the filter has to be periodically or continuously regenerated, for example by oxidation of carbon in CO2. In this work we study active regeneration that uses NO2 as oxidant by using a catalyst that combine a NOX storage material and oxidation catalyst. This material will store the NOX at temperatures below 300°C as nitrates and release it at the temperature range of regeneration (500-600°C). An oxidation catalyst increases the efficiency of the system.

For this purpose a model NOX storage system 20% MeNO3 supported on ZrO2 (90 m2/g), where Me = Mg, Ca, Sr, Ba (alkaline earth metals) and La, Ce, Pr (rare-earth metals) and Pt as oxidant was prepared. This support was chosen because earth metals and ZrO2 are known to form nitrates upon reaction with NOX.

Textural properties of the catalytic systems were studied by TGA. NOX storage/release cycles were analyzed by FTIR-MS and TPD-MS. Soot oxidation was studied by TPD-MS and six-flow reactor. It was used loose contact mixtures between the soot and catalysts.

It was found that introducing stored nitrates into the system decrease the temperature of soot oxidation more than 70°C in comparison with uncatalysed soot oxidation. The best storage capacity was achieved for Ca and Sr-based systems, 91% and 61% respectively in TPD-MS. The combination of bulk nitrates and platinum increases soot oxidation, as well as the selectivity of soot conversion to CO2. It was stated that platinum has almost no effect on NOX storage capacity but can decrease the temperature of nitrates decomposition/desorption.

Sr based-system was chosen as the final catalyst to optimize in terms of NOX storage and soot oxidation performance. It was found that pretreating the catalyst in reducing atmosphere (air + 5% H2 in argon), storage capacity increase to more than 85% and the amount of NOX desorbed between 200 and 500°C also increased. Different activation procedures were used. The best results were achieved by the “standard” method. Good stability was obtained. ZrO2 supported system follows the same trend as for Al2O3 system. However, zirconia support can store more nitrates.