Low Dimensional Materials and Catalysts

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Active sites and aging properties of metal catalyst particles

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The toxic gases NO and NO_2 (NO_x) form during combustion in vehicles equipped with diesel or lean-burn gasoline engines. These engines have better fuel economy and are therefore preferred considering that the consumption of fossil fuels needs to be reduced due to economical and environmental aspects. However, existing standard three-way catalysts cannot reduce the NO_x species that are formed. They are harmful and are, for example, responsible for acid rain, forming ground-level ozone (which in turn is the major constituent of smog) and are also increasing the risk of respiratory allergies. Methods are developed to remove the NO_x species by exhaust gas after-treatment [1,2]. Also there are efforts to replace gasoline and diesel fuels with natural gas to reduce the net CO_2 emission. The typical levels of unburned methane in the exhausts from engines fuelled by natural gas must be lowered to comply with the emission legislations. Methods for efficient oxidation of methane at low temperature are therefore being developed [3]. During use, the efficiency of the catalysts is degraded where the mechanism responsible for the degradation can be either of thermal or chemical nature. Crucial aspects of the performance of the catalysts are the active sites and also ageing properties. The characteristics and number density of the active sites depend the size and morphology of the catalyst particles which in turn are affected by interface between the noble metal particles and the oxide supports, see Figure 1. This work addresses the effect of the interface between noble metal particles and oxide supports on the efficiency of the catalysts and on the ageing properties, see Figure 2, in different gas compositions. The activity, ageing and dispersion measurements are carried out in bench reactors. The mechanisms of degradation and the structure of the particles and interfaces are studied using electron microscopy and in particular high resolution transmission electron microscopy (TEM) and scanning TEM (STEM). High angle annular dark field (HAADF) STEM imaging using a probe corrected Titan TEM instrument provides Z-contrast information revealing the distribution of the noble metals on the oxide support with atomic resolution. Complementary information is extracted using monochromated electron energy loss spectroscopy (EELS) in the Titan TEM and x-ray diffractometry (including measurements at the European Synchrotron Radiation Facility).

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2. X. Auvray, T. Pingel, E. Olsson and L. Olsson, Appl. Catal. B: Environm. 129 (2013), p. 517.
3. D. Bounechada, S. Fouladvand, L. Kylhammar, T. Pingel, E. Olsson, M. Skoglundh, J. Gustafson, E. Lundgren, M. Di Michiel, M.A. Newton and P.-A. Carlsson, Phys. Chem. Chem. Phys. DOI:10.1039/C3CP44289F.

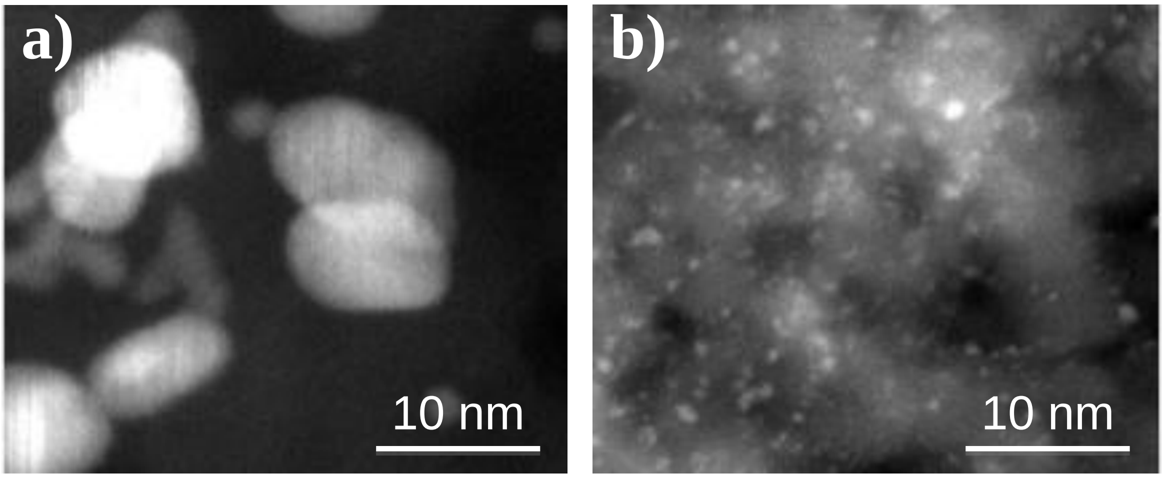

Figure 1. STEM HAADF images showing Pt particles on a) SiO_2 support and b) Al_2O_3 support. The samples were fabricated using identical parameters and the difference in Pt particle size is due to the difference in interaction at the interface between the particles and support.

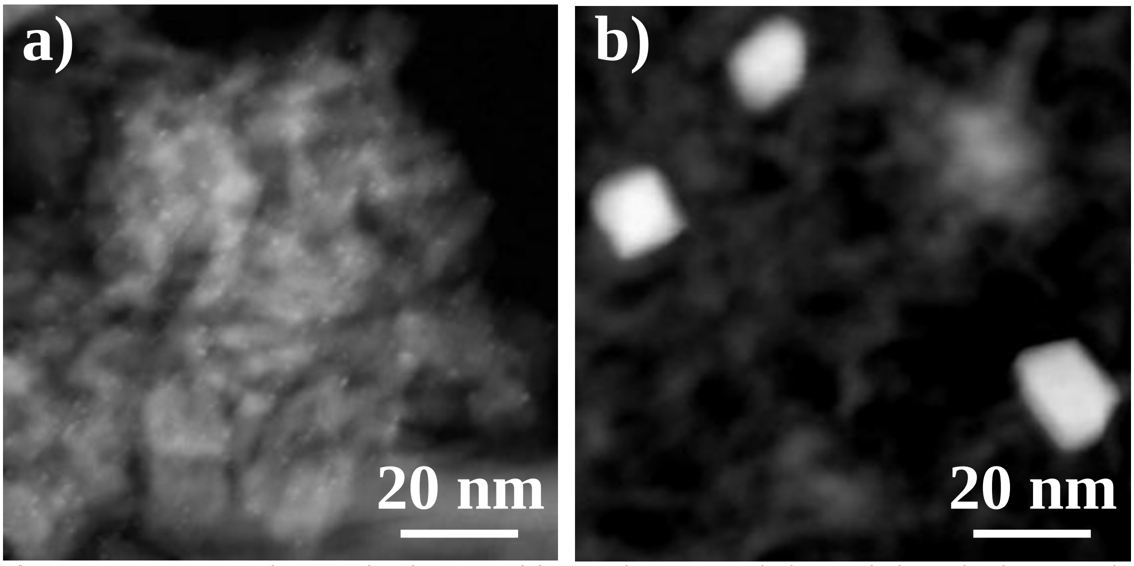

Figure 2. STEM HAADF images showing Pt particles on Al_2O_3 support before and after aging in a gas mixture of O_2 and SO_2 at an elevated temperature. a) As received sample. b) Aged in 10% O_2 and 30 ppm SO_2 at 900 °C for 2h.