# Catalytic Methane Reduction in the Exhaust Gas of Combustion Engines

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***Abstract***

Catalytic methane oxidation contributes significantly to the reduction of the hydrocarbon exhaust of internal combustion engines. In contrast to other gaseous hydrocarbons, significant methane conversion usually occurs only at temperatures above 400°C. In order to continue our previous investigations in this field, we synthesized different batches of single, binary and ternary oxides of cerium, zirconium and praseodymium doped with Pd and Pd-Pt nanoparticles. The primary characterisation of the synthesized materials was carried out by high-throughput impedance spectroscopy (HT-IS) and by in-situ X-ray diffraction (XRD) in the temperature range between 20°C and 850°C. Based on noble metal coated γ-alumina, we produced real wash coats by coating cordierite honeycomb substrates (400cpsi, 33x30x75 mm, see Fig.1) with the mixed oxides synthesized in this study.

The catalytic characteristics of 12 annealed catalyst samples (5h, 750°C, 10% $H_2O$ in air) were monitored using a synthetic gas test bench. According to these measurements, the methane light-off of the Pt-Pd systems appeared at temperatures below 315 °C. To check this result we measured propylene light-off under the same experimental conditions for reference. These measurements confirm that the propylene light-off temperatures of the present prototype catalysts are in the temperature range between 115°C and 135°C, as known for other state-of-the-art automotive catalysts.

For more detailed characterization, a focused ion beam (FIB) lamella from one of the prototype catalysts was prepared and subsequently studied by in-situ transmission electron microscopy (TEM) at elevated temperatures, both under vacuum and in a $CH_4/O_2$ (1:2) atmosphere up to 8 mbar. This study allowed us to track the behaviour of the noble metal nanoparticles, which are assumed to be the most active catalytic sites, up to temperatures of 650°C. The results of the in-situ TEM investigation prove the strong mobility of the noble metal nanoparticles on the γ-alumina surface above 500°C (see frames in Fig. 2.a and b). Moreover, the observed individual steps of particle movement yielded new insights into the special ageing mechanism that takes place in the present system under study.

**Figure 1**: real mini catalyst

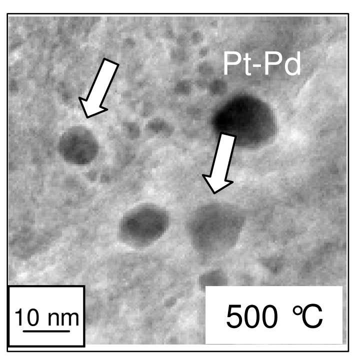


**Figure 2a**: in-situ TEM @ 500 °C
P: 7.3 mbar

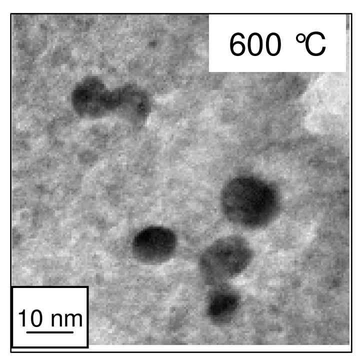


**Figure 2b**: in-situ TEM @ 600 °C
P: 7.3 mbar