

In situ Raman microspectroscopic analysis of soot during temperature-programmed oxidation

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Soot particles generated at incomplete combustions of fossil or bio fuels can influence the human health by causing diseases like lung cancer and have direct and indirect effects on the environment and the climate, *e.g.* as cloud condensation nuclei or interference with the earth's radiation balance. To protect the environment and the people, the EU sets limits for the emission of diesel particles by law. To reach these limitations, cars and trucks are equipped with diesel particle filters (DPF) in order to trap soot particles in the exhaust. However, these DPFs have to be regenerated after a certain amount of soot is trapped inside the filter. In this regeneration step, the soot is oxidized to gaseous products to clear the DPF and remove the soot. As a regeneration at low additional energy demand would be, it would be desirable to produce a reactive soot in the exhaust, which can be oxidized easily but controlled. As it is known that the reactivity of the soot, *i.e.* the temperature at which the soot can be oxidized, depends on the soot's structure (Knauer *et al.*, 2009), effective tools for the determination of the soot reactivity and its structure are necessary to allow a better soot characterization and consequent DPF regeneration optimization.

Currently, two of the most effective tools for soot characterization are temperature-programmed oxidation (TPO) and Raman microspectroscopy (RM). TPO is a tool for soot reactivity determination as the soot is burned in a defined environment with a constant temperature ramp and the oxidation products are quantified by FTIR (Schmid *et al.*, 2011). RM instead provides information about the soot nanostructure as spectral parameters of the Raman spectra are connected to the nanostructure of the soot (Ivleva *et al.*, 2007, Schmid *et al.*, 2011). The aim of the project is to combine these two powerful individual tools in one setup and follow the changes of the nanostructure of the soot *in situ* during heating or oxidation experiments.

Therefore, a measurement device was developed and homebuilt providing the possibility to perform TPO and RM measurements combined in one setup. The measurement cell's inside is heated up to 1000 °C, while the frame is water-cooled, so that the cell can be placed under the objective of a Raman microscope and the soot can be analyzed by RM through a quartz glass window *in situ* during the oxidation. Different possible temperature programs and various gases or gas mixtures provide multiple measurement conditions for thermophoretically deposited soot samples of diverse origin.

With this cell different kinds of test aerosols like spark discharge soot and propane soot have already been analyzed. The reactivity of the soot samples was determined by TPO while the change in the soot nanostructure and the fluorescent background of the sample was analyzed *in situ* by RM. In addition, the heating of some soot samples in inert gas under the Raman microscope was performed. The next step is to perform TPO or isothermal oxidation experiments of spark discharge soot and propane soot generated at different air to fuel ratios with different (oxidative) gas compositions and temperature profiles while monitoring the emission products by FTIR and following the change in the soot structure by RM *in situ* to gain comprehensive results on the soot oxidation with these techniques.

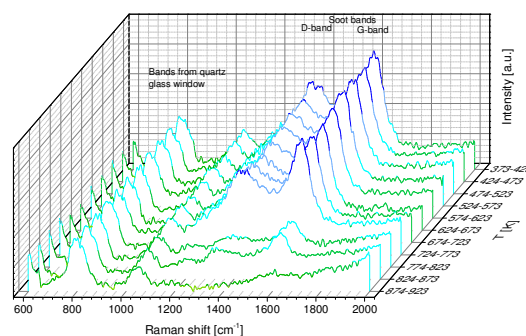


Figure 1. Changes in the spark discharge soot's Raman spectra during the oxidation in 5% O₂ in N₂ at 373-973 K heated with 5 K/min.

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