

15th ETH-Conference on Combustion Generated Nanoparticles

Lintao Zeng, Alfred P. Weber

Institute of particle technology, Clausthal University of Technology

Leibnizstraße 19 D - 38678 Clausthal-Zellerfeld, Germany

Tel: 0049/5323 / 72-2309 Fax: 0049/5323 / 72-2830

E-mail: zeng@mvt.tu-clausthal.de

Catalytic soot oxidation mechanisms in structured layers of nanoparticles

Although investigated intensively within the last decade, efficient continuous regeneration of catalytic coated filter has not been achieved so far. In addition to the lack of tight contacts between catalyst and soot particles, incomplete understanding of the transport mechanisms of oxidizing species may prevent the rational design of appropriate filter geometries. The oxygen transfer mechanism (Oxygen Spillover) has been widely accepted as the general mechanism for the catalytic soot oxidation. According to this mechanism, oxygen activated by platinum may migrate over different routes to the soot. For the transport mechanism Neri et al. suggested that active oxygen migrates by surface diffusion to the soot [1], while Baumgarten and Schuck proposed diffusion in the gas phase [2]. Therefore, in this work the attention is placed on the pathways of active oxygen from Pt to soot, via the gas phase, support surface or both. The investigation would be very helpful for understanding the catalytic soot oxidation processes and improving the regeneration of diesel particulate filter (DPF).

In order to clarify the importance of the different transport mechanisms, the structured layers shown in Fig. 1 were employed. The layers were oxidized in three different combinations of gas phase diffusion, convective transport and surface diffusion, as detailed in Fig.1. By comparing the soot conversion rates of the different setups, the main transport mechanism was identified.

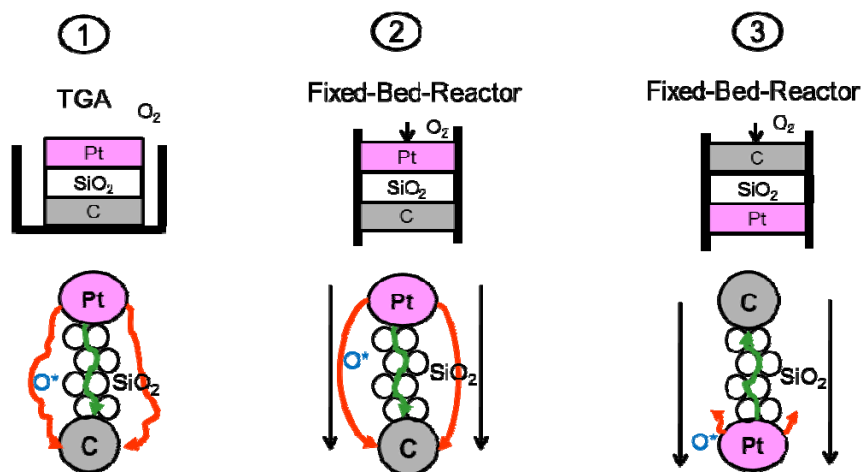


Fig. 1: Soot oxidation by (left) surface diffusion and molecular gas diffusion, (middle) surface diffusion supported by convective transport, (right) pure surface diffusion.

Experiments are carried out with well-defined Pt-SiO₂-C structured layers, in which the carbon layer is separated by an inert SiO₂ layer with variable thickness from the Pt layer. The structured layers are generated by filtering the respective aerosols onto sintered quartz or membrane filters. Then they are oxidized with a temperature ramp of 20K/min in TGA (Fig. 1 left) or in a fixed bed reactor with gas flow in parallel direction (Fig. 1 middle) or in opposite direction to surface diffusion of active oxygen from platinum to soot (Fig.1 right). The CO₂ concentration in the exhaust is measured with FTIR and translated into soot conversion.

The results in Fig. 2 show that the active oxygen may migrate via gas phase and surface diffusion from platinum to soot, even though there is no direct contact between both. For a given conversion (e.g. 20%) the reduction of

reaction temperature represents the catalytic effect of platinum. The effect becomes weaker, if the distance between soot and platinum increases.

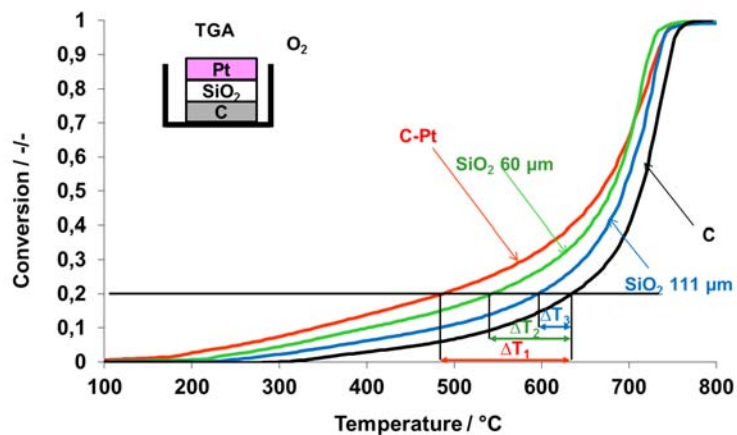


Fig. 2 Conversion of soot as a function of temperature (structured layers with different SiO₂ layer thickness oxidized in TGA)

The results in Fig. 3 indicate which transport mechanisms is dominant. There is almost no difference between non-catalytic soot oxidation (II) and catalytic soot oxidation by surface diffusion (III). On the contrary, the conversion rate of soot was clearly higher if the transport of active oxygen from platinum to soot was increased by convection (I). The results propose that the active oxygen is transported mainly through the gas phase.

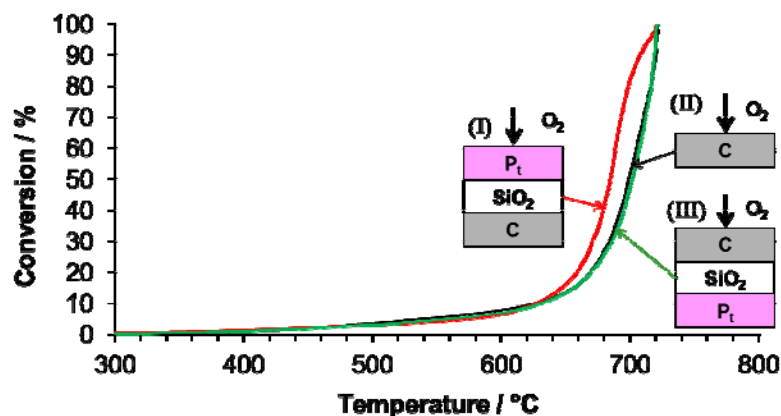


Fig. 3 Conversion of soot as a function of temperature (structured layers oxidized in Fixed-Bed-Reactor)

The experimental results reveal that the main transport of active oxygen takes place in the gas phase. The results can be applied in improving the regeneration of Diesel particle filters (DPF), for example, choosing appropriate geometries, which can increase the transport of active oxygen in gas phase from catalysts to soot.

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References

- [1] G.Neri, L. Bonaccorsi, A. Donato, C. Milone, M. Musolino, A. Visco., Catalytic combustion of diesel soot over metal oxide catalysts, *Appl. Cata. B: Environmental*, vol.11, 217-231, 1997.
- [2] E. Baumgarten and A. Schuck, Investigations about gas phase oxygen spillover, *React. Kinet. Catal. Lett.* Vol. 62, No. 2, 209-216, 1997.

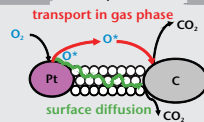


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Institute of Particle Technology, Clausthal University of Technology
Contact: zeng@mvt.tu-clausthal.de phone: +49-5323-722117

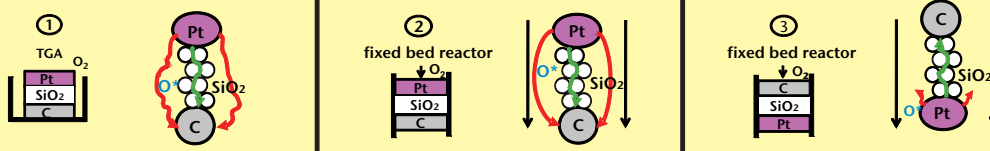
Introduction

- Oxygen spillover as general mechanism in catalytic soot oxidation.
- Surface diffusion [1] and gas phase diffusion [2] proposed as transport mechanism.
- Identification of dominant transport mechanism.
- Possible application for improved geometry of diesel particulate filter (DPF).



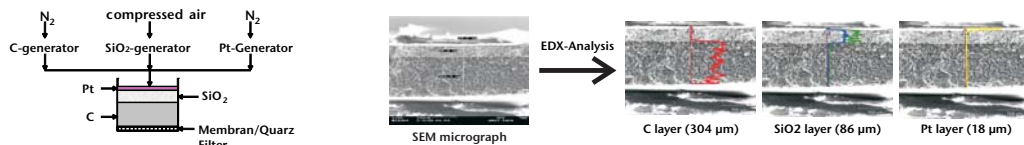
Principle

In order to clarify the importance of the different transport mechanisms, the structured layers shown in the following three schemes were employed. The layers were oxidized in three different combinations of gas phase diffusion, convective transport and surface diffusion, as detailed in schemes. By comparing the soot conversion rates of the different setups, the main transport mechanism was identified.



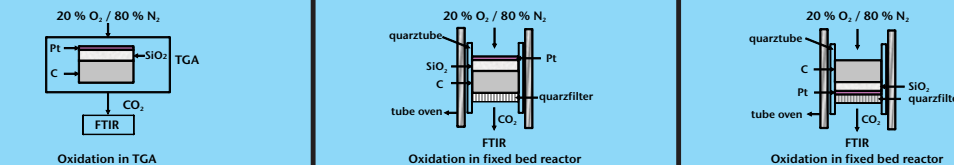
Preparation of structured layers

Experiments are carried out with well-defined Pt-SiO₂-C structured layers, in which the soot layer is separated by an inert SiO₂ layer with variable thickness from the Pt layer as shown in the following scheme and SEM-Micrograph. The structured layers are generated by filtering the respective aerosols onto sintered quartz or membrane filters.



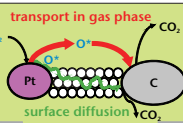
Oxidation

Then the structured layers are oxidized with a temperature ramp of 20 K/min in TGA (down left) or in a fixed bed reactor with gas flow in parallel direction (down center) or in opposite direction to surface diffusion of active oxygen from platinum to soot (down right). The CO₂ concentration in the exhaust is measured with FTIR and translated into soot conversion.



Results

The results show that the active oxygen may migrate via gas phase and surface diffusion from platinum to soot (down left), even though there is no direct contact between both. For a given conversion (e.g. 20 %) the reduction of reaction temperature represents the catalytic effect of platinum. The effect becomes weaker, if the distance between soot and platinum increases [3]. The diagram down to the right indicates which transport mechanisms is dominant. There is almost no difference between non-catalytic soot oxidation (II) and catalytic soot oxidation by surface diffusion (III). On the contrary, the conversion rate of soot was clearly higher if the transport of active oxygen from platinum to soot was increased by convection (I). The results propose that the active oxygen is transported mainly through the gas phase.



Conclusion and outlook

- Gas phase transport as main mechanism.
- Catalyst to be placed upstream of soot layer.

Acknowledgment: The financial support of this work by the DFG under the grant WE 2331/6-3 is highly appreciated.

References

- [1] G.Neri, L. Bonaccorsi, A. Donato, C. Milone, M. Musolino, A. Visco., Catalytic combustion of diesel soot over metal oxide catalysts, Appl. Cata. B: Environmental, Vol.11, 217-231, 1997.
- [2] E. Baumgarten and A. Schuck, Investigations about gas phase oxygen spillover, React. Kinet. Catal. Lett. Vol. 62, No. 2, 209-216, 1997.
- [3] L. Zeng and A. Weber, Spillover-bedingte Reichweite von aktivierten Sauerstoffatomen in der katalytischen Rußoxidation an Nanopartikel-Schichtsystemen, to be published in Chem. Ing. Tech., Vol. 88, 2011.