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Quality of CONTACT in catalytic soot oxidation

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Contact intensity between carbon and catalyst particles affects the efficiency of the oxidation process. We have performed experiments to quantify the effects of the contact intensity. The research was based on the comparison between three different contact types of carbon and platinum catalysts. The catalytic activity of platinum is compared between the following particle systems in the gas phase:

An intensive contact between carbon and platinum particles can be generated in a vapor mixture using a spark discharge generator with platinum and carbon electrodes. The condition would lead to a multi-contact between the mentioned particles, which is called here as "*Fuel Borne Catalyst (FBC)"*. The process is a superposition of nucleation events and physical vapor deposition. A change of experimental arrangement allows to produce a single contact between particles, named *"Doublet"*. In the third type of contact the platinum particles are mixed with carbon agglomerate in the gas phase which yields the *"Random Diffusion Multi-Contact".*

In this investigation both the mixing of the particles and the oxidation are perfomred in the gas phase. The conversion of carbon because of the reaction is measured by FTIR except for the system with Pt/C doublets, where the oxidation rate was measured with TEM analysis. In this case the oxidation rate is measured at different temperature by determination of the carbon reduced projection area [1,2].

Diagram 1. shows the conversion (left) and changes of agglomerate size (right) vs. reaction temperature for different particle systems.

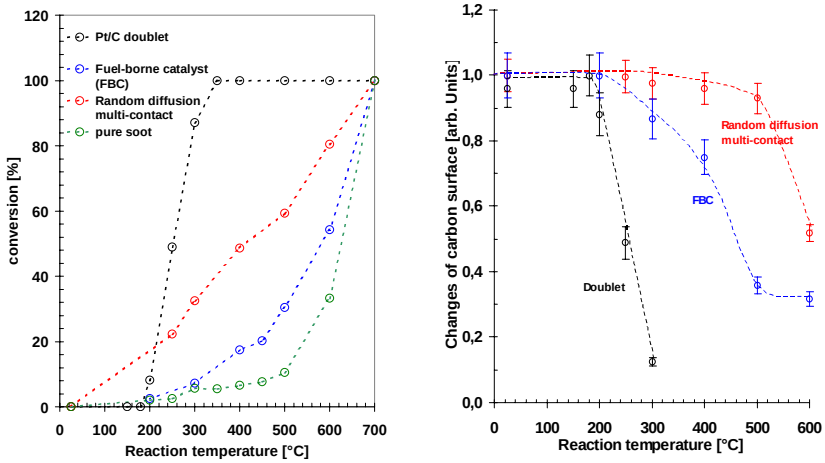

Diagram 1. left: conversion of carbon in catalytic oxidation, right: changes of carbon surface vs. reaction temperature of different contact particle systems

For the catalytic oxidation of carbon in all mixed particle systems a similar value of activation energy, which is several times lower than measured values from the oxidation in the supported system, is observed. From previous investigations it is well

known that the catalyst has no effect on reduction of activation energy in catalytic carbon oxidation. In the earlier investigations, where carbon oxidation was all done in the supported systems, the influencing variables such as transport limitation and gas transport mechanism dominate the reaction. In the present study the measured activation energy for the gasborne particles is lower than reported from the literature.

In order to confirm our new results, the thermal oxidation of two pure carbon sources in the gas phase and as bulk powder is investigated. The synthetic soot from flame process (Printex U) and carbon generated by spark erosion (SD-soot) are used for the investigation on thermal soot oxidation.

Both kinetic parameters of oxidation, E_a and k_0, for bulk powders are in agreement with the literature (140 kJ/mol, 10^7 s^{-1}) [3,4,5]. The calculated value for the activation energy of gasborne soot particles is about 55 kJ/mol. The pre exponential factor is several orders of magnitude higher than those of previous investigations.

Diagram 2. shows the Arrhenius diagram for thermal oxidation of SD-soot and Printex U in bulk form and aerosol and also for catalytic oxidation of FBC.

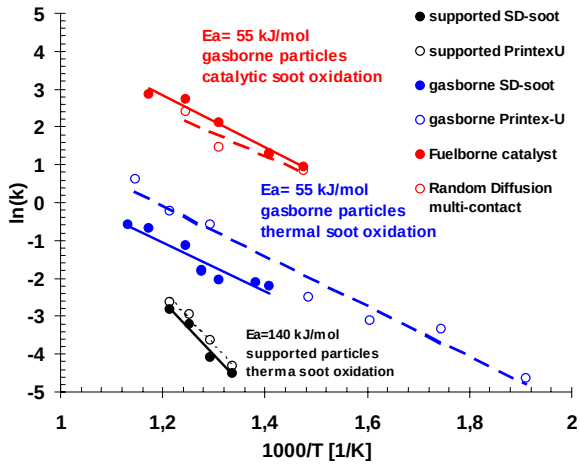

Diagram 2. Arrhenius plot for thermal and catalytic oxidation of supported and gasborne carbon particles

Generally the velocity of reaction for gasborne particles is higher than that of supported particles, which confirms our results. In the catalytic oxidation of our “FBC” and “Random Diffusion Multi-Contact” the same value for E_a is observed. Finally the oxidation rates of all particle systems are compared with the value of pre exponential factor k_0.

References:
[1] Jung et. al, Combust. Flame (2004)
[2] Seipenbusch et. al, Appl. Catal. (2005)
[3] K. Hinot; Technical University of Karlsruhe; Dissertation (2006)
[4] B. Stanmore; Carbon, vol 39 (2001)
[5] J.O.Müller; Catalysis Today, vol 102-103 (2005)

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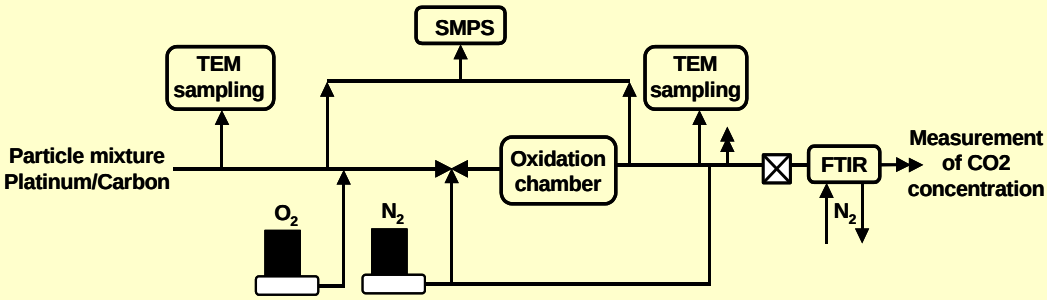
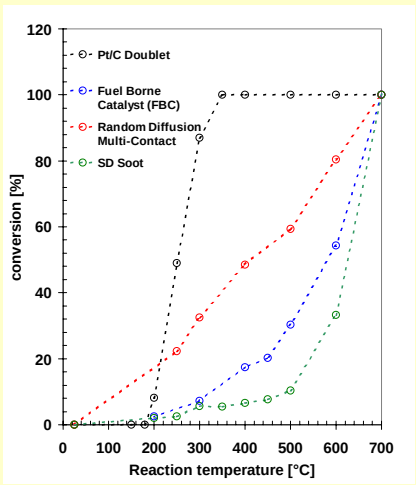
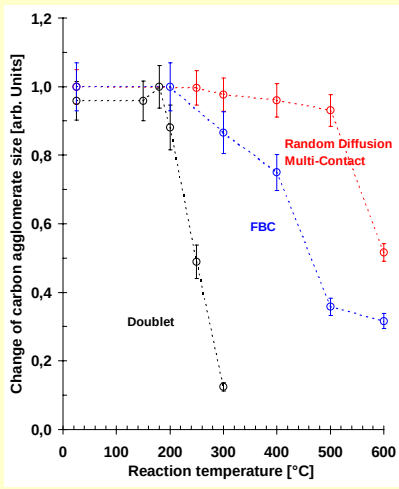
Experimental Setup



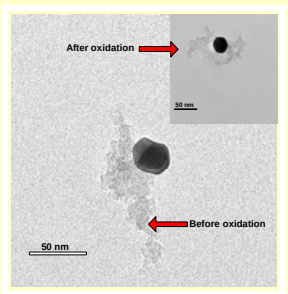
The platinum and carbon nanoparticles are basically generated by spark erosion. All oxidation reactions for the gasborne particles are done in 20 vol% O_2 and in a temperature range of 250 to 600°C. The produced CO_2 concentration from the conversion of carbon was measured in situ by FTIR. Changes of agglomerate size and surface of contact between Pt and C particles are measured by Scanning Mobility Particle Sizer (SMPS) system and TEM analysis, respectively.

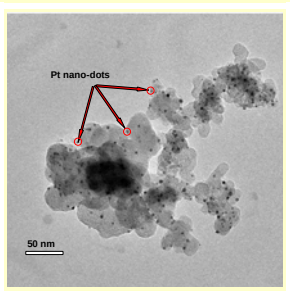
Gas Phase Oxidation

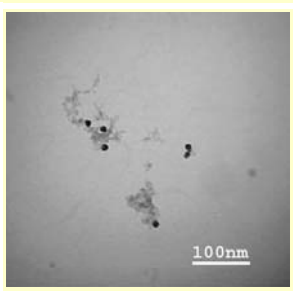




In the left diagram the conversion of carbon for different particle systems is shown. On the right hand the change of carbon agglomerate size, measured by Scanning Mobility Particle Sizer (SMPS), for the same particle systems is presented.







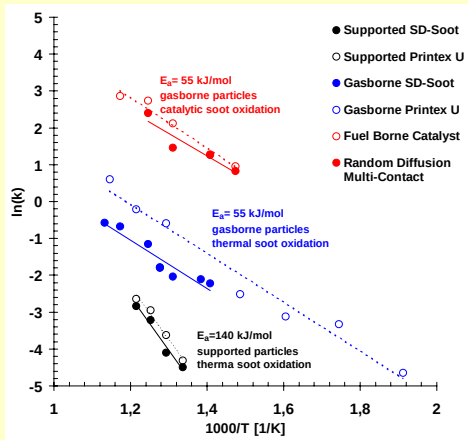
Platinum/Carbon Doublets

Fuel Borne Catalyst (FBC)

Random Diffusion Multi-Contact

The conversion of carbon in the reaction is measured by FTIR except for the system with Pt/C doublet, where the oxidation rate was measured with TEM analysis [3,4]. In this case, the oxidation rate is calculated with the reduction of projection area of primary particles. The Doublet particle system is the super active and the FBC is the least reactive one. Although the high number of Pt-nanodots in FBC denotes a large catalyst surface, the Random Diffusion Multi-Contact" shows a better reactivity.

Activation Energy



In this work the synthetic soot from flame process (PrintexU) and carbon generated by spark erosion (SD-soot) are used for thermal soot oxidation. Both calculated values for oxidation, E_a and k_0, for bulk powder are in agreement with the literature (140 kJ/mol, 10^7 s^{-1}) [1,2]. The calculated activation energy E_a in all different gasborne particle systems is about 55 kJ/mol.

The pre exponential factor k_0 is more than an order of magnitude higher than those of previous investigations. Generally the reaction rate for gasborne particles is higher than that of supported particles, which confirms our results.

In the catalytic oxidation of FBC system and the Random Diffusion Multi-Contact system the same value for E_a is observed. Finally the oxidation rates of all particle systems are compared with the value of pre exponential factor k_0.

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[3] Jung et. al,Combust. Flame (2004); [4] Seipenbusch et. al, Appl. Catal. (2005)

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