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Factors Contributing to Activity in Catalytic Soot Oxidation

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1. Introduction

Soot particles in the exhaust from diesel vehicles are likely to cause lung cancer upon inhalation [1]. The soot particles are therefore typically removed from the exhaust gas by filtration through a ceramic filter [2]. It is necessary with periodic regeneration of the filter, where the filter temperature is increased, and the soot is oxidized. The growing back pressure due to the soot deposits and the temperature increase required for filter regeneration are associated with increased fuel consumption [3]. To limit the increase in fuel consumption it is desirable to develop soot oxidation catalysts that can lower the regeneration temperature - ideally down to the normal temperature of the exhaust gas [2,3]. To facilitate the development of improved catalysts it is of great importance to identify the factors that determine the catalytic activity. Catalytic soot oxidation is a gas/solid/solid reaction, and it has been established that the intimacy in the contact between soot and catalyst is of great importance [2]. In tests with soot and catalyst crushed together (so-called tight contact) the oxidation occurs at significantly lower temperature than with the solids stirred together (so-called loose contact), and both mechanisms are presumably relevant in an actual filter.

2. Experimental

The catalytic materials used in the present studies include both commercial oxides and metals, impregnated samples and especially samples prepared by flame spray pyrolysis according to the method described elsewhere [4]. The catalytic activity has been assessed by temperature programmed oxidation using both a flow reactor [5] and thermogravimetric analyses [4]. Apart from characterizing the employed catalysts by XRD and BET measurements we have also used environmental transmission electron microscopy to study the possibly important role of mobility of the catalytically active phase [6].

3. Results and discussion

A number of factors are observed to be of great importance for the catalytic oxidation. As expected the catalyst surface area is observed to be very important for the oxidation temperature [4]. However, the magnitude of the catalytic surface is not the only parameter of importance. The inherent activity of the surface is strongly dependent on the strength of the oxygen-catalyst bond as measured by the heat of oxygen chemisorption on the catalyst. For soot oxidation in both tight and loose contact with the catalyst the rate constants for a number of catalytic materials outline a so-called volcano curve when depicted as a function of the heats of oxygen chemisorption for the catalytic materials (figure 1). This can be ascribed to the fact that an optimal catalyst binds oxygen strongly enough to activate molecular oxygen, but weakly enough to donate surface oxygen to carbon. We have explored various methods for utilizing the gained insight into the importance of the oxygen-catalyst bond strength to develop improved catalysts by interpolation, doping or combination methods. As an example we have, inspired by the fact that Fe_2O_3 and Cr_2O_3 are found on either side of the volcano curve in figure 1, found that $\text{Fe}_x\text{Cr}_y\text{O}_z$ binary oxides show improved activity compared to either of the pure oxides. The soot oxidation is also strongly influenced by the gas composition. The presence

of very reactive NO₂ in addition to O₂ lowers the oxidation temperature significantly in loose contact with a catalyst but shows only a limited effect on the oxidation rate in tight contact with a catalyst.

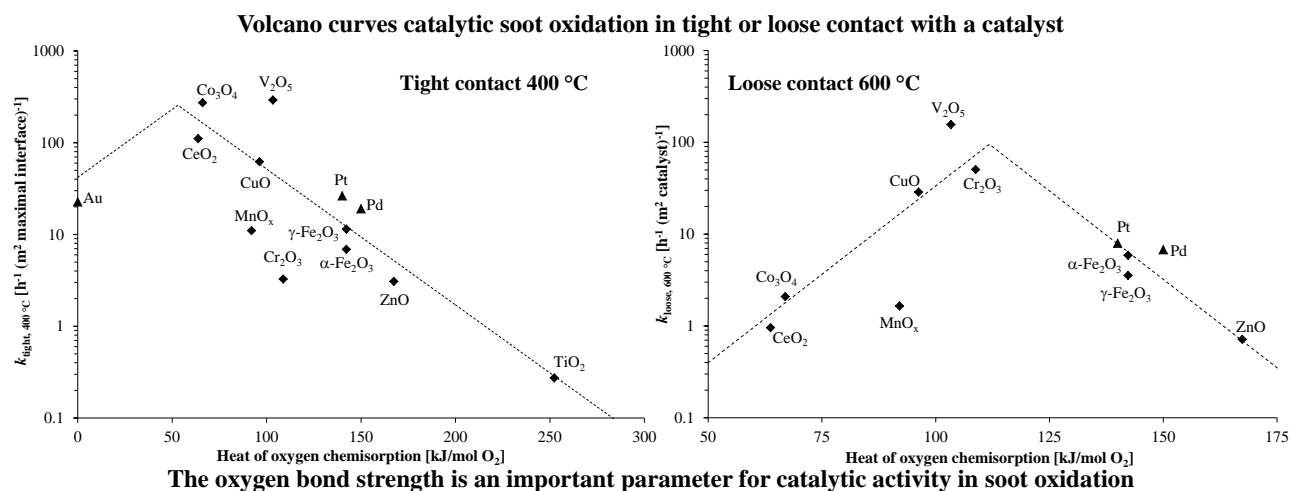


Figure 1. The logarithm to the rate constant for tight contact soot oxidation at 400 °C (left) and loose contact soot oxidation (right) as a function of the heat of oxygen chemisorption on the catalyst. The data outline a volcano curve in both cases [5].

Finally, our findings also illustrate the importance of the contact between catalyst and soot and its relation to catalyst mobility. The high activity of silver nanoparticles in loose contact oxidation can for example be correlated to the high mobility of the silver, which facilitates the creation of additional soot/catalyst interface (figure 2).

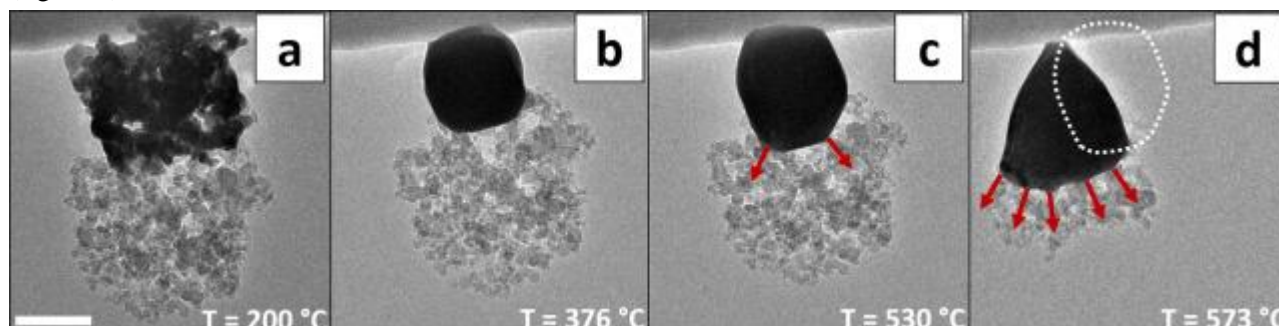


Figure 2. TEM images of a loose contact mixture of soot and silver during in-situ heating in 3 mbar oxygen. The images illustrate how the cohesive force at the soot/silver interface as a result of the catalyst's mobility expands this interface at which the oxidation reaction is focused [6].

4. Conclusions

This work contributes to the understanding of the factors that determine the rate of catalytic soot oxidation and illustrate the importance of the catalyst's oxygen bond strength, the mobility of the catalyst, the catalyst particle size and the composition of the oxidizing gas. The establishment of the factors determining activity in catalytic soot oxidation will not only be a significant fundamental advance in the understanding of the catalytic reaction but also be a prerequisite for the development of improved soot oxidation catalysts that can improve the fuel economy of diesel vehicles.

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