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Technical note

A comparison of diesel soot oxidation rates measured with two different isothermal set-ups

Simone I. Seher^{a,*}, Michaela N. Ess^b, Henrike Bladt^b, Reinhard Niessner^b, Gerhart Eigenberger^a, Ulrich Nieken^a^a Institute of Chemical Process Engineering, University of Stuttgart, Böblinger Str. 78, 70199 Stuttgart, Germany^b Institute of Hydrochemistry, Chair of Analytical Chemistry, Technische Universität München, Marchioninistr. 17, 81377 Munich, Germany

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ABSTRACT

Soot aerosol particles which are emitted by incomplete combustion e.g. in diesel engines are known to have a big influence on the environment and the human health. A detailed understanding of their elimination through trapping and oxidation in diesel soot filters is therefore of great importance. Isothermal experiments of diesel soot oxidation are very advantageous for establishing detailed reaction kinetics. Nevertheless, most soot oxidation studies in the literature rely on non-isothermal burn-off experiments. In this study we present and compare two different experimental set-ups for determining isothermal soot oxidation rates. Such experiments gain importance in order to identify engine operation conditions and/or diesel fuel compositions, which produce more reactive soot, which is easier to oxidise. The first set-up, the so-called flat bed reactor (FBR), was originally developed for determining exhaust purification kinetics, using segments of exhaust monoliths with realistic through-flow under reproducible isothermal conditions. It has been adapted to study soot oxidation in segments of commercial diesel soot filters. The second set-up was originally designed for studying diesel soot oxidation in temperature programmed oxidation (TPO) experiments and requires a lower experimental effort. Although the two set-ups are very different, basic results of the oxidation experiments obtained turn out to be quite comparable, in particular under active regeneration conditions. This allows us to reduce the experimental effort by replacing some of the isothermal FBR experiments by isothermal TPO.

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1. Introduction

Aerosol particles of natural and anthropogenic sources are ubiquitous in our atmosphere and influence climate, environment and human health in many ways depending on their origin and properties. One main source of anthropogenic aerosol with high impact on the environment and climate is the incomplete combustion of fuels which produces soot aerosol particles, mainly composed of carbon, that contribute to the high level of fine and ultrafine particles especially in the urban areas. Because of the negative impact of diesel exhaust, which has been classified as being cancerogenic for humans by the WHO (2014), legal limits of maximum soot emission have been introduced in most countries. Initially, most car manufacturers were able to meet these limits with engine adjustments at the expense of increased NO_x emissions,

* Corresponding author.

E-mail address: simone.seher@icvt.uni-stuttgart.de (S.I. Seher).

exploiting the soot-NO_x trade-off. However, stronger NO_x emissions limits and the limit for the particle number, which was introduced in Europe with EURO 5b in 2011 in addition to the mass limit, have meanwhile enforced the introduction of diesel particulate filters (DPF) as a standard method for the reduction of particulate matter.

Intensive research during the last decades (Adler, 2005; Bardon et al., 2004; Howitt & Montierth, 1981; Ogyu, Ohno, Hong, & Komori, 2004; Olson & Martins, 2005) resulted in reliable DPF systems with a filtration rate over 95% (Adler, 2005) and material properties which resist the thermal and mechanical stress during filter regeneration. But efficient filter regeneration, minimising additional fuel consumption, remains a main challenge. Thus, aerosol characterisation of the diesel soot is important for further reduction of the harmful emissions. One powerful diesel soot characterisation method is the off-line oxidation of the soot particles emitted by diesel engines and collected on filters to simulate the regeneration of a DPF in the laboratory.

During regeneration, soot is oxidised under the influence of O₂ at temperatures above 500 °C (active regeneration) or under the influence of O₂ and NO₂ at temperatures around 350 °C (passive regeneration). Passive regeneration can proceed during standard motor operation and is therefore called continuous regeneration as well. However, passive regeneration at moderate temperatures is significantly slower than active regeneration. As a consequence, active regeneration is additionally necessary in certain intervals, leading to increased fuel consumption. An interesting option to reduce the number and time of active regenerations and the related fuel consumption is the generation of a more reactive soot, which on the one hand improves the efficacy of the passive regeneration and on the other hand shortens the active regeneration time. Finding conditions, such as certain engine operating points or special compositions of diesel fuel which produce more reactive soot, is consequently an important issue. As a prerequisite for this purpose, the oxidation of soot produced under different specific conditions has to be analysed efficiently and reliably.

One possible approach is the temperature programmed oxidation (TPO), in which the regeneration temperature rises with a constant rate, leading to the classification of a soot reactivity index (Schmid, Grob, Niessner, & Ivleva, 2011). However, for establishing more detailed and reliable soot oxidation kinetics, the separation of temperature effects from other effects, such as diminishing soot mass during regeneration or a varying oxidant concentration, is necessary. Because of the exothermal soot combustion, techniques to reduce or avoid heat accumulation during oxidation have been developed. Some rely on dilution of soot with inerts, others on oxygen step-response or on a combination of both (Yezerets et al., 2005). But these methods use a packed bed of soot particles instead of a soot layer deposited on a DPF. Studies with soot, generated in special burners and deposited on commercial DPF filter segments, seem more reliable if isothermality is achieved by efficient heat dissipation (Hauff, Tuttlies, Eigenberger, & Nieken, 2012; Peck & Becker, 2009; Schmeißer, de Riva Pérez, Tuttlies, & Eigenberger, 2007). But the question remains, how close the burner soot represents real engine soot.

In this study we therefore use soot, generated by a commercial diesel engine and deposited under reproducible conditions, either in DPF segments or on a flat metal fibre filter. This soot is oxidised under isothermal conditions, either in a FBR or with the TPO set-up.

2. Experiments

Two different set-ups, described in detail in the following sections, are used for the isothermal oxidation of soot. The soot regenerated with these set-ups was generated in a joint cooperation project with a EURO-5 diesel engine (Mercedes-Benz OM651-EU5) by “Lehrstuhl für Technische Thermodynamik und Transportprozesse”/Bayreuth Engine research Center, University of Bayreuth (LTTT/BERC). The filters were placed behind the diesel oxidation catalyst (DOC) and their retainers were electrically trace heated at 200 °C while sampling. For the experiments reported below, the engine was fuelled with B7 diesel (standard diesel with a maximum of 7% of bio diesel) and was run at a defined and constant operating point during sampling. Therefore, the same soot was used for both isothermal set-ups. However, two different types of filters, shown in Fig. 1, were utilised for sampling and subsequent oxidation experiments in the two set-ups. A two-row segment made from

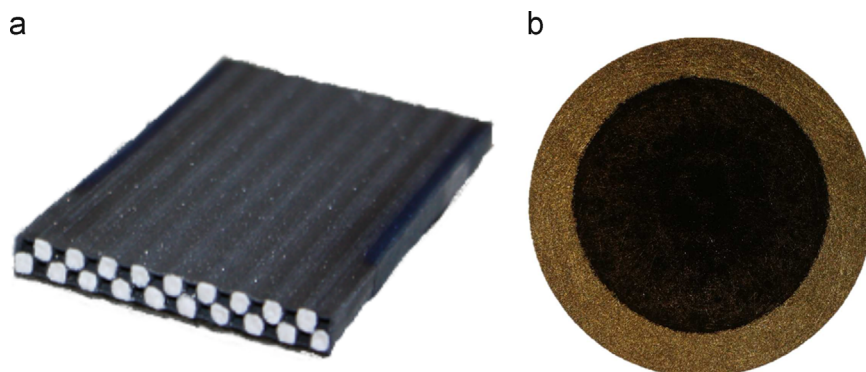


Fig. 1. (a) DPF segment used for the FBR; (b) metal fibre filter used for the TPO.

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