

Impact of soot structure on oxygen reactivity

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Highlights

- Soot oxidation reactivity study.
- In Situ surface area measurement with progress of oxidation.
- Measurement of soot nanostructure with Raman spectroscopy and HRTEM imaging.

1. Introduction

In order to minimize the emission of soot, particulate filters must be used in diesel exhaust systems. These filters must be regularly regenerated by oxidation to remove the deposited soot and reduce the pressure drop. Knowledge of soot reactivity is essential for an optimal regeneration strategy. During gasification with oxygen, the soot undergoes structural changes. The initial morphology of the particles and their development influence the course of the heterogeneous reaction. In this study, the reactivity of soot is analyzed using a set of experimental methods.

2. Methods

A Combustion Aerosol Standard (CAST) soot generator is used to generate soot particles with different size distributions, which are deposited on a filter. The size distribution of the agglomerates is measured with a scanning mobility sizer, while the nanostructure of the primary particles is analysed with HRTEM and Raman spectroscopy. Information on the reactivity of the soot is obtained by temperature-programmed oxidation. The surface area of the soot structure after stepwise oxidation is measured by nitrogen adsorption [1, 2] to determine the surface-specific reaction kinetics [1]. The methodology is illustrated in Figure 1.

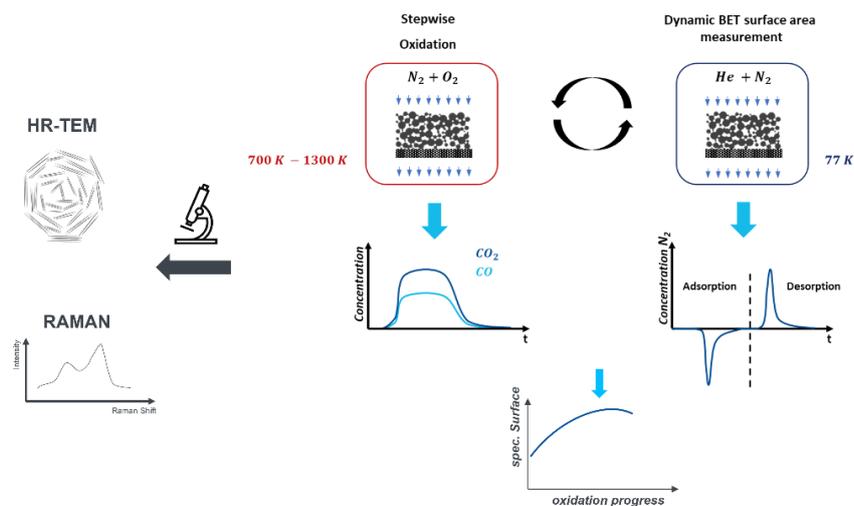


Figure 1: Schematic illustration of the experimental procedure.

3. Results and discussion

The results of the temperature programmed oxidation suggest that the reactivity increases with decreasing mobility diameter (from D_1 to D_4), as it is shown in Figure 2. In Figure 3 the specific surface development with progress of oxidation is reported. The reactivity cannot be correlated with the initial specific surface area, as the most reactive species D_4 has the lowest initial specific surface area. The higher reactivity might result from its amorphous internal structure, exhibiting shorter, disordered fringes as can be observed in the HRTEM images. In comparison, the less reactive species show a core-shell structure, with turbostratic ordered longer fringes. With progression of oxidation, all samples show an extreme increase in specific surface area, which might have a substantial impact on the reaction progression.

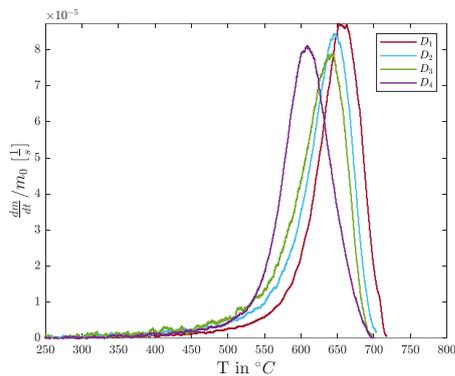


Figure 2: Temperature programmed oxidation.

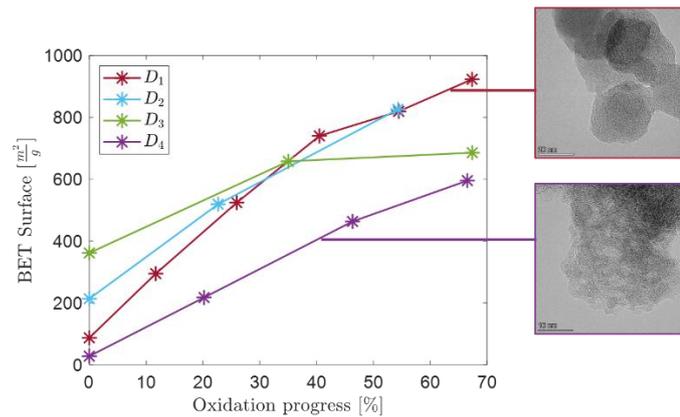


Figure 3: Specific surface with progress of oxidation for different soot samples. On the right: TEM images.

4. Conclusions

Since reactivity of soot cannot be explained by the available surface area, the nanostructure of the particles must be examined. To describe the reaction kinetics, the evolution of the surface must be followed.

References

- [1] A. Strzelec, T. J. Toops und C. S. Daw, „Oxygen Reactivity of Devolatilized Diesel Engine Particulates from Conventional and Biodiesel Fuels,“ *Energy & Fuels*, Bd. 27, Nr. 7, pp. 3944-3951, 2013
- [2] S. Seher, „Experimentelle Charakterisierung und Modellierung der Rußoxidation,“ Logos Verlag Berlin GmbH, Berlin, 2017.

Keywords

Heterogenous reaction; Soot; Oxidation; BET-Surface