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Investigation of soot oxidation by coupling LII, SAXS and scattering measurements

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ABSTRACT

This work presents an *in-situ* characterization of the oxidation of soot particles in flames by coupling three different techniques. Small Angle X-ray Scattering and Static Light Scattering were used in order to provide information on the size of the primary spheres and the aggregates respectively. Laser Induced Incandescence was also used to determine the soot volume fraction in the flame. Flame temperatures and soot residence time were also determined. These techniques were combined to provide a complete description of the impact of the oxidation process on soot aggregates (aggregate and primary sphere densities, size distributions). In order to limit the phenomena to oxidation, soot was generated upstream by a miniCAST generator and injected into a non-sooting flame. Amongst other results, it is shown that primary sphere diameter reduction is accompanied by an increase of the geometric standard deviation. This effect can be modeled by considering the transition from a volume (diffusion) to a surface (reaction) oxidation process.

1. Introduction

Soot emissions from combustion processes have important environmental and health consequences and so the characterization of soot particles and their formation mechanisms are subjects of ongoing research [1–3]. Soot oxidation [4–7] is a key process in the elimination of soot nanoparticles produced during the combustion and the proper control of oxidation, by for example using optimized fuel/air ratios and ensuring proper mixing, is essential in reducing automobile, aircraft and marine engine emissions.

Most of the experimental studies related to the oxidation of soot particles are *ex-situ*, based on the study of the behavior of deposited materials heated in furnaces [8–12]. If these investigations are of interest for the optimization of different catalytic processes including the regeneration of particulate filters [13], they do not yield information on the physical processes involved in the oxidation of soot particles under flame conditions where the OH radical is a major oxidative species and can dominate the role played by O_2 [14,15]. However, the reduction of soot particle emissions from different combustion systems can also be envisaged by an optimization of the combustion process itself. Numerical modeling is a promising tool to optimize combustors; nevertheless, the main mechanisms involved in soot formation (mainly surface growth and oxidation) need to be better understood [16]. Even if O_2 and OH are considered to be the most oxidizing species, the role played by each one is not yet perfectly understood [14,17,18]. It is generally admitted that OH is more reactive than O_2 thus more likely to react on the soot particle surface, while O_2 can penetrate further into the particles, favoring their internal burning [18–20]. The depth in the attacked particle is not known nor is the precise mechanism producing the aggregate fragmentation (rupture of the necking between primary spheres or internal burning) [20–23]. Indeed, the precise mechanisms for reactions at the particle surface are not clearly understood either [24]. For these reasons, many studies

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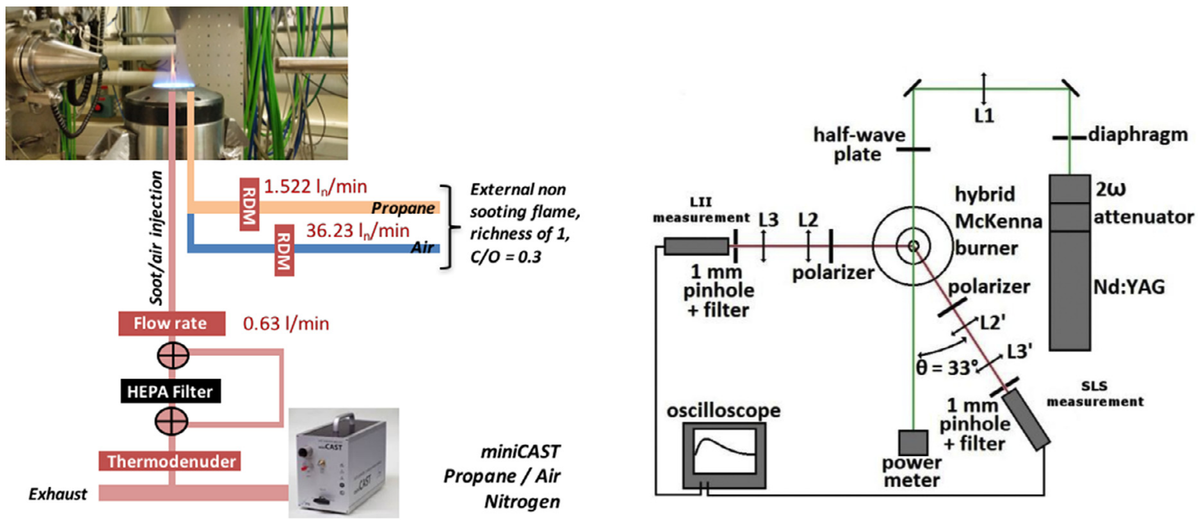

A: Injection of soot produced by miniCAST at the center of the non-sooting flame (modified McKenna Burner). The picture illustrates the SAXS experiment at the SOLEIL synchrotron (Swing beamline).

B: Experimental apparatus for the LII and SLS experiments (seen from above).

Fig. 1. Experimental setup.

have been performed in flames to characterize the oxidation process using *ex-situ* techniques [10–12,20,22,25–28] or *in-situ* optical techniques [15,18,19,29–32] generally based on laser light extinction, scattering and the depolarization ratio.

Most of these studies, however, cannot isolate oxidation from surface growth processes occurring within sooting flames. One interesting way to overcome this drawback and to focus on the oxidation process in isolation, is to inject previously produced soot particles into a non-sooting flame (i.e. a "*two stage burner*") as previously performed by Neoh et al. [19], Echavarria et al. [20] and very recently by Ghiassi et al. [28] and Sirignano et al. [33]. Indeed, in this case, the behavior of the soot particles is not representative of natural flames, nevertheless, it allows the oxidation process to be isolated from surface growth occurring simultaneously, for a better understanding of the soot life-cycle. The final aim of such experiments is to be able to produce accurate oxidation models for numerical soot formation codes.

With the exception of Neoh et al. however, all this work was *ex-situ* involving the sampling of the particles. Neoh et al. [19] performed optical and *in-situ* measurements on a two-stage burner by coupling light scattering and extinction but their results were based on Mie theory, thus considering soot as spherical particles. Unfortunately, it has been shown that Mie theory is not relevant for fractal aggregates [31,34]. Moreover, the use of visible light does not allow the very small scales to be investigated which are typical of those of the primary spheres (around 30 nm).

Small Angle X-ray Scattering (SAXS) is a technique which can provide information on the size and shape of nanoparticles in the range of a few to tens of nanometers. This technique has already been used for the *in-situ* characterization of soot [35–40] but these studies were not focused on the specific examination of the oxidation process. Nevertheless, most of the SAXS experiments performed up until now, do not yield information concerning primary particle diameter and aggregate size to be obtained at the same time, when the latter exceeds 100 nm. This information can be obtained by means of complementary measurements based on visible Static Light Scattering (SLS). Another powerful optical and *in-situ* technique for characterizing the local soot volume fraction in flames, is Laser Induced Incandescence (LII) [41] and for these reasons, the current work has combined SAXS, SLS and LII for the characterization of the oxidation of soot particles in a two-stage burner.

The main improvement in terms of diagnostics used in comparison with the work of Neoh et al. [19] is the coupling of three complementary techniques and the use of an optical model, dedicated to fractal aggregates. Using these three techniques, the primary particle size distribution, number density, aggregate size, soot volume fraction and surface area can be determined in a single study. The impact of oxidation on the primary sphere internal microstructure cannot be investigated by these techniques; nevertheless, this aspect has already been examined in previous studies [9,42–44].

2. Experimental apparatus

In the present experiment, the two-stage burning strategy is achieved by injecting well controlled and characterized soot particles into the centerline of a non-sooting, premixed flame. The experimental arrangement is shown in Fig. 1. Soot is first produced by a miniCAST burner. A thermodenuder (TSI 3065) is used to ensure that volatile species will not condense onto the soot particles after their generation before their injection into a non-sooting flame. A modified version of the McKenna burner with a central tube is used in order to make possible the injection of the soot into the centerline of a non-sooting premixed flame. This strategy has already been used in order to inject gases [45], metal catalyst precursors [46], or different sprays [47] into flames. The McKenna burner presents the advantage of generating stable 1D flames favoring their modeling.

A bypass allows either soot and its surrounding gas to be delivered into the center of a McKenna burner or only the surrounding gas by removing the soot particles by means of a high-efficiency filter (MSA Safety). This procedure was used for the subtraction of the background signal for SAXS measurements. The composition of this surrounding gas has been determined using a Testo