



Soot oxidation-induced fragmentation: Part 2: Experimental investigation of the mechanism of fragmentation



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ABSTRACT

Experiments in a two-stage burner, with ethylene as the fuel, were conducted to further understand the mechanisms associated with the fragmentation of soot particles. Experiments were grouped in the following three categories, based on the main objective of the corresponding experiment: (i) the effect of different inert gases, Ar, He, N₂ (ii) the effect of an increase in N₂ dilution and O₂ concentration (iii) and the effect of particle mobility size on the fragmentation. It was shown that the onset of the fragmentation depended on the presence of large aggregates in the flames. In addition, in the presence of aggregates, the rate of fragmentation was found to be inversely proportional to the peak temperature. It was postulated that at lower flame temperature where the soot burnout was delayed, O₂ molecules were more likely to diffuse into the bridges between particles and burn them. Also, a role for OH oxidation during fragmentation was postulated by probability analysis, since fragmentation was experimentally shown to continue in the region of the flame where OH was present.

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1. Introduction

Soot is formed in a flame under complex steps including the molecular precursor formation, particle inception, coagulation, and growth [1–3]. As result of collisions between particles, aggregates consisting of a large number of primary particles can be formed [4,5]. Following this, aggregates are destroyed through two parallel processes: oxidation and fragmentation [6]. Fragmentation is the process of breaking-up particle aggregates into smaller ones, reflected by the observable increase of ultrafine particles. The fragmentation of soot in the flame was first experimentally observed by Neoh et al. [7,8], where soot oxidation and fragmentation was studied in a two-stage burner. Soot was produced in a first-stage premixed burner and then oxidized in a second stage. They saw evidence of fragmentation at higher fractional carbon burnout, around 0.7–0.8 for a methane–air premixed system. Two hypotheses were presented; that fragmentation was a result of oxygen diffusion in bridges between particles which then burn and break apart; or, that oxygen diffuses into a primary particle, causing internal burning, increasing particle porosity, and eventually breaking it apart. Garo et al. [9] also saw evidence of soot fragmentation, occurring at approximately 0.75 fractional burnout for a methane–air diffusion flame. Puri and

coworkers' [10] observations in a laminar ethylene diffusion flame showed that when soot particles are transported further downstream, where coagulation was not dominant, the degree of particle aggregation started to decrease. They concluded that this observation was related to the soot oxidation-induced fragmentation. Xu et al. [11] reported that soot aggregate fragmentation occurred in the oxidation region in their experimental study of a laminar C₂H₄/air diffusion flames at atmospheric pressure. Recently, soot fragmentation was also studied by Echavarría et al. [12] with an experimental configuration similar to Neoh et al. [8]. A scanning mobility particle sizer (SMPS) was used to obtain particle size distributions (PSDs) and to characterize fragmentation in an ethylene flame.

Numerical models have also shown the importance of fragmentation to correctly predict particle burnout and particle size in flames. Zhang et al. [13] showed that, without considering fragmentation, there is a discrepancy between model prediction and experimental measurement of particle mobility diameter during oxidation. He assumed that the fragmentation was the result of breaking the weak bonds connecting primary particles in an aggregate structure. Harris and Maricq [14] used an arbitrarily chosen fragmentation rate to predict the steady-state soot particle size distribution. They showed that the addition of soot aggregate fragmentation to the Schmoluchowski aerosol coagulation model significantly improved their model predictions; without fragmentation, the model over-predicted the number of particles at larger particle diameters. Mueller et al. [6] developed a model within the hybrid method of moments (HMOM) to predict

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Table 1
Experimental conditions.

No.	ϕ_1^a	ϕ_{Overall}^b	ϕ^c	Secondary oxidizer composition
Case 1 (base case)	1.98	0.9	3.77	21%O ₂ –79%N ₂
Case 2	1.98	0.9	3.77	21%O ₂ –79%Ar
Case 3	1.98	0.9	3.77	21%O ₂ –79%He
Case 4	1.98	0.9	3.77	60%O ₂ –40%N ₂
Case 5	1.98	1.15	3.77	21%O ₂ –79%N ₂
Case 6	1.80	1.10	3.77	21%O ₂ –79%N ₂

^a Equivalence ratio in the first burner.

^b Overall equivalence ratio.

^c Cold gas velocity, cm/s (STP).

particle size distribution by taking into account the fragmentation of aggregates. They considered a mechanism of the fragmentation based on the idea of internal burning. More recently, Sirignano et al. [15,16] used a discrete sectional model to give a further proof of the importance of fragmentation in determining the final amount and the morphology of the particles in a counter flow diffusion flame as well as a premixed flames.

The lack of data on soot fragmentation has been a barrier in identifying the appropriate mechanisms. A better understanding of the soot fragmentation processes can improve the existing models for predicting size distribution and mean properties of particles as they undergo oxidation. The goal of this study is to systematically perform a set of experiments to identify the mechanism of fragmentation. The experiments can be classified as follows,

- 1- The effect of flame temperature on fragmentation by using various diluent gases (N₂, Ar, He) (Section 3.1).
- 2- The effect of flame temperature on fragmentation by altering the nitrogen-to-oxygen ratio (Section 3.2).
- 3- The effect of fuel lean and fuel rich condition on fragmentation (Section 3.2).
- 4- The effect of variation in the mobility particle size on fragmentation (Section 3.3).

2. Experimental set-up

The experimental setup is schematically shown in Fig. 1. The experiments were carried out in a two-stage burner similar to that used in previous studies [12,17,18]. An ethylene stream (A-3) and air stream (A-1) were mixed in the manifold (M-1) and then passed to the premixed section of the first stage burner (stainless steel pipe, 51 mm ID, Schedule 80, 127-mm long). A fuel/air mixture was burned under a fuel-rich condition (see Table 1) in a laminar premixed flame at atmospheric pressure in the first stage resulting in soot formation in the form of primary particles and aggregates, along with a gas mainly composed of H₂, CO, CO₂, N₂, and H₂O. The secondary oxidizer stream (A-2) was introduced into the mixture. Products were oxidized under slightly lean or rich condition, in the second premixed flat-flame burner. The two-stage burner separates the formation process (first burner) from oxidation (second burner). The flame was stabilized through a tube bundle (1/16" ID, 1" long) and a nitrogen shroud was used to isolate the flame from surrounding air. While the cold-gas velocity was constant, the equivalence ratio in the first burner was adjusted by changing the ratio of fuel stream (A-3) to air stream (A-1) and the overall equivalence ratio was defined by the ratio of fuel to total oxidizer (oxygen in the bottom and top burners) fed into the burner over the stoichiometric ratio:

$$\phi_{\text{overall}} = \frac{\left(\frac{\text{Fuel(A-3)}}{\text{Oxygen(A-1)+Oxygen(A-2)}} \right)_{\text{Actual}}}{\left(\frac{\text{Fuel}}{\text{oxygen}} \right)_{\text{Stoichiometric}}} \quad (1)$$

To investigate soot fragmentation, we carried out a wide range of experiments (reported in Table 1). These experiments were

designed to investigate the effect of equivalence ratio in the first burner (ϕ_1), the overall equivalence ratio (ϕ_{overall}), and the composition of secondary oxidizing stream on the fragmentation. Changing ϕ_1 was achieved by adjusting the ratio of fuel stream (A-3) to the oxidizer stream (A-1); this directly affected the soot formation in the first burner and consequently the particle mobility size that appeared in the secondary burner. The comparison between the Cases 5 and 6 demonstrated the result of changing the equivalence ratio in the first burner when the other parameters were kept the same. Changing ϕ_{overall} impacted the flame temperature due to adding more or less oxygen (rich or lean flame). Cases 1 and 5 were performed to show the effect of ϕ_{overall} on the fragmentation when the other parameters were the same. The composition of the secondary oxidizer stream was varied in two ways (i) mixing the same amount of O₂ with different inert gases such as N₂, Ar, and He (ii) using different concentrations of O₂/N₂ mixture. The different inert gases (Cases 1–3) were tested to study the diffusion and heat capacity effects on the fragmentation. In addition, different concentrations of O₂/N₂ (Cases 1 and 4) allowed for investigating the effect of nitrogen dilution which also changed the flame temperature.

Temperature and particle size distributions (PSDs) were found for all cases and specific gas-phase compounds were measured for Cases 1–3. All measurements were performed in the centerline of the flame at different heights above the secondary burner (HABs).

The flame temperature was measured using an uncoated 0.2032-mm Pt/Rh 70%/30% thermocouple (Type B). The radiation correction for the temperature was similar to that of McEnally et al. [19].

PSDs were found with online sampling using a scanning mobility particle sizer (SMPS) including a TSI 3080 classifier and a 3025 ultrafine condensation particle counter coupled to a 3085 nano-DMA. The SMPS was optimized to operate in the 3–135 nm range with a sheath flow of 15 L/min and an aerosol sample flow of 1.5 L/min. The SMPS sampling method was similar to that used by Zhao et al. [20]. The validity and reliability of the SMPS sampling procedure and data acquisition have been extensively discussed [20,21]. The sampling probe was placed horizontally and the orifice (0.24 mm) was faced down toward the incoming flame gas above the burner. The probe was mounted on a translation stage with the accuracy of probe positioning estimated to be 0.1 mm. The soot-laden gas sample was immediately diluted by nitrogen at the rate of 30 L/min (STP) to quench oxidation reactions, and minimize particle coagulation and thermophoretic deposition. Zhao et al. [20] showed that particle diffusion losses and particle coagulation can be minimized by systematically increasing the dilution ratio to a critical value where the particles size distribution function becomes independent of the dilution ratio. The pressure difference across the probe orifice was adjusted with the aid of two U manometers to obtain a desired dilution ratio. The corrections for penetration efficiency into the probe and probe orifice and diffusion losses during transport were applied following the procedure presented by Minutolo et al. [22].

The evolution of the combustion gas-phase species was isokinetically sampled by a system involving a water-cooled probe which was coupled to a particle filter and a chiller for removing particles and water before entering an online micro gas-chromatography analyzer micro-GC (VARIAN, CP-4900). Similar to the SMPS sampling probe, the GC probe was pointed down toward the incoming flame gas above the burner. It was mounted on a translation stage then its vertical position could be varied with respect to the burner. The accuracy of probe positioning was estimated to be around 0.1 mm.

3. Results and discussion

3.1. The effect of different inert gases on fragmentation

To understand the role of inert gases in the soot oxidation, the effect of adding N₂, Ar, and He (Cases 1–3) was investigated. These

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