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# Further details on particle inception and growth in premixed flames

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## Abstract

Soot inception in flames remains mostly unknown. What are, if any, the differences between inception particles formed in non-sooting and sooting flames, and to what extent soot formation is governed primarily by coagulation or by a different composition/aromatization of inception particles are open questions.

In this work, the initial particle growth occurring in premixed flames of ethylene has been investigated. On line size-selected photoionization efficiency of inception particles produced in various flames has been measured. Structural/chemical differences of both nucleating and growing particles have been additionally investigated by off-line cyclic voltammetry, Raman spectroscopy, and light absorption.

Results show that the size of aromatic domains within particles slightly increases when moving from flames in which the particle size distribution remains nearly constant and mono-modal in the nanometer range along the flame height, to flames in which particle coagulation gives rise to the formation of a second mode constituted by larger but still nanometric particles. Only in particles of this latter mode,  $d \approx 10$  nm, the first appearance of stacking of polyaromatic units is observed.

On this basis, it has been hypothesized that two graphitization processes occur during the first stages of soot inception: a slight increase of in plane aromatic islands in primary particles and the formation of aromatic plane stacks in coagulated primary particles. This second type of coagulating-graphitization process explains the closure of the band gap observed in the grown particles respect to the primary ones, and hence the change of optical properties, towards the typical values observed in soot.

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## 1. Introduction

The mechanism of particle inception in flames still remains elusive [1,2]; we are unable to describe the gas-to-particle transformation at an elementary level comparable with that reached in

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gas phase chemistry and to predict the evolution of the chemical structure and composition of the nascent particles.

On a phenomenological basis, particle nucleation starts predominantly through the clustering of PAHs which leads to the formation of particles with sizes of few nanometers [3–6]. The size and chemical structure of the PAHs that are able to clusterize and the binding force that keeps the initial dimers, trimers, and clusters of PAHs together are still unknown.

Experimental evidences based on in-situ optical spectroscopy [7] and off-line chemical and morphological characterization of the sampled material [8] have shown that in slightly-rich premixed flames, besides moderate size gas-phase PAHs, high molecular mass carbon structures containing small aromatic islands with in plane dimensions of about 1 nm are formed. These macromolecules have masses of the order of 1000–10,000 u corresponding to equivalent spherical sizes of 1–3 nm and contain chain-like bridges between aromatic islands or dangling bonds, which allow them to survive at flame temperature [9]. The transitions from macromolecules to particles and from inception particles to soot nuclei are not straightforward because of the limits of most experimental techniques [10], and because a precise definition of soot nuclei has not yet been given.

One may speculate that particle nucleation/graphitization proceeds via a physical interaction of the polyaromatic units belonging to the macromolecules (van der Waals attractive forces) with gas-phase PAHs or with other macromolecules with the consequent formation of coherent aromatic domains or basic structure units, BSU. Obviously, the molecular make-up of the resulting carbonaceous clusters, or nanoparticles, is expected to be different depending on how wide are the aromatic islands in the macromolecules and which types of bonds held together the aromatic islands.

Unravelling the question of particle inception will allow to find out what are, if any, differences between ultrafine particles produced in “clean” combustion systems and in sooting flames. Moreover, it will allow understanding if soot propensity is mainly governed by the rate of particle inception, or by the composition and graphitization degree of the inception particles that affect particle coagulation and growth.

Therefore, the objective of this work is to follow the initial step of particle growth occurring in premixed flames of ethylene by investigating the structural/chemical differences of inception particles. The experimental methods used to shed light on the mechanism of particle nucleation in premixed flames are photoionization of particles selected on the base of their mobility diameter, and analyses of the particles collected on filters

by Raman spectroscopy, UV-visible light absorption, and cyclic-voltammetry.

## 2. Experimental system

Atmospheric pressure laminar premixed ethylene/air flames (cold gas velocity of 10 cm/s) were stabilized on a syntherized bronze McKenna burner having a diameter of 6 cm. Four flames with *C/O* ratios of 0.61, 0.63, 0.65, 0.67 (equivalence ratios 1.83, 1.89, 1.95 and 2.01) were investigated.

Temperature was measured along the flame axis by a thermocouple with a fast-insertion procedure and corrected for gas radiation [11]. Maximum temperature varied from 1770 K in the *C/O* = 0.61 flame to 1680 K in the richer flame; measurement uncertainty remained within 50 K.

Combustion products were extracted from the flames at different heights above the burner (HAB) and particles were analyzed for the determination of their size distribution and chemical and morphological characteristics.

For on-line particle sampling and photoionization measurements of size-selected particles, flame products were sampled through a small orifice into a dilution tube probe operated with  $N_2$  as the diluent. To prevent particle coagulation, the flow of  $N_2$  was maintained higher than the critical value below which particle coagulation starts. Details of the experimental set-up are reported elsewhere [12] (see also [supplemental material Fig. S1 and supplemental text](#)).

Number particle size distributions, PSD, were measured by a differential mobility analyzer (DMA TapCon 3/150) equipped with a Faraday cup electrometer.

The aerosol photoionization experiment was performed positioning, in the sampling line, a photoionization cell made of a 16 cm long, grounded, metal tube with ID = 19.5 mm with two quartz windows on the two sides of the cell. The neutral particles at the entrance of the photoionization cell and the photoionized particles were selected in size by the classifier of the DMA and counted by the Faraday cup electrometer. The photoionization charging efficiency (*CE*) was determined by the ratio of  $N_{p_i}^+$ , i.e., the number of the particles charged via photoionization, and  $N^0$ , i.e., the number of neutral particles.

Off-line analyses were performed on the particles sampled with a tubular probe as that used for size selected particle photoionization measurements and collected on a filter. A dilution ratio lower than that employed for the photoionization experiment was used for sample collection because the only requirement of the dilution was to quench flame reactions and to avoid condensation of gas phase compounds on particle surface and not to avoid particle coagulation.

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