



# On the particle evolution in iron pentacarbonyl loaded counterflow methane–air flame

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## ARTICLE INFO

### Article history:

Received 16 November 2017

Revised 2 April 2018

Accepted 2 April 2018

### Keywords:

Particle evolution  
Iron pentacarbonyl  
Counterflow flame  
Soot precursor  
Catalytic oxidation  
Diesel particulate filter

## ABSTRACT

The present work analyzes these particle evolution processes along the plane axis in an iron precursor (iron pentacarbonyl) loaded methane/air counterflow diffusion flame. The addition of iron pentacarbonyl into methane led to the formation of iron-based nanoparticles in the flame, together with the pre-existing soot particles. These two types of nanoparticles were found to be distinct in their shapes, chemical compositions, geometric mean particle diameters, total particle number concentrations, and particle size distributions. Nanoparticles produced in the flame were sampled from various axial and radial locations by means of a vacuum pump and their particle size distributions were characterized using a scanning mobility particle sizer (SMPS). The representative nanoparticle samples were also collected on the probe and examined using scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDAX). SMPS, SEM and EDAX analysis revealed the nucleation, growth, agglomeration and interaction of these nanoparticles formed in the flame, evidenced by the changes of particle morphology, averaged particle size and elemental composition. Adding iron precursors was found to promote particle inception, leading to a greater total particle number concentration but a smaller mean particle diameter. Near the flame location, combustion of soot particles was accelerated due to the catalytic role of iron-based nanoparticles, which agrees the observation of primarily smaller iron-based nanoparticles dominating in the particle population. These findings shed light in studying the engine performance when the fuel borne catalysts are injected for abating particulate matters (PM) during regeneration of Diesel Particulate Filters (DPFs).

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## 1. Introduction

Using catalytic additives, together with a diesel particulate filter (DPF), to reduce the emission of particulate matters (PM) from engines has recently drawn much attention due to increasing economic and environmental concerns on fossil fuel powered transportation. For diesel engines, the most common and effective approach to mitigate the PM emission is using a DPF. A modern DPF has an efficiency in the range of 95–99% of the total number of captured PM and approximately 90–95% of the total mass of captured PM, including a 95–99.9% reduction of soot and a 60–90% reduction in the range of the soluble organic fraction emissions [1]. Using some catalyst within a DPF can effectively reduce the desired PM oxidation temperature to below the engine exhaust temperature, which removes the need of external heating. One type

of DPFs utilizes fuel borne catalysts (FBCs) [1–3], which are added at a low concentration directly to the engine fuel, during the DPF regeneration. To operate a DPF with a FBC effectively, the catalyst/fuel ratio (usually a few hundred ppm) needs to be well controlled according to the loading of PM in DPF, engine operation conditions, exhaust temperature and composition, types of the catalyst and fuel, etc. The low price, environmental friendliness and high reactivity of iron/iron-oxide based catalysts have been widely studied in recent DPF development [2]. Since iron and iron oxide can be captured in DPFs, the total number concentration and particulate mass of PM<sub>2.5</sub> and PM<sub>10</sub> are regulated at the exit of these DPFs.

Most of the recent experimental studies on the impact of metallic additives on engine emission reduction focused on measurements of gaseous and particulate air pollutants in the engine exhaust, with or without a PDF, lacking of an in-depth analysis of the heterogeneous reaction pathways associated with the evolution of flame generated nanoparticles in the combustion chamber. A few articles recommend, using iron fuel borne catalysts in

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## Nomenclature

SMPS	Scanning Mobility Particle Sizer
EDAX	Energy Dispersive Spectroscopy
HACA	Hydrogen Abstraction Carbon Addition
$P_1$	pressure upstream of the orifice ( $N/m^2$ )
$P_2$	pressure downstream of the orifice ( $N/m^2$ )
$\Delta P$	pressure difference at the orifice ( $N/m^2$ )
$P_o$	pressure at the orifice ( $N/m^2$ )
$a$	distance from upstream location to orifice (m)
$L$	length of the steel tube (m)
$v_o$	sample flow at the orifice (l/min)
$c$	constant
$\mu$	viscosity ( $N\ s/m^2$ )
$d_o$	diameter of the orifice (mm)
$P_{atm}$	atmospheric pressure ( $N/m^2$ )
$t$	thickness of orifice (mm)
$f_D$	dilution ratio
$k$	constant
$v_d$	diluent nitrogen flow rate (l/min)
$N$	total particle density (number of particles/cm <sup>3</sup> )
$n_i$	particle density in each monodispersed channel (number of particles/cm <sup>3</sup> )
$D_{p,u}$	upper limit of the monodisperse channel (nm)
$D_{p,l}$	lower limit of the monodisperse channel (nm)
$D_g$	geometric mean diameter (nm)
$\bar{K}$	average coagulation coefficient (cm <sup>3</sup> /s)
$T$	residence time in sampling tube (s)

engines equipped with DPFs can effectively reduce the PM emission by trapping and oxidizing soot and regenerating the filter [4,5]. Same studies also suggest, when these additives are sometimes used in an engine without a DPF, they contribute to ambient air emissions. Other researchers directly quantified the influence of metal additives on the PM emissions from engines and laboratorial flames. Kannan *et al.* [6] used ferric chloride ( $FeCl_3$ ) as a FBC at an optimum engine operating condition fueled with waste cooking palm oil based biodiesel, and found carbon monoxide (CO), total hydrocarbon (THC) and smoke emission of FBC added biodiesel decreased by 52.6%, 26.6% and 6.9%, respectively, compared to biodiesel without FBC. Nash *et al.* [4] observed, on a single prototype diesel generator, using ferrocene ( $Fe(C_5H_5)_2$ ) as a diesel fuel additive with varying iron concentrations from 0 to 200 ppm could result in ~30–40% decreases in total particle mass, total particle volume, and black carbon emissions. However, the total particle number concentrations were increased by approximately a factor of 5, which was believed to associate with 10–30 nm iron particles liberated during soot oxidation. Skillas *et al.* [7] confirmed a decrease in particulate matter emission by 25–42% when a variety of metal additives were tested on a diesel, heavy-duty, and 4-cylinders engine. Miller *et al.* [8] analyzed the impact of doped diesel fuel, with 60 ppm of iron, on 1.5-l diesel engine, and observed a drop of 20–40% in the total carbon amount in the samples collected from the exhaust. They deduced a threshold iron to carbon ratio of 0.013 for this engine, below which there was no effect of iron precursor on soot morphology. They also identified two different growth modes of these metal nanoparticles attached to carbon agglomerates, primary iron nanoparticles (5–10 nm) and coagulated iron agglomerates (20–200 nm). Ma *et al.* [9] measured a 3.7% fuel saving by adding ferrous thiocyanate in a diesel engine at 3200 rpm. Similar results were demonstrated by others when ferrous compounds like ferrous picrate and iron chloride were used [6,10]. Most recently, the laser-induced fluorescence absorption spectroscopy were utilized in an isoctane dif-

fusion laminar flame seeded with 4000 ppm iron pentacarbonyl, to investigate the evolution of iron species in the flame. Their results were controversial to most engine studies, by suggesting the dominant iron species throughout the primary flame region was iron atoms, rather than any form of iron oxide [11]. Nevertheless, the lack of fundamental understanding of the particle evolution in combustion chambers, especially through experimental particle sizing directly from different locations of the flame, has hindered the process characterization and design optimization of a DPF.

Formation of catalytic metal nanoparticles in combustion has been extensively investigated when transition metal is used in aerosol flame synthesis of metal or metal oxide nanostructures [12]. If iron pentacarbonyl is used as the metal precursor, thanks to its known decomposition mechanisms, its vapor undergoes heating in the flame, leading to its disintegration into gas phase iron atoms [13]. These atoms then form clusters in a reducing environment, or can react with oxidizing species to form condensed-phase iron oxides particles, which act as a heterogeneous catalyst converting the chemical species in the flame, such as CO and  $H_2$ , to desired nanostructures such as carbon nanotubes [14,15]. As there are multiple factors affecting the particle evolution process in the flame, it is necessary to isolate and study these factors separately. The use of a non-premixed flame simplifies the analysis since the time scales of the chemical reactions are much shorter than those of diffusion and convection processes [16]. Moreover, non-premixed flames are representative of the actual applications as they are common in combustion devices like gas turbines and diesel engines [16]. A turbulent diffusion flame, in the combustion systems mentioned above, can be inherently viewed as a collection of multiple laminar flamelets. Such flamelets in a turbulent flame can be simulated by a collection of counterflow diffusion flames, which can represent the conditions and transport phenomenon in the flamelets [17]. A counterflow diffusion flame involves the injection of fuel and oxidizer from opposite directions into the burner to create a stagnant plane with two distinctive regions: fuel rich and fuel lean on the fuel and oxidizer inlets, respectively. Using this type of flame geometry provides the flexibility of adjusting the spatial gradients of temperature and reactant concentrations. Another advantage of the counterflow flame is the ability to control the temperature gradient.

Techniques used for measuring the size distribution of nanoparticles comprise of both intrusive and non-intrusive techniques. The non-intrusive techniques generally involve laser excitation followed by measuring the extent of extinction and scattering [18,19] and laser induced incandescence (LII) [20,21]. Extinction measurement techniques are restricted by their requirement for a line of sight approach and, although they have been used to measure soot volume fraction and some size dependent parameters [22], they are unable to ascertain a particle size distribution. LII requires knowledge of the refractive indices for the particles, which are greatly dependent on the fuel [20] and on the wavelength of the laser [23] used. Photo ionization mass spectroscopy (PIMS) supersedes other techniques in terms of sensitivity. It has a detection limit of atoms and molecules; however, its upper range is limited to just 6 nm [24]. The development of the scanning mobility particle scanner (SMPS) provides another valuable tool for analyzing particle size distribution and total particle concentration, in almost real-time, without incorporating any prior assumptions or disadvantages such as vacuum operation leading to losing the adsorbed organic compounds on soot surfaces [25]. While a significant number of previous research efforts have focused on soot particle size measurement in premixed flames [25–28], by achieving probe sampling using an inert gas, followed by characterization, less literature is available about particle size in a diffusion flame. Burtscher *et al.* [29] and Hepp and Siegmann [30] took samples from a methane diffusion flame using quartz microprobe attached

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