



# Interaction between iron based compound and soot particles in diffusion flame



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## ABSTRACT

Metallic fuel additives have been considered for soot emission control over the last few decades. However, the exact mechanisms of soot reduction are poorly understood and still remain controversial. In response to the need for identifying the state of the iron additive in the diffusion flame, laser-induced fluorescence and absorption spectroscopy were performed in a laboratory-scale isooctane diffusion laminar flame seeded with 4000 ppm iron pentacarbonyl as the metallic additive. The results of the spectroscopic techniques reveal that the dominant iron species throughout the primary flame region was Fe atoms, rather than any form of iron oxide. Moreover, elemental iron was observed to diminish through the soot oxidation region. The primary conclusion is that the catalytic effect of Fe atoms and possibly iron oxides enhanced soot oxidation in the burnout regime of the flame, thereby reducing the overall soot emissions.

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## 1. Introduction

Soot particles have been well known as one of main products resulting from incomplete combustion of fossil fuels, they critically affect human health and the environment. Because of extremely small size of these particles normally termed in particulate matter (PM) (e.g. less than 10  $\mu\text{m}$  in diameter), they are capable of penetrating and accumulating in the respiratory system. It is supported by a study that particulate pollutants increase the incidence of cardiopulmonary diseases and ischemic heart attack [1]. Soot particles are also rich in amorphous carbon and polycyclic aromatic hydrocarbons (PAHs), and are known to be mutagenic and carcinogenic [2,3]. In addition, they have a harmful influence on the environment in many ways. For example, soot particles lead to atmospheric haze resulting in reduction of visibility, and they also may play a role in acid rain, which may be responsible for a range of problems. When settling on soil and water, soot particles change the nutrient and chemical balance that are responsible for depleting ecosystems and ruining sensitive forests and farm crops. According to studies, soot is twice as potent as carbon dioxide in contributing to global warming resulting from the greenhouse

effect because it can darken snow and ice that results in absorption of solar energy rather than reflection [4]. Such harmful impacts of soot can affect the broad areas because it can travel long distance from the sources mostly existing internal combustion engines [5,6].

Numerous efforts have been made to reduce the soot emission from engines. The soot emission could be efficiently reduced with excessive combustion temperature, which however results in the rise of nitrogen oxide (NO<sub>x</sub>) emission. Diesel particulate filter has been employed although it is not cost-effective. Novel combustion strategies such as homogeneous charge compression ignition or low-temperature combustion have shown promising results in terms of near-zero NO<sub>x</sub> and soot emissions, but these strategies are applicable in a narrow operating range, especially low load and engine speed [7–10]. Metallic additives have been also applied to soot emission control in a few decades; however, the correct mechanism on soot reduction is not well-understood and still very controversial. The mechanism of action of metallic fuel additives have been outlined in three different theories. Firstly, the fuel additive may affect nucleation mechanisms of soot formation in the early stage of soot particle inception. Secondly, the additive may enhance soot burnout as a result of rapid elimination of soot precursors attributed to increase in hydroxyl radicals. Thirdly, the additive may accelerate the soot oxidation rate by occlusion within the soot phase. Obviously, all three mechanisms may be closely

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interrelated. The global and local effects of transition metallic additives on soot reduction were evaluated in many studies using a variety of techniques from simple visual observations to novel laser diagnostic measurements. Linteris et al. reported soot reduction effects of manganese and tin containing compounds by analyzing the burning velocity of methane/air flames [11]. In comparisons of the reduction efficiency with other suppressants, manganese-based additives showed about a factor of two less than that of iron-based additives, but twice as effective as bromine-based additives. The soot reduction effect is supported by a study of Wei and Lee [12]. The overall 40% of reduction was obtained in the pyrolysis reaction with manganese in a laboratory quartz reactor. It was concluded that the addition of manganese sulfate into the high temperature pyrolysis of PS inhibited the formation of PAHs in the reaction. However, Feitelberg et al. found an adverse effect, namely that the additive increased soot volume fraction by approximately 50% in a study of a premixed ethylene flame seeded with manganese added in 140 ppm concentrations [13]. It was expected that manganese would exist in the gaseous phase as free metal atoms at high temperatures and form solid MnO through precipitation at residence times. Hayhurst and Jones also investigated the effects of metallic additives on ionization in premixed acetylene/oxygen/argon flames, and they found that manganese addition did not affect ion concentrations and soot particle size due to their relatively high ionization potentials that leads to the low rates of soot nucleation and particle growth [14].

While manganese is a known neurotoxin, iron has relatively low toxicity; therefore, many combustion applications and laboratory studies have concentrated on the iron based additives such as ferrocene [ $\text{Fe}(\text{C}_5\text{H}_5)_2$ ] and iron pentacarbonyl [ $\text{Fe}(\text{CO})_5$ ]. In a study of a laminar premixed ethylene flame seeded with 0.015–0.46%  $\text{Fe}(\text{C}_5\text{H}_5)_2$  wt of the fuel, Ritrievi et al. studied the effects of the addition of  $\text{Fe}(\text{C}_5\text{H}_5)_2$  on inception and growth of soot particles [15]. As particles moved from inception to growth regime, the average size of soot particles in seeded flames was smaller at inception zone, but it was even larger at growth zone compared to that in the unseeded flames. In addition, Mossbauer spectroscopy was used to determine the chemical state of the iron in the particles, and iron oxide, FeO, was found to be the stable species on the given flame conditions. It was concluded that the carbon deposited on the particles was used for the direct reduction of FeO to metallic Fe. Feitelberg et al. also found that the additives increased overall soot formation in studies of a laminar premixed ethylene flame seeded with  $\text{Fe}(\text{C}_5\text{H}_5)_2$  [13]. This agreed well with other work [15]; however, any additive effect at an early soot inception region was not observed. To conclude, the role of the iron additives was not to affect soot particle inception but to increase the rate of gas-solid reactions leading to increase in the total mass of soot. Hahn assessed the role of the metallic additive in a premixed propane/oxygen flame with  $\text{Fe}(\text{CO})_5$  added in concentrations of 0.32% by weight of iron to the fuel, and it was found that the iron additive had an effect to increase the overall amount of soot in the flame [16]. X-ray photoelectron spectroscopy was used to investigate the state of the iron in sampled particles. Instead of significant quantities of elemental Fe or FeO identified in other works [13,15], the form of iron oxide,  $\text{Fe}_2\text{O}_3$ , was a dominant species in the extracted soot particles. McMillin et al., observed FeO LIF signal at the primary reaction zone in the premixed flame experimentally, and their numerical model shows the FeO is dominant species throughout the flame [17,18].

In addition to premixed flame studies, there are soot suppression studies using iron based additives in diffusion flames. Bonczyk studied the effect of the metallic additive on soot production with a pre-vaporized isooctane/air diffusion flame seeded with  $\text{Fe}(\text{C}_5\text{H}_5)_2$  added in 0.3% by weight of fuel, and it was found that the metal

additive contributed to not only soot enhancement in soot inception zone but also soot reduction in burnout zone [19]. Soot samples were collected post-flame and subjected to an Auger-type chemical analysis so that the species of iron present in the soot could be determined. Based on the Auger data, a condensate from the seeded flame with 0.3%  $\text{Fe}(\text{C}_5\text{H}_5)_2$  was determined to be  $\text{Fe}_2\text{O}_3$  containing only negligible amounts of carbon. In contrast, the condensate was carbon retaining less than 2% of elemental iron when the percentage of  $\text{Fe}(\text{C}_5\text{H}_5)_2$  in the fuel was reduced to under 0.001%. With respect to a qualitative illustration on the soot enhancement by additive in the early residence time, the result supported conclusions drawn from other works [15,20] that soot enhancement was a result of an increase in nucleation sites provided by solid  $\text{Fe}_x\text{O}_y$  particulates and an increase in the surface activity of particles resulting from a catalytic effect of iron on the carbon deposited on the surfaces of soot particles. The iron oxide catalytically reinforces the removal of carbon by molecular oxygen, which enhances the soot reduction in the burnout zone as well. The similar tendency of soot reduction via addition of  $\text{Fe}(\text{C}_5\text{H}_5)_2$  into ethylene coflowing diffusion flame was found by Zhang and Megaridis [21]. Kasper et al. also reached the same conclusion in a study with  $\text{Fe}(\text{C}_5\text{H}_5)_2$  seeded methane/argon and acetylene/argon flames [22]. Kim et al., analyzed the products from  $\text{Fe}(\text{CO})_5$  seeded ethylene diffusion flame using a Laser Microprobe Mass Spectrometry, and found that Fe dominated over iron oxides such as FeO and  $\text{Fe}_2\text{O}_3$  [23].

As mentioned above, many studies often have yielded different conclusions on the role of the metallic additive on soot reduction effect in spite of the similar type of combustion conditions. It is noted that most analyses were done with sampled (i.e. extracted) soot particles that could be oxidized post sampling process; therefore, more precise mechanism can be extracted using *in situ* methods. The primary objective of this study is to quantitatively explore a role of the metallic additive for soot suppression in an isooctane/air diffusion flame seeded with  $\text{Fe}(\text{CO})_5$  using transmission measurement. In addition, Laser-induced fluorescence spectroscopy (LIF) and absorption spectroscopy were used to identify the chemical state of the iron additive in the flame.

## 2. Experimental apparatus and methods

A lab-scale concentric diffusion burner composed of stainless steel tubing was employed for all experimentations. Fig. 1 represents a schematic of the burner shown from side view and diffusion flame. Gaseous Isooctane ( $\text{C}_8\text{H}_{18}$ ) and nitrogen were supplied through the inner tube of the burner. A solid annular disk was press fit between the inner and outer tubes to maintain concentricity. The disk was perforated with 9, 0.03 cm-diameter holes, and the oxygen flow was fed through the annular disk and out the nine ports at the burner outlet. The exit area of burner was reduced to increase the flow velocity at the burner exit. As a result, a roughly 30 cm long diffusion jet flame was created, and fluctuations of the flame were decreased to an acceptable level. First, the Froude number was 11.8, indicating that the flame was momentum-controlled rather than buoyancy-controlled, which was by design. Second, the Reynolds number was approximately 761.3, denoting that the flame was laminar flow, as it was smaller than the critical value of 2300. Furthermore, the fuel/air equivalence ratio was calculated as 1.08 based on the oxygen and isooctane flow rates. While this value corresponds to fuel rich, significant additional oxygen was expected to diffuse into the flame. In order to investigate the soot characteristics at various positions along the vertical axis of the flame, the burner was controlled with a vertical translation stage which allows the burner to move up and down.

Iron pentacarbonyl [ $\text{Fe}(\text{CO})_5$ ] was selected in this study to

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