



Soot in flame-wall interactions: Views from nanostructure and reactivity

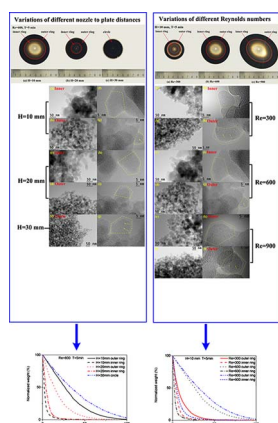


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ABSTRACT

The jet impinging flame is a typical flame-wall interaction configuration. This paper presented an experimental study of laboratory jet impinging ethylene diffusion flames with the special emphasis on characteristics of soot forming on the impinging plate. The soot distribution, nanostructure and reactivity which corresponded with different flame structures were investigated comparatively based on parametric variations of nozzle to plate distance (H), Reynolds number (Re) and collecting time (T). The results showed that impinging flame structures could be classified as cool central core, envelope, and disc flames based on different nozzle to plate distances and Reynolds numbers. A series of concentric soot rings for jet impinging flames were formed on the plate surface. Inner ring soot was the film-like material with the amorphous structure, and outer ring soot consisted of sub-orbicular primary particles with the classic core-shell structure. For soot at $Re = 600$ and $T = 5$ min, the augment of the nozzle to plate distance reduced the content of film-like soot at the inner ring because the extension of the jet free region could promote the oxidation of soot. The crystallization degree of outer ring soot was non-monotonic with the increase of the nozzle to plate distance. It might attribute to the reduction of air entrainment of the flame layer in the wall jet region. Inner ring soot at $H = 10$ mm had the most disordered carbon atoms with the shortest fringe length and the highest fringe tortuosity. It also presented the highest reactivity. Because of the high degree of crystallization with the longest fringe length and the smallest tortuosity, the reactivity of soot sampled at $H = 30$ mm was the lowest. For soot sampled with Reynolds numbers of 300, 600 and 900 at $H = 10$ mm and $T = 5$ min, inner ring soot at $Re = 600$ had the most amorphous structure related with the shortest fringe length and the largest tortuosity. The crystallization degrees of soot from outer rings increased

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with Reynolds numbers. The increase of the impinging flame layer might promote the air entrainment which could accelerate the soot oxidation. In addition, the oxidation reactivity of soot sampled at different Reynolds numbers were in a sequence of $Re = 600$ inner ring $>$ $Re = 300$ inner ring $>$ $Re = 900$ inner ring $>$ $Re = 300$ outer ring $>$ $Re = 600$ outer ring $>$ $Re = 900$ outer ring, which was in consistent with variations of the fringe length and tortuosity. The reactivity of soot from both inner and outer rings decreased with the increase of collecting times at $H = 10$ mm and $Re = 600$, which might result from the increase of the plate temperature. The results also confirmed the relationship between the soot structure and oxidation reactivity that soot with the higher degree of crystallization was more difficult to oxidize.

1. Introduction

Soot as one kind of universal particulate matter is mainly produced by the incomplete combustion of hydrocarbon fuels. The emission of soot could cause serious health problems such as asthma, premature mortality, and lung cancer [1]. It is also considered as a source which is responsible for the global warming and climate changes [2]. It is urgent for researchers to take measures to regulate soot emitted into the atmosphere. Impacts of flame temperatures, oxygen contents, and different additives on soot evolution have been widely studied to find ways to control soot emissions [3–11]. Kumfer et al. [3] found that soot volume fractions increased with the flame temperature. Sirignano et al. [4] showed that the increase of temperature promoted the oxidation of small particles and reduced its concentration. Escudero et al. [5] experimentally investigated the influences of oxygen addition in an ethylene inverse diffusion flame and found that the increase of oxygen addition could promote the soot formation. Burshald et al. [6] presented the soot concentration in diesel flames could decrease with the blending of oxygenated fuels. Liu et al. [7] analyzed the effects of oxygenated biofuels on soot formation in an optical constant volume combustion chamber. The addition of biofuel showed a larger effect of soot suppression at the lower temperature. Chen et al. [8] comparatively studied the influences of four alcoholic isomers of butanol addition on polycyclic aromatic hydrocarbons (PAHs) and soot formation in T20 diesel surrogate co-flow partially premixed flames. Singh et al. [9] comparatively investigated soot formation processes for butanol and butane isomers in counterflow non-premixed flames. It was found that less soot was observed in butanol isomers flames than that in butane flames due to the effect of the hydroxyl group. Liu et al. [10] numerically studied the influences of water vapor addition on soot volume fractions in a laminar ethylene flame. The addition of water vapor could reduce soot inception through the reduction of H radical concentration. Sun et al. [11] revealed that the addition of nitrogen in ethylene flames apparently reduced the volume fraction and particle diameter of soot due to the dilution effects.

Since the nanostructure of soot particles could affect its oxidation reactivity and formation process, researchers have great interest to interpret the influences of soot nanostructure on its oxidation reactivity and formation using high-resolution transmission electron microscopy (HRTEM) and thermogravimetric analysis (TGA) [12–22]. Vander Wal et al. [12] proposed the impacts of soot nanostructure on its reactivity using different fuels under pyrolysis conditions. Huang et al. [15] studied the influence of oxygen addition on soot structure in benzene premixing flames. The HRTEM images showed that the fringe length of soot decreased with the increase of oxygen content in benzene flames. Tian et al. [17] adopted a new analysis method of transmission electron microscopy micrographs to illustrate the morphology of soot aggregates. In the research of Hu et al. [19] the soot morphology, structure and surface properties were analyzed to highlight the effect of catalyst on soot oxidation in an oxygen atmosphere via TEM. The crystalline structure of soot with catalyst showed less ordered under the low temperature range. HRTEM images of soot in the study of Choi et al. [21] presented that the addition of hydrogen in ethylene-air counter diffusion flames decreased the soot nanostructure size and promoted the production of fullerenic nanostructure.

Flame-wall interaction depicts a phenomenon in which wall surfaces and flames interact with each other through the coupling of energy, momentum, and chemistry [23]. It can be universally found in laboratory flames and practical combustion devices. Impinging flames have been widely studied due to diverse interests of researchers on characteristics of combustion near surfaces [24–33]. Dong et al. [25] conducted an experimental flame impingement study on the characteristics of the impingement region of an inverse diffusion flame jet, and comparatively investigated effects of the flame equivalence ratio and nozzle to plate distance on the heat flux and the length of the impingement region. The maximum values of the impinging region length and heat flux were obtained when the reaction zone impinged on the plate uprightly. Li et al. [28] experimentally studied the influences of different impingement plate temperatures on the heat transfer of a liquefied petroleum gas premixed flame. The heat flux was evidently suppressed with the augment in the impingement plate temperature which achieved by the cooling water. In the experimental work performed by Zhen et al. [30], the pollutant emissions of CO and NO_x from impinging swirling and non-swirling inverse diffusion flames were reported. The concentrations of CO and NO_x in the impinging swirling flames were lower than those in corresponding open flames due to the much more ambient air entrainment. Chien et al. [31] provided evidence that the CO emission related with variations of impinging flame structures via planar laser induced fluorescence thermometry. The stagnating flow affected the major transport path of intermediates for CO oxidation. Jarray et al. [32] numerically simulated the heat transfer of a methane-air premixed flame which impinged vertically to a composite panel via the computational fluid dynamics software, and found that the increase of Reynolds numbers promoted the total heat transfer between the panel and flame. Hindasageri et al. [33] proposed a novel method to evaluate the adiabatic wall temperature which was essential in determining the Nusselt number. The results acquired by the combined analytical–numerical method well coincided with the experiments. The majority of flame impingement experiments and numerical calculations focused on the heat transfer/CO/NO_x emissions of different burner configurations or fuels.

In practical engine studies, diesel engines have the higher heat and the lower fuel consumption, but suffer more serious particulate emissions than any other power-production devices. While the shape and the geometry of combustion chambers could induce large changes in engine performance and particulate emission due to flame-wall interactions. Few studies have directly been involved in soot emissions associated with the flame impingement [34–37]. Wang et al. [35] illustrated influences of various nozzle geometry and injection pressure parameters on soot formation and flame structures of the impinging diesel spray, and both the micro-hole nozzle and the ultra-high pressure had the distinct effect on soot reduction. Pickett et al. [36] emphasized the effects of secondary flame-wall interaction on the combustion process and the soot emission of a diesel fuel jet in a constant-volume combustion vessel with confined wall jets.

From the studies above, the effects of flame-wall interactions on soot distributions within the flame have been conducted, but there is very little work to study on the nanostructure of soot influenced by flame-wall interactions [38]. Detailed soot structural and oxidation reactivity information can benefit for a better understanding of soot

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