



Multi-parameter measurements of laminar sooting flames using thermophoretic sampling technique



Zuwei Xu, Haibo Zhao*, Xiaobing Chen, Chun Lou*

State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, Luoyu Road 1037, Wuhan 430074, China

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ABSTRACT

The dual exposure-time thermophoretic sampling (DTTS) method had successfully realized multi-parameter measurements for flame synthesis TiO_2 nanoparticles (Xu and Zhao, 2015), which was further researched and developed to apply to sooting flame diagnosis in this study. However, the traditional sampling probe (TEM grid) cannot satisfy the assumption of convection dominant for heat transfer between probe and flame when moving to a sooting flame that radiation would affect the temperature of the probe dramatically owing to very high emissivity of soot particles. After a comprehensive investigation, we found that the radiative heat transfer between the sampling probe and the sooting flame can be effectively eliminated when tailor-made ultrathin quartz glass slices (UQGSs) were used as novel sampling probes due to a very low emissivity of UQGSs (high radiative transparency). Thereby the interaction between the flame and the UQGS probe conformed to an experiential convective heat transfer - thermophoresis coupling mechanism. As the amount of particles deposited on the probe surface by thermophoresis was a function of soot volume fraction, probe exposure time, gas temperature and flow velocity, two thermophoresis samplings with different exposure time (one is far less than the time constant τ of the UQGS and another is close to τ) in a same position were conducted to simultaneously measure soot volume fraction and flow velocity in the location. The collections of particles with tunable exposure time of probes in the flame were observed and analyzed by a field-emission scanning electron microscope (FESEM) and FESEM-image processing for soot particle size. The novel sampling probe as well as the DTTS technique provided a practical way to perform multivariate measurements of soot volume fraction, flow velocity, and soot particle size distribution.

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1. Introduction

Soot, which is a serious environmental pollutant, also affects dramatically heat transfer characteristics of most practical hydrocarbon/air combustion occasions, such as internal combustion engines, gas turbines, boilers, and fires [1–3]. Consequently, it is important to gain a fundamental understanding of the complex sooting processes in flames. Soot particles produced in hydrocarbon flames as results of complicated physical–chemical processes in which gas-to-particle conversion occurs within a very short time scale, of the order of few milliseconds [2,4]. The chemical kinetics are coupled with particle dynamics for sooting processes from micro atomic scale to mesoscopic particulate scale, considering PAH-based inception and surface chemistry (growth and oxidation), coagulation, and fragmentation [5–8]. The experimental and numer-

ical investigations of the fundamental soot evolution, including the soot precursor chemistry, particle nucleation, mass/size growth and ablation, are undoubtedly the mainstream scientific attempts [9–14]. Although many considerable knowledge of the soot phenomenon was obtained over the last two decades, the complete understanding of the chemical and physical mechanisms of the entire process has not been accomplished yet.

The study of soot formation and destruction mechanisms requires reliable measurements of local soot concentrations and particle/aggregate sizes as well as the temperature-time history of soot particles in flames. The properties of soot history include, but are not limited to, temperature, velocity, exposure time, and size distribution of particles. Soot concentrations and particle/aggregate sizes have been investigated extensively in the past decades. In practice, measurements of distributions of temperature and soot volume fraction in flames are essential for the understanding of soot formation. There is a vast literature related to measurements of temperature and soot volume fraction, particularly with non-intrusive in-situ optical diagnostic methods. Light extinction is a

* Corresponding authors.

E-mail addresses: klinmannzhb@163.com (H. Zhao), lou_chun@sina.com (C. Lou).

path-integrated or line-of-sight technique, which provides field-integrated values of the soot volume fraction [15,16]. Two-color analysis of soot radiative emission is a typical kind of emission technique, which has been used to estimate the average values of soot volume fraction and temperature [17]. Laser-induced incandescence (LII) of soot provides spatially and temporally resolved measurements of soot distribution [18–21]. Multi-wavelength emission measurements have also been employed to measure the spatial distributions of soot temperature and volume fraction [22,23]. However, optical diagnostic techniques, almost without exception, make use of a large number of sophisticated equipment that integrate highly specialized skills and instruments in signal detection, data inversion, information representation [24,25]. These make us have to consider the costs and experimental conditions of research works. Additionally, a calibration procedure, often based on extinction measurements or gravimetric analysis, is normally required.

Recently, a thermocouple thermometer and a thermophoretic probe have been widely used as a calibration-independent technique for absolute flame temperature and particle volume fraction measurements due to less uncertainty. Thermocouple particle densitometry method (TPD) is mainly used in measurement of temperature and soot volume fraction in laminar diffusion flames [26,27]. Although the probe contacts with the flame in the measurement, this method obtains information through the variation of temperature in the probe because of soot deposition. A novel technique is proposed for improvement of the traditional thermocouple measurement recently reported by the current research group [28,29]. The key point of the newly-proposed method is to tailor-make a thermocouple junction that can realize accurate "balance compensation" for heat conduction from the wires, radiation heat loss from the bead, variations of specific heat capacity. There was a time window to maintain invariable thermal inertia coefficient during the initial phase of the thermocouple response.

Measurement and analytical techniques using thermophoretic sampling and transmission electron microscopy (TEM) have greatly improved the knowledge about the properties of soot particles [30]. Thermophoretic sampling is also the most common and easily realized way of collecting particles out of flames, introduced by George et al. [31] and further improved by Dobbins et al. [32] for measuring the size and morphology of soot particles. Köylü et al. [33] proposed the thermophoretic sampling particle diagnostic (TSPD) method based on the theory of particle deposition rate by Eisner and Rosner [27], which can be used to determine not only particle/aggregate size distribution and microstructure but also absolute soot volume fraction. However, the TSPD technique was subject to the following assumptions and restrictions [33]: the gas flow velocity and the probe wall temperature are known constant. In fact, there is spatial distribution of gas flow velocity even in a steady flame, with exact numbers not known in advance, and the temperature of the probe changes over the exposure time in the flame due to heat transfer.

The exposure time of soot particle is directly dependent on stream velocity. The increased stream velocity in the normal flame may reduce the exposure time for soot formation in coflow flames, resulting in a reduction of soot emissions [34]. Therefore, flow velocity is critical to soot dynamic evolution and offers the possibility of studying the fate of soot particles. As known, there is not currently an available measurement solution for flow velocity in sooting flames because most well-developed flow velocity measurements depend on micron-sized tracer particles, such as particle image velocimetry (PIV) [35] and laser Doppler velocimetry (LDV) [34,36]. However, laser-based techniques need expensive dedicated equipment and strongly depend on the tracer particles. In our previous work, multiple internal and external properties of TiO₂ nanoparticles in flame were measured and characterized simultaneously by the dual exposure-time thermophoretic sampling

(DTTS) method that used molybdenum TEM grids as thermophoresis probe [28,37]. The DTTS technique is well-established for the analysis of flow velocity.

In this work, the DTTS technique using ultrathin quartz glass slices (UQGSs) as thermophoresis probes to collect soot particles within two different time intervals at one position of flame is proposed, followed by field-emission scanning electron microscope (FESEM) characterization and FESEM-image processing. It should be noted that TEM is not applicable to observe soot particles on the UQGSs, whereas FESEM is a fairly applicable instrument. Since the electron beam passes through the sample that is being examined by TEM, the sample and holder must be sufficiently thin. Usually, samples are supported on a wafer-thin foil (dozens of nanometres thick) of a TEM grid before being loaded into the TEM. Actually UQGS possesses a thickness of 30 μm and therefore electron beams fail to pass through in a TEM. FESEM focuses on the sample's surface and morphology, thereby meeting the requirement of characterizing soot particles on UQGSs. The novel thermophoretic sampling method is a development of TSPD technique to explore the relationship between soot thermophoresis deposition and convection heat transfer of probes, which can simultaneously detect soot volume fraction and flow velocity. The tailor-made DTTS method for multivariate measurement of sooting flame is examined in a benchmark ethylene/air laminar diffusion flame. The experimental measurements of soot volume fraction and stream velocity at different heights above the burner (HABs) are compared with past measurement results as well as the simulation results using a soot model.

2. Experimental

2.1. Apparatus description

In the present experiment, the Gülder burner flame is chosen for the measurement, which is one of three target flames defined by the LII workshop [20]. There is a standardized flame condition of Gülder burner, which can be used for validation and calibration experiments. The burner consists of a fuel nozzle with a 10.9-mm inner diameter, centered in an air tube 100-mm in diameter, and the annulus air region packs with glass beads and porous metal disks to prevent flame instabilities. The ethylene fuel flow rate is set to the smoke point, 0.194 L/min, and the air coflow is 284 L/min [38,39].

The thermophoretic sampler is composed of a double acting high-speed pneumatic cylinder (Festo, DSNU), a programmable logic controller (PLC), a self-closing tweezers, a well-designed sampling probe – ultrathin quartz glass slice (UQGS, presented in Fig. S1 of the supplementary material (SM)) and other auxiliaries. Figure 1 shows a sketch of this thermophoresis sampler. The pneumatic rod end of the double acting cylinder is connected with the self-closing tweezers, carrying the UQGS to the intended sampling positions for an intended exposure time (t_e). When an UQGS is rapidly inserted into the pre-specified sampling position by the high-speed pneumatic cylinder, nearby soot particles will deposit on the UQGS surface due to thermophoresis, meanwhile the temperature of UQGS will increase due to heat transfer from the flame. The timer function of the PLC is used to determine and control the exposure time at the sampling locations, which can ensure representative sampling and avoid overlapping aggregates on the UQGS (The discussion is presented in SM Section 1). After this sampling process, the central zone of the UQGS with an area less than 0.01 mm² is observed by FESEM to separate the soot samples at the target point, same as the authors' previous method [28].

The UQGS possesses ultrathin structure with a diameter of 3 mm and a thickness of 30 μm, which benefits minimizing the disturbance to sooting flame. The manufacture of the UQGSs is based

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