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In situ laser diagnostics of nanoparticle transport across stagnation plane in a counterflow flame

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ABSTRACT

The transport of nanoparticles in the boundary layer is closely related to particle mixing or deposition. We present an *in situ* imaging of TiO_2 particle volume fraction near gas stagnation plane in a counterflow flame by recently developed phase-selective laser-induced breakdown spectroscopy technique. The concentration boundary layer is well resolved with a spatial resolution of 10 μm. Together with a numerical analysis of particle transport equation, the roles of convection, diffusion and thermophoresis are discussed. The calculated profile of particle volume fraction agrees well with experimental measurements, which indicates that current model of nanoparticle transport is capable to quantitatively predict the concentration profile in boundary layers. Further study shows that altering thermophoretic velocity shifts the concentration boundary layer but does not change the shape of concentration profile. The decaying slope is mainly controlled by diffusion process that is dependent on particle size.

1. Introduction

The transport of nanoparticles in the boundary layer is widely concerned since it is always closely related to mixing or deposition (Friedlander, de la Mora, & Gokoglu, 1988; Inthavong, Tian, & Tu, 2016; Mädler et al., 2006). Different from main flow where inertia dominates, the influences of both diffusion and thermophoresis (when there is a temperature gradient) are also important in boundary layers. Particularly for nano-scale transport, gas-particle scattering could be quite different at highly curved surfaces (Li & Wang, 2004, 2005; Kravchenko & Thachuk, 2014). For instance, the accommodation coefficient, quantifying the interaction between gas molecules and particle surfaces, was found to be dependent on particle size because of non-rigid collisions, which substantially affects the formulation of drag and thermophoretic forces (Li & Wang, 2004, 2005). Experimental efforts are essential to understand simultaneous convective, diffusive and thermophoretic motions of nanoparticles. Most quantitative studies employed a 'penetration technique' to investigate particle thermophoresis and diffusion which are perpendicular to the main flow (Zheng, 2002; Sagot et al., 2009). As an indirect method, the deposition efficiency of aerosol was measured to reveal thermophoretic velocity or diffusion coefficient. *In situ* measurements mainly rely on optical methods e.g. elastic light scattering. Yang and Biswas (1997) used light scattering to study the sintering of TiO_2 nanoparticles in a premixed jet flame. Xing, Koylu, and Rosner (1999) measured morphological evolving of Al_2O_3 nano-aggregates in a counterflow flame by light scattering. Though straightforward, this technique

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could not decouple particle concentration from size since light scattering intensity is a function of both parameters. Besides, elastic scattering often suffers from interferences of gas Rayleigh scattering and wall reflections. Therefore the information revealed from light scattering method is usually only qualitative for particle volume fraction in flames. As for soot (Quay, Lee, Ni, & Santoro, 1994; Vander Wal & Weiland, 1994, Bambha & Hope, 2015) and some other particles with high sublimation temperatures e.g. Si (Sipkens et al., 2014) and MgO (Lehre, Suntz, & Bockhorn, 2005), laser-induced incandescence (LII) is able to reveal information on particle volume fraction and size. However, for low vaporization point particles such as TiO_2, V_2O_5, Fe_2O_3 etc., LII may suffer from undesired ablation (Maffi, Cignoli, Bellomunno, De Iuliis & Zizak, 2008; Vander Wal, 2009). Recently a new diagnostics technique named phase-selective laser-induced breakdown spectroscopy (PS-LIBS) has been developed for *in situ* tracing gas-particle transition and measuring volume fraction of metal oxide nanoparticles (Zhang, Xiong, Li, Dong, Buckley & Stephen, 2013; Ren, Zhang, Li & Law, 2015) in nano-aerosol systems. By setting laser fluence in the gap between breakdown thresholds of gas and particle phase, the nanoparticles are ablated and excited in individual nano-sized plasmas while no breakdown occurs in the gas phase. Distinct from conventional LIBS technique, there is no visible spark or detectable continuum background for PS-LIBS after the laser pulse. Zhang, Li, Ren, Yao, and Stephen (2015) has proved the linearity between atomic spectra intensity and particle volume fraction, which is almost independent of particle size for particles larger than 6 nm. Benefitted from the absence of visible sparks, two-dimensional imaging can be realized with a high spatial resolution. This new technique enables us to look deeper into the transport of nanoparticles in boundary layers.

In this work, we study nanoparticle transport across the stagnation plane of a counterflow flame. This type of flame configuration is chosen because of its quasi one dimensional stagnation flow field. In the vicinity of stagnation plane, the influences of convection, thermophoresis and diffusion are comparable for particle transport. The two-dimensional profile of particle volume fraction in this region is imaged with high spatial resolution by PS-LIBS technique. On the other hand, the particle transport equation with simultaneous convection, diffusion and thermophoresis is numerically solved in the boundary layer. By comparing experimental and theoretical results, the roles of diffusion and thermophoresis are discussed.

2. Methods

2.1. Flame configuration

As shown in Fig. 1(a), a premixed counterflow flame is used to create a quasi-one-dimensional nano-aerosol stagnation flow. Premixed gas of 0.9 L/min methane, 9.0 L/min N_2 and 2.0 L/min O_2 is delivered through the lower nozzle while heated N_2 (600 °C, PID controlled) goes through the upper nozzle, which maintains a stagnation plane between 15 mm gap. The tube is concentric with blue arrows representing working gas and white arrows representing shielding gas. The stagnation plane locates at about z = 10.2 mm above the lower nozzle as shown in Fig. 2 and Fig. 4. We shifted the stagnation plane a little higher from the exact middle position (z = 7.5 mm) because there would be larger space between the flame and stagnation plane for particles to grow and for the convenience of optical imaging. We have verified both experimentally and numerically that this range of stagnation plane did not affect the profile of local particle volume fraction. Liquid precursor titanium isopropoxide (TTIP) is maintained at 368 ± 1 K in a bubbler, with the equilibrium vapor delivered by N_2 carrier gas (0.8 L/min) through the lower nozzle. The downstream gas lines are heated to 500 K to inhibit condensation of precursors. The precursor transforms to TiO_2 nanoparticles by hydrolysis at the flat flame sheet that locates 4 mm below the stagnation plane. Formed nanoparticles then grow up by collision and coalescence while being transported along the streamline.

2.2. Optical imaging

The principle and details of PS-LIBS can be referred to our previous work (Zhang et al., 2013, 2015; Ren et al. 2015). As shown in Fig. 1(b), the optical layout is quite straightforward. The second harmonic (532 nm) of an Nd:YAG laser operating at 10 Hz serves as

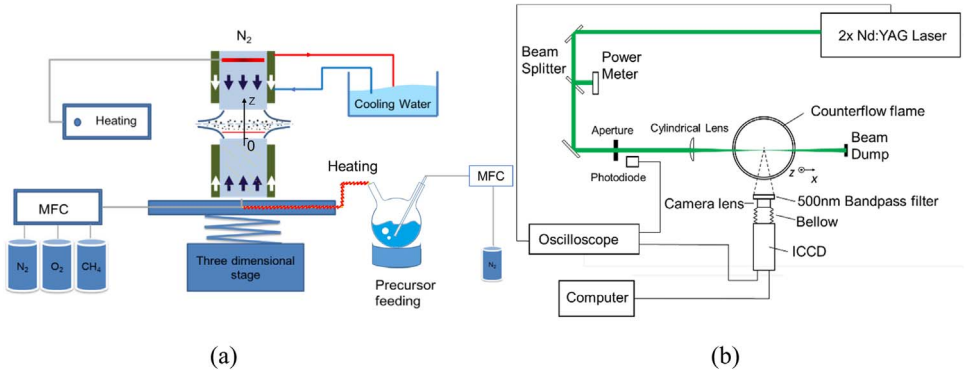

Fig. 1. Diagram for experimental setup. (a) Premixed counterflow flame with precursor feeding; (b) optical layout for two-dimensional imaging of TiO_2 particle volume fraction by PS-LIBS. The blue arrows represent working gas and white arrows represent shielding gas.