In-Situ Characterization of Coal Particle Combustion via Long Working Distance Digital In-Line Holography

Xuecheng Wu,† Longchao Yao,‡ Yingchun Wu,*,† Xiaodan Lin,† Linghong Chen,† Jun Chen,*,‡ Xiang Gao,† and Kefa Cen†

†State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China
‡School of Mechanical Engineering, Purdue University, West Lafayette, Indiana 47907, United States

ABSTRACT: A long working distance digital holographic system is developed to study burning coal particles, which are less than 200 μm in diameter, in a methane-air flame. A general model for the lensed digital holography system is introduced first. An optimal lateral resolution of 3.9 μm is achieved while the object is located at a safe distance (>5 cm) from the optical apparatus. The evolutions of particle morphology and devolatilization products are observed. Sizes and three-dimensional velocities of particles are measured and compared at the burner outlet, 10 and 24 cm above the burner. At the ignition, a condensed phase cloud is observed surrounding the coal particle due to the ejection of devolatilization products. Then the volatile cloud accumulates into stringy tails after detaching from the parent particle. The surface of the coal particle has rough morphology due to formation of porous structure after the reaction. Statistical size distributions at different heights above the burner show a 35% increase of peak size after combustion, and the fraction of the larger particles also significantly increased. Particles spread in the radial direction; meanwhile, particle velocities along the axial direction decrease with the height because of the drag force and gravity.

INTRODUCTION

Coal is one of the major fossil fuels to produce energy due to its low cost and vast deposits. There have been many strategies to utilize coal including pulverized coal combustion, gasification, liquefaction, coal water slurry, and integrated gasification combined cycle,1,2 among which pulverized coal combustion with the coal particles less than 200 μm in size has remained the dominant method for decades. Despite the advantages of coal based energy production, the large amount of greenhouse gas release and air pollutant emissions are becoming an increasingly severe threat to the environment.3,4 Thus, there is an urgent need to improve the energy efficiency and to reduce the pollutant emissions in coal combustion. As coal is a complex mixture of organic and inorganic substances, the combustion process is more complicated than those of liquid and gas fuels. Pulverized coal undergoes pyrolysis, ignition, homogeneous combustion of the volatile matter, and the heterogeneous combustion of the char.5,6 Meanwhile, the air pollutants such as SO2 and NOx are generated in the chemical process, and soot and fly ash are ejected and accumulate into particulate matters.7,8 Understanding the complex physical and chemical process around and in the coal particle is important to reveal coal combustion mechanism, which will further help the development of new combustion models and optimization of combustion conditions. Knowledge of the coal particle morphologies, sizes, velocities, and their evolutions is essential for understanding coal combustion.

A variety of architectures such as tube drop furnace,6,9,10 flat flame burner,11−14 and coal-laden jet burner15−17 have been developed to simulate the high temperature and high heating rate of the combustor in coal-fired boilers. It is convenient to implement optical/laser diagnostic techniques to characterize the flow and particle features with these architectures. Laser Doppler velocimetry (LDV) and particle image velocimetry (PIV) are two standard tools for flow field velocity measurement, and their applications to flame velocity are well proven.18,19 However, the scattered laser signal is frequently contaminated by larger coal particle’s incandescence, which introduces noises in the Mie scattering images and deteriorates the measurement accuracy. To measure coal particle velocity, particle streak velocimetry (PSV)15,20 records a long exposure image of bright burning chars as streaks. Particle displacements are determined by examining the start and end of the streaks. It is more suitable for the stage of heterogeneous char burning when the particles are incandescent. In many research studies, particle morphologies were observed with scanning electronic microscopy (SEM)15,21 before and after the combustion. This helped the study of the particle size evolution and porous structure formation on the particle surface. However, SEM is unable to capture images for in situ coal particles in the flame and thus is not applicable to the dynamic measurement. Direct observation of the coal particle behavior is one of the most important ways to investigate events and parameters of coal combustion. For this purpose, shadowgraphy,14 high-speed photography,6,11,22 and holography17,23,24 were used in the coal combustion study history. Shadowgraphy uses bright white background light so that the coal particles and condensed phase of the volatile matters are recognized as a dark region in the bright background. High-speed photography takes photographs or shadowgraphs at a high frame rate at 1000 Hz or higher and has become more prevalent than ever before with
the fast development of ultrafast cameras and data storage techniques. A dynamic process like volatile flame evolution, coal particle fragmentation, and movement can be recorded by high-speed photography. The drawback of the traditional imaging techniques is that only particles in focus are useful for analysis, while the defocused particles are blurred, making the observation and measurement confined in a limited depth of field. In addition, most optical/laser techniques resolve only one parameter at one time, and the measurement area is usually limited to point wise, or two-dimensional (2D). Combining different techniques in one experiment is a feasible but sophisticated and expensive solution.

Differently, holography uses coherent light as the illumination source. Morphologies, sizes, and locations of three-dimensional (3D) distributed particles are encoded in the interferometric fringes known as hologram and then decoded in a reconstruction process. Holography is a real 3D imaging technique that has the potential to obtain quantitative, multiparameter results of burning coal particles. It was adopted for pulverized coal combustion nearly 40 years ago with interesting phenomena observed. But the optical holography at that time was limited by the tedious recording and reconstruction processes, with only fundamental observations being made. With the fast development of digital cameras and computer science, digital holography has begun to play a more important role in engineering measurements. Holograms are digitally recorded by a digital camera and numerically reconstructed in the computer. In addition to particle measurement, it also shows some potential to retrieve the temperature distribution of a flame. The digital in-line holography (DIH) configuration, which is easy to implement with higher sampling efficiency (a tilted reference beam in off-axis creates denser fringes that require smaller sampling step for the same imaging resolution), has demonstrated its advantages in studying particle dynamics and has been applied to measure solid particles, droplets, crystals, micro-organisms, and cells. DIH was also demonstrated to image burning aluminum drops with high contrast and clear borders. The image distortion caused by the index of refraction gradient due to thermal variation seemed negligible for the laminar flame. Together with two-color imaging pyrometry, dynamic parameters and the temperature of burning aluminum could be measured simultaneously.

Our group recently applied lensless DIH for burning coal characterization, in which the morphology of an individual coal particle and its surrounding flame and wake plume were extracted, and the number concentration of coal particles in a 3D volume was retrieved. In these studies, a charge couple device (CCD) was used to record the hologram digitally. Holograms were reconstructed at different depth locations to refocus coal particles by simulating the wavefront’s backpropagation in the computer. Automatic particle detection and location algorithm allowed for fast measurement. A key challenge of applying DIH to combustion diagnostics is that the camera should be put far away from the harsh and high temperature flame, and thus the imaging resolution is limited by a significantly reduced numerical aperture (NA). Using lenses can not only improve the NA, but increase the effective working distance as well in DIH. So far, many research studies have applied the single-lens model even when a complicated lens combination was used. The resolution limitations from the lens aperture, recording distance, magnification, sensor size, and pixel size together have not been discussed. A quantitative design method of the DIH system with the multilens system is essential to optimize the resolution and working distance.

In this paper, we introduce a design of a long working distance DIH system for investigating combustion behaviors of pulverized coal particles. Spatial resolution of a few micrometers is achieved when the optical apparatus is put more than 15 cm away from the flame. The pyrolysis stage of pulverized coal is studied by observing particle morphology and volatile structure at different residence times. Coal size distribution and 3D velocities are also obtained and compared at different heights above the burner. The work begins with a general model for lensed DIH with generic lens combinations. The experimental setup and method are then described. The resolution and errors of the system are discussed, followed by direct observation of particle morphology and quantitative analysis of particle size and velocity statistics.

### A GENERIC MODEL FOR LENSED DIH SYSTEM

Figure 1a shows a conventional design of a lensless DIH system. A plane wave travels through the particle field and is scattered by particles. Scattered light (object wave, OW) interferes with the undisturbed wave (reference wave, RW) to form a hologram on the digital recording media (e.g., CCD sensor). When a particle is located at a distance $z_a$ from the recording plane and paraxial approximation is employed, the complex amplitude of the wave field at the recording plane is

$$U(u,v) = \frac{\exp(ikz_a)}{kz_a} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} U_o(x,y) \times \exp \left\{ \frac{ik}{2z_a} \left[(x-u)^2 + (y-v)^2\right]\right\} dx dy \quad (1)$$

where $U_o(x,y)$ and $U(u,v)$ are the complex amplitudes at the particle plane ($PP, z = 0$) and sensor plane ($SP, z = z_a$), respectively. $U_o(x,y)$ is usually represented by the transmission

![Figure 1. Schematics of digital in-line holography system: (a) conventional lensless configuration and (b) lensed configuration.](image-url)
function \([1 - O(x,y)]\) when the particle is approximated as an opaque screen (\(\lambda\) is the wavelength, and \(k = 2\pi/\lambda\) is the wavenumber).

Moreover, the measurement accuracy of lensless DIH system is limited by relatively large pixel pitch and overall small chip size. For example, the pixel pitch of a typical CCD sensor is close to 10 \(\mu m \times 10 \mu m\), meaning the smallest measurable particle is around 30 \(\mu m\) assuming a 3 \(\times\) 3 pixel block can describe the particle. The overall chip size is on the order of 1 cm \(\times\) 1 cm, suggesting the recording distance of a hologram should be less than 10 cm if a numerical aperture of 0.05 is required. To investigate small objects, magnifying lenses were consequently adopted in imaging, e.g., secondary droplet sizing,\(^43\) burning aluminum particles,\(^\) and particle-laden flow,\(^45\) in addition to the development of digital holographic microscope.

Previous research revealed that the recorded hologram can be seen as a magnified image of a virtual hologram located at the conjugate plane (CP) of the sensor plane by applying a single-lens model. A more general model containing multiple lenses and the aperture, shown in Figure 1b, is developed here using ray-transfer matrix and the generalized Huygens—Fresnel integral.\(^46,47\) Generalized Huygens—Fresnel integral was applied in DIH in some earlier studies, showing its effectiveness in astigmatic optical systems.\(^46,49\) Fractional transform should be used to reconstruct particles. Hereby we will express the system in an equivalent lensless way, and the traditional angular spectral method is able to reconstruct particle images. The complex amplitude of optical field at SP is

\[
U(u, v) = \frac{\exp(ikL)}{i\lambda L} \int\int_{-\infty}^{\infty} U_r(x, y) \times \exp \left\{ \frac{ik}{2B} \left[ A(x^2 + y^2) + D(u^2 + v^2) - 2(xu + yv) \right] \right\} dx\,dy
\]

(2)

where \(L\) is the axial optical path from PP to SP. \(A, B,\) and \(D\) are the elements of the system ray-transfer matrix \(M = \begin{bmatrix} A & B \\ C & D \end{bmatrix}\) that relates the ray coordinates of PP and SP: \(\begin{bmatrix} \theta_{SP} \\ r_{SP} \end{bmatrix} = M \begin{bmatrix} \theta_{PP} \\ r_{PP} \end{bmatrix}\).

Here \(r\) and \(\theta\) denote a ray’s elevation coordinate and direction coordinate, respectively. By defining \(x' = Ax\) and \(y' = Ay\) and adopting \(AD - BC = 1\) in free space, eq 2 becomes

\[
U(u, v) = \frac{\exp(ikL)}{i\lambda AB} \int\int_{-\infty}^{\infty} \frac{kC}{2A} (u'^2 + v'^2) \left\{ \exp\left( \frac{ik}{2B} \left[ (x' - u')^2 + (y' - v')^2 \right] \right) \right\} dx'\,dy'
\]

(3)

It is interesting to compare eqs 3 and 1, which model the hologram recording process in the lensed and lensless configurations, respectively. First the constant phase terms, \(\exp(ikz_r)\) and \(\exp(ikL)\), can be neglected since they only influence the absolute amplitude. If we introduce an equivalent particle transmission function \(U_{eq} = \frac{1}{A} U_r \left( \frac{z_r}{\lambda}, \frac{z_r}{\lambda} \right)\) and an equivalent recording distance \(z_{eq} = AB\), the hologram recorded by a lensed system can be modeled as one by an equivalent lensless system because the additional phase \(\exp\left( \frac{ikC}{2A} (u'^2 + v'^2) \right)\) does not affect the intensity distribution.

The system ray-transfer matrix can be further expressed as

\[
\begin{bmatrix} A & B \\ C & D \end{bmatrix} = \begin{bmatrix} A_1 & B_1 \\ C_1 & D_1 \end{bmatrix} \begin{bmatrix} 1 & z_{se} \\ 0 & 1 \end{bmatrix}
\]

(4)

in which \(\begin{bmatrix} 1 & z_{se} \\ 0 & 1 \end{bmatrix}\) is the translation matrix (from PP to CP in Figure 1) and \(\begin{bmatrix} A_1 & B_1 \\ C_1 & D_1 \end{bmatrix}\) is the matrix describing ray traveling from CP to SP. From eq 4, it is easy to get \(A = A_1\) and \(C = C_1\).

Since CP is the conjugate plane of SP, one can also get \(B_1 = 0\) and \(B = A_1z_{se}\). So the analytical complex amplitude at SP is expressed in the frequency domain by applying the convolution theorem as

\[
U(u, v) = \exp\left( \frac{ikC}{2A} (u'^2 + v'^2) \right) \times \mathcal{F}^{-1}\left\{ \mathcal{F}[U_r(x, y)] \right\}
\]

(5)

\[
\exp\left( ikz_{eq} \right) \left[ 1 - \frac{\lambda^2}{2} (f_X^2 + f_Y^2) \right] P(f_X, f_Y)
\]

where

\[
U_{eq}(x, y) = \frac{1}{A_1} \left( \frac{x}{A_1}, \frac{y}{A_1} \right)
\]

(6)

and

\[
z_{eq} = A_1^2 z_{se}
\]

(7)

\(\mathcal{F}\) and \(\mathcal{F}^{-1}\) denote Fourier transform (FT) and inverse Fourier transform (IFT), respectively. \(f_X = u/(\lambda A z_{se})\) and \(f_Y = v/(\lambda A z_{se})\) are the corresponding frequency components, and \(P\) is the aperture function at the exit pupil. Thus, the hologram recorded by lensed system can be described by an equivalent lensless system. \(A_1\) has nothing to do with the particle position which theoretically means a uniform spatial magnification for all particles. By transferring to equivalent lensless DIH, we are able to design a DIH system with magnifying lenses including the limit of aperture. In a lensless DIH system, when an \(N \times N\) pixel imaging sensor with the pixel size of \(d_p\) is used, the minimum fringe spacing of a particle located at the center of the field of view is \(d_{min} = \lambda / \sin(\alpha) \approx 2\Delta z_{se} / (Nd_p)\). \(\alpha\) is the angle that represents the direction of the highest frequency. According to the Nyquist theorem, \(2d_p \leq d_{min}\) is required to sample the fringes; otherwise, the aliasing problem will occur.\(^50\)

This results in the relationship \(\Delta z_{se} \geq N d_p^2 / \lambda\) to satisfy the sampling condition. Decreasing \(\Delta z_{se}\) will increase the numerical aperture with \(NA = d_p/(2\Delta z_{se})\), so as to improve the resolution. However, when \(\Delta z_{se}\) is smaller than the critical distance \(z_{cr} = N d_p^2 / \lambda\), the aliasing problem occurs and dense fringes near the edge of sensor can not be sampled properly. Thus, the effective NA and resolution will no longer be improved. At the critical distance, the best resolution proves to be \(\Delta x = d_p\) by applying \(\Delta x \equiv \lambda / (2NA)\) and \(NA = d_p/(2\Delta z_{se})\). Combined with eq 7, the critical \(z_{cr}\) for a general lensed DIH system is \(z_{cr} = N d_p^2 / (A_1^2 \lambda)\) to achieve the best spatial resolution of \(d_p / A_1\). On the other hand, the limit by the aperture could be calculated in the diffraction limited system as in ref S1. The lateral resolution

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will thus be finalized by the stricter limit from both the aperture and the pixel size

$$\Delta x = \begin{cases} \frac{d_p}{A_1}, & z_b \leq \frac{Nd_p^2}{A_1^2} \\ \frac{\lambda A_1 z_b}{Nd_p}, & z_b > \frac{Nd_p^2}{A_1^2} \end{cases}$$

(8)

Our long working distance DIH system here simply adopts a 4f telescope unit consisting of two lenses and an iris as shown in Figure 2a. The focal lengths of $L_1$ and $L_2$ are $f_1$ and $f_2$, respectively. The aperture diameter is changeable by adjusting the iris which is put immediately in front of $L_1$. The rear focal plane of $L_1$ overlaps with the front focal plane of $L_2$ and the SP of a CCD is located at the rear plane of $L_2$, and thus the CP is at the front plane of $L_1$. One advantage of this design is that the working distance is extended from $z_b$ to $f_1 + z_b$ compared to a lensless hologram that has the same resolution by using CCD with pixel size of $d_p/A_1$. It should be noted that the spatial resolution could be improved by using $L_2$ with larger focal length, but the working distance will always be longer than $f_1$.

Additionally, the extra phase term $\exp \left[ \frac{j K C}{2 \lambda} (u^2 + v^2) \right]$ turns out to be 1 in this system because $C = 0$. Though a telescope is not the first time applied in a holographic system, we evaluate the resolution of the design that almost reaches the theoretical one and improve the effectiveness of DIH in studying burning coal particles. We also expect the developed model to provide solutions to more complicated DIH systems with lens combinations such as holographic endoscope for measurements in a boiler in the future.

### EXPERIMENTAL SETUP

A schematic of experimental setup is shown in Figure 2. A double-pulsed Nd:YAG laser beam (532 nm) is spatially filtered and collimated to a plane wave of 50 mm diameter before illuminating the particle field. The width of each laser pulse is 5 ns, which is short enough to “freeze” the fast moving coal particles. The energy of each pulse is chosen as 30 mJ. Then the scattered OW and the undisturbed RW both travel through a 4f imaging unit whose usage is explained at the end of last section. Here the focal lengths are $f_1 = 100$ mm and $f_2 = 200$ mm, respectively, while the iris aperture can vary between 3 mm and 25 mm. The system magnification is 2. A band-pass filter (532 ± 10 nm) is mounted on a CCD camera (2048 × 2048 pixel array with pixel size of 7.4 × 7.4 μm²) to block the unwanted flame radiation. Hologram pairs from the double pulses are captured at a frame rate of 7 Hz by the camera.

Coal particles (diameter less than 200 μm, supplied by a screw feeder) are delivered by air stream and emitted to the center of an annular burner. They are then ignited by the surrounding methane flame. The inset of Figure 2a exhibits the detail structure of the burner. The particle-laden air is fed through the inner tube with an inner diameter $d_0 = 5.5$ mm, at a flow rate of 9.0 standard (298 K; 101,325 Pa) liters per minute (slpm). The methane is introduced though an annular slit with a thickness of 0.5 mm, at a flow rate of 2.0 slpm. The burner is installed on a 3D motorized translation stage, which is able to relocate the burner to control the flame distance and sample volume with an accuracy of 10 μm. The origin of the coordinate system is set at the center of the burner surface as shown in Figure 2a.

Particles of China Shanxi bituminous coal are studied, of which the main properties are listed in Table 1. The coal particles are ground and sieved to a size fraction from 10 to 200 μm, with a peak diameter of about 40 μm, which is similar to the pulverized coal used in thermal power plants. The temperature of the methane flame is first measured with a K-type thermocouple at some different heights as shown in Figure 2b that initiates with 600 K at the burner outlet and peaks 1300 K at the height of 10–15 cm. Different phenomena are seen at various heights in the coal combustion process. A representative photograph (Figure 2b) indicates that coal particles are ignited at $y = 4$ cm and above, where both the homogeneous volatile matter combustion and heterogeneous char burning are observed.

### RESULTS AND DISCUSSION

#### Calibration of the Lensed DIH System

In this design, the critical value of $z_b$ is chosen as $z_{bcr} = 52.7$ mm according to eq 7 and ref 50. The sample volume is located between $z_{bcr}$ and $1.6z_{bcr}$, indicating the working distance (from particle to $L_1$) is 152.7 mm to 192.3 mm. The centerline of the burner outlet is located 170 mm away from $L_1$. The imaging resolution of the system is calibrated with a USAF target set at different $z_b$. As described by eq 8, the resolution remains the same when $z_b$ is smaller than $z_{bcr}$, but it deteriorates in the $z_b > z_{bcr}$ region.

<table>
<thead>
<tr>
<th>Table 1. Properties of the Coal Sample</th>
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<tbody>
<tr>
<td>proximate analysis moisture ash volatile matter fixed carbon</td>
</tr>
<tr>
<td>air-dried, %</td>
</tr>
<tr>
<td>ultimate analysis carbon hydrogen nitrogen sulfur oxygen</td>
</tr>
<tr>
<td>air-dried, %</td>
</tr>
</tbody>
</table>
Figure 3a shows a resolution of 3.91 μm line width (group 7, element 1) at $z_b = 0.75z_{b_{cr}}$, very close to the analytical one of 3.7 μm line width. The reconstructed target becomes very blurred and hard to be recognized when $z_b$ gradually increases to 2.28$z_{b_{cr}}$. Lower resolution will increase the measurement uncertainty. To evaluate the size error and depth position error, we use our model to simulate holograms of sphere particles (regarded as opaque disks) with the same recording parameters of the experimental setup. Particles are reconstructed and extracted from these holograms followed with a comparison with the preset size and position. The size error (normalized by true diameter) and depth position error (normalized by pixel size) are shown in Figure 3b,c. Both the size error and depth position error increase after $z_b > z_{b_{cr}}$. The size error is larger for smaller particles because a smaller particle covers fewer pixels. However, the depth position error shows little relevance with particle size. The measured region in our experiment, $z_{b_{cr}}$ to 1.6$z_{b_{cr}}$, has a relatively low measurement uncertainty.

**Observation of Burning Particles and Volatile Matter Release.** By reconstructing 2000 depth planes from a hologram in a span of 40 mm, 3D distribution of particles and volatile clouds are refocused. Figure 4a,b is a sample hologram (recorded at $y = 10$ cm) and a reconstructed image at $z = 0$ in which no particle is in focus, respectively. The particle in region 1, of about 60 μm, is refocused at $z = -4$ mm with a clear border (Figure 4c). The larger particle in region 2 is refocused at $z = 6.4$ mm, surrounded with an obvious volatile cloud (Figure 4d), where the condensed matter introduces slight blurring around the burning particle. Then the cloud, which encloses fine soot particles less than the resolution, accumulates into a stringy shape tail. Figure 4e shows the top of the tail refocused at $z = 8.8$ mm, indicating its 3D feature.

Figure 5 compiles the reconstructed images of 16 selected burning particles. The volatile matters also display $x - y$ movement (No. 2), because of the nonuniform emission of decomposition product and attachments of volatile clouds are most possibly observed right above the ignition point. The cloud rises up with the gas and gradually detaches from the parent particle (Nos. 3–5) because of the solid–gas slip. In the area 10 cm < $y < 24$ cm, the detached stringy objects are commonly observed. The lengths of stringy objects vary from the longest (several millimeters, e.g., Nos. 6, 9, and 10) to the shortest (tens of micrometers, e.g., No. 16). The coal particles become bare and the char starts to burn. The stringy objects are formed by the detached volatile cloud and move with the gas flow before being oxidized. Some other tails, e.g., Nos. 8 and 12, break up into smaller ones. Previous investigations with shadowgraphy or conventional holography also observed similar phenomena.14,53 This can be explained by the fact that bituminous coals are rich in hydrocarbons and tars which accounts for soot formation.

Figure 6 shows the morphology evolution of coal particles along $y$ direction. Particles larger than 50 μm are collected from five holograms for each of the three cases: Figure 6a, burner outlet ($y \approx 0$); Figure 6b, $y = 10$ cm ($y/d_o \approx 18.2$); Figure 6c, $y = 24$ cm ($y/d_o \approx 43.6$). These particles are selected by the autodetection algorithm with good representativeness of the volatile clouds. Particles at the burner outlet have smooth edges, although their morphologies vary dramatically, which are similar to the raw coal particles. The edges of particles at $y = 10$ cm become ragged, and it can be explained by that the
volatile matters are ejected from the surface of particles in the devolatilization process and the ragged edges roughly reflect the side view of blow holes. The particle morphologies also become more irregular. A larger proportion of particles at $y = 24$ cm display morphologies significantly different from the ones at $y = 10$ cm. The particles in Figure 6, panels b and c are more blurred compared with Figure 6a, it is probably due to the lower hologram quality caused by extensively released volatiles that increase the shadow density. As described in Malek et al., the larger shadow density would decrease the signal-to-noise ratio. The particle velocities at $y = 10$ cm and $y = 24$ cm are about $8 \text{ m/s}$ (detailed below), so the residence times in these two regions are about 13 and 30 ms, respectively. Since the whole process of the devolatilization may last 100 ms, only the early stage of this process is observed in the present study. The fast devolatilization at this stage causes fast changes of particle morphologies. Further studies attaining longer residence time should be carried out in future studies to characterize the entire evolution during the devolatilization process.

The observation of relatively small particles from the cracking of volatile tar tail (e.g., No. 12 of Figure 5) should be attributed to the improved resolution of this system. In our previous research with the same camera but without the long working distance unit, only the volatile cloud and wake in a larger scale were imaged. Similarly, the improved resolution of the long distance system enables us to study the morphology change in Figure 6. The improvement can be easily seen by comparing Figure 6 and another previous study (also Figure 6 in that paper).16

**Particle Size Distribution.** In this study, particle size is defined as the diameter of the circle that has the same particle cross-section area. The distributions of particle sizes at the burner outlet, $y = 10$ cm, and $y = 24$ cm are obtained by 50 independent holograms for each case with a total number of 5593, 3216, and 1396 identified particles, respectively. Figure 7a–c shows the histograms and logarithmic normal fits of the three cases, with peaks at 35 $\mu\text{m}$, 48 $\mu\text{m}$, and 45 $\mu\text{m}$, respectively. A log-normal fit is often used to represent size distributions of pulverized coal particles.55 The fits agree well with the histograms in the measurements. A comparison of the probability density functions (PDFs) (Figure 7d) demonstrates an averaged size increase in the devolatilization process. The obvious size increase at in the residence time of 13 ms (from $y = 10$ cm) suggests that particles experience swelling in the very early stage of the devolatilization process. A very slight decrease of peak size from $y = 10$ cm to $y = 24$ cm might result from char burning out. However, the char burning out time is nearly 10 times as the devolatilization time, and this influence should be too small to be determined in the present study. In addition, the size distribution tendency along the longitudinal direction is of good accordance with that observed from the morphology evolution (Figure 6).

**Particle Velocity.** To extract the particle velocities, the camera is operated in a double-exposure mode and synchronized with the double-pulsed laser by a programmable timing unit. A pair of holograms are sequentially recorded with a time interval of 20 $\mu\text{s}$. Particles in the hologram pair are first identified and then matched in a 2D way by using the Hungarian algorithm,56 which minimizes the total distance to yield the globally most possible pairing of particles in sequential frames. The $x - y$ displacement is calculated by a simple subtraction, while the $z$ displacement was calculated directly with a focus correlation method because of the lower $z$ location accuracy.57 The particle velocity is then derived from the displacement and the time interval ($v_x = \frac{\Delta x}{\Delta t}$, $v_y = \frac{\Delta y}{\Delta t}$, $v_z = \frac{\Delta z}{\Delta t}$). Figure 8 demonstrates the 2D and 3D velocity field from a pair of hologram recorded at the outlet of the burner. In Figure 8a, the base is a depth-of-field extended image with all particles focused using a wavelet-based image fusion algorithm.42 Particles in the first frame are marked by red circles, while those in the second frame are marked by blue crosses, as shown in the inset of Figure 8a. The vectors present the magnitudes and directions of the particle velocities. Figure 8b visualizes the 3D velocity field. Particles are represented by the colored spheres. Both the diameter and the color of the spheres indicate the particle sizes. The vectors show the three components and magnitudes of velocities. 3D particle velocity measurement relies highly on the $z$ location accuracy, which is actually determined by the imaging resolution. We did not report velocity measurement in our two previous research studies16,17 because the uncertainty was relatively high. Empirically, locating and tracking burning coal particles are more difficult than cold particles, and thus needs higher physical resolution. It may be caused by the gas density fluctuation in the flame, as well as the deteriorative signal-to-noise ratio after the condensed-phase volatiles appear. That is
why the particle images at $y = 10$ cm and $y = 24$ cm seem more blurred compared to the nonignited particle at the burner outlet having lower gas temperature.

At each of three cases, particle velocities are examined. The radial direction velocity is characterized by $v_x$ in Figure 9a−c. At the burner outlet, the radial velocity is below 1 m/s for most particles, while the average velocity is around 0, indicating the particles are moving dominantly upward with the gas jet. At $y = 18.2d_0$, the $v_x$ in the outer region (larger $x$) tends to be larger than that in the core region. The outer particles fled from the center at an average velocity of about 0.5 m/s, which suggests an expansion of the flame in radial direction. At $y = 43.6d_0$, however, the $v_x$ difference between the inner and outer regions is not evident. Instead, there is strong velocity fluctuation, induced by unsteady flame motion. One is reminded that due to the limited field of view in $x$ direction the fast particles may exit the sample volume, while slow particles occupy the margin of the field of view. The distributions of axial velocities ($v_y$) are shown in Figure 9d−f. Because the $z$ span is much larger than the $x−y$ span of the measurement volume in hologram reconstruction, we adopt $z$ as the radial direction to characterize $v_y$. At the burner outlet, all particles are confined within the gas jet. Average $v_y$ presented a profile of pipe flow with the center velocity of 8 m/s. The large standard deviation is attributed to the difference of drag force on particles of different size. At $y = 18.2d_0$ particles are more widely spread in the radial direction, due to mixing of burning gas with the ambient air. The average velocity peaks at 7.5 m/s at the center. Particles at $y = 43.6d_0$ spread even wider and the average $v_y$ keeps on decreasing.

CONCLUSIONS

In this study, a model for describing a general lensed DIH system is first developed by expressing it to an equivalent lensless DIH. A 4f unit is adopted to achieve a working distance longer than 15 cm, with spatial resolution of 3.9 μm.
for DIH. Meanwhile, the lens aberration is suppressed. A direct observation of coal particle combustion in a pulverized coal flame shows that volatile cloud is ejected from the coal particle at the ignition stage and then detached from the parent particle due to the velocity difference between the coal particle and the bulk gas. The devolatilization products accumulate into stringy objects that are explained as soot emission from the reaction of hydrocarbons in the literature. Examining the reconstructed particles at different heights above the burner shows that the morphologies change after the combustion process. Ragged borders of particles indicate the formation of blow holes in the devolatilization. Particle size statistics indicate that particles experience swelling at first because of the devolatilization and then reduce slightly in size after very short char burning out. Both the radial and longitudinal velocity are measured at different heights. They visualize the influences of the flow expansion, the drag force, and the gravity on coal particles. The particle morphology, size, spatial distribution, and velocity can be quantitatively measured in the 3D volume.

This work demonstrates the effectiveness of a long working distance DIH system for particle measurement in a harsh environment like the methane flame. With this method, we improve the measurement accuracy while retaining a safe working distance. The progress hereby presented enables us to study the burning coal particles in three aspects compared to previous works: First, smaller soot agglomeration (∼10 μm) from volatile combustion can be observed. Second, morphology change before and after devolatilization is shown, and it will be helpful to characterize the volatile bubbling. Third, 3D velocity of coal particles is obtained with the improved resolution. The data are valuable for optimizing flow condition and validating models. We also expect the techniques described in this work can be used to develop a noninvasive holographic probe for measuring in situ parameters in the coal-fired burner or fuel engine in the near future.

Figure 9. Measured velocities of particles at different heights: (a–c) radial components \( v_x \), and (d–f) axial component \( v_y \). Scattered dots represent the results from individual particles. Curves are the averaged value at a chosen \( x \) or \( z \) location while the error bars give the rms value.

### AUTHOR INFORMATION

**Corresponding Authors**
*(Y.W.)* E-mail: wyycgsp@zju.edu.cn.
*(J.C.)* E-mail: junchen@purdue.edu.

**ORCID**

Xuecheng Wu: 0000-0001-9897-8776
Yingchun Wu: 0000-0001-8914-1931

### Notes

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### REFERENCES


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