Measurement of the number density of droplets in an aerosol by laser-induced breakdown method

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Abstract Laser-induced breakdown method is proposed as a diagnostic method to evaluate the absolute number density of droplets in an aerosol ejected from spray nozzles with high temporal resolution (∼several ns) and spatial resolution (∼100 μm). This method utilizes that the laser-induced breakdown threshold intensity of liquid droplets is much lower than that of gases. The volume, in which laser intensity exceeds the breakdown threshold of droplets, is evaluated from laser focus diameter and Rayleigh length, and intensity ratio between actual laser irradiation and the breakdown threshold of droplets. The number of droplets in this volume is evaluated from the experimentally measured breakdown probability using Poisson distribution formula. The experimental measurement result of steady state ejected aerosol by laser-induced breakdown method is reasonably agree with another measurement result, which is evaluated from the precipitation rate, droplets diameter distribution, and droplets velocity. We also applied this method to an intermittently ejected aerosol as a simulation of a diesel engine. The temporal density variation and spatially density distribution are clearly measured by this measurement method. This temporal density variation is clearly explained by the results of the steady state aerosol measurement results. Those are the number density of steady state aerosol, aerosol velocity as a function of valve pressure and temporal variation of valve pressure. The pressure dependences of breakdown threshold intensities of gases were experimentally measured for the measurement of fuel aerosol in high pressure condition in order to avoid the explosion by laser-induced breakdown. The threshold intensities of each gas decrease exponentially as a function of the gas pressure. N₂ gas is the best candidate gas for fuel aerosols because of the high breakdown threshold at high pressure condition and the same breakdown threshold intensity as the air breakdown. The breakdown threshold intensities of N₂ gas at 2.0 MPa was 7 times larger than that of the water droplets, which is large enough to diagnose the number density in fuel aerosol by this measurement method. These experimental results indicate that the laser-induced breakdown method can be applied as a diagnostic method of number density of liquid droplets in aerosol especially for fuel droplets in a diesel engine.

1. Introduction

Liquid droplets from spray nozzles are employed in many engineering, agricultural and industrial applications for performing functions such as painting, cleaning, and spreading. Many types of spray nozzles have been developed for different type applications. Simultaneously, some laser-based systems are utilized to understand their characters such as droplets diameter, velocity, and number density. Laser diffraction size analyzers (LDSAs), Phase Doppler velocimeters and Particle image velocimeters (PIVs) are the typical examples. They have been developed as commercially available installments. Number densities of droplets in aerosols were also evaluated by the visualization techniques with visible lasers. Schrielen, shadowgraph and laser scattering measurements are typical examples [Nishida 1987, Won 1992, Shiratani 1996]. Computer tomography technique was also applied to measure three-dimensional density distribution [Kawamura 1988]. However, those techniques can’t measure the absolute number density directly, can’t be applied for high density or large scale aerosols, and can’t be applied for high speed ejected aerosol.

We proposed and experimentally demonstrated a novel method to measure number density of droplets in aerosol using the laser-induced breakdown technique [Yashiro 2010a]. This method
utilizes large difference of the laser-induced breakdown thresholds between liquid droplets and air. The laser-induced breakdown threshold intensities were reported for various kinds of liquid droplets and gases, that the breakdown thresholds of droplets are a few 10 times lower than that of air [Pinnick 1988]. When a pulse laser is focused on a spot in atmosphere condition below the laser-induced breakdown threshold intensity of air, any generation of breakdown indicates that there are droplets or particles present in the laser focus spot. This measurement configuration has high temporal resolution determined by the laser pulse duration (several ns) and high spatial resolution determined by the laser focal parameters and intensity ratio between breakdown threshold of droplets and actual irradiation. This high temporal and spatial resolution can be utilized as a diagnostic device for number density measurement of droplets especially for intermittently ejected aerosol. Common rail systems as diesel engines have been developed, in which fuel aerosol is injected several times in a diesel engine in each combustion cycle with high speed as sound velocity. Diesel aerosol injection characters such as timing, amount, droplets size, number of repetitions, and density distribution are crucial technologies that govern the engine performance. We propose this measurement method can be applied to evaluate the temporally variation of number density distribution especially for an intermittently ejected aerosol such as a diesel engine. In order to demonstrate this measurement method, we have done three types of experiments. In this article we describe these experimental results and discussion about the laser-induced breakdown method. First is the experiment of demonstration to confirm the accuracy of the laser-induced breakdown method for number density measurement in a steady state aerosol [Yashiro 2010a]. Second is the application to an intermittently ejected water aerosol by this method as a simulation of diesel engine [Yashiro 2010b]. Last is the comparison of various breakdown threshold intensities of gases as a function of their pressure and water droplets as a simulation of fuel aerosol in high pressure gases [Yashiro 2011].

2. Principle of diagnostic method

Principle of diagnostic method was reported by our previous paper [Yashiro 2010c]. In this paper we assume that the spatial laser profile is a lowest-order Gaussian beam, and we evaluate the spatial intensity $I_{(r,x)}$ as a function of $r$ and $x$ in cylindrical coordinates using follow equation.

$$I_{(r,x)} = I_0 \left[ \frac{r_0}{\omega_{(r,x)}} \right]^2 \exp \left\{ -2 \left[ \frac{r}{\omega_{(r,x)}} \right] \right\}, \quad (1)$$

$$\omega_{(x)} = r_0 \sqrt{1 + \left( \frac{x}{x_0} \right)^2}, \quad (2)$$

Where $r_0$ is the focal radius at the optimal focus position and $x_0$ is half the Rayleigh length. When the actual laser intensity $I_0$ is $\alpha$ times greater than $I_{th}$ ($\alpha \geq 1$), $V_{th}$ is given by

$$V_{th} = -\pi \int_0^{x_1} r_0 \left[ 1 + \left( \frac{x}{x_0} \right)^2 \right] \ln \left[ \frac{1 + (x/x_0)^2}{\alpha} \right], \quad (3)$$

$$x_1 = x_0 \sqrt{\alpha - 1}. \quad (4)$$

Fig. 1 Schematic intensity distribution of a focused laser beam. Each number shows the intensity ratio $\alpha$ between actual laser irradiation and $I_{th}$. Each figure indicated by an arrow is the region of $V_{th}$ for each $\alpha$ [Yashiro 2010c].
A schematic diagram of the laser intensity distribution of Eq. (1) is illustrated in Fig. 1. When the laser intensity is the same as the breakdown threshold intensity ($\alpha=1$), $V_{th}$ is just the point at the coordinate origin. As increasing of laser energy ($\alpha \geq 1$), $V_{th}$ spreads to the periphery. Then, with increasing of laser energy, the total number of droplets in $V_{th}$ also increases.

The number of droplets in $V_{th}$ is evaluated from the experimentally measured laser-induced breakdown probability $P_b$ using Poisson distribution formula. $P_b$ for $k$ droplets in this volume can be calculated as a function of average number of droplets $\lambda$. Total breakdown probability of breakdown $\sum P_{bk}$ is given by following and experimentally measured:

$$P_{bk} = \frac{\exp(-\lambda)\lambda^k}{k!},$$  \hspace{1cm} (5)

$$\sum_{k=1}^{\infty} P_{bk} = 1 - P_{b0}. \hspace{1cm} (6)$$

From the average number of droplets $\lambda$ in the volume $V_{th}$ and the $V_{th}$, which is calculated by the intensity ratio between actual laser intensity and the breakdown threshold intensity, we can evaluate the number density of droplets in an aerosol.

3. Diagnostic of number density in a steady state aerosol

A schematic of the experimental arrangement of a steady state aerosol is illustrated in Fig. 2. In this experiment, two atomizing nozzles are employed. Water droplets are ejected continuously from these nozzles with air to a pressure of 0.1 MPa. Water flow rate through the nozzles were controlled at 10, 20 and 40 ml/min. Water droplets were ejected and spread at an angle relative to $Z$ axis as shown in Fig. 2.

A Q-switched Nd:YAG (yttrium aluminum garnet) laser [wavelength 1.06 $\mu$m, pulse duration 3 ns FWHM (full width at half maximum)] was used to generate laser-induced breakdown in this aerosol. A laser beam was expanded by optical system and focused by an f=1m plano-concave lens at a diameter of 146 $\mu$m (FW/2 ) and a Reyleigh length of 20 mm. The laser was focused under 400 mm vertically below the nozzle.

A photodiode is set at a distance of about 100 mm from the focal point to observe the visible breakdown emission. The scattering laser light in the aerosol is suppressed by a laser mirror and filters in front of the photodiode to be negligible level. The other photodiode is set as a monitor of the laser beam and this signal is used as a trigger of the oscilloscope to observe the generation of the breakdown. In this experiment, the breakdown probability is evaluated by 100 laser shots. The instability of the laser output is less than 5%.

Fig. 3 shows the $P_b$ as a function of laser intensity for 10, 20 and 40 ml/min. The
breakdown probability of atmospheric air as a function of laser intensity was also shown in Fig. 3. The difference of the breakdown thresholds between water droplets and air was about 20-fold. The breakdown threshold of the water droplets was about 8.0 x 10^9 W/cm². The $P_b$ increases monotonically as the laser intensity is increased as shown in Fig. 3. The theoretical breakdown probabilities for some number densities, which is explained in Eq. (5) and (6), are drawn as solid lines in Fig. 3 as a function of laser irradiance. The number densities of droplets at water flow rates of 10, 20 and 40 ml/min are evaluated from these lines at 4 x 10^3 (-25 to +25%), 9 x 10^3 (-33 to +11%) and 2 x 10^4 (-50 to 50%), respectively.

The accuracy of the measured number density by laser-induced breakdown is compared with another experimental measurement method, which is evaluated from the precipitation rate, droplet diameter distribution, and droplet velocity. The precipitation rate was measured at 6.4 x 10^{-4} cm⁻³ at the measurement point. The velocity of the water droplets was measured to be 5.5 m/s by a PIV. The diameter distribution and the medium diameter $D_{50}$ of the water droplets ejected by air pressure 0.1 MPa and a water flow rate at 30 ml/min were measured by an LDSA. The number of droplet, which was calculated by taking the diameter distribution into consideration, was about 4.4 times greater than that calculated from $D_{50}$. Because the number of droplets is proportional to the cube of the droplets diameter, the total number of droplets is strongly affected by the small-diameter droplets. We used this factor of 4.4 in order to estimate the actual number density from $D_{50}$ for different water flow rates. When the water flow rate is 20 ml/min and air pressure is 0.1 MPa, $D_{50}$ is 14.3 µm. The number density of droplets is calculated at 3.3 x 10^3 cm⁻³. The number density of droplets determined by the laser-induced breakdown is only 2.7 times larger than that determined by other method. The measurement error in the number density is strongly influenced by other measurement errors such as $D_{50}$, diameter distribution, etc. Therefore, we concluded that the number density of droplets evaluated from the laser-induced breakdown probability is in reasonable agreement with the other measurement result.

4. Diagnostic of number density in an intermittent aerosol

Schematic of experimental of an intermittent aerosol is shown in Fig. 4. The essential configuration of experimental arrangement is the same as shown in Fig. 2. A Q-switched Nd:YAG laser [wavelength 1.06 µm, pulse duration 10 ns (FWHM)] is used to generate the laser-induced breakdown using an f=200 mm plano-convex lens. The focus parameters of the laser were measured by a laser beam profiler to have a diameter of 40 µm (FW/e²) and a Rayleigh length of 940 µm.
Aerosol is ejected intermittently from an atomizing nozzle, which is smaller type than those used in Fig. 2, into atmosphere as shown in Fig. 4. The water flow rate through the nozzle is fixed at 10 ml/min. Air is intermittently supplied to the nozzle via an electrical valve at 10 Hz. The maximum pressure applied to the nozzle is 0.47 MPa. When the air pressure is low, the air flow is not strong enough to atomize water into micrometer-sized droplets. The number of droplets generated is negligibly small for air pressure under 0.05 MPa. The number of droplets depends strongly on the diameter of droplets. Therefore, this system supplies the aerosol intermittently for the number density. Synchronization between the intermittent aerosol and the laser is controlled by the digital pulse generator. This electrical pulse opens the valve with a duration of 50 ms at a repetitional rate of 10 Hz. This pulse generator also triggers the laser at 10 Hz. The time variation of the number density in an intermittently ejected aerosol is measured by changing the delay of laser irradiation.

Photodiode is set to observe the visible breakdown emission. The number density of the droplets is evaluated by measuring the laser-induced breakdown probability for 100 laser shots. The measurement was done below 100 mm from the nozzle as shown in Fig. 4. In this experiment the exit of the nozzle is defined as the coordinate origin and vertical direction as the Z axis (down is positive), the laser propagation direction as X axis as Fig. 2. The temporal variation of the number density and spatial distribution are experimentally measured.

The temporal variation of number density of droplets in an intermittent aerosol at 100 mm below the nozzle (0,0,100) is shown in Fig. 5. The black circle points show the directly measured results of the number density and solid line shows the fitting curve of these results. The accuracy of this measurement results were compared with that evaluated from the steady state aerosol configuration. The temporal variation of number density and spatial distribution are experimentally measured.

Fig. 4  Schematic of experimental arrangement of intermittently ejected aerosol and measurement system [Yashiro 2010b]. Copyright 2010 The Optical Society (OSA).

Fig. 5  Temporal variation of number density of water droplets in an intermittent aerosol at (0,0,100). Black circles show the experimental measurement results and solid line shows the fitting curve. Dashed curve is evaluated from the measurement of steady state aerosol [Yashiro 2010b]. Copyright 2010 The Optical Society (OSA).
variation of the number density of droplets is plotted as a dotted line that is evaluated from the temporal variation of the air pressure at the nozzle, number density of droplets in a steady state aerosol as a function of the air pressure and the flow speed at the measurement point as a function of air pressure. In this evaluation the flow speed of droplets is assumed the same as that of air. The directly measured result as a solid line is coincident with the result evaluated by the continuously ejected aerosol as a dotted line. The difference of the total values between the directly measured result (solid line) and the estimation from the steady state results (dotted line) is due to experimental error in controlling the water flow rate through the nozzle. It should be possible to attain much better agreement between the experimental results and estimates based on the steady state results by treating the acceleration length of each droplet in the air flow as a function of the droplet diameter and considering the spatial distribution of the air flow.

Fig. 6(a) and (b) show the results for spatially resolved measurements of the number density of water droplets in an intermittent aerosol along the vertical direction (Z axis) and along the horizontal direction (Y axis), respectively. The number density at (0, 0, 100) is the same as that shown in Fig. 5. As shown in Fig. 6(a), the total number density of droplets decreases with distance from the nozzle. This reduction is mainly due to diffusion of the aerosol at an angle to the Z axis. On the other hand, the droplet number density decreases as the distance from the center along the Y axis increases. The start of the rise time of the number density of droplets occurs at almost the same time at each observation point because the distance from the nozzle to each observation point is almost the same. These experimental results for the temporal and spatial distribution of the number density successfully demonstrate that the laser-induced breakdown method measures the dynamics of the distribution of droplets in the aerosol.

5. Measurement of breakdown threshold intensities of high-pressure gases and water droplets

The breakdown threshold intensities of gases decrease as increasing of their pressure [Phuoc 2000, Musing 2007]. On the other hand, the breakdown threshold for liquid droplets is independent of the surrounding gas pressure [Chylek 1990]. This indicates that the difference of the breakdown threshold intensities between liquid droplets and gases becomes narrower as increasing of the gas pressure. Therefore, we must confirm the difference of the breakdown thresholds precisely using the same laser focus configuration, whether this measurement method is available to diagnose in a high pressure aerosol such as a fuel aerosol in an engine. Moreover, laser-induced breakdown causes ignition or explosion in a fuel aerosol, which make it difficult to diagnose a fuel aerosol [Phuoc 2000]. We must find a gas, other than air or oxygen, that has a
high breakdown threshold at high pressure.

The breakdown threshold of several gases as a function of gas pressure and liquid droplets are measured in the same laser focus configuration. We used a YAG laser (1064 nm, 10 ns) to compare the breakdown threshold between high-pressure gases and water droplets. The YAG laser is operated at 10 Hz to generate the laser-induced breakdown. A high pressure gas cell is utilized to measure the breakdown threshold intensity. The laser beam was focused on the center of a high-pressure cell and aerosol using a focusing lens. The focused beam diameters of the laser was measured by a beam profiler at a focus diameter of 19.5 µm (FW/2e) with an f=220 plano-concave lens. Fresh gas was continuously supplied to this cell during the laser irradiation. Four kinds of gases (pure dust free air, N₂, He and Ar) are selected to measure the breakdown threshold intensities as a function of gas pressure at room temperature. The air in a diesel engine is compressed and air temperature and pressure increase immediately prior to the ignition. However, the threshold intensity of breakdown depends on only the molecular density of the gas. The temperature of compressed air is negligible low in comparison with the plasma temperature caused by the laser-induced breakdown. The gas pressure is proportional to the gas density when the gas temperature is constant. We used gas pressure instead of molecular density. The other measurement configurations are the same in Fig. 2 and 4.

Fig. 7 shows the breakdown threshold intensity as a function of gas pressure for the laser. This figure shows that, for four gases, breakdown threshold intensity \( I \) is proportional to \( P^{\gamma} \), where \( P \) is the gas pressure at room temperature and \( \gamma \) is a constant. In this experimental configuration, the \( \gamma \) values for N₂, Ar and He gases are -0.45, -0.58, and -0.97, respectively. The breakdown threshold intensity of air appears to be the same as that of N₂ gas, whereas the breakdown threshold intensity of He gas is about twice that of N₂ gas at 0.1 MPa. The breakdown threshold of N₂ gas becomes higher than that of He gas over 0.4 MPa. At 2.0 MPa, the breakdown threshold intensity of He (2.3x10^10 W/cm²) is considerably lower than that of N₂ gas (4.3x10^10 W/cm²). Ar gas will be higher than that of He gas at a pressure of about 3 or 4 MPa. N₂ gas is a suitable gas for estimation the air breakdown threshold intensity while avoiding the laser ignition of fuel aerosol.

The absorption mechanism of ns-pulse laser was described in many papers as inverse Bremsstrahlung absorption [Hughes 1975]. The ionization potential of a He atom (24.56 eV) is much higher than those of Ar atom (15.76 eV), and a N atom (14.53 eV). However, He atom has a metastable level (1s2p³P). The ionization level is only 4.8 eV from this level. Therefore, He atoms in the metastable level can be easily ionized at low electron temperature. At low gas pressure, the population of the metastable level is not sufficiently large to absorb the laser energy. Thus, most of He atoms are ionized from the ground state. In contrast, at high pressure, the population of the metastable level is large, so that a small number of He atoms are ionized from the
ground state. Thus, a plasma by laser-induced breakdown can be easily produced at low electron temperature and the breakdown is generated at low laser intensity.

The breakdown threshold intensity of water droplets is also measured with the same laser focusing configuration to be 6.0x10^9 W/cm^2 in air at atmospheric pressure. Chylek et al. reported that the breakdown threshold intensity of droplets was independent of the surrounding gas pressure in lower pressure gas than atmosphere [Chylek 1990]. We assume that this pressure dependence is the same at higher pressure of up to 2.0 MPa. The breakdown threshold intensity of N_2 gas of 2.0 MPa (4.3x10^10 W/cm^2) is 7-fold larger than that of water droplets. We reported that larger intensity ratio between high pressure N_2 at 2.0 MPa and water droplets are observed when we used probe lasers of shorter wavelength laser at 532 nm or short pulse laser at 3 ns pulse duration [Yashiro 2011]. We reported that when the laser intensity for diagnostic is over three times greater than the breakdown threshold of droplets, the measurement error for the number density of droplets is reduced [Yashiro 2010c]. Pinnick et al. reported that the breakdown threshold intensities for several kinds of liquid droplet [Pinnick 1988]. Fuel liquid droplets such as ethanol and diesel have much lower breakdown threshold intensities for laser-induced breakdown than that of water. This result also confirms that the laser-induced breakdown method is applicable for measuring the number density even in high-pressure fuel aerosols.

6. Summary

We proposed a novel measurement method for diagnostic of number density of droplets in an aerosol by laser-induced breakdown method. Measurement of number density of liquid droplets was experimentally demonstrated in a steady state aerosol by the laser-induced breakdown method. The result measured by this method was reasonably coincident with the other measurement result. The laser-induced breakdown method was also applied to an intermittent aerosol as a simulation of fuel aerosol in a diesel engine. The temporal variation of the number density of droplets was clearly measured and obviously explained by the experimental results of steady state aerosol. Moreover, spatial distribution of the number density was also measured as predictable results. These spatial density profiles also indicate the accuracy of the measurement method. The breakdown threshold intensities as a function of gas pressure and droplets were experimentally measured for air, N_2, He and Ar gases. The breakdown thresholds of gases decreased exponentially as increasing the gas pressure. N_2 gas was the best candidate as a buffer gas because of the high breakdown threshold at high pressure and the same breakdown threshold intensity of air. Moreover, fuel droplets in N_2 gas can avoid explosion, which disturbs the measurement by the laser-induced breakdown method. These experimental results indicate that the laser-induced breakdown method will be a good diagnostic method for number density distribution in an aerosol especially for a diesel engine with high spatial and temporal resolution.

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References