Measurement of Soot Particle Size Distribution in a Laminar Diffusion Flame Using Multiwavelength Polarization Ratio Method

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Abstract

The major source of particulate matters in atmosphere is soot particles generated in fossil fuel flames. With the development of scientific research and diagnostic technology, the control of particulate matter in the flame has been attracting more attention. Developing soot particle diagnostic techniques is necessary for reduction of soot particle emission. In this study, a development soot particle measurement method which is multi-wavelength polarization ratio (MPR) method based on Mie scattering theory is introduced, improved and validated.

In Chapter 1, the background and progress of research is introduced. The various definitions of particle diameter, common particle size distribution and widely use soot particle size measurement methods are introduced. Characteristics and measurement ranges of several measurement methods are discussed.

In Chapter 2, based on the Mie scattering theory, the multi-wavelength polarization ratio computation theory and the procedure for the estimation of the required parameters are also introduced. Previous study of MPR method, its limitations and development of present study are introduced. The measurement principles, experimental devices and experimental conditions are to be explained. The flame height is set at 30 mm in order to compare with other researchers’ data. The scattered light intensities were obtained by polarization charge coupled device (CCD) cameras set at a certain scattering angle. The determination process for known parameters which are particle number, geometric mean diameter, geometric mean standard deviation and complex refractive index are introduced in detail.

In Chapter 3, some famous and experienced methods such as light extinction method (LEM), portable aerosol mobility spectrometer (PAMS) and a transmission electron microscope (TEM) are used to validate the accuracy of MPR method. The measurement principles, experiment se-up and experimental conditions are introduced.

In Chapter 4, it is the results and discussion part. The geometric mean diameter $D_g$ and temperature distribution in the radial direction at $z = 20$ mm and $z = 25$ mm are introduced. The lower limit of measurement has been expanded to ten nm order through developing the calculation method. A comparison of the results for $z = 20$ mm and for $z = 25$ mm shows that the geometric mean diameter increases as moving downstream of the flame. Particle number distributions in the radial direction at $z = 20$ mm and $z = 25$ mm are introduced. A comparison of the results for $z = 20$ mm and for
$z = 25 \text{ mm}$ shows that the particle number decreases moving downstream of the flame. The reason is considered to be the coagulation of soot particles and oxidation.

The measurement accuracy of the MPR method is discussed in detail. The soot volume fraction is calculated and compared with the data obtained using LEM. While the experimental results are consistent with previous researches, the discrepancy between these two methods increases with the downstream distance. The reason is considered to be value of fitting coefficient which is based on $z = 20 \text{ mm}$ data. The particle size distribution is compared with the result obtained by a PAMS. The data obtained from the MPR method is smaller as compared with PAMS. The reason considered is the effect of aggregations. Polystyrene standard particles of 46 nm and 269 nm in ultrapure water under five different number densities were used to validate the accuracy of the MPR method too. For both standard particles, the MPR method overestimates the diameter when compared with the TEM. This could be attributed to the leak of polarized light at polarizers on each pixel of CCD sensor.

In Chapter 5, the correction methods which are used to improve the accuracy of the measurement were introduced in details. Finally, the conclusions are presented in Chapter 6.

**Keywords:** Polarization, Mie scattering theory, Particle size distribution, Particle number, Refractive index, Soot volume fraction
CONTENTS

Nomenclature ........................................................................................................................................ 6

Chapter 1  Introduction .......................................................................................................................... 9
  1.1  Background ..................................................................................................................................... 9
  1.2  The purpose of research ............................................................................................................. 12
  1.3  The research process of polarization ratio method ................................................................. 13
  1.4  A brief introduction to soot particle diameter ............................................................................. 1
    1.4.1  Geometrically equivalent diameter ....................................................................................... 1
    1.4.2  Optical equivalent diameter .................................................................................................. 1
    1.4.3  Electromigration diameter ................................................................................................... 2
    1.4.4  Aerodynamic diameter ........................................................................................................... 2
    1.4.5  Application of particle diameters .......................................................................................... 3
  1.5  Particle size distribution .............................................................................................................. 5
    1.5.1  Normal distribution ............................................................................................................... 5
    1.5.2  Lognormal deviation distribution ........................................................................................... 5
    1.5.3  Rosin-Rammler distribution .................................................................................................. 6
  1.6  Soot particle measurement methods .......................................................................................... 6
    1.6.1  Portable aerosol mobility spectrometer (PAMS) ................................................................. 8
    1.6.2  Thermophoretic sampling particle diagnostics followed by
electron microscopy analysis TSPD-TEM .......................................................................................... 9
    1.6.3  Light diffraction (LD) method ............................................................................................... 10
    1.6.4  Light extinction method (LEM) ............................................................................................. 11
    1.6.5  Dynamic light scattering (DLS) method ................................................................................ 11
    1.6.6  Laser induced incandescence (LII) method ......................................................................... 12

REFERENCES ............................................................................................................................................. 15

Chapter 2  Measurement principle and experimental set-up .............................................................. 19
  2.1  Measurement principle of the MPR method .............................................................................. 19
    2.1.1  Previous study-single wavelength polarization ratio method ..... 20
    2.1.2  Previous study - Multi-wavelength polarization ratio method based
on polarization ratio .......................................................................................................................... 22
    2.1.3  Present study - Multi-wavelength polarization ratio method based
on scattered light intensities ......................................................................................................... 24
2.1.4 Determination of unknown parameters ........................................ 26
2.1.5 Determination of the optical constant ....................................... 29
2.1.6 Calculation of the soot volume fraction in the radial direction ... 32
2.2 Experimental set-up of the multi-wavelength polarization ratio
   method .......................................................................................... 33
2.2.1 Butane burner system .............................................................. 33
2.2.2 Optical set-up .......................................................................... 36
2.2.3 Temperature measurement ........................................................ 42

2.3 The experimental procedure .......................................................... 43
2.3.1 Scattered light images of soot particles ..................................... 45
2.3.2 Function of fitting scattered light intensity ................................. 47

REFERENCES ................................................................................. 49

Chapter 3 Principles and experimental set-ups of validation methods ...... 50
3.1 Light extinction method for soot particle measurement ............... 50
3.2 Portable Aerosol Mobility Spectrometer (PAMS) for soot particle
   measurement .................................................................................. 52
3.3 Standard particles for validation .................................................... 55

REFERENCES ................................................................................. 57

Chapter 4 Soot particle size distribution and comparison results with
validation methods ............................................................................ 58
4.1 The geometric mean diameter and temperature distribution .......... 58
4.2 Particle number distribution ......................................................... 61
4.3 Soot volume fraction distribution and comparison with LEM ....... 62
4.4 Comparison of particle size distribution between PMR and PAMS .... 65
4.5 Comparison of mean particle size between MPR and TEM .......... 68

REFERENCES ................................................................................. 70

Chapter 5 Correction methods for improving measurement accuracy ...... 71
5.1 Scattering angle correction ......................................................... 71
5.2 Correction by sensitivity curve .................................................... 73
5.2.1 Experiments to obtain sensitive curve correction .................... 73
5.2.2 Obtaining the intensity calibration curve ................................. 74

Chapter 6 Conclusions ..................................................................... 79
Acknowledgments ........................................................................... 80
### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon$</td>
<td>Sum of residuals</td>
</tr>
<tr>
<td>$\tau$</td>
<td>Exposure time</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Polarization ratio</td>
</tr>
<tr>
<td>$D$</td>
<td>Particle diameter</td>
</tr>
<tr>
<td>$C$</td>
<td>Optical constant</td>
</tr>
<tr>
<td>$n$</td>
<td>Particle number</td>
</tr>
<tr>
<td>$\theta$</td>
<td>Scattering angle</td>
</tr>
<tr>
<td>$E$</td>
<td>Electric field intensity</td>
</tr>
<tr>
<td>$R$</td>
<td>Fuel nozzle radius</td>
</tr>
<tr>
<td>$e$</td>
<td>Primitive charge (1.6E-19 C)</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>Particle size parameter</td>
</tr>
<tr>
<td>$m$</td>
<td>Complex refractive index</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>Wavelength of the incident light</td>
</tr>
<tr>
<td>$z$</td>
<td>Flame height above the burner</td>
</tr>
<tr>
<td>$r$</td>
<td>Lateral measurement distance</td>
</tr>
<tr>
<td>$B$</td>
<td>Granular mechanical mobility</td>
</tr>
<tr>
<td>$\mu_g$</td>
<td>Gas viscosity,</td>
</tr>
<tr>
<td>$D_f$</td>
<td>Fractal dimension</td>
</tr>
<tr>
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<td>Turning radius</td>
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<tr>
<td>SVF</td>
<td>Soot volume fraction</td>
</tr>
<tr>
<td>$n_e$</td>
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<tr>
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<td>Slip correction coefficient</td>
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<tr>
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<tr>
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<td>Electrical low pressure impactor</td>
</tr>
<tr>
<td>TEM</td>
<td>Transmission electron microscope</td>
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DLS Dynamic light scattering method
PAMS Portable aerosol mobility spectrometer
$Q_{\text{ext}}$ Extinction coefficient of single particle
$Q_{\text{abc}}$ Absorption coefficient of single particle
$Q_{\text{s,agg}}$ Scattering cross section of aggregates
$Q_{\text{scat}}$ Scattering cross section of single particle
$Q_{\text{a,agg}}$ Absorption cross section of aggregates
$Q_{\text{abs}}$ Absorption cross section of single particle
LII Laser induced incandescence method
MPR Multi-wavelength polarization ratio method
TSPD Thermophoretic sampling particle diagnostics
$N_p$ Particle numbers of constituent particle for aggregate
$T$ Transmissivities of the two wavelength lights through the flame
$\nu_{TE}$ Velocity of charged particles relative to the current-carrying gas.
$i_1$ Perpendicular component of scattered light intensity for single particle
$i_2$ Parallel component of scattered light intensity for single particle
$I_{1,\lambda_1}$ Perpendicular components of theoretical scattered light intensities having wavelength of $\lambda_1$
$I_{2,\lambda_1}$ Parallel components of theoretical scattered light intensities having wavelength of $\lambda_1$
$I_{1,\lambda_2}$ Perpendicular components of theoretical scattered light intensities having wavelength of $\lambda_2$
$I_{2,\lambda_2}$ Parallel components of theoretical scattered light intensities having wavelength of $\lambda_2$
$P_{1,\lambda_1}^M$ Perpendicular components of measured scattered light intensities having wavelength of $\lambda_1$
$P_{2,\lambda_1}^M$ Parallel components of measured scattered light intensities having wavelength of $\lambda_1$
$P_{1,\lambda_2}^M$ Perpendicular components of measured scattered light intensities having wavelength of $\lambda_2$
$P_{2,\lambda_2}^M$ Parallel components of measured scattered light intensities having wavelength of $\lambda_2$
$\varepsilon_{11,\lambda_1}$ Residual between measured value and theoretical value of
perpendicular components having wavelength of $\lambda_1$  

$\varepsilon_{I2,\lambda1}$ Residual between measured value and theoretical value of parallel components having wavelength of $\lambda_1$  

$\varepsilon_{I1,\lambda2}$ Residual between measured value and theoretical value of perpendicular components having wavelength of $\lambda_2$  

$\varepsilon_{I2,\lambda2}$ Residual between measured value and theoretical value of parallel components having wavelength of $\lambda_2$
Chapter 1  Introduction

1.1  Background

Combustion converts the chemical energy contained in the fossil fuels into thermal energy [1]. Combustion of fossil fuels is the most important form of energy utilization in the present and supports the normal operation and development of human society [2]. However, fossil fuel combustion produces pollutant emissions such as carbon dioxide (CO$_2$) Nitrogen oxides (NO$_x$), sulfur oxides (SO$_x$), carbon monoxide (CO), Polycyclic aromatic hydrocarbons (PAH) and particulate matter (PM), including soot particles [3-6].

Soot particles usually result from incomplete combustion. The particles with aerodynamic diameter lower than 10 µm are known as PM10. Soot particles emitted from the diesel engines can enter human body through nose and mouth. Likewise, the particles with aerodynamic diameter lower than 2.5 µm are known as PM 2.5, and they can enter the human lungs and the blood circulation system [4]. Soot particles show strong absorbability and are capable of carrying several toxic trace elements. Particles with diameters of 0.01 ~ 0.1 µm and primary particle aggregates can enter the alveoli [5-6]. Additionally, soot particles present in the atmosphere are long lasting, usually for around a month. The diffuse reflectivity of the soot particle surface makes the glacier capable of absorbing more sunlight, which could speed up its melting, as shown in Fig. 1.2 [7]. Moreover, soot particle optical properties contribute to the greenhouse effect [8].

Therefore, from the perspective of environmental protection and social development, the effective control of soot formation is essential. To understand the soot particle formation mechanism, information on the primary particles is necessary. Then, obtaining the information about particle size, particle size distribution and soot volume fraction is important to understand soot particle formation mechanism.
Figure 1.1 Soot particles are deposited in the alveoli [6].

Figure 1.2 Melting glaciers [7].
Several studies have been conducted on the mechanism of soot particle formation; and its physical and chemical properties have been investigated [9-21]. Figure 1.3 shows the soot particles from the formation process of cyclic hydrocarbon to particle aggregates [16, 22].

Fuel pyrolysis results in the production of precursors for soot. At some point the PAH is large enough to develop into a nuclear soot particle [22]. Surface growth is the process of adding mass to the surface of a nucleated soot particle. Coagulation occurs when particles collide, thereby decreasing the particles number and holding the mass of soot particles constant. Agglomeration occurs when primary particles stick together [22].

![Soot formation process diagram](image)

Figure 1.3  Soot formation process [16, 22].

Primary soot particle size appears to vary depending on the experimental conditions such as the flame type, temperature and concentration of oxygen. However, most primary particles sizes reported range from 10 to 30 nm in laminar diffusion flame [22].

To understand the soot particle formation mechanism is essential to reduce soot particle emission. Developing soot measurement techniques is important to understand soot formation and growth mechanism. Soot particle measurement technologies can be classified into intrusive measurement methods and non-intrusive methods.

By using of intrusive methods like portable aerosol mobility spectrometer (PAMS), transmission electron microscopy (TEM), thermophoretic sampling particles diagnostics (TSPD), it is possible to measure particle size on the order of tens nm. Intrusive methods have several advantages and have been widely used until now. However, sampling is necessary in the intrusive measurement. The sampling process can also affect the structure of the combustion field and soot particle formation.
For most of non-intrusive measurement methods, like light diffraction method (LD), light extinction method (LEM), the dynamic light scattering (DLS) method, laser-induced incandescence (LII) and other laser used methods, flame structure is not affected and in-situ measurement can be achieved. There are a lot of advantages like high accuracy and high resolution of space. However, it is difficult to determine the value of complex refractive index and cannot obtain the particle size distribution for these non-intrusive measurement methods.

As compared with intrusive measurement methods, the information of spatial distribution and time evolution can be obtained by using multi-wavelength polarization ratio (MPR) method. As compared with some famous non-intrusive measurement methods such as LII and LEM. MPR method can determine particle number, geometric mean diameter, determine standard deviation and complex refractive index.

In the present study, a coflow laminar diffusion flame is used. It is very difficult to analyze the variation of soot particles in engine spray flame, due to the unstable high flow field and pressure variation. Therefore, the flame should have characteristics which are simple flame structure and easy to investigate soot particle formation, growth, oxidation and aggregation process. By using laminar diffusion flame, it is able to obtain time course of soot growth from primary soot particles to secondary soot particles and the measurement at each cross section of the flame can be produced. Butane was selected as the fuel. According to butane is a simple hydrocarbon, it is also one of the most widely used fuels in hydrocarbon fuels.

1.2 The purpose of research

There are two proposes in the thesis. The first purpose is to make sure the smallest measurement limit. We want to knows to know how small soot particles can we measure by using MPR method. Can we measure the nuclide soot particles? That is the problem. MPR method can measure soot particles which are from 20 nm to some tens-nm order. However, it is difficult to measure soot particles which ranges around 10 nm order.

The second purpose is to make sure the largest measurement limit of MPR method. By using MPR method, how large soot particles can we measure. Can we measure the aggregates? Is the aggregates affect on data measured by using MPR method? The author wants to ensure that.
1.3 The research process of polarization ratio method

Polarization ratio method is not a new method in soot particle measurement field. Kunugi et al [23] used a photomultiplier tube to measure the intensities of scattered light from soot particles in diffusion flame of hydrocarbons in air. From a comparison of the measured ratio of the two light-scattering intensities for the perpendicular and parallel polarization components with the predictions from Mie's theory, the particle size and number density of soot particles were determined.

D’Alessio [24] summarized the results of polarization ratio measurements for smoke produced by a variety of fuel chemistries and burner types and found the results to be consistent with predictions based on Mie theory and based on Rayleigh theory both for a prolate spheroid and a linear chain agglomerate.

Nishida et al [25] examined the means for determining the particle size from the Mie scattered light theory, and measured the scattered light intensity ratio and the transmittance of the laser to determine the soot particle size and number density in a laminar diffusion flame. Measurements were made and presented in correspondence with measurements of temperature, soot weight concentration and various gas composition concentration distribution. Nevertheless, due the nature of the single wavelength of the laser source, the single wavelength polarization ratio method is incapable of distinguishing the width of the particle size distribution.

Dobbins et al. [26] modeled the agglomerate structure as a porous sphere and were able to obtain fair agreement between the measured and predicted polarization ratio at 90°. By measuring the polarization ratio, Presser et al [27] researched the effect of swirl on droplet transport processes, which is examined in a pressure-atomized, hollow-cone kerosene spray. Polarization ratio method provided spatially resolved measurements on the local values of droplet mean size and number density in dense regions of the nonburning spray. Iannone et al [28] has been developed a light scattered devise which is based on polarization ratio technique for the discrimination of coal and fly ash. The scattering angle is set at an angle in the forward direction (60°). The results reported showed the possibility to measure the carbon percentage within fly ash by exploiting the different scattering properties of the particles. Measurement result showed that it has been, indeed, demonstrated that there is a good correlation between the ratio of the scattered intensities and the carbon content. In particular, a clear
decrease of the polarization ratio is seen when the carbon percentage rises within the fly ash. In addition, the reproducibility of the proposed method is investigated and the carbon content can be measured with an uncertainty of about 1%.

However, single wavelength polarization ratio method [23-28] cannot distinguish the width of the particle size distribution due to the nature of the single wavelength of the laser source. In order to solve this problem, a new particle measurement method which can measure soot particles distribution and distinguish the width of particle size distribution has been proposed [29-32].

Araki et al [29] used three kinds of He-Ne lasers with wavelengths of 543.5, 594.1, and 632.8 nm to pass through the test particles in purified water. The scattered images were acquired with a digital CCD camera set in parallel and perpendicular directions to the polarization plane of the incident light. The measurement range of the particle size can be extended up to the order of 1 μm, and that the accuracy of the results lies between 0.1 % and 7.6 %, when compared with the results of a centrifugal sedimentation method.

However, the method proposed in [29] had two problems. The first one is the influence of finite collecting angles of the superimposing scattered lights, and in the preset study, the fourth wavelength is utilized to exclude the effect of the superposition. The other problem is the effect of laser power stability, and by acquiring reference light intensity simultaneously, the effect of fluctuation of laser power is canceled. Inoue et al [30] used multi-wavelength polarization ratio to solve two problems of previous research [29]. Four different wavelength lasers whose wavelengths are 532.3, 543.5, 594.1 and 632.8 nm. The four wavelengths were utilized to exclude the effect of finite collecting angles of the superimposing scattered lights. By acquiring the reference light intensity simultaneously, the effect of the fluctuation of the laser power was canceled. The measurement range in diameter was further extended up to 3 μm with an error of 7.6 % at maximum. Furthermore, the number density of particles was newly measured with a maximum error of 32%. How soot particles can absorb the light and the chemical structure of flame field is not clear. It is difficult to determine the value of the complex refractive index exactly. For most of optical measurement methods, including the single wavelength polarization ratio method, the complex refractive index was obtained from previous research. However, the values of complex refractive index are different under different experiment conditions; as it is the standard deviation.
Therefore, developing a method which can measure the soot particle size and determine the value of complex refractive index is necessary.

Compared with previous studies of multi-wavelength polarization ratio method [29-30], Koizumi et al [31-32] improved the computational method of multiwavelength polarization ratio method. By using the scattered light intensities instead of polarization ratio as a function of geometric mean diameter. The geometric mean diameter, the geometric standard deviation, the particle number and the complex refractive index in a butane laminar coflow diffusion flame are determined simultaneously and the lower limit of measurement has been reduced to 1 nm order.
1.4 A brief introduction to soot particle diameter

Particle diameter is the most basic geometric characteristic of soot particles. Tapered particles are represented by their bottom diameter and height. Cubes are represented by their side lengths. However, in the actual measurement, most soot particles have irregular shapes. Therefore, it is difficult to describe their particle size accurately. The particle size of a spherical particle with a unit density can be expressed simply as a geometric diameter. The equivalent diameter is often used to represent the particle size for irregular particles [33-36]. There are several methods to define the particle diameter. Each of method has its own application background and definition. In the following sub-section, detailed definitions of diameter according to the measurement principle are introduced for soot particle measurement.

1.4.1 Geometrically equivalent diameter

Regarding particle measurement, the geometrically equivalent diameter is frequently used to represent the particle diameter. It is a virtual value rather than a real diameter. The geometrically equivalent diameter can be equivalent to the projected area diameter, the equivalent surface diameter or the equivalent volume diameter [34-36]. As measuring instruments operate based on different principles, the geometrically equivalent diameters varies according with the measuring instrument.

For example, in the electron microscope measurement, the diameter of the particle is represented by the maximum width of particle projection. The soot particles are collected in a copper mesh and the two-dimensional image information of soot particles is obtained by the electron microscope. It is required to convert the projection area and the main projection parameters such as the length through empirical relationships into three-dimensional structure parameters of the real soot particles.

1.4.2 Optical equivalent diameter

Optical equivalent diameter is defined as the particle diameter of the standard particle with the same instrument detection response signal as the measured particle [35]. Optical instruments detect particles by their interaction with laser light. Optical equivalent diameter measurement is widely used in static light scattering method, laser
diffraction method, dynamic light scattering method, light extinction method, multi-wavelength polarization method and other optical measurement methods. In measurement methods that use laser, the existing commercial instruments usually rely on the standard particle methods to determine the optical equivalent diameter. However, these methods tend to be affected significantly by the morphology and complex refractive index of the measured particles. The measured value is converted to the particle size using a physical law, such as Mie theory, developed when assuming a specific particle shape, such as a sphere, and the specific physical conditions.

1.4.3 Electromigration diameter

The method of measuring the particle diameter distribution according to the electromobility of aerosol particles appeared in 1902 [37]. Until now, it has become an important mean to measure particle size. Electromobility is defined as the velocity of a particle with a charge of a unit element in a unit strength electric field.

\[ Z_p = \frac{v_{TE}}{E} = n_e eB = \frac{n_e e C_c}{3 \pi \mu g d_m} \]  

(1-1)

Here, \( v_{TE} \) is the velocity of charged particles relative to the current-carrying gas. \( E \) is the electric field intensity, \( n_e \) is electrically charged particles number, \( e \) is the primitive charge \((1.6E-19 \text{ C})\), \( \mu_g \) is the gas viscosity, and \( C_c \) is the slip correction coefficient. The parameter \( B \) is the granular mechanical mobility, which is with related to the size of the spherical particles in the air drag force. For spherical particles, \( B \) can be used to measure the relative ease of motion of the particles in the air flow. Therefore, the mobility of particle size \( d_m \) can be determined according to \( B \). For the non-spherical particles, \( d_m \) is actually an equivalent diameter; in other words, a spherical particle with the diameter of \( d_m \) and the measured non-spherical particles are subjected to the same drag force in the air flow. This diameter is widely used in the electric mobility method in devices such as SMPS, PAMS, SEMS [38].

1.4.4 Aerodynamic diameter

An electronic low-pressure impactor is usually employed to measure the aerodynamic particle size. The aerodynamic diameter is defined as the diameter of a sphere with standard density that settles at the same terminal velocity as the particle of interest [39]. The difference between the aerodynamic diameter and the
electromigration diameter is that although they both rely on charged particles measurement, the aerodynamic diameter is based on the principle of inertial deposition. In other words, it is based on the relaxation time to differentiate particle diameters:

\[ \tau = mB \]  

In the equation, \( m \) represents the particle mass, while \( B \) refers to the mechanical mobility. The aerodynamic particle size of spherical particles with a density \( \rho_0 \) of 1g/cm\(^3\) is expressed as follows:

\[ \rho_0 = \frac{d^2_a C_c}{18 \mu_g} \]  

Spherical particles with this particle size have the same relaxation time as the measured particles. Nevertheless, electrostatic measurement is conducted only for obtaining the mass and quantity concentration of the particles. 

### 1.4.5 Application of particle diameters

Table 1-1 summarizes the definitions of particle diameter and applications in several measurement methods. The principles of different methods will be introduced in the next part. The geometrically equivalent diameters are usually used in optical microscopes, electron microscopes, image analysis method and so on. The optical equivalent diameter is widely used in static light scattering method, laser diffraction method, dynamic light scattering method, light extinction method, multi-wavelength polarization method and other optical measurement methods. Electromigration diameter is widely used in the electric mobility method in devices such as SMPS, PAMS, SEMS. Aerodynamic diameter is used in electrical low-pressure impactor (ELPI). Stokes diameter is widely used in the sedimentation methods.
<table>
<thead>
<tr>
<th>Measurement method</th>
<th>Measurement range</th>
<th>Definition diameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electron microscope</td>
<td>10 nm 100 nm 1 μm</td>
<td>Geometrically equivalent diameter</td>
</tr>
<tr>
<td>Portable Aerosol Mobility Spectrometer (PAMS)</td>
<td>1 μm 10 μm 100 μm</td>
<td>Electromigration diameter</td>
</tr>
<tr>
<td>Centrifugal liquid sedimentation (CLS) method</td>
<td>100 μm 1 mm</td>
<td>Stokes diameter</td>
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<td>Light diffraction (LD) method</td>
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<td>Optical equivalent diameter</td>
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<td>Dynamic light scattering (DLS) method</td>
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<td>Optical equivalent diameter</td>
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<tr>
<td>Laser induced incandescence (LII) method</td>
<td>10 μm 100 μm</td>
<td>Optical equivalent diameter</td>
</tr>
<tr>
<td>Multi-wavelength polarization ratio (MPR) method</td>
<td>1 μm 10 μm</td>
<td>Optical equivalent diameter</td>
</tr>
</tbody>
</table>
1.5 Particle size distribution

Soot particles are distributed in groups in a discrete way. Nevertheless, there are a large number of particles that can be treated as continuous. The size distribution of polydisperse grain system consisting of particles of different size shows the form of a distribution with a single peak or multiple peaks.

In the actual measurement methods, the particle size is not completely consistent with a certain distribution function. In some cases, it is not clear in advance which distribution function it approximately conforms to. Therefore, in order to determine which distribution function is most appropriate for the measured particle system, the sum of the squares of the measured and calculated values is frequently used. Currently, most distribution functions of two-parameter type are adopted, one of which is the characteristic size parameter to represent the particle size, and the other is the distribution parameter to represent the particle size distribution of the particle group. The commonly used particle size distribution functions are introduced below.

1.5.1 Normal distribution

The normal distribution, also known as Gaussian distribution, is expressed as follows [40]:

\[
f(D) = \frac{1}{\sqrt{2\pi}\sigma} \exp \left[ -\frac{1}{2} \left( \frac{D - \bar{D}}{\sigma} \right)^2 \right]
\]  

(1-4)

Where, \( D \) is the diameter of soot particle, \( \bar{D} \) represents the mean diameters of soot particles. The parameter \( \sigma \) refers to distribution parameter, it is the width of distribution. As the standard normal distribution function is a symmetric function, the smaller the distribution parameter \( \sigma \) is, the narrower the distribution is. When \( \sigma < 0.2 \), the particle swarm can be viewed as a monodisperse particle system.

1.5.2 Lognormal deviation distribution

The distribution of actual particles is barely symmetrical. Therefore, the normal distribution is not so common in practice. The logarithmic normal distribution function is more commonly used. The lognormal distribution function is derived by taking the logarithm of all the parameters in the normal distribution function, with the form shown
below [40]:

\[ f(D) = \frac{1}{\sqrt{2\pi}\sigma_g D} \exp \left[ -\frac{1}{2} \left( \frac{\log_{10} D - \log_{10} \sigma_g}{\log_{10} \sigma_g} \right)^2 \right] \quad (1-5) \]

The probability distribution function, \( f(D) \), where \( \sigma_g \) indicates the geometric standard deviation and \( D_g \) denotes the geometric mean diameter.

### 1.5.3 Rosin-Rammler distribution

Known as the \( R-R \) distribution function [40], the Rosin-Rammler distribution function was first proposed by Rosin and Rammler in 1963 after a study on the particle size of pulverized coal. It is a particular case of Webster's probability distribution. Most coal mine dust is suitable for this distribution, expressed by the following equation:

\[ V(D) = 1 - \exp\left[ -(D/D_g)^k \right] \quad (1-6) \]

Where \( D \) represents particle diameter and \( V \) indicates the cumulative volume percentage of particles with diameter less than \( D \) in the particle swarm. \( D_g \) is referred to as the characteristic size parameter, indicating that the volume of particles less than this size accounts for 63.21% of the total volume of particles. The size of \( D \) value generally indicates the size of the entire particle system. \( k \) refers to the particle group size distribution parameter, which is dimensionless and indicates the dispersion degree of particle size. The smaller \( k \) is, the wider the dispersion degree is. Conversely, the larger \( k \) is, the more likely the particles are to be monodispersed.

### 1.6 Soot particle measurement methods

There are a lot of hypotheses about the reaction paths of the various stages in the formation of soot particles. However, there is no unified soot particle generation mechanism; and it is still needed to use different methods and techniques of soot particles generated test simulation and diagnosis. The detection of soot particles plays an important role in the exploration and verification of the physicochemical reaction mechanism during soot generation. Many methods for soot particle sizing have been proposed [41-56]. Currently, soot particles detection methods can be divided into two categories: intrusive and non-intrusive methods as shown in figure 1.4 [41].

In intrusive measurement methods, the measurement device makes direct contact with the particles, for example, the portable aerosol mobility spectrometer (PAMS)
Method [42-43] involves a sampling measurement instrument for soot particles and usually supplemented by a dilution method. Soot particles have frequently been collected by sampling methods for subsequent observation and analysis with the assistance of electron microscope. Thermophoretic sampling particles diagnostics (TSPD) is one of the most commonly used sampling methods [44]; while the collected soot particles are usually examined by scanning electron microscope (SEM) [45] or transmission electron microscopy (TEM) [44]. Thermocouple particle densitometry (TPD) [46] is applied to measure flame temperature and soot volume fraction at the same time. Nevertheless, it is a challenge for intrusive measurements to achieve high temporal and spatial resolutions [41]. Moreover, they will cause disruption to the measured flame. Therefore, the non-intrusive optical method is most widely used for soot measurement [41].

Optical measurement techniques are divided into two categories [31]. One kind of measurement techniques are based on lasers which mainly includes light diffraction method (LD) [47], light extinction method (LEM) [48-50], the dynamic light scattering (DLS) method [51], laser-induced incandescence (LII) [52-53], multi-wavelength polarization ratio method [29-32] and other laser used methods.

Another kind of measurement technique is based on the flame emission spectrum. It directly detects integral values of soot emission and then calculates temperature and soot volume fraction from the flame emission spectrum. A typical emission technique is two-color method [56]. The principles of these soot measurement techniques will be described in the following sub-sections.
1.6.1 Portable aerosol mobility spectrometer (PAMS)

The layout of the various components of the prototype instrument and the flow scheme is shown in Figure 1.5 [43]. The key components include a miniature dual-corona bipolar charger, a differential mobility electrical classifier, and a condensation particle counter [42]. A bipolar charger is used for establishing a known equilibrium charge distribution. After being charged, the particles are transferred into a differential mobility analyzer (DMA), where they are categorized on the basis of their electrical mobility. With different voltages applied, different electrical mobilities are obtained in a DMA. A condensation particle counter (CPC) is used to count the mobility-classified particles downstream of the DMA [42-43].

PAMS method is applied to obtain each soot particle size and particle number. PAMS merits are high system integration, high industrialization degree and easily portable. Nevertheless, the main drawback is the dilution of collected sample. It is ineffective in restoring real morphology of particles within the flame, which is a challenge to all dilution sampling method. There are no effective technological or theoretical measures to address this issue. The PAMS measurement range goes from 10 to 1μm order [43].
Figure 1.5  (a) Flow scheme and layout of different components of PAMS; (b) assembly of charger, DMA, and CPC units; (c) prototype PAMS instrument. Flow rates shown are in L/min [43].

1.6.2 Thermophoretic sampling particle diagnostics followed by electron microscopy analysis TSPD-TEM

Figure 1.6 shows the soot particles and aggregates obtained from a flame using a thermophoretic sampling particle diagnostics (TSPD) [44]. The sample is observed using a transmission electron microscope (TEM), and the morphology and size of the soot particles are analyzed [44].

The TSPD-TEM method [44] is capable of obtaining the morphology, aggregation and particle size regardless of the position and time in the flame. The direct and instantaneous sampling enables researchers to observe instantaneously the morphology of soot particles and aggregates. Besides, the particle morphology will be maintained in the flame throughout the process of sampling and the. Similar to the laser induced incandescence method, TSPD-TEM is applied in the calibration of non-intrusive methods that only provide relative soot volume fractions [44]. However, this method is limited to a single point measurement; and therefore, it cannot be used to obtain spatially and temporally resolved results for unstable flames. By using TSPD-TEM, the lower limit of measurement can reach 0.1~ 0.2 nm [56].
The gradients in temperature and velocity impose restrictions on the accuracy of TSPD, limiting the accuracy for the calculation of the Nusselt number.

1.6.3 Light diffraction (LD) method

The laser diffraction method [47] is used to analyze light diffraction phenomena in a parallel laser beam in a particle dispersion liquid. The diffraction pattern can be observed on the focal plane of the lens when the light diffracted by the particles dispersed in the liquid is condensed by the lens. The smaller the particle size, the larger the diffraction angle of light, and vice versa. Therefore, the light intensity distribution on the focal plane of the lens is mixed with diffracted light from particles of various sizes.

The particle size distribution and average particle size are calculated using Fraunhofer's diffraction theory from the diffracted light intensity distribution by taking...
advantage of the fact that the change in the diffraction pattern concerning the angle decreases as the particle size decreases. Based on the Mie scattering theory, the scattered light also exhibits the scattering intensity pattern at each angle according to the particle size. Therefore, the light intensity pattern is observed as a mixed pattern of diffracted light and scattered light of particles. Besides, the average particle size and the particle size distribution can be obtained simultaneously by analyzing the angle-dependent light intensity pattern. Moreover, this method is the most widely used and most developed type of the various scattering particle measuring methods [56]. The upper limit of the light diffraction method can reach up to 3 mm, and the lower limit of measurement is 0.01 μm [56]. Thus, the diffraction method can measure only large particles and has limitations to measure particles with a mean diameter of less than 1 μm.

1.6.4 Light extinction method (LEM)

Rather than the scattered light intensity signal, the attenuation of the transmitted light intensity relative to the incident light intensity is the foundation for the development of the light extinction method (LEM) [48-50]. Not only can the technique measure the particle size, it can also measure the soot volume fraction.

The principle is that the scattering and absorption of the particles causes the intensity of the beam to be attenuated when the beam passes through a medium which is containing particles. The extent of attenuation is associated with the size and concentration of particles, thus providing a scale for particle measurement. Distinct from other methods of light scattering, the full scattering method measures the transmitted light instead of the scattered light of particles.

The principle is simple, the measurement is convenient and quick, the requirements for instrumental equipment are low, the data collection and processing process is simple. Its measurement range is relatively wide, with a lower limit of tens of nanometers and an upper limit of about 10 μm [48-50].

1.6.5 Dynamic light scattering (DLS) method

The application of DLS to flames has been reviewed briefly by Lamprecht in 1999 [51]. Rayleigh scattering occurs when the particles are much smaller than the incident wavelength. If the light source is a laser, the phenomenon which scattered light
intensities fluctuates with time can be obtained in a certain direction. This is because particles in the liquid or flame are follow the Brownian motion. The particle velocity of Brownian motion is related to particle size (Stokes - Einstein equation). Large particles move slowly and small particles move fast. If large particles are measured, the intensity of the scattered light will fluctuate slowly as they move slowly. Similarly, if the small particles will be measured, the intensity of the scattered light will fluctuate rapidly because they are moving fast. The particle size and its distribution can be calculated by the fluctuation of light intensity and the correlation function of light intensity.

The dynamic light scattering (DLS) method [51] has been applied for ultrafine particle measurement in various applications. As its measurements are based on the Brownian motion of particles, it is capable of measuring ultrafine particles as small as a few nanometers. The measurement range of DLS method is from 1 nm to 1 μm order [56].

The measurement results are highly accurate for monodisperse and narrow distribution grain systems. For wide distribution and multi-peak distribution grain systems, however, the measurement results are not as accurate. DLS measurements show high sensitivity to temperature and solvent viscosity. Therefore, it is necessary to keep the temperature constant and determine solvent viscosity for obtaining reliable results of DLS experiment. DLS is also restricted to transparent sample preparation.

1.6.6 Laser induced incandescence (LII) method

With regard to the measurements of soot volume fraction, laser-induced incandescence has been accepted as a primary technique [52-55]. LII method is considered most effective in measuring soot volume fraction for conducting internal combustion research. It is aimed at determining the two-dimensional (2-D) distributions of soot volume fraction and primary particle size.

According to the principle of LII method, soot particles are subject to heating through the absorption of laser radiation by raising the temperature to approximately 4000 K. The temperature radiation achieved in this process is known as LII signal.

After laser heating, the primary-particle sizes or primary particle size distributions with pulsed LII are determined on the basis of the particle-cooling rate, which contributes to a primary-particle-size dependent decay rate of the LII signal, as
measured by means of time-resolved detection (TiRe-LII).

This technique demonstrates its capability of spatial and temporal soot measurements; and therefore, it is applicable to practical combustion systems. However, the cost of the equipment is relatively high. In addition, a theoretical model is required for particle sizing. Analysis of the data shows that for a stable measurement target, without flickering, a measurement error of less than 20 ppb can be achieved, resulting in a measurement range of tens of ppbs for soot volume fraction measurement [55]. Lower limit of measurement of LII method can reach 0.1 nm order [53].

Common particle size measurement methods were summarized. The evaluations of each measurement method are summarized in table1-2 [41-56].
<table>
<thead>
<tr>
<th>Measurement method</th>
<th>Measurement range</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
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</table>
| Electron microscope                         | 10 nm  100 nm     | 1. Observe morphology of soot particle.  
2. Accurately and directly obtain particle size. | 1. Can not obtain the spatially and temporally resolved data.  
2. Disturb the measured flame. |
| Portable Aerosol Mobility Spectrometer (PAMS)| 1 μm  10 μm  100 μm | 1. Can obtain each classified particle size and particle number.  
2. High integration, portable carrying. | Can not obtain the spatially and temporally resolved data. |
| Centrifugal liquid sedimentation (CLS) method|                   | High size resolving power, enabling populations of single particle to be distinguished from populations of dimers and multimers. | Long measurement time for submicron particles and correction of absorption coefficient is necessary. |
| Light diffraction (LD) method               |                   | 1. Easy measurement.  
2. Wide measurement range. | Complex refractive index is necessary and submicron particles are not accurate. |
| Light extinction method (LEM)               |                   | 1. The device structure is simple.  
2. High precision. | The morphology of soot particles is not considered. |
| Dynamic light scattering (DLS) method       |                   | Is no independent on complex refractive index. | Easy to depend on scattering intensity and affected by dust. |
| Laser induced incandescence (LII) method    |                   | 1. Two-dimensional measurement.  
2. High temporal resolution and high spatial resolution. | 1. Complex refractive index can not be determined.  
2. A known and uniform soot volume fraction is needed. |
| Multi-wavelength polarization ratio (MPR) method |                   | 1. Particle size distribution can be determined  
2. The complex refractive index can be determined | Expensive set-up and complex procedures. |
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Chapter 2  Measurement principle and experimental set-up

2.1 Measurement principle of the MPR method

MPR method is based on Mie scattering theory. The Mie scattering theory [1] was developed on the basis of the electromagnetic scattering theory proposed by Mie in 1908. It is the premise of various light scattering theories commonly applied in the measurement of particle concentration and particle size distribution. Maxwell’s equations enable the interaction between the light and matter to be expressed mathematically. The solution of these equations requires information about the geometry of the inhomogeneous particle and its electric, magnetic properties and optical properties with respect to its surroundings. For homogeneous spheres, for example, it is necessary to determine particle diameter, complex index of refraction and the wavelength of the incident light. Once these equations are solved, it can be established how a specific particle absorbs and scatters the incident electromagnetic wave on it. The reverse of this idea is workable as well. That is to say, particle physical and optical properties can be determined from an inverse analysis using this relation like the Maxwell equations if the exact relation for the interaction between light and particle is known.

In case that the particles are significantly smaller than the wavelength of the light incident on them, the Mie scattering theory is further simplified to the Rayleigh approximation. Due to its relative simplicity and availability, the Mie scattering theory has been commonly applied to characterize particles in different physical systems; even though the particles are not homogeneous spheres. It can be expected that this mathematical simplification leads to physically inaccurate predictions. In various applications, this error may be small, insignificant or comparable with the signal-to-noise ratio of the detection system.

However, there are some circumstances where more precise monitoring is required for particle properties. Apart from gaseous components, the solid particles such as soot particles and fly ash particles also exist in the combustion field. Therefore, the Mie scattering theory can also be applied to the study of optical and radiation characteristics
exhibited by such materials. The solution process of Mie scattering theory is highly complex. Rather than elaborating here, only the most relevant conclusions regarding the Mie scattering theory will be presented.

2.1.1 Previous study-single wavelength polarization ratio method

Figure 2.1 shows the light scattering of a single particle for an incident light of random polarization. The observation plane is shaped by the incident light axis and the observer, the camera in this case. The scattered light involves component $i_1$ and component $i_2$. The component $i_1$ is perpendicular to the observation plane, while the component $i_2$ is parallel to the observation plane and can be expressed by the following equation.

\[ i_1 \propto \tau_{\lambda} C_{\lambda} f_{i_1}(D, m, \lambda, \theta) \quad (2-1) \]

\[ i_2 \propto \tau_{\lambda} C_{\lambda} f_{i_2}(D, m, \lambda, \theta) \quad (2-2) \]

Herein, the parameter $\tau$ refers to the exposure time of the polarization CCD cameras, the parameter $C_{\lambda}$ denotes the optical constant which the factor is depending on the particle number concentration and on the instrument constant, $D$ represents the particle diameter, $m$ is the complex refractive index, $\lambda$ is the wavelength of incident light and $\theta$ means the scattering angle.

Figure 2.1 Light scattering from a single particle for incident light of random polarization.
The polarization ratio is defined as component $i_1$ divided by component $i_2$ which can be expressed by the following equation.

$$\rho = \frac{i_2}{i_1} = \frac{\tau_{\lambda} C_\lambda f_{i_1,\lambda}(D,m,\lambda,\theta)}{\tau_{\lambda} C_\lambda f_{i_2,\lambda}(D,m,\lambda,\theta)}$$  \hspace{1cm} (2-3)

In Eq.2-3, $i_1$ and $i_2$ can be measured, the parameters of $\tau_{\lambda}$, $C_\lambda$, $m$, $\lambda$ and $\theta$ are known values. Therefore, the particle diameter can be obtained.

The intensity functions depend upon both the complex refractive index $m$ and the size parameter $\alpha$. The value of $m$ could be obtained from calculation. The size parameter $\alpha$ is calculated using the following formula.

$$\alpha = \frac{\pi D}{\lambda}$$  \hspace{1cm} (2-4)

In real particle size measurement, the measurement target is particle group. When the above theory is applied to a particle group having a particle size distribution, the scattered light intensities of each component are expressed by the following equations.

$$I_1 = \sum i_1 = \tau CN f_{i_1}(D_g,\sigma_g, m, \lambda, \theta)$$  \hspace{1cm} (2-5)

$$I_2 = \sum i_2 = \tau CN f_{i_2}(D_g,\sigma_g, m, \lambda, \theta)$$  \hspace{1cm} (2-6)

Herein, $N$ is the particle number. The polarization ratio which is in particle group is shown as flows.

$$\rho = \frac{i_2}{i_1} = \frac{\tau CN f_{i_2}(D_g,\sigma_g, m, \lambda, \theta)}{\tau CN f_{i_1}(D_g,\sigma_g, m, \lambda, \theta)}$$  \hspace{1cm} (2-7)

With regard to the probability distribution function, $f(D)$, it is found that particle size distribution conforms to a lognormal distribution, which is expressed as [2]:

$$f(D) = \frac{1}{\sqrt{2\pi}\sigma_g D} \exp \left[ -\frac{1}{2} \left( \frac{\log_{10} D - \log_{10} D_g}{\log_{10} \sigma_g} \right)^2 \right]$$  \hspace{1cm} (2-8)

Where $\sigma_g$ indicates the geometric standard deviation and $D_g$ denotes the geometric mean diameter.

Figure 2-2 shows the relationship between the polarization ratio and the geometric mean diameter in the single polarization ratio method. In the figure 2-2, the parallel axis is the geometric mean diameter of soot particles, vertical axis is the polarization ratio. It can be found that polarization ratio is as a function of geometric mean diameter. Herein, the solid curves are the theoretical polarization ratios which can be calculated based on Mie scattering theory when the complex index of soot particles is $m = 1.9 - 0.63i$ and the scattering angle is $\theta = 60^\circ$. The dotted straight line is the measured
polarization ratio which can be obtained by experiment. The intersection point between theoretical polarization ratio and measured polarization ratio is the geometric mean diameter. The parameters of complex refractive index and incident wavelength are known values. However, the geometric standard deviation is unknown parameters. Therefore, the geometric mean diameter cannot be determined.

\[ \rho = \frac{i_2}{i_1} = \frac{\tau_{\lambda} C_\lambda N f_{i_2}(D_g, \sigma_g, m, \lambda, \theta)}{\tau_{\lambda} C_\lambda N f_{i_1}(D_g, \sigma_g, m, \lambda, \theta)} \]  \hspace{1cm} (2-9)

In figure 2.3, there are multiple intersection points between the theoretical values and the measured values at each geometric standard deviation. Herein, purple lines are values while incident wavelength is 405 nm and blue lines are values while incident wavelength is 488 nm. It can be found the geometric mean diameter obtained by each wavelength is different. By comparing difference between two geometric mean

2.1.2 Previous study - Multi-wavelength polarization ratio method based on polarization ratio

In particle group measurement, single wavelength polarization ration cannot determine geometric mean diameters of soot particles. Therefore, two different wavelengths laser sources were applied. In multi-wavelength polarization ratio method, the polarization ratio is expressed as follows equation.

\[ \rho = \frac{i_2}{i_1} = \frac{\tau_{\lambda} C_\lambda N f_{i_2}(D_g, \sigma_g, m, \lambda, \theta)}{\tau_{\lambda} C_\lambda N f_{i_1}(D_g, \sigma_g, m, \lambda, \theta)} \]  \hspace{1cm} (2-9)

In figure 2.3, there are multiple intersection points between the theoretical values and the measured values at each geometric standard deviation. Herein, purple lines are values while incident wavelength is 405 nm and blue lines are values while incident wavelength is 488 nm. It can be found the geometric mean diameter obtained by each wavelength is different. By comparing difference between two geometric mean
diameters under different geometric standard deviation. The minimum difference combination will be selected. The corresponding geometric mean diameter and geometric mean particle size can be uniquely determined.

Figure 2.3  Schematic of multi-wavelength polarization ratio method.

Figure 2.4  Problem of multi-wavelength polarization ratio method.
By using previous multi-wavelength polarization ratio method, the soot particles which range from 20 nm to tens nm order can be measured. It can be found from figure 2.4, while $D_g = 15$ nm, the corresponding polarization ratio is 0.253, while $D_g = 30$ nm, the corresponding polarization ratio is 0.264, the variation of polarization ratio is about 4%, the value of variation of polarization ratio is very tiny. In multi-wavelength polarization ratio method, the determination of geometric mean diameter is based on the variation of polarization ratio. The tiny variation of polarization ratio is difficult to determine the geometric mean diameter. It also can be found that the curve which is the function between polarization ratio and geometric mean diameter does not fluctuate much while soot particle diameters around 10 nm to 20 nm. However, the primary particle diameters range from 10 nm to 30 nm. It is necessary to expand the lower measurement limit.

Through calculation, it can be found 5% $I_2$ varies resulting in 10% polarization ratio varies which was shown as black line in figure 2.4, which indicates that scattered light intensities are more sensitive to the geometric mean diameter than polarization ratio.

2.1.3 Present study - Multi-wavelength polarization ratio method based on scattered light intensities

In order to expand lower measurement limit. The scattered light intensities are used to instead of polarization ratio as a function of geometric mean diameter. Two pairs of equations describing the scattered light intensities can be obtained which are expressed as follows.

$$I_{1,\lambda} = \sum I_{1,\lambda} = \tau_{\lambda_1} C_{\lambda_1} n f_{\lambda_1}(D_g, \sigma_g, m, \lambda_1, \theta)$$

$$I_{2,\lambda} = \sum I_{2,\lambda} = \tau_{\lambda_1} C_{\lambda_1} n f_{\lambda_2}(D_g, \sigma_g, m, \lambda_1, \theta)$$

(2-10)

$$I_{1,\lambda_2} = \sum I_{1,\lambda_2} = \tau_{\lambda_2} C_{\lambda_2} n f_{\lambda_1}(D_g, \sigma_g, m, \lambda_2, \theta)$$

$$I_{2,\lambda_2} = \sum I_{2,\lambda_2} = \tau_{\lambda_2} C_{\lambda_2} n f_{\lambda_2}(D_g, \sigma_g, m, \lambda_2, \theta)$$

(2-11)

Here, $I_{1,\lambda}$, $I_{2,\lambda}$, $I_{1,\lambda_2}$ and $I_{2,\lambda_2}$ represent the theoretical scattered light intensities of MPR method applied to describe the perpendicular components and parallel components under two different wavelength lights, respectively.

The residuals between the theoretical values of the scattered light intensities and the measured values are expressed as follows.
Where $I_{M,1}$, $\lambda_1$, $I_{M,2}$, $\lambda_2$, $I_{M,1}$, $\lambda_2$ and $I_{M,2}$, $\lambda_2$ represent the measured values of the scattered light intensities. The values are obtained from experiment. The superscript $M$ represents the measured value. Eq. 2-10 and Eq. 2-11 are incorporated into Eq. 2-12 and 2-13 as follows.

\[
\begin{align*}
\epsilon_{I_1,\lambda_1} &= \left[ I_{M,1,\lambda_1} - I_{1,\lambda_1} \right]^2 \\
\epsilon_{I_2,\lambda_1} &= \left[ I_{M,2,\lambda_1} - I_{2,\lambda_1} \right]^2 \\
\epsilon_{I_1,\lambda_2} &= \left[ I_{M,1,\lambda_2} - I_{1,\lambda_2} \right]^2 \\
\epsilon_{I_2,\lambda_2} &= \left[ I_{M,2,\lambda_2} - I_{2,\lambda_2} \right]^2 
\end{align*}
\]

(2-12)

(2-13)

The sum of the total residuals $\epsilon$ is shown by the following equation.

\[
\epsilon = \epsilon_{I_1,\lambda_1} + \epsilon_{I_2,\lambda_1} + \epsilon_{I_1,\lambda_2} + \epsilon_{I_2,\lambda_2}
\]

(2-16)

The optical constants $C_{\lambda_1}$ and $C_{\lambda_2}$ can be determined using polystyrene standard particles with diameters of 22 nm. The unknown parameters include the particle number $n$, the geometrical mean diameter $D_g$, the geometric mean standard deviation $\sigma_g$, and the complex refractive index $m$. When $\epsilon$ reaches its minimum, the iterative calculation has converged. In the meantime, the corresponding unknown parameters can be determined.

Figure 2.5 shows the relationship between scattered light intensities and geometric mean diameter at the complex refractive index $m = 1.9 - 0.6i$. The vertical axis represents the intensity of scattered light and the horizontal axis is an indication of the geometric mean diameter. The curves mean the theoretical scattered light intensities which can be calculated based on Mie scattering theory. The straight lines mean the experiment data which can obtained by measuring. The solid lines mean $I_1$ polarized component which is perpendicular to the observation plane. The imaginary lines mean $I_1$ polarized component which is parallel to the observation plane. Blue lines are light intensities obtained from a laser having wavelength of 488 nm. Purple lines are light intensities obtained from a laser having wavelength of 405 nm. The intersection point between solid line and imaginary line in one color is the value obtained by MPR method. Two different wavelengths were used in MPR method. Therefore, two values
of geometric mean diameter can be obtained in one geometric standard deviation. The group which difference between two values is minimum will be selected, the corresponding geometric mean diameter and geometric standard deviation are the values obtained by MPR method.

![Graph showing scattered light intensity as a function of geometric mean diameter with two different wavelengths of incident light.](image)

**Figure 2.5** Scattered light intensity as a function of geometric mean diameter with two different wavelength of incident light.

### 2.1.4 Determination of unknown parameters

Two different lasers with wavelengths of 405 and 488 nm are used, since two different polarization components of scattered lights, perpendicular to and parallel to the polarization plane, are acquired by using of the polarization cameras. In total, four combinations of equations, namely Eqs. 2-14 and 2-15, are obtained. The measured values of the scattered light intensities are substituted into Eqs. 2-14 and 2-15. The theoretical values are calculated by substituting temporal values into the four unknown parameters. The residual between the measured and theoretical values changes according to the values for four unknown parameters. By using an iterative procedure, the values for four unknown parameters are changed so that the sum of the residuals given in Eq.2-16 is minimized, which corresponds to the proper combination of the four parameters.
Figure 2.6  Four layers of unknown parameters of theoretical scattered light intensities.

Figure 2.6 shows the four layers of unknown parameters of theoretical scattered light intensities. These consist of the particle number $n$, the geometric mean diameter $D_g$, the geometric standard deviation $\log \sigma_g$ and the complex refractive index $m$. The range of these values are determined under several constraints. The value of $\log \sigma_g$ is assumed to be $0.05 < \log \sigma_g < 0.25$ based on previous studies [3-5]. Regarding the complex refractive index, the range of the real part is assumed to be set from 1.5 to 2.0 and the sum of the real and imaginary parts is assumed to be kept at 2.5 based on a previous work of numerical calculations [3].

Figure 2.7 illustrates the isosurface of $\varepsilon$ at $z = 20$ mm. The values of $\varepsilon$ are demonstrated in a color scale. The horizontal axis is the geometric standard deviation $\log \sigma_g$ and the vertical axis is the complex reference index. The range of $\log \sigma_g$ and $m$ were introduced in last part which are based on previous study [3-5]. The condition of convergence of iterative calculation is to get the minimum value of $\varepsilon$. Through iterative calculation, it was found that while $m = 1.9 - 0.6i$ with the value of $\log \sigma_g$ is 0.20, the value of $\varepsilon$ is smaller than others. Therefore, to find a smaller $\varepsilon$, the combination which $m = 1.9 - 0.6i$ with $\log \sigma = 0.175$ and $\log \sigma_g = 0.225$ were also calculated based on iterative computation. The value of $\varepsilon$ is smaller while $\log \sigma = 0.225$. Herein, the value of $\log \sigma$ is given with two significant digits. Finally, while $m = 1.9 - 0.6i$ with $\log \sigma_g = 0.23$, the minimum value of $\varepsilon$ was obtained. The point in the circle indicates the
minimum value of $\varepsilon$. In the meanwhile, the corresponding values of $D_g$ and $n$ were also determined.

Figure 2.7 Sum of the squared residuals as a function of $m$ and $\log \sigma_g$ at $z = 20$ mm.

Figure 2.8 Sum of the squared residuals as a function of $m$ and $\log \sigma_g$ at $z = 25$ mm.
Figure 2.8 illustrates the isosurface of $\varepsilon$ at $z = 25$ mm. For the complex refractive index of $m = 1.6-0.9i$ with the geometric standard deviation $\log \sigma_g = 0.23$, the minimum value of $\varepsilon$ was obtained.

### 2.1.5 Determination of the optical constant

This section introduces the procedure to determine the optical constant $C_{405}$ in the butane laminar coflow diffusion flame. The optical constant is a coefficient that is used to correct the detection sensitivity of the cameras. In the experiment, the output intensities of each CCD camera are different. The difference is because of distance between camera and flame, aperture, quantum efficiency of CCD camera, transmissivity of interference filters. Therefore, the scattered light intensities should be corrected to reduce the effect of the experimental device. The value of the optical constant varies according with the laser wavelength.

First, it was considered the effect of the experimental device under the condition in which there is no absorption effect. Second, since the MPR method can be applied to measure primary soot particles and the diameter of primary soot particles is in the range 30 nm [1], the MPR method was used to estimate the optical diameter of polystyrene standard particles of 22 nm. Finally, the effect of particle composition was adjusted for soot particles. The differences in the optical constant between the two experimental conditions for the flame and for the standard particles in purified water corresponds to the absorption of soot particles. By using LEM, the transmissivities of the two wavelength lights through the flame can be obtained.

Figure 2.9 shows the quartz tube filled with purified water and polystyrene standard particles. Schematic of experiment setup for optical constant measurement is shown in Figure 2.10. The experimental conditions and experimental device used for the experiment using standard particles were almost identical to the MPR system used for soot particle measurement. Two lasers with the wavelengths 405 nm and 488 nm were used. The scattering angle was set at 60°. The main difference between the two experiments is that the burner is replaced by a quartz cell filled with purified water and polystyrene standard particles.
The equation used to determine the optical constant of soot particles is presented below.

\[
\frac{C_{405}}{C_{488}} = \frac{C_{p,405} \times T_{405}}{C_{p,488} \times T_{488}} \tag{2-17}
\]

Where \(C_{405}\) and \(C_{488}\) are the optical constants corresponding to the soot particles in the under wavelengths of incident light of 405 nm and 488 nm, respectively. \(C_{p,405}\) and \(C_{p,488}\) refer to the optical constants of the standard particles. \(T_{405}\) and \(T_{488}\) are the transmissivities of the two wavelength lights through the flame and can be obtained using the LEM system. The optical constants of standard particles \(C_{p,405}\) and \(C_{p,488}\) are defined as:

\[
\frac{C_{p,405}}{C_{p,488}} = \frac{I_{1,405}^M}{I_{1,488}^M} \times \frac{I_{1,488}^M}{I_{1,405}^M} \times \frac{I_{1,405}^M}{I_{1,488}^M} \tag{2-18}
\]

Herein, \(I_{1,405}^M\) and \(I_{1,488}^M\) are the measured scattered light intensities of standard particles by MPR system. \(I_{1,405}\) and \(I_{1,488}\) are the theoretical scattered light intensities of standard particles obtained by MPR method. Here, the value of \(C_{p,488}\) is set to 1. Accordingly, the value of \(C_{p,405}\) can be determined in Eq. 2-17. In Eq. 2-17, the value of \(C_{488}\) is also set to 1, and the value of \(C_{p,405}\) has been obtained from Eq. 2-18. As a result, \(C_{405}\) can be determined. The calculated optical constant \(C_{405}\) was used for all
particle size and particle number density conditions.

Aimed at determining the optical constant $C_\lambda$, standard particles (MORITEX, 3020A) were utilized. Standard particle properties are presented in Table 2-1. The values of parameters in the optical constant experiment are shown in Table 2-2.

Table 2-1  Standard particles for determining optical constant.

<table>
<thead>
<tr>
<th>Material</th>
<th>Polystyrene (Transparent)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Refractive index $m$</td>
<td>1.59</td>
</tr>
<tr>
<td>(Relative refractive index)</td>
<td>(1.20 in water)</td>
</tr>
<tr>
<td>Geometry</td>
<td>Spherical</td>
</tr>
<tr>
<td>Arithmetic mean diameter $D_{10}$</td>
<td>22 nm</td>
</tr>
<tr>
<td>Number density</td>
<td>$2.75 \times 10^{13}$ 1/cm$^3$</td>
</tr>
</tbody>
</table>
Table 2-2  Specifications of determining optical constant.

<table>
<thead>
<tr>
<th></th>
<th>$z = 20$ mm</th>
<th>$z = 25$ mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_{p, 405}$</td>
<td>1.2197</td>
<td>1.2197</td>
</tr>
<tr>
<td>$C_{p, 488}$</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$C_{488}$</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$T_{405}$</td>
<td>0.8111</td>
<td>0.8789</td>
</tr>
<tr>
<td>$T_{488}$</td>
<td>0.8284</td>
<td>0.9000</td>
</tr>
<tr>
<td>$C_{405}$</td>
<td>1.1942</td>
<td>1.1923</td>
</tr>
</tbody>
</table>

### 2.1.6 Calculation of the soot volume fraction in the radial direction

In this research, the Abel transform was used to obtain the soot volume fraction in the radial direction of the flame. It was assumed that the cross section of the flame is a perfect circle. Figure 2.11 is a horizontal cross-sectional view of a certain height of the axisymmetric coflow diffusion flame. The soot volume fraction in each region was calculated. It was assumed that the soot volume fraction distribution in each circle is homogenous and constant.

![Figure 2.11  Laser measurement of axisymmetric coflow Flame.](image)
The determination method of soot volume fraction will be introduced as follows:

$$\Delta f_k = \frac{1}{\sqrt{2\pi \alpha_k \ln \sigma_g}} \exp \left[ -\frac{1}{2} \left( \frac{\ln \alpha_k - \ln \sigma_g}{\ln \sigma_g} \right)^2 \right]$$

(2-19)

Here, $\Delta f_k$ is frequency of $k$-th particle size parameter $\alpha_k$, $\sigma_g$ is standard deviation, $\alpha_g$ is geometric mean particle size parameter. The particle size was calculated from $\alpha_k = 0$ to $\alpha_k = 10$ with a step size of 0.01. The number of particles was calculated for the radial direction. The soot volume fraction (SVF) at each radius was calculated by using geometric mean particle diameter, geometric standard deviation and number of particles.

2.2 Experimental set-up of the multi-wavelength polarization ratio method

2.2.1 Butane burner system

In this research, an axisymmetric, laminar, coflow, diffusion flame was used, as shown in figure 2.12. In MPR research, a certain length of exposure time on the order of 0.1 second is needed. It cannot be used in a turbulent flame. The premixed flame is a low-soot flame and widely used in PAHs research. The laminar diffusion flame is widely used for soot particles measurement. The measurement target is soot particles not the precursor for soot particles. Therefore, laminar diffusion flame is used in the present study.

![Direct image of burner and butane laminar diffusion flame.](image)  
*Figure 2.12 Direct image of burner and butane laminar diffusion flame.*
The burner consists of a fuel nozzle with an inner diameter of 5 mm and a co-annular air nozzle with a diameter of 50 mm. Hereafter, the fuel nozzle radius is denoted as $R$. The flame height is set to 30 mm. The $z$- and $r$-axes are set along the center axis and the lateral direction of the flame, respectively.

Figure 2.13 Particle size distribution measured by PAMS.

Measurements have been done at from heights above the fuel nozzle $z = 10$ mm to $z = 30$ mm. Figure 2.13 is the particle size distribution from the PAMS measurement. While $z = 30$ mm, soot particle diameter has a peak at around 100 nm, which means that most particles are secondary particles [6] and were introduced in Chapter 1. This
measurement position is the tip of the flame. In this location, the flame shakes frequently, it is difficult to obtain a good reproducibility. While at \( z = 10 \) mm, it is very difficult to obtain scattered light intensities of soot particles because of the weak scattered light intensities of small particle sizes. While \( z = 20 \) mm, soot particle diameter has a peak at around 40 nm which means that most particles are primary particles. The author wants to know the process which primary particles grow up into secondary particles. Therefore, the measurement at \( z = 20 \) and 25 mm are selected in the present study.

The entire burner system is positioned on a stage and can be scanned in the vertical and horizontal directions. The test fuel is butane stored in a fuel bottle in liquid phase. In order to keep the vapor pressure of fuel constant, the fuel bottle is settled in a constant-temperature water bath set at 5°C.

The gas phase fuel is supplied through a pipeline. The fuel flow rate is measured using the assistance of a thermal flow meter (KOFLOC, Model 3760). A precision needle valve was employed to adjust the mass flown rate. For the purpose of keeping the fuel density constant, the fuel temperature was set at 35°C using a silicon cord heater and a temperature controller. Moreover, the air flow rate was measured using a thermal flow meter (KOFLOC, Model 3105).

The experimental conditions for the butane laminar coflow diffusion flame are presented in Table 2-3.

| Table 2-3  Experimental conditions for butane laminar coflow diffusion flame. |
|------------|-----------------|
| Fuel nozzle diameter | 5.0 mm |
| Air nozzle diameter | 50 mm |
| Butane volumetric flow rate | \( 6.8 \times 10^{-7} \) m\(^3\)/s |
| Butane flow velocity | 0.037 m/s |
| Air volumetric flow rate | \( 2 \times 10^{-4} \) m\(^3\)/s |
| Air flow velocity | 0.11 m/s |
| Fuel temperature | 35°C |
| Air temperature | 35°C |
Figure 2.14  Schematic diagram of the burner profile.

Figure 2.14 shows a schematic diagram of the burner, which is placed inside some layers of cellular materials. Under the action of cellular materials, evenly from above the burner, the air flows uniformly from all directions around the fuel nozzle to ensure the flame velocity, in order to make flame stable.

### 2.2.2 Optical set-up

A schematic diagram of the optical setup is shown in figure 2.15. Two diode lasers with wavelengths of 405 nm and 488 nm are used. The two lasers are emitted simultaneously on the same axis. Two polarization CCD cameras (4D-Technology, Polar Cam 7001-00161) are set symmetrically at 60° with respect to the laser axis. The interference filters centered at 405 nm (VPF-50C-03-25-40500) with 3 nm HWHM and 488 nm (VPF-50C-01-45-48800) with 1 nm HWHM are set in front of the two cameras, 405 nm corresponding to the camera 1 and 488 nm to the camera 2. The wavelength of OH, whose peak is at around 308 nm [32], has no effect on our result. The wavelength of CH, whose peak is at about 430 nm [32], the effect can be neglected too. A polarizer was used to make the incident light linearly polarized. The orientation of the polarization is set at 45° with respect to the observation plane. By setting the orientation at 45°, the incident light intensities of perpendicular and parallel
components become the same. The technical data of the filters are indicated in figure 2.16 and figure 2.17. A polarizer was applied to make the incident light linearly polarized. The orientation of the polarization was set at 45° relative to the observation plane. By setting the orientation at 45°, the incident light intensities of perpendicular and parallel components are made identical.

Figure 2.15  Schematic of optical setup.
Figure 2.16  Filter of wavelength of 405 nm.

Figure 2.17  Filter of wavelength of 488 nm.
A multi-wavelength continuous-wave diode laser system (Japan Laser, SP0206) was applied in the experiments. The parameters of diode laser are shown in table 2-4 [7].

<table>
<thead>
<tr>
<th>Model</th>
<th>JL-SP0206</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>405 nm</td>
</tr>
<tr>
<td></td>
<td>488 nm</td>
</tr>
<tr>
<td></td>
<td>638 nm</td>
</tr>
<tr>
<td>Power</td>
<td>300 mW (@ 405 nm)</td>
</tr>
<tr>
<td></td>
<td>200 mW (@ 488 nm)</td>
</tr>
<tr>
<td></td>
<td>150 mW (@ 638 nm)</td>
</tr>
<tr>
<td>Extinction ratio</td>
<td>100:1</td>
</tr>
<tr>
<td>Laser beam diameter</td>
<td>1.2 mm</td>
</tr>
<tr>
<td>Frequency</td>
<td>47~63 Hz</td>
</tr>
<tr>
<td>Size of laser head (D× W×H)</td>
<td>293 mm×220 mm×64 mm</td>
</tr>
</tbody>
</table>

According to the technical specifications, the Glan Laser polarizer is capable of providing an enhanced laser damage threshold for high power lasers and high energy laser pulses. The transmission loss is reduced to the minimal, and a high extinction ratio below $5 \times 10^{-5}$ can be obtained. The calcite type which can be used in the visible to the infrared region is available, as is $\alpha$-BBO crystal type that is usable in the ultraviolet region.

A GL10-A Polarizer and SM1PM10 Mount were mounted on a SPH-50-ARS Rotation Mount. They were used to adjust the direction of polarization of waveplates and polarizer optics. The mount has 360 degrees of smooth rotation.

Two polarization CCD cameras (4D-Technology, Polar Cam 7001-00161) were employed in this research, with a 648 × 488 resolution and a 12-bit depth. The cameras were installed symmetrically with respect to the optical axis, the two cameras were deployed to perform simultaneous measurement under two wavelengths. The lens used
was a Nikon AF Nikon 50 mm \( f/1.8D \), which was set at a position where the optical axis of the lens had a scattering angle \( \theta = 60 \) deg. With the use of a close-up ring, the focus was adjusted at a distance of approximately 240 mm from the principal point of the lens to the fuel nozzle. Taking into consideration the possibility of two-dimensional measurement, it is desirable to set the scattering angle \( \theta = 90 \) deg. However, the \( I_2 \) component of the soot scattered light intensity at the scattering angle \( \theta = 90 \) deg is about 1/1000 to 1/10000 compared to the \( I_1 \) component, it is not possible to ensure a sufficient S / N ratio. In this case, the scattering angle was determined by making references to the research report of Nishida et al [8].

![Figure 2.18 Images of Polarization CCD cameras.](image)

Figure 2.18 and figure 2.19 show the polarization camera and its micro polarizers attached to each pixel. In this polarization camera, a polarization element is attached to the surface of the CCD sensor. The polarizer has four angles, which are attached to each pixel. Therefore, four polarization components can be obtained from a single photo. For measurement, the micro polarizers for 0° and 90° were used. Depending on the size distribution pattern, the scattered light from soot particles contains the
perpendicular and parallel components of polarization. The polarizers on the CCD (4D-Technology, Polar Cam 7001-00161) elements show four different orientations, with two polarization components obtainable from a single image.

Figure 2.19  Polarization camera and micro polarizers attached on each pixel.

Figure 2.20  Scattered light images for two polarized components.
Figure 2.20 presents the scattered light images showing two different polarization components taken at $z = 20$ mm. The wavelength of the incident light is 405 nm. The left image is the perpendicular component $I_1$ and the right one is the parallel component $I_2$. The scattered light intensities for these components are calculated from the images shown in figure 2.21.

### 2.2.3 Temperature measurement

Figure 2.22 shows a photograph of a thermocouple (Class R, manufactured by SAKAGUCHI E.H VOC CORP. with wire diameter 0.1 mm) located 20 mm above the butane burner. The thermocouple measuring sphere has a diameter of 0.334 mm.

When a thermocouple is inserted in a direction perpendicular to the flame flow field, the wire becomes hotter than the measuring sphere, due to the wire passing through the flame surface. Since the heat capacity of the strand is lower than that of the measuring sphere, the temperature shows an upward trend. As a result, heat is transmitted from the strand to the measuring sphere. In order to prevent the heat conduction from the wire, the thermocouple was installed parallel to the flow field. Therefore, the temperature difference between the wire and the measuring sphere was reduced.
Before the soot particle measurement experiment, two other experiments should be done. The first one, is performed to create the sensitivity curves and correct the scattered angle. The sensitivity curves are used to correct the scattered light intensities about the exposure time of the camera. The detailed creating sensitivity curves and scattering angle correction method will be introduced in Chapter 5. The second experiment is performed to obtain the optical constant, which was introduced.

The experimental procedure to obtain the sensitivity curves is as follows: The laser lights from a diode laser pass through a Gran laser prism (GLPB2-10-26SN-3 / 7) which is installed in front of the diode laser. Then, the linearly polarized lights which are inclined at 45° compared to the horizontal plane can be obtained. The laser generated by the semiconductor laser becomes a linearly polarized light through the polarizer, and the angle between the polarizing plane and the horizontal plane is 60°. In this case, the polarization plane of the laser light can be regarded as a combination of parallel component and perpendicular component to the to the observation plane.
with a ratio of 1:1.

The laser lights pass through a lens group. The lens group consist of a planoconcave lens (SLB-30-80NM) and a planoconvex lens (SLB-30-90PM). The focal length $f = 80$ mm of the planoconcave lens is used to expand the laser beam diameter. After that, the planoconvex lens with the focal length $f = 80$ mm is used to reduce the expanded laser beam. The distance between the two lenses is 47 mm. A 0.4 mm diameter laser beam can be obtained by using this lens group. The laser was set to intersect the flame at heights of $z = 20$ mm and $z = 25$ mm.

Then the installation accuracy of the cameras is adjusted. The installation accuracy here refers to the camera height, the tilted angle of camera to the horizontal plane and inclination in the roll direction. For this reason, cameras were installed on a newly designed stage which can adjust the camera height, tilted angle and roll direction. The scattering angle is corrected next. The calibration plate is placed on the fuel nozzle and take pictures with CCD cameras. Through the analysis of the calibration plate image, the real scattering angle can be obtained and then correcting the real scattering angle using a computational program.

Then, the two CCD cameras are used to take pictures of the flame through which the laser is passing. To reduce the error, 30 images are taken. Additionally, an interference filter with (VPF-50C-03-25-40500) with FWHM of 3 nm and an interference filter (VPF-50C-01-45-48800) with FWHM of 1 nm are installed in front of the CCD cameras respectively, in order to avoid the effect of the addition of the wide-band bright flame light on the scattered light. After a set of taking photos, the interference filters are exchanged.

The last part is the analysis of data. First, by using the U7483-01 Intensities distribution measured software pictures of the flame are analyzed. Then, the information of scattered light intensities can be obtained. Second, the measurement area is determined. For the measurement area, the longitudinal direction of the image is set as $z$ coordinate, and the transverse direction is set as $r$ coordinate. Each $r$ coordinate represents one pixel, of the pixel which is has the maximum scattered light intensity will be found. Set 50% of the maximum value as the threshold. The scattering intensity above is assumed to be a normal distribution and fitted; taking the central value as the measured value. Third, it is assumed that the scattered light intensity in the flame changes continuously. A sixth order function is used to fit the distribution of
scattered light intensities. At last, the unknown parameters which are particle number, geometric mean diameter, geometric standard deviation and complex refractive and index can be determined through iterative computation.

2.3.1 Scattered light images of soot particles

Two unaveraged scattered images at two different flame heights are given as figure 2.23 and figure 2.24. A single scattered image has a great fluctuation in the distribution of the location of the soot particle due to the flame is not quiescent state. For a diffusion flame, a single scattered image cannot be used to compare the generation characteristics of soot particles under working conditions. Generally, more images are required to be averaged before comparison. In this research, 30 images of the flame were taken at $z = 20$ mm and $z = 25$ mm.

![Figure 2.23 Scattered light of soot particles at $z = 25$ mm](image)
Figure 2.24  Scattered light of soot particles at $z = 25$ mm.

Figure 2.25  Scattered light intensities at $z = 20$mm.
Figure 2.26 Scattered light intensities at $z = 25$ mm.

Figure 2.25 and figure 2.26 show the scattered light intensities which are obtained by analyzing the images at $z = 20$ mm and the scattered light image at $z = 25$ mm. Laser light is emitted from the left of the image. At $z = 20$ mm, the scattered light intensity is minimum near the flame center axis and increases towards the edge. The change in the scattered light intensity is continuous and convex downward. The intensity of the scattered light is maximum near the flame surface, and decreases sharply outside. At $z = 25$ mm, the scattered light intensity increases toward the flame center. The brightness change is convex upward. At $z = 25$ mm, the scattered light intensity of flame center is about 1/4 compared to $z = 20$ mm. The scattered light intensity is proportional to the number of soot particles and the projected area. Therefore, it is considered that the place where the scattered light intensity is large has a large soot particle size or a large number of particles.

### 2.3.2 Function of fitting scattered light intensity

It is assumed that the intensity of the scattered light in the flame changes on a continuous basis; therefore, function fitting was conducted. The function used was a 6th order function. For $z = 20$ mm, fitting was carried out in the range of 8.6-12.1 mm for $z = 25$ mm, 8.86-11.88 mm for purple which wavelength is 405 nm and 8.8-11.9
mm for blue which wavelength is 488 nm, as shown in figure 2.27 and figure 2.28. The order of the function and the fitting range used were those where the value of the correlation function R representing the variation approached to 1. When the fitting range was extended to the flame front, the function failed to follow the experimental value, suggesting that the change near the flame front is not continuous.

Figure 2.27 The scattered light intensities at $z = 20$ mm.

Figure 2.28 The scattered light intensities at $z = 25$ mm.
REFERENCES

7. www.japanlaser.co.jp.
Chapter 3  Principles and experimental set-ups of validation methods

3.1 Light extinction method for soot particle measurement

To validate the MPR method, the soot volume fraction obtained using the MPR method was compared with the data obtained using the LEM [1-4] at \( z = 20 \) mm and \( z = 25 \) mm. Moreover, the Abel transform [5-6] was used to calculate the soot volume fraction used in the LEM computation. The schematic of LEM system is shown in figure 3.1. The diode laser source with a 638 nm wavelength is applied in the LEM system.

\[
\frac{I}{I_0} = \exp(-Q_{ext} \pi D^2 NL/4)
\]

Figure 3.1 The schematic of light extinction method.

The principle of the light extinction method states that the time intensity of a beam of light passing through the carbon cloud will attenuate, and the relationship between the emitted light intensity and the incident light intensity can be expressed as [2]:

Here, \( I_0 \) is intensity of incident light, \( I / I_0 = \tau \) is the transmitted light, \( L \) is the optical path length of the light through the particle swarm, \( \tau \) is the transmissivity, \( Q_{ext} \) is the
extinction coefficient of single particle based on incident wavelength, particle diameter, complex refractive index, \( D \) is the particle diameter and \( N \) is the particle number density. In the coflow diffusion flame, the soot particle size ranges from 20 nm to 50 nm which is smaller than the incident wavelength \([2]\). The particle parameter \( \alpha \) is 0.1 ~ 0.25 which is less than 1\([2]\).

When \( \alpha < 1 \), the Rayleigh equation can be used, so scattering occurs when electromagnetic waves are incident on soot particles. Attenuation due to scattering is about 0.2% of absorption \([2]\). Therefore, the parameters \( Q_{\text{ext}} = Q_{\text{abc}} \) in soot research \([2]\).

Since \( \alpha \ll 1 \) is not strictly true for soot particles, \( Q_{\text{abc}} \) is examined using Mie's theoretical solution. The first item and the second item in Mie's solution are as follows:

\[
Q_{\text{abs}} = 12 \frac{\pi D}{\lambda} \left[ \frac{2nk}{(n^2 + k^2)^2 + 4(1 + (n^2 - k^2))} \right]
+ \left( \frac{\pi D}{\lambda} \right)^3 2nk \left[ \frac{2}{15} - \frac{5}{12((n^2+k^2)^2+2.25+3(n^2-k^2))} \right]
\]  (3-2)

The first term in the right is consistent with Rayleigh's solution. From 400 to 700 nm in the visible wavelength range to the long infrared range, if \( D \) is 50 nm, Rayleigh's equation gives a good approximation to the exact solution. In the case where the Rayleigh equation can be used, the transmittance and the soot particle volume fraction will be obtained. Substituting the soot volume fraction \( SVF = \pi D^3 N / 6 \) into Eq.3-1, it can be obtained the following equation:

\[
SVF = -\frac{2}{3} \frac{D}{L Q_{\text{abs}}} \ln \tau_{\lambda}
\]  (3-3)

By substituting Eq. 3-3 in the first term of Eq.3-2, the parameter \( D \) can be eliminated, and the following equation can be obtained:

\[
SVF = -\frac{2}{3} \frac{\lambda \rho_s}{\pi L f(n,k)} \ln \tau_{\lambda}
\]  (3-4)

The equation about \( f(n,k) \) will be introduce as follows:

\[
f(n,k) = \frac{24nk}{(n^2+k^2)^2 + 4(1+(n^2-k^2))}
\]  (3-5)

Where, \( \rho_s \) is the soot particle densities (1.8g/cc) \([2]\), the transmissivity \( \tau \) and the length of the optical path could be measured while the incident wavelength is \( \lambda \) known value. In this measurement, Abel transform was used to obtain the soot volume fraction in the radial direction in the flame.
3.2 Portable Aerosol Mobility Spectrometer (PAMS) for soot particle measurement

For the purpose of validating the MPR method, a PAMS [7-9] was employed for the measurement of the particle size distribution followed by a comparison with the results of the MPR method. The MPR method was used to measure primary soot particles in the flame. Consequently, it is necessary to ensure the measured particles are primary particles. The PAMS system was also used to distinguish primary particles and secondary particles. By using the PAMS system, the result showed that at the flame height $z = 20$ mm, most of the soot particle diameters range from 24-55 nm. While flame height $z = 25$ mm, most of the soot particle diameters range from 42 -177 nm. Therefore, it is considered that while at $z = 20$ mm, most of soot particles are primary soot particles, at $z = 25$ mm, most of soot particles are aggregates.

The schematic of PAMS is shown in figure 3.2. Figure 3.3 shows the image of PAMS device. The principle of PAMS was introduced in chapter 1. The specifications of PAMS used in this research are shown in table 3-1.

![Flow Diagram](image)

Figure 3.2 Schematic of PAMS used in the present research [9].

52
Figure 3.3  The image of PAMS.

| Measurement mode          | 1. Single Size mode  
                          | 2. Scanning mode       |
|---------------------------|----------------------|
| Diameter range            | 10~863nm              
                          | High resolution: 10~433nm |
                          | Wide: 14.5~863nm       |
| Channel                   | High resolution: 27ch 
                          | Wide: 14ch              |
| Scanning time             | High resolution: 27ch 
                          | Wide: 14ch              |
| Maximum particle number density | 100,000 particle/cc |
| Flow rate                 | Inlet: 0.7LPM         
                          | Sample: 0.05LPM        |
| Size                      | W230×H230×D150mm     
                          | 4.5kg (without battery) |
A schematic of the soot particle sampling system is presented in figure 3.4. There are 4 measurement positions which flame heights are \( z = 10, 15, 20, 25, 30 \) mm for PAMS. The data of \( z = 20, 25 \) were compared with the MPR method. The nitrogen flow rate used for diluting the aerosol was measured by a thermal flowmeter on the upstream side of the sampling probe and a thermal flow meter on the ejector side. The nitrogen flow rate is \( 1.7 \times 10^{-5} \text{ m}^3/\text{s} \) from the upstream side and \( 2.3 \times 10^{-4} \text{ m}^3/\text{s} \) for the ejector. In addition, the dilution ratio in the flow path system at the flow rate above is \( 5.6 \times 10^3 \). The dilution of nitrogen to the sampling probe is carried out to freeze the soot particle reaction. The experimental conditions of the experiment using PAMS are shown in Table 3-2.

**Table 3-2  Experimental conditions of PAMS**

<table>
<thead>
<tr>
<th>Measurement position [mm]</th>
<th>10, 15, 20, 25, 30</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass flow rate of ( \text{N}_2 ) at Ejector [L/min]</td>
<td>14</td>
</tr>
<tr>
<td>Mass flow rate of ( \text{N}_2 ) at Probe [L/min]</td>
<td>1.0</td>
</tr>
<tr>
<td>Dilution ratio</td>
<td>( 5.6 \times 10^3 )</td>
</tr>
<tr>
<td>Measurement time [s]</td>
<td>145 (wait time: 10s, 5s×27ch)</td>
</tr>
</tbody>
</table>
3.3 Standard particles for validation

In the present research, the polystyrene standard particles were used two times. First time was used to obtain the optical constant with 22nm diameters. Second time, the polystyrene standard particles of 46 nm and 269 nm in purified water under five different number densities were used to validate the accuracy of MPR method. The test particles are made of polystyrene and are spherical and transparent. The refractive index is $n_p = 1.59$ [10]. The arithmetic mean particle size of the test particles and the standard deviation of the particle size are determined by the transmission electron microscope (TEM). Since the refractive index of water is $n_w = 1.33$, the relative refractive index of the test particles with respect to the medium is $m_p = 1.20$ [10].

![Figure 3.5 Dense and sparse limits in multi-wavelength polarization ratio method [10].](image)

Table 3-3 shows the characteristics of the test particles used for measurement. The number density was set to a value considered to be within the measurement range of the multi-wavelength polarization ratio method, as shown in figure 3.5 [10].

The preparation method of the test particles is introduced below. A stoppered flask was placed on an electronic balance and tared. Then, a small amount of polystyrene standard particles was added, and the mass was measured. After that, standard particles
were diluted with purified water and weighed. The prepared test polystyrene particles were transferred to a quartz cell with a syringe and sealed. The material of the sample tube is synthetic quartz. The outer diameter of the quartz cell is 5 mm and the height is 250 mm. It was confirmed that the quartz cells have a sufficient transmittance for all the lasers used in this research.

In this study, the camera is set at about 60 ° from the laser traveling direction. In a rectangular quartz cell, the polarization plane is not preserved when scattered light passes through the cell. Therefore, a cylindrical sample tube was adopted. In the case of a cylindrical type, the scattered light at the center of the circle is transmitted perpendicularly to the plane of the sample tube, so the polarization plane is preserved. The experimental set-up is the same as the experimental set-up used for the optical constant determination method shown in figure 2.6.

<table>
<thead>
<tr>
<th>Particle sample No.</th>
<th>Arithmetic mean diameter D&lt;sub&gt;10&lt;/sub&gt;</th>
<th>Number density</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>46 nm</td>
<td>3.08 × 10&lt;sup&gt;11&lt;/sup&gt; 1/ cm&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
<tr>
<td>2</td>
<td>46 nm</td>
<td>3.02 × 10&lt;sup&gt;12&lt;/sup&gt; 1/ cm&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
<tr>
<td>3</td>
<td>269 nm</td>
<td>1.82 × 10&lt;sup&gt;6&lt;/sup&gt; 1/ cm&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
<tr>
<td>4</td>
<td>269 nm</td>
<td>2.57 × 10&lt;sup&gt;8&lt;/sup&gt; 1/ cm&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
<tr>
<td>5</td>
<td>269 nm</td>
<td>1.96 × 10&lt;sup&gt;9&lt;/sup&gt; 1/ cm&lt;sup&gt;3&lt;/sup&gt;</td>
</tr>
</tbody>
</table>
REFERENCES

Chapter 4  Soot particle size distribution and comparison results with validation methods

4.1 The geometric mean diameter and temperature distribution

The geometric mean diameter $D_g$ and temperature distribution in the radial direction at $z = 20$ mm and $z = 25$ mm are shown in figure 4.1 and figure 4.2, respectively. The figures present the right half of the butane laminar coflow diffusion flame. When $z = 20$ mm, the complex refractive index $m = 1.9 - 0.6i$ and $\log \sigma_g = 0.23$ settings are utilized. When $z = 25$ mm, the complex refractive index $m = 1.6 - 0.9i$ and $\log \sigma_g = 0.23$ settings are utilized.

![Figure 4.1](image)

Figure 4.1  Radial distributions of the geometric mean diameter at $z = 20$ mm and $z = 25$ mm.

At a flame height $z = 20$ mm, the geometric mean diameter is around 10 nm near the flame centerline. The range of geometric mean diameter varies from 10 nm to 13 nm.
It can be found that the diameter of soot particles gradually increases from the flame center to the flame edge. This trend matches the trend of the temperature distribution at the same flame height. It is considered that in the radial direction, due to the sufficient fuel, the continuous increase of the flame temperature and the weak oxidation of the flame from the center to the edge occur. The growth reaction on the surface of the soot particles dominates. This results in a gradual increase of the soot particle size in the radial direction.

![Figure 4.2](image-url)

**Figure 4.2** $O_2$ concentration distribution in flames on radial axis [1].

At a flame height of $z = 25$ mm, the geometric mean diameter is 19 nm near the flame centerline. The range of geometric mean diameter goes from 17 nm to 19 nm. With the increase of the flame height, the flame temperature keeps increasing, which promotes the growth reaction on the surface of soot particles. A comparison of the results for $z = 20$ mm and for $z = 25$ mm shows that the geometric mean diameter increases as moving downstream of the flame. The variation of geometric mean diameters near the flame center is more obvious than near the flame sheet. This is considered to be the effect of oxidation concentration. Figure 4.2 shows the radial distribution of oxygen concentration in the flame at $z = 20$ mm which is from Zama et
where the oxygen concentration in the flame peaks on the central axis of the flame. The oxygen concentration in the radial direction decreased the oxygen concentration in the air as it moved toward the center of the flame, and was reduced to the minimum near the boundary between the air and the flame. In the center of the flame, in addition to oxidization, has a very active growth.

Figure 4.3 shows the temperature distribution at $z = 20$ mm and $z = 25$ mm. The vertical axis is the temperature, and the horizontal axis is the dimensionless radial distance $r/R$. The temperature rises from the flame center to the flame edge and peaks at the flame edge. Near the center of the flame, the temperatures at $z = 20$ mm and $z = 25$ mm are about 1429 K and 1544 K. With the increase of the flame height, the temperature in the center of the flame increases, which agree with Saito et al [2]. The maximum radial temperature continuously contracts to the axis of the flame center. This may be because the distance between the flame edge and the flame center became shorter as reducing.

![Figure 4.3 Temperature distribution at $z = 20$ mm and $z = 25$ mm.](image)
4.2 Particle number distribution

Particle number distributions in the radial direction at $z = 20$ mm and $z = 25$ mm are shown in figure 4.4. The vertical axis is particle and the horizontal axis is the dimensionless radial distance $r / R$. At $z = 20$ mm, the range of particle number goes from $3.1 \times 10^5$ to $5.9 \times 10^5$. The peak value appears near the flame center and the minimum appears the flame edge. In figure 4.3, it can be seen that near the flame center, for a normalized radial distance lower than 0.3, the particle number did not vary too much. As the radial distance increases, the particle number decreases gradually. It is considered that the aggregation of soot particles gradually increases in the radial direction. At $z = 25$ mm, the range of the particle number varies from $3.6 \times 10^4$ to $5.0 \times 10^4$.

A comparison of the results for $z = 20$ mm and for $z = 25$ mm shows that the particle number decreases moving downstream of the flame. The particle number at $z =$25 mm is about 1/10 of the values at $z =$20 mm. This finding agrees with the results from Zama et al [1]. The reason is considered to be the coagulation of soot particles. The surface growth reaction of the soot particles plays a leading role within the zone from $z = 20$ mm to $z = 25$ mm.

![Particle number distribution](image)

Figure 4.4 Particle number distribution of radial direction at $z = 20$ mm an $z = 25$ mm.
4.3 Soot volume fraction distribution and comparison with LEM

To validate the MPR method, the soot volume fraction obtained by the MPR method is compared with the data obtained using the LEM [3-5] at $z = 20$ mm and $z = 25$ mm. Moreover, the Abel transform [6-7] was used to calculate the soot volume fraction used in the LEM computation.

![Comparison of soot volume fractions (SVF) measured by MPR method and LEM.](image)

Figure 4.5 Comparison of soot volume fractions (SVF) measured by MPR method and LEM.
The diode laser source with a 638 nm wavelength is applied in the LEM system. The burner system was introduced in the chapter 3. The light intensities before and after through the flame can be obtained by using a laser power meter.

Figure 4.5 shows the comparison of the soot volume fractions measured by the two methods at $z = 20$ mm and $z = 25$ mm. The horizontal axis is the flame height and the vertical axis is the normalized soot volume fraction. While $z = 20$ mm, the soot volume fraction measured by MPR is about $0.4 \times 10^{-8}$, while $z = 25$ mm, the soot volume fraction measured by MPR is around $1.1 \times 10^{-8}$. Moving upwards from $z = 20$ mm to $z = 25$ mm, the overall soot volume fractions decrease, which suggests that the oxidation is increasingly prominent. This soot volume fraction trend is the same as results found by Smooke et al [8]. The soot oxidation was found to be OH dominated, both on the centerline and on the wings [9]. Figure 4.6 from Hayashida et al [10] shows the axial distribution of OH. In the bottom graph of figure 4.5, it can be found, OH concentration goes up as moving downstream of flame. Especially, while $z = 20 \sim 30$ mm, the increase of OH concentration is very significant. The OH concentration variation is as the same as soot volume fraction variation.

---

Figure 4.6 Variations of $d_p$ and $L_\alpha$ (top); and axial distributions of PAHs, soot, OH and temperature (bottom) [10].
In the radial direction, even though the value of the soot volume fraction continues growing near the flame centerline, the value of the soot volume fraction decreases at the radially outward location. At $z = 20$ mm, the soot volume fractions obtained by the two methods exhibit similar distribution trends and values. At $z = 25$ mm, a significant difference between the two methods can be found. The reason is considered that the value of soot volume fraction obtained by MPR method is a relative value. MPR measurement is based on scattered light intensities. Therefore, the absolute value of SVF measured by MPR cannot be obtained. However, SVF measured by LEM is an absolute value. In order to compare SVF between two methods, SVF measured by MPR method was adjusted. A coefficient was used and this coefficient determined based on data measured at $z = 20$ mm. Therefore, the SVF measured by two methods at $z = 20$ mm show a good consistence.

Figure 4.7  The comparison of SVF of MPR, LEM and LII [1] method.

Figure 4.7 shows the MPR, LEM and LII [1] measured values of the normalized soot volume fraction as a function of radius. The horizontal axis is the flame height and the vertical axis is the normalized soot volume fraction. The soot volume fraction
measured by LEM was at $z = 10, 15, 20, 25$ mm and measured by MPR was at $z = 20$ mm and 25 mm. The flame height is 30 mm. Soot volume fraction obtained by LII method and MPR method are based on light intensities of soot particles. Therefore, the value of SVF is relative value. In order to compare SVF obtained by LII, LEM and MPR method, the SVF were normalized by the data measured at $z = 20$ mm. It can be found the SVF measured by three method showed a good correlation. As compared with LEM and LII, the SVF obtained from three methods show a good agreement, which indicates that the volume of particles can be measured correctly.

4.4 Comparison of particle size distribution between PMR and PAMS

Figure 4.8 shows the particle size distribution measured by the PAMS and the MPR method at $z = 20$ mm. The vertical axis is the normalized particle number, and the horizontal axis indicates the particle size. PAMS results contain a bimodal particle size distribution that peaks at 42 nm and 129 nm. The distribution on the side of the larger particles results from the soot coagulation inside of the collection tube.

![Figure 4.8](image.png)

Figure 4.8 Comparison of particle size distributions measured by the MPR method and the PAMS at $z = 20$ mm.
Figure 4.9 shows a comparison of particle size distributions measured by the MPR method and the PAMS at z = 25 mm. The diameter with maximum value particle number by using MPR method is about 23 nm and by using PAMS is about 73 nm.

The particle size measurement obtained from the MPR method is smaller than PAMS measurements. Data measured by PAMS may overestimate than real particle size. While z = 20 mm, the author also has no idea why MPR data is smaller than PAMS data. It may be the measurement error. We are not sure that. While z = 25 mm, the underestimate reason is considered to be the effect of aggregation.

There is no perfect measurement method to measure real soot particle size, and the emphasis is on comparison between two methods at present. In that sense, in addition to improving the measurement accuracy, it is necessary to accurately determine the optical constant $C_\lambda$ that corrects the scattered light intensity. In chapter 3, it was discussed that the values of total sum residual from the determined geometric standard deviation and its nearby value have a small difference. The minimum value of $\varepsilon$ is $7.38 \times 10^{-3}$. While log $\sigma_g = 0.20$ and log $\sigma_g = 0.25$, the values of $\varepsilon$ are $7.76 \times 10^{-3}$ and $7.46 \times 10^{-3}$. The differences are 5.1% and 1.1% respectively. Therefore, the value of optical constant is very important to obtain accurate results.
The secondary particles which is aggerate has effect on MPR data according to the irregularity shape. The author estimated the effect of secondary soot particles on the measured diameter based on professor Takahashi [11] and professor Sorensen [12].

The angular distribution of light scattering by irregularly shaped single particles is non-symmetrical with respect to the incident light direction. As a whole, the light scattering distribution shows the same performance as spherical particles [11].

The agglomerated particle with a lot of single soot particle which has light absorption. Calculations are as follows.

\[
 q = \frac{4\pi}{\lambda} \sin \left( \frac{\theta}{2} \right)
\]  

(4-1)

Here, \( q^{-1} \) is light scattering vector, the value is around 0.013 [11,12], \( \lambda \) is the wavelength of incident light, \( \theta \) is the scattering angle. The equations of scattering cross section and absorption cross section of aggregate are as follows.

\[
 Q_{s,agg} = N_p^2 Q_{scat} \left(1 + \frac{2}{3D_f} q^2 R_g \right)^{-D_f/2} 
\]

(4-2)

Where, \( Q_{s,agg} \) is the scattering cross section of aggregates, \( N_p \) is particle numbers of constituent particle for aggregate, here \( N_p \) is assume 17. \( Q_{scat} \) is scattering cross section of single particle. \( D_f \) means Fractal dimension, here the value of \( D_f \) is 1.7 [11], \( R_g \) is turning radius and the value of \( R_g \) is 80 [12]. It can be found that the scattering cross section of aggregates is proportional to the square of the constituent particles number. Scattering cross section of aggregates increases means aggregates diameter increases. Therefore, the conclusion which aggerate has effect on single particle data can be obtained.

For the absorption cross section of aggregates, the equation is shown as follows.

\[
 Q_{a,agg} = N_p Q_{abs}
\]

(4-3)

Here, \( Q_{a,agg} \) means absorption cross section of aggregates, \( Q_{abs} \) is absorption cross section of single particle. It can be found \( Q_{a,agg} \) is proportional to the constituent particles number. For LEM, the SVF calculation of aggregates is similarly for single particle, there is no effect on SVF values obtained by LEM.

The equation about sing particle scattering cross section is as follows.

\[
 Q_{scat} = \frac{128\pi^6 a^6}{3} \frac{m^2-1}{m^2+2} 
\]

(4-4)

\[
 Q_{s, agg} = N_p^2 Q_{scat} \left(1 + \frac{2}{3D_f} q^2 R_g^2 \right)^{-D_f/2} 
\]

(4-5)
Where $Q_{scat}$ is a single particle cross section, $a$ is single particle radius and the value is assumed as 15 in the present study. $m$ is the relative refractive index which is $m = 1.9 - 0.6i$. Through calculation, it is assumed that an aggregate composed of 17 particles, typical values for secondary particles, with a diameter of 30 nm is judged as a single particle with a diameter of 88 nm.

### 4.5 Comparison of mean particle size between MPR and TEM

Figure 4.10 shows the comparison of results for the MPR and TEM methods. The horizontal axis indicates the mean diameter obtained by TEM and the vertical axis the value for the MPR method. Two kinds of polystyrene standard particles having nominal diameters of 46 and 269 nm, measured by TEM, are used to validate the accuracy of the MPR method. The polystyrene particles are suspended in pure water in a quartz cell. The number density of the polystyrene particles is varied as well.

![Figure 4.10 Comparison of mean particle sizes of MPR and TEM.](image)
For the smaller particle, nominal diameter of 46 nm, the MPR method overestimates the diameter when compared with TEM. The error is around 170 %. For the larger particle, nominal diameter of 269 nm, the error against the TEM becomes much smaller whereas, but it still overestimates the diameter. The error decreased to become around 20 to 40 %. For both standard particles, the MPR method overestimates the diameter and with the decrease in the diameter the error increases. This could be attributed to the leak of polarized light at polarizers on each pixel of the CCD sensor. The extinction ratio of the polarizer of this camera is on the order of 1 % and this makes the measured intensities of the two polarization components uniform, which corresponds to the overestimation in the particle diameter in this range. This problem can be solved by applying correction algorithm under development.
REFERENCES

Chapter 5  Correction methods for improving measurement accuracy

In the MPR method, one of the most important parameters is the scattered light intensity. Almost all the experimental computations are performed according to the scattered light intensity. The experimental data are very sensitive to the scattered light intensity. The scattered light intensities will be different under different scattering angles. Since the scattering angle affects the value of the \( I_2 \), the value of polarization ratio will be affected. Through calculation, a 10% change in \( I_2 \) can lead to a 4% variation in the polarization. About 1 deg scattering angle error affects the final polarization ratio as much as 7%. Therefore, obtaining a correct value of scattered light intensity is necessary.

5.1  Scattering angle correction

In this section, it will be introduced the correction of the scattering angle. The calibration plate (Edmond # 59-210) is placed on the fuel nozzle and some images will be taken with the CCD camera which was shown in figure 5.1. Then analyzing the taken images by using U7483-01, intensities distribution is measured using a software. The left and right edges of the scattering angle \( \theta \) are obtained from the calibration plate which can show the dot positions. Since the \( I_2 \) component of the scattered light intensity is proportional to \( (\cos \theta)^2 \), it is corrected by the following equation:

\[
I_{2 \text{corrected}} = I_2(\theta) \left( \frac{\cos 60 \text{deg.}}{\cos \theta} \right)^2
\]  

(5-1)

Herein, \( I_{2 \text{corrected}} \) is the corrected intensities. \( I_2(\theta) \) is the original scattered light intensities. According to Eq. 5-1, the effective equation is as follows:

\[
I_{2 \text{corrected}} = I_2 \left\{ \frac{\left[ \cos \left( \frac{\pi}{180} \times 60 \right) \right]^2}{\left\{ \cos \left( \frac{\pi}{180} \times \left[ \theta_R - \left( \theta_R - \theta_L \right) \frac{324}{\text{actual coordinates}} \right] \right\}^2} \right\}
\]  

(5-2)
Through calculation, the angles were obtained. The corrected scattering angles of two cameras were shown in table 5-1.

![Diagram of scattering angle correction](image)

**Figure 5.1** Schematic of scattering angle correction.

<table>
<thead>
<tr>
<th>Correction parameters</th>
<th>Camera1</th>
<th>Camera2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Purple 405</td>
<td>Blue 488</td>
</tr>
<tr>
<td>$\theta_R$ = Scattering angle of right side</td>
<td>61.68</td>
<td>61.62</td>
</tr>
<tr>
<td>$\theta_L$ = Scattering angle of left side</td>
<td>57.43</td>
<td>57.26</td>
</tr>
<tr>
<td>$\theta$ = Scattering angle of center</td>
<td>59.56</td>
<td>59.44</td>
</tr>
</tbody>
</table>
5.2 Correction by sensitivity curve

5.2.1 Experiments to obtain sensitive curve correction

By using an integrating sphere, CCD cameras and two LED light sources with a wavelength of 406 nm and 498 nm to create uniform light, the sensitivity curves were constructed by transforming the light intensities to exposure time. In the present study, two lasers having 405 nm wavelength and 488 nm wavelength are used. It is very difficult to obtain the light intensities form the diode laser by using integrating sphere directly. Therefore, LED light source with 406 nm was replaced the diode laser having 405 nm wavelength and LED light source with 498 nm was replaced the diode laser having 488 nm wavelength. Different color lasers have different effect on the leakage of polarized light. The schematic diagram of the experimental set-up is shown in figure 5.2. Light from the integrating sphere was linearly polarized by a polarizer. The polarizing plate was turned at 10 ° intervals for shooting. When the image was taken with the integrating sphere rotated by 180 °, a change was observed in the luminance distribution at the edges of the screen. As the edges of the screen were not used for analysis, no problem arose. The exposure times were 8, 16, 32, 64, 128, 256, 512, 1024, 2048 μs.

![Schematic diagram of the experimental set-up](image)


Figure 5.2 The schematic diagram of extinction ratio of polarization.
Figure 5.3 is a single image taken using the integrating sphere, divided for each polarizer. The direction of the polarizer is 0, 45, 135, 90° from the upper left. In the experiment of the polarization ratio, $I_1$ component is obtained in the vertical direction at 0° and $I_2$ component in the horizontal direction at 90°. The color bar indicates the value above the average intensity of one pixel before division. The difference between the average intensity of each image is small. The intensity of lower right cell which is $I_2$ component is large. It suggests polarization leakage. Moreover, it is difficult to guarantee that $I_1$ component and $I_2$ component have the same leak rate.

However, if the polarization leakage matches, it means that the camera has not been able to separate the polarization component uniformly. The ideal condition is that the measured intensities of the two polarization components are uniform.

### 5.2.2 Obtaining the intensity calibration curve

Since the sensitivity of each pixel in CCD camera is also different, a calibration curve has been made in order to know each input to the camera sensors and get each sensitivity curve for the light intensity and exposure time.

Figure 5.4 shows the calibration curve fitted with a cubic function. The vertical axis
is the exposure time and the horizontal axis is the input light intensities. By changing the exposure time and identifying the corresponding input intensity, a sensitivity curve function was obtained by approximating each pixel with a cubic function. As a result, there is a possibility to mitigate the potential impact of such variation on sensitivity for each pixel.

By converting the intensity of scattered light into the exposure time using the sensitivity curve which were shown in figure 5.4, correction was conducted. The vertical axis is the exposure time and the horizontal axis is the input light intensities.

It can be found that it is almost a linear function and the contribution of the higher-order coefficients is insignificant because the value of the coefficient $a_2$ is $7.36 \times 10^{-6}$ and the value of the coefficient $a_3$ is $-5.74 \times 10^{-10}$. At the same time the value of the coefficient $a_1$ is 0.67.

![Figure 5.4 Correction curve at cubic function.](image)

$$y = \sum \alpha_n x^n$$

$a_0 = -9.00485609e-01$

$a_1 = 6.72168359e-01$

$a_2 = 7.56019503e-06$

$a_3 = -5.74322431e-10$

$2.79008887e-01$

$|r| = 9.99999908e-01$
Figure 5.5  Correction curve at cubic function at low exposure time.

It can be found there are some gaps at low exposure time. Therefore, the function at the low exposure time which is less than 400 μs was shown in figure 5.5. By enlarging the area near zero which is the low exposure time condition, it was found that the light intensities are underestimated on the order of 1%. 

\[ y = \sum \alpha_n x^n \]
\[ \alpha_0 = -9.00485609 \times 10^{-1} \]
\[ \alpha_1 = 6.72166359 \times 10^{-1} \]
\[ \alpha_2 = 7.36019503 \times 10^{-6} \]
\[ \alpha_3 = -5.74322431 \times 10^{-10} \]
\[ 2.79008887 \times 10^{-1} \]
\[ |r| = 9.99999906 \times 10^{-1} \]
Figure 5.6 Cubic function of spatial distribution of a1.
Figure 5.6 shows the cubic function of spatial distribution of the coefficient $a_1$. The distribution of the coefficient $a_1$ is shown in the top for the $I_1$ component and in the bottom for the $I_2$ component. Since the actual fitting is performed for each pixel, the result may differ from the fitting with the average light intensities of the whole image. The color bar is within the range of $0.672 \pm 10\%$, which is the average value of the coefficient $a_1$. All cells are generally within this range. Variation in polarizer transmittance (about $\pm 10\%$) is expressed by the coefficient $a_1$. 
Chapter 6  Conclusions

Developing MPR method has been demonstrated for this application and validated in the context of the measurement of particle size distribution, soot volume fraction and complex refractive index. Results confirmed the feasibility of MPR method for soot particle measurement in a coflow laminar diffusion flame. Main conclusions of this research are presented below:

1. The smallest limit of measurement is determined based on the measurement accuracy of the light intensities. Taking into account the measurement accuracy, the smallest limit lies on the order of 10 nm.

2. The largest limit of measurement is affected by the existence of aggregates. Under a certain assumption, the radius of gyration is underestimated by 45%. Thus, with the increasing in the fraction of aggregates, MPR tends to underestimate the particle size.

3. Based on the light scattering theory, the SVF measured by LEM is not affected by the existence of aggregates. The SVF measured by MPR showed a good agreement with that obtained by LEM.
I am very grateful to my supervisor, Pro. Mikiya ARAKI. Thanks for his priceless guidance. Thanks for his careful guidance of my graduation thesis, which greatly improved my understanding of research and taught me a lot of specific research skills. Professor ARAKI instructed me how to do research as a real researcher, especially the researching attitude.

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Thanks to Gunma University, who has given me the educational resources in Japan, enjoy a more advanced lifestyle, given me a rational look at the world, given me a delicate sense. I sincerely hope that Gunma University can become a world-class university in the 21st century.

At last I have to extend my thanks to my parents and my wife, Ning ZU, because they are always there for me.