Assessment of the factors influencing PN emission in a DISI engine under cold-start condition

by

Changhoon Oh

B.Sc., Korea Advanced Institute of Science and Technology (2008)
M.Sc., Korea Advanced Institute of Science and Technology (2010)

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Signature redacted

Department of Mechanical Engineering
Aug 19, 2016

Signature redacted

Wai K. Cheng
Professor of Mechanical Engineering
Thesis Supervisor

Signature redacted

Rohan Abeyaratne
Professor of Mechanical Engineering Chairman, Department Committee on Graduate Theses
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Abstract

Due to their advantages in higher fuel efficiency and torque compared to conventional port fuel injection (PFI) engines, direct injection spark ignition (DISI) engines have become dominant in gasoline-fueled engines. However, DISI engines have a significant drawback in particulate matter (PM) emission: the PM emission of DISI engines is at least an order of magnitude higher than that of PFI engines. The objective of this study is to investigate PM emission in DISI engines, mainly focusing on particulate number (PN) emission. The study aims to assess, respectively, the plausible PM formation mechanisms: non-fuel originated sources (e.g., lubricant), flame propagation in rich mixture and the pyrolysis of the vapor from liquid fuel film. Through a series of experiments, it has been found that non-fuel contribution is less important than the other two mechanisms. For all operating conditions, the absolute amount of the non-fuel contribution is much smaller than the total emission. In case of PM generated by flame propagation in rich mixture, there is a threshold air-fuel equivalence ratio below which PM starts to form rapidly. The threshold is influenced by the combustion temperature. PM starts to form at lower equivalence ratio when the combustion temperature was lower. Contrary to the PM generated from flame propagation in fuel-rich mixture case, that from the liquid fuel film is suppressed by lowering the combustion temperature. Transmission electron microscopy (TEM) imaging shows that the sizes of primary particles and agglomerated particles become larger as engine load increases, but particulates from different mechanisms have different morphology.

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Title: Professor of Mechanical Engineering
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Chapter 1

Introduction

1.1 Background

Internal combustion engines are widely used in transportation and power generation sectors due to their advantages in reliability and cost competitiveness. However, using internal combustion engines generates air pollution problems. Although there have been continuous improvements in emission abatement technology, emission from internal combustion engine powered vehicles, such as unburned hydrocarbons (HC), carbon dioxide (CO), nitrogen oxides (NOx) and particulate matter (PM), are still significant contributors of air pollution.

Compared to these pollutants, carbon dioxides (CO₂) received less attention until the late 20th century because they are not as hazardous as other pollutants to humans. However, CO₂ became one of the most important topics in transportation sector after its impact on global warming was revealed. Reducing carbon dioxide emission requires an approach different from methods that treat other pollutants because carbon dioxide is a product of complete combustion, while other pollutants are byproducts of combustion. Because of this difference, the solutions used for treating other pollutants, such as chemical conversion and physical capturing, cannot be applied to the CO₂ emission problem. It is hard to treat CO₂ by using chemical reactions because CO₂ itself is a very stable product. Physical capture of CO₂ is practically impossible for a vehicle since the amount of CO₂ generated is much larger
than the other pollutants. A typical light duty vehicle generates approximately 150 - 300 grams of CO₂ per km (distance traveled), while generating other pollutants in the order of few milligrams per km. Consequently, the only reasonable solution is to improve fuel efficiency, burning less fuel per distance traveled.

Continuous efforts to reduce CO₂ emission from light-duty vehicles led to the introduction of direct injection spark ignition (DISI) engine technology in the late 1990s. Unlike conventional port fuel injection (PFI) engines which mix fuel and air at the intake manifolds, fuel is directly injected into the cylinders in DISI engines. This difference provides DISI engines with better fuel economy and torque characteristics; injected fuel lowers the temperature of the mixture in the cylinder when it evaporates, leading a higher volumetric efficiency and better resistance to knock. Because of these advantages, DISI engines have been quickly replacing PFI engines over the past 20 years [7], especially for boosted engines. However, DISI engines have their drawback in PM emission. PM emission from DISI engine is substantially higher than that of PFI engines, especially when the engine is not fully warmed up [8, 9]. Because both the fuel economy and emission regulations are strengthened each year, reducing the PM emission of DISI engines is necessary for automobile manufacturers. Although numerous studies have been conducted to reduce PM emission in DISI engines, many fundamental questions about the PM formation mechanism still remain unanswered due to the lack of understanding of PM formation processes.

1.2 Literature Review

The PM emission problem of DISI engines has been recognized and studied from the early stages of development. This chapter briefly summarizes previous work relevant to PM emission in DISI engines to situate the contribution of this thesis.

1.2.1 PM emission legislation

Since the harmful effects of soot (Carbonaceous PM) have been known to people for a long time, the history of PM regulations is longer than most people think. There was

In the amendments of Clean Air Act in 1970, the United States government authorized the development of regulations to limit emissions from stationary (factories and power plants) and mobile (automobiles) sources. National Ambient Air Quality Standards (NAAQS) were established as a result. PM was selected as one of six criteria pollutants monitored and controlled by NAAQS with CO, NO₂, sulfur dioxide (SO₂), HC and photochemical oxidants 1. In the case of automotive PM emissions, the first regulation in 1970 was based on opacity, and concerned only heavy duty diesels [12]. A more complicated diesel PM emission standard was established in 1980. This new regulation was firstly applied to passenger cars and light-duty trucks, and then expanded to heavy-duty engines and trucks in 1985 [10].

In Europe, the first PM regulation was legislated in 1989. This regulation controls the mass of diesel particulates that are collected during the test cycles (EC Directives 88/436/EEC, 91/441/EEC and 91/542/EEC) [13]. The PM emission level allowed by these regulations has become stricter due to elevated concerns about health and environmental issues. Current regulations require much lower level of PM emission compared to those early regulations. For example, in European standards, the PM emission limit of light-duty diesel vehicles was 0.14g/km for Euro 1 (legislated in 1992), but the current Euro 6 (legislated in 2014) requires PM emission of less than 0.005 g/km for the same category vehicles.

As the details of PM's harmful impact are uncovered, the interest has moved to smaller particulates. Clean Air Act 1970 regulates total particulate emission, including coarse particles. However, in 1987, Environmental Protection Agency (EPA) switched from regulating total particles to particulates that have a diameter of 10 μm or smaller (PM₁₀). Furthermore, epidemiological studies in the 1990s suggested that regulations based on PM₁₀ criterion are not effective enough since they cannot catch

---

1 Chemical species produced by reactions between NOx and volatile organic compounds (VOCs). Ozone (O₃) is considered to be the most important.
fine particles which can penetrate deep into human respiratory system.

In response to these findings, the EPA proposed the PM$_{2.5}$ (particulates equal to or less than 2.5 $\mu m$ of diameter) standards in 1996. At first, several industrial groups and researchers claimed that there was no direct evidence that such fine particles are adverse to human health, but various studies have shown that PM$_{2.5}$ and public health are clearly related [14, 15]. In addition, several studies in the late 1990s suggested that the response to particles from nontoxic materials correlates better with the particle surface area rather than its mass, suggesting that ultrafine particles (diameters less than 0.1 $\mu m$) and nano particles (diameters less than 0.05 $\mu m$)$^2$ are more important to health effects[16, 17]. However, the extensive reviews done by EPA in 2009 claimed that the PM$_{2.5}$ definition still showed the best correlation with to adverse health effects and PM exposure for both short-term and long-term cases [18].

In the automotive industry, increased attention to ultrafine particles has brought another form of regulation—particulate numbers. Previous PM regulations were mostly mass-based regulations. (g/km for light-duty vehicles and g/kWh for heavy duty trucks). However, mass based regulations cannot catch ultrafine particle emissions since the mass contribution of particulates is relatively smaller than that of bigger particles [19]. In order to solve this discrepancy, the Particulate Measurement Protocol (PMP), a research initiative supported by the United Nation’s Economic Commission for Europe - Group of Experts on Pollution and Energy (UNECE-GRPE), was launched in the 2000s [18]. With the effort of working groups on PMP, particulate number (PN) in the range of 23 to 2500 nm has been suggested as a complementary measurement method.

As a result, a PN regulation has been included with a conventional mass based PM regulation from the Euro 5b standard, which was legislated in 2011 and only regulated the PN emission from light duty diesel vehicles. However, Euro 6, the most recent regulation, endorsed a PN regulation that also limits PN for passenger cars equipped with DISI engines. The regulation limits the number of particulates per

---

2 Classification of particulate based on size can vary depending on the literature.
km travelled $6 \times 10^{11}$ particles/km), and this limit is the same for both DISI engines and light duty diesel engines. For DISI engines, PN regulation is considered to be more challenging than mass based regulations. In fact, various studies have reported that many DISI engines emit more PNs than modern diesel engines equipped with particulate filters [9, 20, 21].

The United States has neither a PM regulation for SI engines, nor a plan to adopt PN regulations yet. However, EPA tier III regulation requires a stricter mass limit than European standards. In terms of state regulation, California is considering the introduction of a PN regulation with the PN limit based upon the assumption that 1 mg of PM emission per mile is equivalent to $10^{12}$ (nonvolatile) particles/mile [18].

1.2.2 PM measurement

Dissimilar to other exhaust emissions, PM is neither physically nor chemically homogeneous. Therefore, PM measuring methods have to be different from other pollutants measuring methods. When PM regulations were based on the mass of particulate with relatively loose limits, "measuring PM" itself was not a difficult task. First, PM from motor vehicle exhaust (light duty vehicles) or engine exhaust (heavy-duty diesels) was collected onto filters through a constant volume sampling (CVS) system. Then, the weight was measured after post-processing [22]. However, as regulations became stricter, and the focus moved to small-size particles, the development of reliable PM measuring technology has become an important issue. In 2007, EPA revealed that the PM measurement process may show different results depending on experimental conditions, such as dilution ratio at the tunnel, residence time and filter face velocity, because these factors can generate differences in gas - particle partitioning during the sampling process [23].

Counting particulates is even more difficult because particulates can be composed of different sets of chemical elements ranging in size from a few nanometers to a few micrometers. The official PMP protocol measures the number of solid (non-volatile) particulates within the range of 23 nm - 2.5$\mu$m) by using an optics based method called a condensation particle counter (CPC). PMP protocol regulates the total num-
ber of solid particulates only with a particles/km based limit. In this protocol, volatile fractions are removed by preheating samples to 300°C [20], and size distributions are not considered. Volatile fractions are omitted due to the repeatability issue; counting volatile particles is more challenging compared to counting solid particles since the measurement process is strongly depending on experimental conditions, such as dilution ratio or sampling positions. Therefore, further study is needed to legislate volatile particles [18].

Because the European standard is currently the only standard that has a PN limit, the CPC is the only device that has a legal standing. However, PN measurement devices that have a wider range of detection - in terms of size limit, availability of measuring volatile particles and time resolution - have been developed for research purposes. Since the behavior of particles is mostly characterized by size rather than chemical compositions, most particle detection devices sort out particles depending on their aerodynamic diameter. Various particle detection devices with different sampling systems and measuring principles are available, and each device has its own pros and cons. Generally speaking, in situ measurements are considered to be better than ex situ measurements, especially in transient conditions, but the former are less developed [10]. Sometimes more than two devices are simultaneously used to obtain more detailed results.

PN measuring devices can be categorized into a few groups depending on their operating principles. Impactors, the most primitive type, use the inertial mobility of particles. Traditional impactors were able to offer only mass-based information, but electrical low-pressure impactors (ELPI) can offer the number distribution by measuring the electrical current caused by impacts. These impactor-type devices have an advantage in measuring time, but have a problem in detection ranges. Since they rely on inertial forces generated by particles, it is hard to detect very small particles. Originally, the lower limit was about 400 nm, far larger than particulates from modern engines. Although this was lowered to around 30 nm in modern ELPIs by lowering operating pressure, 30 nm limit is still not low enough to cover all particulates from automobile emissions. In addition, lowering operating pressure may cause problems
in measuring volatile particles due to evaporation [24].

Another type of detector uses electrical mobility of particles. Devices like the electrical aerosol analyzer (EAA), scanning mobility particle sizer (SMPS), differential mobility particle sizer (DMPS) and differential mobility analyzer (DMA) belong to this category. These devices first electrically charge the particulates, and then sort out particles according to their electrical motilities by using electro-static forces. Compared to impactors, they can detect much smaller particles (up to 5nm) but may have some inaccuracy due to non-uniform charging of particles [24, 10]. SMPSs had been widely used in the automobile industry, but they are less frequently used now due to the fact that they can be used for steady-state experiments only. As transient PN emission became more important, PN measurement devices that can offer real-time results are more commonly used today. The device that used in this thesis, Cambustion DMS 500 Mk II, also belongs to this type. Details of above are provided in Chapter 2.

Less commonly used than the methods mentioned, laser induced incandescence (LII) can be used for particle measurements. LII can offer high temporal resolutions and does not require a dilution system. However, this method heats up samples before measurement, and therefore may cause the evaporation of organic fractions or morphological changes of solid particles [25].

Since different instruments measure different properties of particulates, there are always discrepancies among the results. In addition, results from the same device with the same experimental conditions can be different depending on environmental conditions, such as humidity and the ambient temperature. In order to compensate for these inaccuracies, results from different type of devices can be used complementarily, but this requires careful attention when interpreting the data. Generally speaking, the official PMP protocol is considered to have the best reproducibility and repeatability, sometimes even better than the conventional mass based method [26]. However, the PMP protocol gives the least information: it gives only the number of particles collected over a size range during the entire test, and offers no information on size-spectra or changes in PN emission as a function of time.
1.2.3 Characteristics of automotive PM emissions

Classification of particulates

Due to the severe heterogeneity of PM, it is impossible to perfectly classify each particulate. Therefore, PMs are categorized into several groups depending on their sizes and chemical composition. Even this classification is often not clear, but gives some insights into the PM formation process occurring inside the cylinders.

In terms of size, the PM from internal combustion engines can be categorized into three groups: nucleation mode, accumulation mode and coarse mode [19]. These modes encompass particles ranging from a few nanometers to several micrometers in diameter. It is reported that all three modes have log-normal distribution, but the peak and the shape of the each mode change significantly with operating conditions.

Nucleation mode particles are formed from nucleated volatile fractions, and have the smallest sizes. Little was known about nucleation mode particles until the late 1990s because it was hard to detect them. They are formed as the combustion process produced primary particles, which are solids at the exhaust condition, or as the nucleated particles from volatile hydrocarbon species as the charge cools down in the expansion and exhaust process. The latter particles are considered to be volatile.

Accumulation mode particles are made of carbonaceous cores called spherules. Spherules have similar sizes regardless of operating conditions, ranging from 20 to 50 nm. However, the size of agglomerates, chunks of spherules, shows a wider deviation because the number of spherules contained in particles can vary from tens to thousands. Since this agglomerate absorbs liquid or semi-liquid material well due to its porous structure, the surface of an accumulation mode particle is usually coated with volatile or semi-volatile layers, while its back-bone is mainly carbonaceous.

Unlike nucleation mode or accumulation mode particles, coarse mode particles are not directly generated through the in-cylinder combustion process. Coarse mode particles are made of other particles through a process of storage and release in the downstream exhaust system, or through material disintegration. Therefore, coarse mode particles were not considered in this study, which focused on in-cylinder process
combustion.

Figures 1-1 shows a typical mass and number weighted distribution of each mode in engine exhaust [19]. As can be seen, nucleation mode particles are dominant in terms of number, and accumulation mode particles are dominant in terms of mass, while coarse mode particles are insignificant in both cases. The distribution of nucleation mode particles is strongly affected by how the exhaust gas is treated due to the volatility of particles. Compared to nucleation mode particles, accumulation mode particulates are less likely to be affected by sampling conditions, and are therefore better for characterizing PM emissions from engines [10].

![Mass and number weighted distributions of particulates in engine exhaust](image)

Figure 1-1: Mass and number weighted distributions of particulates in engine exhaust

In terms of composition-wise classification, there are five categorizes: carbonaceous (soot), ash, organic, sulphate and nitrate particulates. Carbonaceous particulates and ash are insoluble, while the others are soluble to water. Ash is inorganic incombustible compounds. Most of these are metals (Fe, Cr, Cu, Zn, Ca) but a few of them contain non-metal elements (Si, P, S, Cl). The majority of sulphate and nitrate are sulphuric acid nitric acid dissolved into water vapor, but a small portion exists as metal-sulphates or metal-nitrates. Organic fraction encompasses major chemical families such as esters, aromatics and alcohols. Soot is mostly composed of carbon atoms with small amount of hydrogens.
Soot formation mechanism

Since a large portion of the PM from DISI engine is soot, this study mainly focused on soot emissions, rather than the other four groups of particulates. This section explains the fundamentals of the soot formation process inside of the cylinders. Simply speaking, soot is formed in flames through pyrolysis (thermolysis of organic compounds, such as fuel molecules, at high temperature and the absence of oxygen atoms) [27]. Simplified diagram for the soot formation process is shown in Figures 1-2. However, the details are much more complicated. Many questions remain unanswered. Therefore, researchers have used empirical relations expressed by global variables such as the mass fraction of fuel burned, combustion temperature, and values obtained from the heat release rate. Semi-empirical relations based on Arrhenius relationships are widely used as well [28].

![Chemical process Physical process](image)

Figure 1-2: Particulate formation mechanism

The difficulty in understanding the soot formation process comes from its complexity. Fuel molecules, with less than 30 carbon atoms with a hydrogen/carbon ratio around 2, are converted to spherules that contain more than thousands of carbon atoms with a negligible portion of hydrogen atoms within a few milliseconds of reaction time. More than hundreds of chemical relations are involved during the process. However, researchers have found that the soot formation process can be divided into four stages: (1) inception, (2) surface growth, (3) agglomeration and (4) oxidation [29].

In the inception stage, fuel molecules are broken up by pyrolytic reactions. The broken molecules are then rearranged into precursor molecules, such as polycyclic aromatic hydrocarbon (PAH) or acetylenes ($C_2H_2$). Aliphatic molecules first form
simple aromatic molecules like benzene through cyclisation reactions, and then continue to grow using acetylenes as feedstock. Since carbons are added and hydrogens are removed during the reactions, this process is called the hydrogen abstraction and carbon addition (HACA) process, and the formed particles are called nuclei. The relative concentration of aromatic molecules and acetylene molecules determines the reaction pathway in the HACA process. If the concentration of acetylene is much higher than that of aromatics, PAH grows by the addition of acetylene molecules. Conversely, PAH is more likely to grow by the condensation of existing aromatic molecules if aromatics are abundant [30]. Nuclei have a mean diameter of one or two nanometers, composed of approximately fifty carbon atoms. The number of nuclei generated in this stage determines the initial number of soot particles.

Once enough nuclei are formed, additional particles start to attached nuclei, moving to the surface growth stage. It is hard to determine the exact transition from the inception stage to the surface growth stage. Detecting transition even depends on the measurement methods used to detect the formation of nuclei. However, it is known that the zones where inception and surface growth occur are different in the flame [31]. Through the surface growth stage, the mass of soot is greatly increased, while the total number of soot particles remains almost constant. Particles generated in this process are called spherules. Spherules grow up to a mean diameter of 20 - 50 nm. Outer layer of spherules, formed during the surface growth stage, has layered micro-structures. In contrast, cores formed during the inception stage have amorphous shape. It is not known whether the inception stage or the surface growth stage that determines the final amounts (number of particles) of soot. However, in terms of mass, it is generally accepted the most of soot is generated during the surface growth stage[29].

The maximum size of spherules has been observed to a few tens of nanometers, regardless of conditions. Beyond this limit, spherules start to grow by adhering to each other. This growth process is called agglomeration. Unlike the previous two stages, which are chemical processes, the agglomeration process is a physical process. In the agglomeration stage, the number of particles decreases while the entire mass
is conserved. The underlying physics of agglomeration is relatively well known and extensively modeled, compared to other stages. Revised Smoluchowski equations are generally used to explain the number decreases in agglomeration stages. The age of the spherules is an important factor in the agglomeration stage. Solidified (mature) spherules exhibit point contacts, while viscid (young) spherules are more likely to be fused together. The final morphology of particulates is determined during the agglomeration stage. A point contact between particles generates a chain-like structure, while fusing forms a clustered particle [32].

Although all of these three stages are chained events, they are not completely separated in time. At some points, the inception process competes with the surface growth process for soot precursors, and the agglomeration process sometimes occurs between very young spherules that are still possible by the surface growth.

The oxidation stage, on the other hand, is a soot removing process. Like the previous three stages of soot production, the oxidation process can happen simultaneously with other processes in the flame. This nature generates difficulties in experimentally separating soot formation and soot oxidation because the chain of soot production events can be terminated at any stage. In general, more than 90 % of soot formed through combustion is oxidized by the combustion process. There are several species involved in this process such as O₂, O, OH, H, CO₂ and H₂O [33]. In the case of engine combustion, the peak concentration of OH radical correlates well to the onset of soot depletion [34]. However, the dominant oxidants can be different depending on spatial and temporal aspects of abundance. In terms of time, soot particles are initially attacked by OH radicals. H₂O and CO₂ start to play a role after OH radicals have been consumed, while the later part of the oxidation process is governed by O₂ molecules. Spatially, exposure of soot particles to OH is limited to the narrow region of the flame. Outside of the flame, O₂ is more important due to its abundance [33]. Similar to the soot production process, semi-empirical equations based on Arrhenius form are widely used to model soot oxidation, such as Nagle and Strickland-Constable model [35]. In general, the oxidation of spherules is considered to be kinetically controlled [36], but physical aspects, such as mixing of air and soot or movement of the
soot cloud, can be important depending on operating conditions [37]. This means that the oxidation process can be either kinetically controlled or mixing-controlled, limiting the regimes where each model can apply.

1.2.4 PM emissions in DISI engines

As discussed in Section 1.2.1, gasoline engines had been exempted from PM regulations. In terms of mass, PM emission from conventional PFI engines is approximately 1% of that from diesel engines. However, the situation changed after gasoline engines adopted a direct injection technology. Several studies have shown that the amount of PM generated from early DISI engines is closer to that from diesel engines, rather than that from PFI engines.

The main difference between DISI engines and PFI engines with respect to soot formation comes from the air-fuel mixture preparation process. By directly injecting fuel into the combustion chamber, DISI engines can enjoy the "charge cooling effect", resulting in improved fuel economy and torque compared to PFI engines. However, this diesel-like mixture preparation process generates more favoring conditions for soot generation. Since DISI engines have a significantly reduced time for air-fuel mixing, they are more likely to have locally rich region where soot is generated [38].

PM formation is greatly affected by engine operating parameters. An overall air/fuel ratio may have an impact on PM emission since incomplete fuel conversion in fuel-rich conditions may cause severe soot formation. However, many studies have shown that soot formation is more closely related to the injection timing rather than the overall air/fuel ratio since the injection timing directly affects the mixing process of the fuel and air. In [39, 40], PM emission greatly varied as injection timing changes, while there were much smaller deviations as a function of overall air/fuel ratio around the stoichiometric condition. Generally, retarding the injection timing increases PM emission because reduced mixing time results in less homogeneous charge at the moment of ignition. On the other hand, if the fuel injection timing is too early, PM emission increases as well due to the piston or valve wetting. Since a very small deviation in injection timing can cause wall impingement or inhomogeneous mixture
preparation, the optimization of injection timing is essential for PM reduction.

In terms of mixture formation methods, a spray guided method has less PM emission, especially in terms of number, than a wall guided method because it has less chance of wall or valve impingement. A study done by Price et al.[41] showed that the spray guided system has much smaller peaks in accumulation mode PN emissions, but has slightly higher nucleation mode PN emission compared to the wall guided system. This opposite trend in accumulation mode and nucleation mode PN emissions can be observed in the same hardware. The work of Pei et al.[42] found that advancing the fuel injection timing reduces the total PN emission. The nucleation mode particles were slightly increased by advancing the fuel injection timing, but the increase was compensated for significantly reduced accumulation mode particles.

Combustion phasing is another factor that determines the soot formation. If all other conditions are the same, retarding the ignition timing tends to decrease PM emissions. This trend applies for both homogeneous and stratified charge operation. Both the number and mean diameter of particles simultaneously decrease as the spark timing retards [41, 42]. The underlying mechanism is not clear, but one possible explanation is that the higher exhaust temperature can promote the oxidation of particulates. However, retarding combustion phasing deteriorates fuel efficiency when the 50% of mass fraction burned is prolonged more than than 6-7 CAD from MBT timing [39].

This thesis specifically focuses on PM emission under cold-start condition where PM emission is the highest. Similar to other gaseous emissions such as HC and CO, the majority of the tailpipe PM emission comes under cold-start condition. A vehicle test with a new European drive cycle (NEDC) showed that the highest PN emission occurs within the first 100 seconds of the test [8].

Under cold-start condition, an in-cylinder environment offers an unfavorable condition for fuel vaporization, causing significant variations in the flame growth and fuel spray structures[43]. Through the optical engine and single cylinder engine test, Xu et al.[44] found that the formation of a robust rich fuel-air mixture around the spark plug is the key for lower cyclic variation and more stable combustion, while
vaporization of fuel could be improved by injection optimization.

The fact that fuel is directly injected into the cylinder makes another PM formation mechanism in DISI engines – liquid fuel film. For PFI engines, the formation in-cylinder liquid fuel film is minimized with the aid of backflow. Backflow facilitates the evaporation of liquid fuel film by distributing the fuel film in the intake port and valve region to a large surface area in the intake manifold. However, in DISI engines, in-cylinder liquid fuel film is more likely to form since liquid fuel spray is directly injected into the combustion chamber [45]. This problem becomes worse under cold-start condition because fuel is less likely to evaporate. Stevens and Steeper investigated the role of liquid fuel film on soot formation [46]. They characterized the combustion of liquid fuel film at the piston top, referred to pool-fire via direct imaging. If fuel film exists on the piston-top during the combustion of the premixed charge, it ignites to create piston-top pool-fire. With a laser scattering experiment, it was found that soot is produced by the pool fire, and this soot can survive and throughout the exhaust stroke. This means that pool fire is a potential source of PM emission in DISI engines. A further study performed by a research group at Karlsruhe Institute of Technology [47] reconfirmed that the pool fire is the main cause of soot formation in the homogeneous charge operation, while soot is both formed in local rich mixture regions of combustion chamber and on the piston due to pool fire.

Ketterer and Cheng [4] suggested a more detailed explanation for this pool-fire. They argued that the heat conduction from the burned gases and heat release from reactions occurred at the rich-mixture region, located above the liquid film on the piston, sustain soot formation in this region during expansion stroke. Radiation from the particles in this region appears as the pool-fire although there is no traditional "fire" since there is not sufficient oxygen available. Mainly focused on hydrocarbon emission, but Contanzo and Heywood [48] offer insight into the nature of fuel film as well. When the other conditions are the same, hydrocarbon emission always increased if liquid fuel films exist in the combustion chamber. Furthermore, it was also found that the wall temperature is a governing factor when differences in the amount of liquid fuel actually on the combustion chamber surfaces are accounted for, implying
that the contribution of liquid fuel film could be more significant under cold-start condition.

It is not considered as significantly as other mechanisms, but coking of fuel can be another reason for PM emission in DISI engines. Carbon deposits can form at the tip of the injectors in DISI engines because the tip of the fuel injector is always exposed to flame. During the fuel injection process, fuel vapor is stored at this porous deposit. Then, stored fuel vapor is subsequently released and combusted in the injector diffusion flame, becoming soot [49].

Although PFI engines are exempted from PM regulations due to their low emission level, this does not mean that PFI engines are completely free from PM emission issues. Several studies indicated that PFI engines showed much smaller PN during steady-state operation compared to diesels engines, but can emit an increased fraction of nano particles depending on operation conditions. A stable baseline PN concentration is an order of $10^5$ particles/cc for PFI engines, however, spikes in PN are frequently observed during engine operations. These spikes can have more than 100 times higher PN concentration compared to the baseline. This is mainly due to the increased fraction of volatile particles less than 30 nm of mean diameter. In addition, it is also observed that PN concentration significantly increased when engines were operated under high-load, transient or cold-start condition [7]. The Euro 6 regulation requires a limit of $6\times10^{11}$ particles/km as determined by PMP.

### 1.2.5 Mitigation Strategy

Efforts to reduce PM emission in DISI engines have been directed at soot fractions. Roughly speaking, the mitigation strategies can be divided by two categories. The first one aims to minimize soot formation inside of the combustion chamber, and the second one focuses on oxidizing soot in the exhaust system. Particle filtration systems as exhaust gas treatment are also being developed.

As discussed above 1.2.4, the fuel injection process strongly influences the mixture formation, determining the homogeneity of the mixture and the presence of liquid fuel droplets and films. Various advanced injection methods such as post injection
or multi-pulse injection have been suggested to reduce soot formation. Under cold-start condition, an injection strategy which improves the combustion stability can effectively reduce soot formation as well [9]. Optimizing fuel pressure is especially important at low coolant temperature. Excessively low injection pressure results in the poor atomization of fuel, and excessively high pressure causes increased wall or piston impingement. Additional improvement can be achieved by changing the injector geometry, such as varying the spray angle or the number of holes.

In addition to the fuel injection process, the optimization of the air intake process can improve air-fuel mixing and flame front propagation as well. Many studies proved that optimizing valve overlap combined with exhaust gas recirculation (EGR) can reduce PM emission [39, 50]. Hedge et al.[51] showed that high EGR levels with the 1.6L DISI engine reduced both PM mass and number in a variety of operating conditions; brake specific soot mass was reduced by at least 60% for all cases, and PN was reduced by 40% or more in most operating conditions. Both the external and internal EGR method have been tested, and it was found that internal EGR was more effective for PM reduction compared to external EGR in light load, low speed conditions. However, external EGR can be to used to reduce PM emissions when the engine is operated with non-optimal operating conditions, such as fuel-rich condition under the transient operation[51].

Because PM emission is much higher under cold-start condition than warmed up phases, any methods that accelerate the warming up process of engines or reduce the light-off time of the three-way catalyst system can contribute to reduction of PM emission. Retarding spark timing is one simple way to quickly warm up the engine, but it can only be used for cold-idle condition because retarding the spark timing deteriorates fuel economy of vehicles. In addition, as much of the enthalpy is dissipated into the exhaust, it takes longer time to heat up the combustion chamber. Since fuel droplets contact with the cold combustion chamber walls cannot evaporate sell, and fuel vapor can condense back into liquid, retarding spark timing may lead a high PM emission during cold-start condition [8]. Instead, heating up fuel or intake air can be used as alternative methods to warm up the engine, but the additional
energy required for heating up the engine should be accounted for a fair comparison.

Like other emissions, aftertreatment systems can be used to reduce PM emissions. Secondary air injection (SAI) is a method that provides additional air to the exhaust stream under fuel-rich operating condition, enabling post oxidation of the exhaust gas prior to the catalyst. Lee and Heywood [52] found that SAI can reduce significant amounts of PM and the light-off time of the catalyst. The feed gas of secondary oxidation is CO and HC, but the oxidation of feed gases offers radicals that can oxidize PM in the exhaust stream. The amount of PM reduced (in terms of volume) was proportional to the amount of heat release in the secondary oxidation [53].

Like diesel engine particulate filters (DPF), gasoline particulate filters (GPF) are being researched as well. In principle, modern DPF can be used for gasoline engines as well. However, due to smaller particles from DISI engines compared to those from Diesel engines, GPF requires a finer filter characteristics in terms of cell density and mean pore sizes, leading to a higher back pressure that may cause a stronger negative impact on vehicle performance and fuel consumption [9]. Several GPFs have been developed and demonstrated their performance [54]. Compared to a conventional DPF, a GPF has difficulty in achieving a stable regeneration in the filter because of the difference between operational conditions of gasoline and diesel engines. The temperature of exhaust gas is too low to burn up stacked particles in low load and lean operation, while there is insufficient amount of oxygen in exhaust gas in high load and stoichiometric conditions. In order to solve this problem, the development of catalysts that can exploit H₂O and CO₂ as oxidants is being researched [55]. In any case, adding an aftertreatment system is a less favorable option for automotive makers because it inevitably increases the manufacturing cost of the power-train system.

Outside of the hardware perspective, PM reduction via changing fuel formulation is being researched as well. Similar to diesel combustion, several studies have reported that gasoline with oxygenate compounds, such as alcohol or ether, generates less PM [54, 2]. Another benefit that can be obtained from oxygenate blending is better knock resistance, which enables a higher compression ratio because most oxygenate compounds, such as methanol or ethanol, have higher research octane number (RON)
values compared to regular gasolines. Therefore, blending oxygenates boosts the fuel’s octane rating, offering a better knock margin for engine operations. However, the fuel economy might be deteriorated due to the lower heating value (LHV) of oxygenates. For example, ethanol, the most commonly used oxygenate compound in US, has a much higher RON value than gasoline (107 vs 92-98), but its LHV is only 61% of that of gasoline (26.9 MJ/kg vs 44.0 MJ/kg) [36].

Because it is known that the physical and chemical properties of gasoline have an impact on PM emissions, various models have been developed to correlate PM emission and fuel properties or compositions. Honda’s PM index model[56] is based on a weight fraction, vapor pressure, and a double bond equivalent (DBE) value of each component in the fuels. This model showed that low vapor pressure species are related to PM emissions, due to their combustion behavior. In terms of component, aromatic compounds with a high boiling point and a high DBE value have stronger impacts on PM emission than low DBE components such as paraffins. With a single-cylinder optical spray guided DISI engine experiment, Leach et al.[57] modified this PM index, making the PN index. PN index encompasses the change of DBE and vapor pressure of fuel mix by using Raoult’s law and the universal functional activity coefficient (UNIFAC) method.

1.3 Research objectives and approach

This study is a part of the bigger project which has been performed at the Sloan Automotive Laboratory at the Massachusetts Institute of Technology to understand and reduce the emission problems in DISI engines, especially under the cold-start condition.

This thesis is intended to investigate the in-cylinder soot formation process in DISI engines under the cold-start condition. Here, 'cold-start condition' refers the engine operating conditions where coolant temperature is low (around 25 °C), while other engine components are under thermal equilibrium, and maintaining steady state temperature during the experiments. Therefore, the term 'cold-start condition' in
this thesis is different from 'cranking-start condition' which means all the engine components are as cold as ambient temperature before the start of operation, and then warms up as firing proceeds.

The main difficulty of understanding soot formation comes from the nature of combustion. Several factors simultaneously contribute to soot formation, but it is very hard to control each factor independently because most of them are coupled. Therefore, most of previous studies were qualitative, leaving various contributing factors coupled. However, this study aims to analyze various possible soot formation mechanisms independently by performing a series of experiments that are designed to decouple various factors.

Decoupling all the physical and chemical factors will really require various optical diagnostics and simulations, which needs significant amount of capital and time. Instead, the study tries to focus on the fundamentals and include features such as the capability to run with pre-vaporized and fully homogeneous to help decouple the fundamental effects like sensitivity to gas phase enrichment, combustion or flame temperature, influence or oil, a role of liquid fuel films on soot formation.

Three primary mechanisms chosen for this study are (1) soot generated from non-fuel sources (e.g. lubricants), (2) flame propagation in locally rich mixture, and (3) the pyrolysis of fuel-rich vapor liquid fuel film on the piston top. These mechanisms are selected from the basic principle that "soot is formed via pyrolysis", which means that soot is generated under the absence of oxygen with elevated temperature. Figure 1-3 shows the conceptual diagram of three mechanisms.

Before investigating each mechanism individually, the general trend of PN emission was investigated with various operating parameters with a conventional direct injection system in Chapter 3.

The first and second mechanisms was studied in Chapter 4. Since both mechanisms assume soot formation under the absence of liquid fuel film, fully premixed charge experiments were performed. Low C/H ratio premixed gaseous fuel was used to investigate the first mechanism, while premixed fuel-rich gasoline was used for the second. In Chapter 5, the role of liquid fuel film on soot formation was addressed with
dual-fuel experiments. Finally, the morphology of particles from different mechanisms was described in Chapter 6.

With better understanding of soot formation process, it is expected that this thesis can contribute to designing DISI engines that has reduced PM emissions under cold-start conditions without sacrificing fuel efficiency or engine performance.
Chapter 2

Experimental setup and methodology

This chapter provides a detailed description of the experiments conducted in this study. This thesis mainly focuses on experimental approach. Most of the experimental results are given in terms of particulate number, which is considered to be more important than particulate mass for DISI engines. The focus is mainly on the accumulation mode particles, but sometimes the nucleation mode particle data are also used to interpret the results. Additionally, an electron microscope was used to see the morphology of particulates from formation different mechanisms.

In the first section, the experimental equipment is detailed, including the base engine, particle measurement device, transmission electron microscope and auxiliary sensors. The second section explains underlying logic of designing experiments conducted for this study as well as corresponding experimental procedures.

2.1 Experimental setup

2.1.1 Base engine

An up-to-date 2.0 L four cylinder turbocharged DISI engine, manufactured by General Motors and equipped in various vehicles including the Cadillac ATS, CTS, CT6, Chevrolet Malibu, was chosen for this study. The engine is a spray guided engine, using side-mounted electromagnetic injectors which has which has 6 holes with a 52°
cone angle. The injector is inserted with a 25° inclination from the horizontal. The engine also features a centrally mounted spark plug and 4 valves per cylinder. The basic specifications of the engine are shown in Table 2.1.

<table>
<thead>
<tr>
<th>Code Name</th>
<th>LTG</th>
</tr>
</thead>
<tbody>
<tr>
<td>Engine type</td>
<td>In-line 4 cylinder</td>
</tr>
<tr>
<td>Displacement volume</td>
<td>1,998 cc</td>
</tr>
<tr>
<td>Bore × Stroke</td>
<td>86.0 mm</td>
</tr>
<tr>
<td>Compression ratio</td>
<td>86.0 mm</td>
</tr>
<tr>
<td>Maximum power</td>
<td>272 hp (203 kW) @ 5500 rpm</td>
</tr>
<tr>
<td>Maximum torque</td>
<td>400 Nm @ 3000 - 4600 rpm</td>
</tr>
</tbody>
</table>

The engine has hydraulically actuated variable valve timing (VVT) on both the intake and exhaust camshafts, but cam timings were fixed at the parked position in this study. Further details about the valve timings are shown in Figure 2.2.

| Intake valve opening (IVO) | 11 CAD ATDC |
| Intake valve closing (IVC) | 61 CAD ABDC |
| Maximum intake valve lift | 10.4 mm @ 126 CAD ATDC |
| Exhaust valve opening (EVO) | 52 CAD BBDC |
| Exhaust valve closing (EVC) | 10 CAD BTDC |
| Maximum intake valve lift | 10.5 mm @ 125 CAD ATDC |

The engine was installed and mated to a Froude-Consine AG-80 eddy current dynamometer, and connected to an electric motor via a driver shaft. The shaft has rubber coupling to absorb fluctuations in engine torque. A custom made enlarged flywheel was used to minimize speed fluctuations. An in-house developed C-code was used for fuel injection, allowing full customization of the operating parameters such as injection and spark timings, injection duration.
The engine was cooled using a heat exchanger, which uses city water to absorb heat. The coolant circuit circulated a 50/50 mixture of water and ethylene glycol through the engine's original cooling circuit. For lubricant oil, the OEM oil cooler originally installed on the engine was used, with the coolant supplied from the main engine cooling circuit.

Figure 2-1 shows the schematic diagram of the experimental set-up before modifications. The details of the test hardware modification are listed in Section 2.2.2

2.1.2 Test fuel

EPA Tier II EEE certification gasoline produced by Haltermann Solutions (HF0437) with a carbon/hydrogen ratio 1.82 was used for this study. The fuel's RON and MON were 96.1 and 88.5, respectively. Figure 2.3 summarizes detailed specifications for the test fuel.
Table 2.3: Technical specification for the test fuel

<table>
<thead>
<tr>
<th>Test</th>
<th>Units</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distillation</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Initial boiling point (IBP)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10 %</td>
<td>OC</td>
<td>30</td>
</tr>
<tr>
<td>20 %</td>
<td></td>
<td>50</td>
</tr>
<tr>
<td>30 %</td>
<td></td>
<td>62</td>
</tr>
<tr>
<td>40 %</td>
<td></td>
<td>74</td>
</tr>
<tr>
<td>50 %</td>
<td></td>
<td>92</td>
</tr>
<tr>
<td>60 %</td>
<td>°C</td>
<td>104</td>
</tr>
<tr>
<td>70 %</td>
<td></td>
<td>111</td>
</tr>
<tr>
<td>80 %</td>
<td></td>
<td>116</td>
</tr>
<tr>
<td>90 %</td>
<td></td>
<td>126</td>
</tr>
<tr>
<td>End point (EP)</td>
<td></td>
<td>157</td>
</tr>
<tr>
<td>Density</td>
<td>kg/L</td>
<td>0.742</td>
</tr>
<tr>
<td>Reid vapor pressure</td>
<td>kPa</td>
<td>62.7</td>
</tr>
<tr>
<td>Carbon</td>
<td></td>
<td>0.868</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>wt. fraction</td>
<td>0.132</td>
</tr>
<tr>
<td>Oxygen</td>
<td></td>
<td>0.000</td>
</tr>
<tr>
<td>Aromatic</td>
<td></td>
<td>29</td>
</tr>
<tr>
<td>Olefin</td>
<td>vol. %</td>
<td>1</td>
</tr>
<tr>
<td>Saturate</td>
<td></td>
<td>70</td>
</tr>
</tbody>
</table>
2.1.3 Particulate measurement system

All PN concentrations data in this thesis were measured with the DMS 500, therefore this section briefly describes the operating principle of the DMS 500. The DMS 500 classifies and counts the particles based on their electrical mobility. Before the measurement process, sampled exhaust gas is diluted by DMS 500’s built-in 2 stage dilution system. The first dilution occurs right after the sampling port, by using mass flow controlled high efficiency particulate arrestance (HEPA) filtered compressed air, while the second dilution occurs at a rotating disc type diluter located at the end of sampling line. The sampling line is kept at 150°C to prevent condensing of volatile particles. Figure 2-2 shows the schematic diagram of the dilution system.

![Schematic diagram of DMS 500's dilution system](image)

Figure 2-2: Schematic diagram of the DMS 500's dilution system [1]

After dilution, particles are charged while flowing through a unipolar corona discharge unit. The unit puts a charge proportional to the surface area of particles. Charged particles are then introduced into a classification section where a radial electrical field exists. This field causes particles to drift through a sheath flow toward the electrometer detectors. Particles are detected at different distances down the column, depending upon their aerodynamic drag/charge ratio. Outputs from the 22 electrometers are processed in real time at 10Hz. This operating principle of DMS
500 is shown in Figure 2-3.

![Figure 2-3: Particle detection mechanism of DMS 500 [1]](image)

From the value measured with electrometers, a software built for DMS 500 calculates 38 channels of size data and 6 representative values for the entire particle spectrum (mean diameter, the geometric standard deviation of the mean diameter and particle number concentration of the nucleation mode and those of accumulation mode). The software uses a sophisticated Bayesian statistical algorithm that considers the base level of noise level, and only returns genuine modes above this level. The actual interface of the DMS 500 is shown in Figure 2-4.

![Figure 2-4: User interface of the DMS 500](image)

Finally, it should be noted that all DMS data is strictly valid at standard temper-
ature and pressure (STP., i.e. 0 °C and 1 atm. absolute pressure pressure). Therefore volumetric exhaust flow measurements should be converted to STP before any calculation.

**Notes: Plotting the particle size distributions**

In the DMS 500 (and most particle size instruments), the total number of particles per standard cubic centimeter (#/cc) is obtained by integrating the lognormal distribution for each mode, as given in Equation (2.1), where \( N \) is the volume concentration of the number of particles, \( D_p \) is the particle size and \( \frac{dN}{d\log(D_p)} \) is the concentration of particles for a the size range represented by \( D_p \).

\[
N = \int_{5nm}^{1000nm} \frac{dN}{d\log(D_p)} d\log(D_p) \tag{2.1}
\]

As can be seen, the total volumetric concentration is given as an integral form based on a logarithm of \( D_p \). Therefore, particle instruments plot size data at discretized intervals along the x axis, with intervals of \( d\log(D_p) \) and uses \( \frac{dN}{d\log(D_p)} \) for y axis, as shown in 2-4. This is a conventionally used method in the aerosol field, and this thesis uses the same method as well. However, it is worth mentioning that the PN spectrum can be altered if the y axis is plotted in \( \frac{dN}{d(D_p)} \) by using Equation (2.2).

\[
\frac{dN}{d\log(D_p)} = \frac{\log(10)}{D_p} \frac{dN}{d(D_p)} \tag{2.2}
\]

Plotting this way would not change the information content of the plot. However, plotting in different scales may change the shape of the spectrum, as shown in Figure 2-5. The plot of \( \frac{dN}{d\log(D_p)} \) shows clearly a peak at 70 nm, while the same data, plotted again as \( \frac{dN}{dD_p} \), does not show a peak at all.

The most likely diameter is interpreted as the diameter at which the occurrence probability of particle is the highest. Therefore, it might be correct to plot the y axis in linear scale, and select the value where \( \frac{dN}{dD_p} \) is the maximum. If \( \frac{dN}{d\log(D_p)} \) is used as the y axis, the location of the maximum would not be the same as the \( \frac{dN}{dD_p} \) case.
As mentioned, all PN spectrum data in this thesis uses dN/dlog(Dp) as the y axis since this is a convention that has been used for decades, but it might be interesting to explore whether it is possible to apply the dN/d(Dp) plotting scheme in the aerosol field.

### 2.1.4 Transmission electron microscope (TEM)

In addition to PN concentration measurement, electron microscopy was used to investigate the morphology of particles. JEOL 2010 Advanced high performance transmission electron microscope, installed at the MIT Center for Materials Science and Engineering (CMSE) was used. The microscope has an ultimate point-to-point resolution of 0.19 nm with the ability to image lattice fringes at 0.14 nm resolution. In this study, the instrument was operated at 200 kV with a lanthanum hexaboride cathode (LaB$_6$). Images were taken with a charge-coupled device (CCD) camera, manufactured by AMT. After taking images, post-modifications were made to increase contrast of gray scale images.

Particles were sampled with two different methods, the direct sampling and filter sampling. Each method has its own characteristics, as summarized in Table 2.4. However, the analysis mainly focused on direct sampling method because it gives better information about morphology of particles.
Table 2.4: Differences between sampling methods

<table>
<thead>
<tr>
<th>Sampling method</th>
<th>Direct sampling</th>
<th>Filter sampling</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sampling time</td>
<td>1-3 mins</td>
<td>20 mins</td>
</tr>
<tr>
<td>Particle loading</td>
<td>Thermophoresis</td>
<td>Van der Waals force</td>
</tr>
<tr>
<td>principle</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TEM grids</td>
<td>Carbon film + Copper</td>
<td>Copper</td>
</tr>
<tr>
<td>Advantage</td>
<td>Able to capture</td>
<td>High magnification rate</td>
</tr>
<tr>
<td></td>
<td>an individual particle</td>
<td></td>
</tr>
<tr>
<td>Disadvantage</td>
<td>Low image contrast</td>
<td>Particle agglomeration</td>
</tr>
</tbody>
</table>

Direct sampling method, also known as thermophoretic sampling technique, was firstly used by researcher at Argonne National Laboratory [6]. Further details about TEM imaging technique are described in Chapter 6.

2.2 Methodology

2.2.1 Selecting experimental conditions

This thesis specifically focused on PN emission under cold-start condition. Since the term cold-start condition does not specify any operating points in terms of engine load or engine speed, it is necessary to choose proper operating conditions for designing experiments.

Figure 2-6 shows the PN emission of a DISI engine-equipped vehicle in the federal test procedure - 75 (FTP-75) cycle, measured with the CPC. The dashed blue shows the speed of vehicle, and the black solid line shows the corresponding PN emission [2].
As can be seen, most PN emission comes from the cold-start phase (0–575 seconds), and PN emission peaks at the first acceleration of FTP-75 cycle. Therefore, the fast idle and the first acceleration in FTP-75 cycle were chosen for representative periods in cold start conditions.

With vehicle speed, engine RPM, and torque data from actual vehicle test in FTP-75 cycle (Figure 2-7), it was found that RPM and torque at the end point of the first acceleration in FTP-75 are approximately 1800 and 125 Nm, respectively.
For the test engine (with 2.0 L displacement volume), 125 Nm of torque corresponds to approximately 7.6 bar of net indicated mean effective pressure (NIMEP). Therefore, the highest RPM and NIMEP for experiments were set to 1800 and 8 bar, respectively. Since the fast idle condition corresponds to 1200 RPM and 2 bar NIMEP, two more operating points are added between two end points.

Engine operating conditions are illustrated in Figure 2-8. Most experiments performed in this thesis were performed along the line in Figure 2-8, or the combination of four different RPM and NIMEP.

![Figure 2-8: Experimental matrix](image)

Unless it is specified, the spark timing was always set to the maximum brake torque (MBT) timing. Other experimental conditions are described in Table 2.5.

**Table 2.5: Experimental conditions**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Data collection</td>
<td>100 cycle average</td>
</tr>
<tr>
<td>CA&lt;sub&gt;50&lt;/sub&gt;</td>
<td>7 ± 1 CAD ATDC</td>
</tr>
<tr>
<td>λ</td>
<td>1.00 ± 0.01</td>
</tr>
<tr>
<td>Coolant temperature</td>
<td>25 ± 2.5 °C</td>
</tr>
<tr>
<td>Intake air temperature</td>
<td>25 ± 2.5 °C</td>
</tr>
<tr>
<td>Coefficient of variation (COV) of IMEP</td>
<td>less than 10 %</td>
</tr>
</tbody>
</table>
### 2.2.2 Hardware modification

The original engine was a commercially available turbocharged engine, but several modifications were made for research, especially in intake, exhaust, fuel delivery system. In addition, sensors were installed to monitor the status of the test system.

**Intake and exhaust system**

The custom intake and exhaust manifolds were built for single cylinder operation. For precise control of intake pressure, the original throttle valve was removed and two passive valves were installed in parallel. The original turbocharger was replaced to an external supercharging system, enabling independent control of boost pressure regardless of engine operating conditions. As shown in Figure 2-9, the external supercharging system consisted of two surge tanks and one electrically driven air compressor. To minimize temperature change caused by compression, a heat exchanger is connected to the downstream of the compressor. A nitrogen dilution system was installed to control the oxygen mole fraction of intake mixture.

![Schematic diagram of the intake and exhaust system. The original setup (left) and modified setup (right)](image)

Figure 2-9: Schematic diagram of the intake and exhaust system. The original setup (left) and modified setup (right)

The premixing chamber(Figure 2-10) was installed for premixed gasoline injection. The chamber was located between the intake manifold and throttle valves. Air enters
the direction orthogonal to the chamber, increasing the swirl intensity of charge. To aid vaporization of fuel spray, three bar-type heaters were attached at the outer surface of the chamber.

![Figure 2-10: Premixing chamber (before installation)](image)

**Fuel delivery methods**

Figure 2-11 illustrates the fuel delivery system of the test engine. Custom made injector drivers were used to control injectors. The drivers were controlled by the main computer. When the computer sends an injection signal at designated crank angle, then the driver triggers the injector to inject fuel.

![Figure 2-11: Schematic diagram of the fuel delivery system. The original setup (left) and modified setup (right)](image)

Three different fuel delivery systems were used in this study: (1) gasoline direct
injection, (2) gasoline premixed injection, and (3) premixed gaseous fuel. Sometimes two of these injection fuel delivery methods were simultaneously used depending on the experiments.

Although the original engine had a mechanical fuel pump for direct injection, it was replaced with a new fuel supply system consisting of a hydro-pneumatic accumulator to maintain the fuel pressure regardless of engine speeds. The accumulator was pressurized by compressed nitrogen, feeding the fuel rail directly with desired pressure. Fuel was filled into the accumulator using an automotive low-pressure electric fuel pump. The fuel injection pressure could be varied from 30 to 150 bar by changing the pressure of a nitrogen vessel. A port fuel injector, originally used for Mazda 6 vehicles model year 2003–2006, was mounted at the premixing chamber. The port fuel injector directly supplied the fuel from the low-pressure fuel pump, and injection pressure was fixed to 3.5 bar for all cases.

While both of the injectors used for direct injection and premixed injection were electrically controlled, the gaseous fuels (methane or hydrogen) directly flowed to the premixed chamber, but the flow rate was controlled by a mechanical pressure regulator attached to the gas vessel.

Because of differences between the efficiency of each fuel delivery system, manifold absolute pressure (MAP) has to be differentiated in order to obtain the same NIMEP. Due to the absence of charge-cooling effect, MAP of premixed gasoline injection system needed to be 5-7 % higher than that of the gasoline direct injection system. In the case of premixed methane, MAP needed to be 5-10 % higher than that of gasoline premixed injection system due to the lower volumetric heating value of methane.

2.2.3 Sensors and auxiliary instruments

Crank angle degree measurement
The crankshaft position was measured using the optical encoder, which is connected to the front of the crankshaft. The encoder's bottom dead center (BDC) signal was aligned with that of cylinder #1 (firing cylinder) on the engine. The engine speed was also calculated from this BDC signal.
Lambda sensor
Lambda value was not controlled by a feedback control in this study, but was monitored using a Horiba MEXA-700 lambda meter and an universal exhaust gas oxygen (UEGO) sensor. The sensor was mounted in the downstream of the custom-made exhaust manifold.

Pressure sensors
The in-cylinder pressure was measured using a Kistler 6052C piezoelectric pressure transducer. The signal was amplified and converted to an analog signal through a type 5010b charge amplifier. This transducer and charge amplifier combination experiences an offset drift, requiring the signal to be pegged to a value provided from another pressure measurement.

In this study, pegging was done by the intake manifold pressure. The original set up has an intake MAP sensor right behind the throttle body, which is not suitable for pegging. For accurate pegging, the MAP sensor was implemented at the intake runner for cylinder #1, approximately 5 cm upstream of the intake manifold. The pegging was done at the beginning of the compression stroke, where the intake manifold and the cylinder contents are in mechanical equilibrium due to the low piston and flow speeds.

Fuel pressure was measured by Omegadyne pressure sensor (PX309-5KG5V). The sensor was located approximately 10 cm upstream of the fuel rail.

Thermocouple
A number of K-type thermocouples were mounted on the system to monitor the status of the system. These included thermocouples in the external supercharging system, intake manifold, premixing chamber and external coolant circuit. Thermocouples mounted on the premixing chamber and coolant circuit were also used for feedback control of each subunit.

Data acquisition (DAQ) system
Data from most of these instruments mentioned above were collected using a National Instruments data acquisition system along with a custom made National Instruments LabView program. Data were recorded every crank angle degree, and one data set
contains data from 100 cycles. This DAQ system independently operates with the DMS 500.

The schematic diagram of the engine and instrumentation setup is illustrated Figure 2-12.

![Schematic Diagram](image)

Figure 2-12: Schematic diagram of the experimental setup and the sensor locations

2.2.4 Experimental Procedures

The term cold-start condition used in this study refers the status where all the engine components maintain constant temperature while the coolant temperature is fixed at 25 °C. Therefore, the engine needs to be in steady state before any measurements.

Figure 2-13 shows the change in the accumulation mode PN for direct and pre-mixed gasoline injection during the fast idle operation. The engine was motored before firing, and firing started at 0 second and never stopped. Data were taken every 10 seconds, and normalized by the PN emission at the end of experiments (500 seconds).
Figure 2-13: Change in the accumulation mode PN emission during the cold idle operation

As shown in Figure 2-13, it takes approximately 1 minute for stabilizing PN emission for the direct injection case, and 6 minutes for the premixed injection cases. Therefore, all the data in this study were measured after those stabilizing periods. It is guessed that the time required for PN emission to reach the steady state value is much longer in premixed injection case because the absolute PN emission level is much lower in premixed injection than that of direct injection.

One data set contains at least three data points which were obtained from averaged PN concentration for 10 seconds. The entire measurement process was repeated if any fluctuation between data points was found in a single data set.

Before firing, the engine was motored to reach desired speed and the throttle valve was adjusted. Due to mechanical friction caused by non-firing three cylinders, motor assist was required during firing for low load operation (lower than 6 bar NIMEP). In case of high load operation, the motor was turned off after firing starts.

For the particle sampling experiments, two TEM grids were assembled to a grid holder before the engine operation. The holder was inserted into the sampling port after the engine operation was stabilized, then particles were sampled for a few minutes, depending on the PN concentration of exhaust gas. The holder was removed from the port after the engine stopped.
The engine was motored for a few minutes before/after firing to blow out any residuals remain in the cylinder or the exhaust port. The entire procedure is summarized in Figure 2-14.

![Experimental Procedure Diagram](image)

Figure 2-14: Experimental procedure
Chapter 3

Direct injection experiments

As a starting point, this chapter summarizes the PN emission pattern of DISI engines under various operating conditions (e.g., fuel injection timing, RPM, NIMEP, oxygen mole fraction).

For the first part, the influence of fuel injection timing on PN emission was measured for various NIMEP and RPM. The optimization of fuel injection timing and related findings are described in the second part. The influence of oxygen mole fraction on soot formation, investigated using a nitrogen dilution system, is described in the last part.

3.1 Experiments description

Figure 3-1 shows the schematic diagram of the experimental setup for this chapter. Only gasoline direct injection method was used for the work reported in this chapter, and fuel injection pressure was fixed at 50 bar. Nitrogen dilution was used to determine the influence of combustion temperature. Lambda and the spark timings were always fixed to 1.00 and MBT timing, respectively.

The engine speed was controlled by the motor (for low load) or the dynamometer (for high load). For cases where NIMEP = 8 bar, supercharging was required. The fuel injection duration was varied from 900 to 2800 µs. Engine operating parameters depending on engine load are summarized below.
Table 3.1: Engine operating parameter

<table>
<thead>
<tr>
<th>NIMEP</th>
<th>MAP (bar)</th>
<th>Injection duration (μs)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.0</td>
<td>0.40-0.42</td>
<td>980-1050</td>
<td>Motor assist required</td>
</tr>
<tr>
<td>4.0</td>
<td>0.61-0.63</td>
<td>1600-1800</td>
<td>Motor assist required</td>
</tr>
<tr>
<td>6.0</td>
<td>0.79-0.82</td>
<td>2100-2300</td>
<td></td>
</tr>
<tr>
<td>8.0</td>
<td>0.98-1.02</td>
<td>2600-2800</td>
<td>Supercharging required</td>
</tr>
</tbody>
</table>

As can be seen in Table 3.1, the required MAP was higher than ordinary operating conditions at high NIMEP because the cam phasers were inactivated, resulting in lower volumetric efficiency for operation under high load.

3.2 Experimental result

3.2.1 Influence of RPM, NIMEP on PN emission

It was already known that fuel injection timing is a key factor for PN formation in DISI engines because it determines the quality of the mixture formation in DISI engines [4]. Therefore, injection sweep experiments were performed at various RPM and NIMEP.

For injection sweep experiments, start of fuel injection (SOI) was chosen as a
parameter. SOI was varied from the top dead center (TDC) intake to TDC compression. The sweep interval was 20 CAD from the TDC intake to the bottom dead center (BDC) compression, 10 CAD from BDC compression to 90 ABDC compression, and 5 CAD from 90 ABDC compression to TDC compression. Injection timing was limited by the combustion stability; if COV was higher than 10%, it was considered as very unstable combustion, and the data was discarded. SOI timings and corresponding motoring pressure are illustrated in Figure

![Figure 3-2: fuel injection condition, cam timings and motoring pressure](image)

Because the fuel injection timing affects the fuel conversion efficiency, NIMEP was also influenced by fuel injection timing. Operation was at constant fuel consumption, so to keep lambda constant, the throttle was changed to compensate for change in the volumetric efficiency. As SOI deviated from the optimal timing (SOI that shows the highest NIMEP), NIMEP decreased. However, no compensation was made for the NIMEP change, meaning that NIMEP in this experiment refers to the maximum NIMEP for each case.

In general, the operating regime was wider at lower speed, and lower load conditions, but it was always possible to obtain very stable combustion (COV <3%, and NIMEP drop<10%) when the SOI was between TDC intake and BDC compression.

In addition to four representative operating points, two additional points were
chosen to investigate the influence of RPM or NIMEP independently, as shown in Figure 3-3.

Figure 3-3: Experimental conditions for the injection sweep experiment

Figure 3-4 shows the particle number concentration and mean diameter for fast idle (1200 RPM and 2 bar NIMEP) condition. It can be seen that the accumulation mode PN significantly changed as the SOI timing varied. The change of nucleation mode PN was relatively smaller compared to the accumulation mode PN, but it had the opposite trend than that of the accumulation mode PN.

Figure 3-4: PN emission as a function of SOI

To investigate the trend of PN emission as a function of fuel injection, the injection timings were divided into three different groups, as shown in Figure 3-5. Early injection was defined from SOI timings TDC to 60 ATDC, where the accumulation mode PN was high. For these SOI timings, strong piston impingement was expected. From 80 ATDC and later SOI timings until combustion became very unstable was
grouped as mid injection timing. During mid-injection, piston impingement became weaker compared to early injection cases. A very narrow regime where combustion became stable again during late compression stroke was defined as late injection. For late injection, piston impingement became strong again, and the mixture stratification was expected because the time for mixing was very limited compared to other cases.

![Figure 3-5: Classification of fuel injection timing](image)

Figure 3-6 shows the PN spectra of each injection timing group. The nucleation mode peak was bigger for SOI timings that resulted in low accumulation mode PN emission (Figure 3-6b), and the nucleation mode peak was smaller for SOI timings that had higher PN emissions (Figure 3-6a and 3-6c).

A fraction of the nucleation mode particles were absorbed to the accumulation mode particles for SOI timings where the accumulation mode PN was higher. This is because volatile particles can be absorbed to solid accumulation mode particles. However, it is expected that the portion of absorbed particle was not significant. Because the amount of materials in a particle is proportional to the cube of the particle diameter, it takes approximately a thousand 10 nm particles to change the size of a 100 nm particle. Therefore, given the fact that the order of nucleation mode particles is not changed much, the fraction of absorbed volatile particles is small.

The accumulation mode PN shows approximately two orders of magnitude differ-
ence as a function of SOI. Generally speaking, the accumulation mode PN emission becomes lower as SOI timing changes from TDC intake to BDC compression. It is guessed that PN emission was higher for early SOI timings due to the severe liquid fuel impingement on the piston, and PN emission was reduced as SOI was retarded because the fuel was sprayed into air in these timings, resulting in reduced piston impingement.

However, PN emission started to increase again when the SOI timing was retarded more than the BDC compression. This was mainly due to the reduced mixing time, which increased the inhomogeneity of the mixture. Inhomogeneous mixtures increased PN emission because they caused poor combustion stability, and generated locally fuel-rich mixtures. In such cases, there was a large amount of hydrocarbons for particle surface growth, with little oxygen for particle oxidation.

It was not possible to maintain stable combustion when the SOI timing was retarded more than 70 CAD ABDC compression (gray area). However, the combustion
becomes stable again with very high PN when the SOI was close to BDC compression. In this case, the stability was improved through the interaction of fuel spray with the piston crown, because the bowl on the piston delivered some of the injected fuel toward the spark plug, resulting in a rich mixture around the spark plug. This fuel-rich mixture enables robust combustion, but the impingement of liquid fuel on the piston crown promotes soot formation, similar to early SOI cases.

Regarding mean particle diameter, both modes showed a similar pattern. When the total number of particulates is lower, mean diameter was also smaller. The mean diameter differs up to factor of two, but the geometric standard deviation was similar (1.5-2.0 nm) for all SOI timings.

Figure 3-7: Changes in PN emission as a function of SOI

Figure 3-8: Changes in particle mean diameter as a function of SOI
PN emissions under different operating conditions are illustrated in Figure 3-7 and Figure 3-8. Because stable combustion with late injection timings was impossible except in two cases (1200 RPM /2 bar NIMEP) and (1400 RPM /4 bar NIMEP), corresponding SOI timings are omitted.

Quantitatively, the change of PN emission (as function of SOI) shows similar trends regardless of RPM/NIMEP. The accumulation mode PN emissions were highest for very early injection timings, and were the lowest when the SOI was between 60 and 100 CAD ATDC. For this low PN emission regime, NIMEP was also highest. For low load cases, PN emissions did not change much after this region, and kept constant unless SOI was retarded further than BDC compression. However, PN emission rapidly increased after SOI = 100 CAD ATDC in high load operations. Except for this region, the PN concentration was proportional to the engine load. On the other hand, there was no specific trend in the case of the nucleation mode PN emission.

Mean diameter and PN emission showed the same trend for the accumulation mode. For SOI timing that shows high PN concentration, the mean diameter was large as well. In the case of the nucleation mode, there was no clear relation between mean diameters and PN concentrations, showing a similar pattern for all cases. In general, mean diameters were approximately 25–30 nm for early SOI timings (TDC intake–80 CAD ATDC), and 7–15 nm for other cases.

### 3.2.2 Optimization of injection timing

For the comparison with the premixed charge experiments in Chapter 4, the optimum fuel injection timings (SOI timing that shows the lowest accumulation mode PN concentration) were investigated. Although the general trend of PN emission as a function of SOI was revealed in previous experiments, it was also found that 20 CAD of interval is not narrow enough to find the minimum possible PN emission, especially when operating under high load.

Figure 3-9 shows the change of PN emission when SOI deviated from the optimal timing. It is shown that 5 CAD deviation of SOI timing increased PN emission by 5x, at most, for high load operation. However, the same deviation does not bring a
significant change in low load operation. Therefore, 5 CAD SOI sweep experiments were performed to find the minimum PN emission at 16 different operating points, as shown in Figure 3-10. Other experimental conditions were the same in the injection sweep experiments.

The minimum PN emissions and corresponding SOI timings for each condition are given in Figure 3-11 and Table 3.1. As shown in Figure 3-11, the accumulation mode PN emissions increased as engine load increased and decreased as RPM increased.

Increased engine load generally increased PN emission because the amount of fuel injected also had to be increased during high load operation. As the amount of fuel injected increases, the fuel spray becomes longer and has stronger momentum, thereby
becoming more likely to contact with the piston or cylinder wall before vaporization. However, engine speed can influence PN emission in both directions. Faster engine speed could increase PN emission because the time for evaporation is reduced, but could decrease PN emission because increased piston speed induces stronger charge motion which helps air-fuel mixing. It is guessed that the latter was greater under these operating conditions.

Table 3.2: Optimum SOI timings (CAD ATDC intake)

<table>
<thead>
<tr>
<th>NIMEP</th>
<th>1200 RPM</th>
<th>1400 RPM</th>
<th>1600 RPM</th>
<th>1800 RPM</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 bar</td>
<td>110-130</td>
<td>115</td>
<td>95</td>
<td>95</td>
</tr>
<tr>
<td>4 bar</td>
<td>80</td>
<td>75</td>
<td>70</td>
<td>70</td>
</tr>
<tr>
<td>6 bar</td>
<td>75</td>
<td>70</td>
<td>70</td>
<td>70</td>
</tr>
<tr>
<td>8 bar</td>
<td>70</td>
<td>65</td>
<td>65</td>
<td>65</td>
</tr>
</tbody>
</table>

The optimal SOI timing advances as RPM increases or engine load increases. The schematic diagram of the geometric relationship between liquid fuel spray and the piston is drawn in Figure 3-12. For the optimal SOI timings for 2 bar NIMEP cases, there is almost no contact between fuel spray and the piston, but fuel spray touches the piston for the high load optimal SOI timings.

LTG, the experimental engine, is a wall-guided DISI engine which uses a piston crown to generate a tumble motion that aids air-fuel mixing. As shown in Figure
3-7, the general trend of the accumulation mode PN emission as a function of SOI is roughly the same because the geometrical relationship between fuel spray and piston does not change much. However, the quantity and the phasing of the SOI dependence changes with load. For low load cases, only a small amount of fuel is injected, hence most of the fuel can vaporize without the tumble motion, as shown in Figure 3-12. Therefore, the liquid fuel film on the piston top is a main source for particulate matter, therefore avoiding contact between fuel spray and the piston was better for reducing PN emissions.

![Diagram of fuel spray-piston interaction in low load operation](image)

**Figure 3-12:** Schematic diagram of fuel spray-piston interaction in low load operation

However, the situation becomes different during high load operation, as illustrated in Figure 3-13. Even if avoiding fuel spray and piston by retarding the SOI timing, fuel spray contacts the cylinder liner, causing severe liner impingement. In addition, the aid of the tumble motion becomes more important as amount of injected fuel increases, thereby the optimal SOI timing advances where fuel spray can contact the piston crown, maximizing the tumble effect. The optimal SOI timing advances as RPM increases because the piston travels faster at higher engine speed.
Because the optimum injection timing is mainly determined by the geometrical relationship between the fuel and piston, it is expected that this optimum timing would not change much under warmed-up conditions. The single-cylinder engine tests performed by a research team at Delphi Powertrain Systems showed that the optimum injection timing is independent of engine temperature [9].

3.2.3 Influence of the nitrogen dilution on PN emission

To investigate the influence of an oxygen mole fraction on the accumulation PN emission, nitrogen dilution was used. When designing experiments, using a cooled EGR was considered for more realistic conditions, but nitrogen dilution was chosen (1) to maintain the oxygen mole fraction for all experimental conditions, and (2) to avoid water condensation issues.

Similar to the experiments in section 3.2.1, injection sweep experiments were performed at six different operating points. The intake oxygen mole fraction was set to 16.7% using nitrogen dilution, which corresponds to the 4:1 dilution ratio (Air : nitrogen). The fuel injection condition was the same, but the span of the injection sweep was much narrower due to increased combustion instability when the mixture was diluted.

In order to operate at the same NIMEP, MAP had to be higher for the diluted charge due to the reduced oxygen mole fraction. However, the amount of fuel injected was 3-5% smaller because of reduced pumping loss. Spark timing was set to MBT,
requiring 15 - 20 CAD of spark advance compared to the fresh-air mixture due to low combustion speed. The other conditions, such as lambda and fuel injection pressure, were the same as in the other experiments.

Figure 3-14: Accumulation mode PN concentration for diluted mixture; at air to oxygen dilution ratio of 4:1 (16.7 % O₂)

Figure 3-14 shows the PN emission patterns for the diluted mixture as a function of SOI. There was no noticeable change in PN emission patterns compared to non-diluted case (Figure 3-7a). PN emission was the highest for very early injection timing, and became lower after SOI = 60 CAD ATDC. The PN emission level maintains a similar level until SOI reaches BDC intake for low load cases, but PN emission started to increase again after SOI = 120 CAD ATDC for high load cases.

Figure 3-15: Normalized accumulation mode PN concentrations
The difference in PN emission between diluted and non-diluted charge is shown in Figure 3-15. This figure shows the values of PN concentrations in diluted charge cases divided by those of non-diluted charge cases for each operating conditions. Therefore, to have values larger than '1' means that the PN emission was increased due to the dilution. As can be seen, dilution worked both ways. In general, dilution increased PN emission for high RPM or high load cases, but reduced PN emission for low RPM cases.

![Figure 3-16: Accumulation mode PN concentration for 1200 RPM](image)

To decouple the influence of RPM and NIMEP, the same experiment was performed at 1200 RPM and four NIMEPs. Figure 3-16 shows the PN emission for both non-diluted and diluted mixtures. The PN emission patterns were similar for both non-diluted and diluted cases. Normalized PN concentration is given in Figure 3-17. When RPM was fixed, it is shown that the dilution reduced PN emission for low load and early injection cases, but increased or did not change PN emissions for other cases.

It is believed that dilution mainly impacts soot formation from the liquid fuel film, because it affects the pyrolysis process that occurs above the liquid fuel film. It is expected that dilution would reduce soot formation by suppressing pyrolysis because dilution reduces the combustion temperature. However, it was found that dilution also could increase the rate of soot formation, as described in Figure 3-18.
Figure 3-17: Normalized accumulation mode PN concentrations

---

Figure 3-18: Effects of nitrogen dilution on soot formation

With the SOI sweep experiments, it is hard to draw a conclusion except for the fact that nitrogen dilution does not change the PN emission patterns of each condition. Relative magnitudes of the effects in Figure 3-18 might be changed, or there could be another soot formation mechanism strengthened due to the dilution. For more detailed investigation, PN emissions from fixed SOI timings were measured at different dilution ratios. The dilution ratio varied from 9:1 to 4:1, as shown in Table 3.3.

SOI timings were fixed to 60 and 90 CAD ATDC intake because these two SOI timings are the earliest and the latest SOI timing that can maintain NIMEP and combustion stability for all operating conditions. Similar to the injection sweep ex-
Table 3.3: Dilution ratios and corresponding oxygen mole fractions

<table>
<thead>
<tr>
<th>Dilution Ratio</th>
<th>Oxygen mole fraction (vol%)</th>
<th>Add nitrogen amount (vol%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No dilution</td>
<td>20.9</td>
<td>0.0</td>
</tr>
<tr>
<td>9:1</td>
<td>18.8</td>
<td>10.0</td>
</tr>
<tr>
<td>6:1</td>
<td>17.9</td>
<td>14.3</td>
</tr>
<tr>
<td>4:1</td>
<td>16.7</td>
<td>20.0</td>
</tr>
</tbody>
</table>

Experiments, lambda and spark timing were set to 1.00 and MBT spark timing for each case. The amount of fuel injected decreases as the dilution ratio increases due to the reduced pumping loss, but the change was less than 7%.

Figure 3-19 shows the change of the accumulation mode PN concentration as a function of oxygen mole fraction.

As can be seen, the accumulation PN emission increased as the dilution ratio increases at 8 bar NIMEP and decreased at 2 and 4 bar NIMEP in Figure 3-19a, while dilution has no influence in Figure 3-19b. It is guessed that dilution did not change the PN emission in SOI = 90 CAD ATDC because the contact between the fuel spray and the piston top was not severe, forming only a small amount of liquid fuel film.

The case in which PN spectrum for SOI = 60 CAD ATDC, is shown in Figure...
In Figure 3-20a and 3-20b, both the nucleation and accumulation mode peak decreased as dilution ratio increased. However, both peaks increased as the dilution ratio increased in Figure 3-20d. In Figure 3-20c, the accumulation peak was not influenced by dilution, but the nucleation mode peak was noticeably larger at the highest dilution ratio.

The effect of dilution might be influenced by operating parameters such as fuel injection pressure, RPM, load and a test engine. An inconsistent effect of dilution depending on engine operating conditions was found at other studies as well, but generally speaking, the EGR is more effective for reducing PN emission at lower loads. Hedge et al. [51], measured the particle mass and number with 25% cooled external EGR under the operating conditions at 2500 RPM / 6.7 bar brake mean effective...
pressure (BMEP) and 3500 RPM / 9.6 bar BMEP. Particle mass was reduced for both cases but particle number was reduced by 91% in 2500 RPM / 6.7 bar BMEP case but increased 1.98 times for 3500 RPM / 9.6 bar BMEP case, although the solid particle number (particles measured at the downstream of oxidation catalyst) was reduced for both cases. Engine experiments performed by Pei et al. [42] showed that the accumulation mode PN emission decreased as the EGR rate increased from 0 to 20% at 2000 RPM and 1 and 3 bar of BMEP conditions, but the amount of abatement was most effective at 1 bar BMEP. For the 3 bar BMEP condition, there was almost no change in the rate from 0% to 15% EGR. Sabathil et al. [39] found a rising PN emission with increased EGR rate with a 2.2 L naturally aspirated DISI engine at 1000 RPM and 4 bar BMEP condition.

To explain the reason for this inconsistency, a few more experiments were performed in various SOI timings. Because SOI = 60 CAD ATDC is very close to the optimum SOI timing given in Table 3.2, the same experiments were performed at the optimum SOI timings for each operating point. In addition, SOI was further advanced than 60 CAD ATDC (20 CAD ATDC), to have more liquid fuel film on the piston top. With 20 CAD ATDC SOI timing, the combustion stability was maintained, but NIMEP dropped by 5–7% compared to the case of SOI = 60 CAD ATDC.

![Graphs showing the effect of SOI and oxygen mole fraction on PN concentration](image)

Figure 3-21: Accumulation mode PN concentration as a function of the oxygen mole fraction

In Figure 3-21a, the PN emission decreased as dilution ratio increases only in the
case of 4 bar NIMEP. In the case with 2 bar NIMEP, the PN emission was not affected by dilution because the optimal timing was late. In Figure 3-21b, the trend of PN emission was similar to Figure 3-19a; showing reduction only in the case of 2 and 4 bar NIMEP.

From these experiments, it is found that the dilution reduces the accumulation mode PN emission in low NIMEP operating condition with early SOI timings. In order to check whether there is any dependence on RPM, the same experiment shown in Figure 3-19 was performed with fixed RPM. Figure 3-22 and 3-23 show the results for 1200 and 1800 RPM, respectively.

![Figure 3-22: Accumulation mode PN concentration as a function of the oxygen mole fraction at RPM = 1200](image1)

(a) SOI = 60 CAD ATDC

(b) SOI = 90 CAD ATDC

![Figure 3-23: Accumulation mode PN concentration as a function of the oxygen mole fraction at RPM = 1800](image2)

(a) SOI = 60 CAD ATDC

(b) SOI = 90 CAD ATDC
Similar to previous results, the accumulation mode PN emission was only affected by dilution with SOI = 60 CAD ATDC. The PN emission decreased in 2 and 4 bar NIMEP cases. In 6 bar and 8 bar cases, the PN emission remained the same at 1200 RPM, but increased as the dilution ratio increased in 1800 RPM.

As the last step, the impact of dilution was tested at various SOI timings under fast idle condition. The fast idle condition, 1200 RPM with 2 bar NIMEP, was chosen because the positive impact of dilution (reducing PN emission) was largest under this condition. SOI timings and corresponding the accumulation mode PN emissions are shown in Figure 3-24.

![Figure 3-24: Accumulation mode PN concentration as a function of the oxygen mole fraction with various SOI timings at fast idle condition; 1200 RPM and 2 bar NIMEP](image)

The results shown in Figure 3-24 reconfirms the previous results. Dilution affected soot formation only if there was strong piston impingement of fuel spray. This result proves that nitrogen dilution affects the soot formation from liquid fuel film, but cannot explain why it reduces PN emission only in cases with low NIMEP.

The main reason for why it is hard to draw a consistent conclusion is that all possible soot formation mechanisms are involved in direct injection experiments. Further investigation on the contribution of soot formation from the pyrolysis of the fuel vapor derived from liquid fuel films will be addressed in Chapter 5.
3.3 Findings

As the first step of the thesis, this chapter investigates the PN emission of DISI engines with a conventional gasoline direct injection system. No modification was made on the fuel delivery system except that the fuel pressure was fixed to 50 bar by nitrogen pressurization. Fuel injection timing (SOI; start of injection) was chosen as the main parameter. The accumulation mode PN emission as a function of SOI timing was measured under the various operating conditions. In addition, nitrogen dilution experiments were performed to determine the influence of combustion temperature. Through a series of experiments, the following things were revealed.

Injection sweep experiments
The accumulation mode PN emission varied more than two orders of magnitude as SOI varied. The nucleation mode PN emission was not strongly influenced by SOI timings, but there was a slight trade-off relationship between the nucleation mode and accumulation mode PN emission. The accumulation mode PN emission was higher at early SOI timings, and was reduced as SOI timing was retarded around to the late intake stroke due to the reduced piston impingement. However, the PN emission started to increase again when SOI timing was further retarded due to the reduced time for air-fuel mixing. This trend was not strongly dependent on RPM and NIMEP because the geometrical relationship between fuel spray and piston crown/cylinder wall was not changed. However, a window of SOI timings that records low PN emission was much narrower at high load operation. Observation suggests that the nucleation mode particles are formed in the nominal flame, where the fuel/air ratio is not far from stoichiometric. Therefore, the formation of nucleation mode particles is not sensitive to the injection timing. The accumulation mode particles, however, are formed from the much richer regions created by the fuel vapor derived from liquid fuel films. Thus, the particle formation is sensitive to the fuel injection timing. When fuel is injected around near lower part of the intake stroke, there is not much liquid fuel film on the walls, hence very little accumulation mode PN is observed.
SOI timing optimization experiments
The optimum injection timing, SOI timing that shows the lowest PN emission, advances as RPM increases and engine load increases. The absolute amount of the accumulation mode PN emission (at these optimum timings) increases as engine load increases because more fuel is introduced. However, it decreases as RPM increases because the better fuel air mixing associated with the faster charge motion at higher speeds is more critical than the less available mixing time. Under at high load operating condition, a slight deviation (5–10 CAD) from the optimum SOI timing results in a significant increase of the accumulation mode PN emission, but the effect of deviation is less significant at low load operation. Under the fast-idle condition, reducing piston impingement is important for reducing PN emission, but maximizing air-fuel mixing becomes more important as engine speed and load increases.

Nitrogen dilution experiments
Nitrogen dilution does not change the PN emission pattern determined by the SOI timings. Similar to the non-diluted charge experiments, the PN emission is lowest when SOI timings are between 60 and 100 CAD ATDC. However, the absolute amount of PN emission is changed due to the nitrogen dilution, and the effect of dilution on the PN emission depends on operating conditions. For 60 ATDC intake or earlier SOI timings, dilution reduces PN emission at lower NIMEP, but increases PN emission for higher NIMEP. This trend is stronger when the SOI timing is close to the optimal injection timing and RPM is high. For later fuel injection timings (SOI timing 90 ATDC intake or later), dilution does not substantially affect the accumulation mode PN.
Chapter 4

Premixed charge experiments

In Chapter 3, general trends of PN emission in DISI engines as a function of various operating parameters were investigated. Because gasoline direct injection process involves all PN formation mechanisms (non-fuel contribution, flame propagation in locally rich mixture and liquid fuel film), it is difficult to quantify the contribution of each mechanism.

To decouple some of these fundamental effects which contribute to PN formation, fully homogeneous (premixed) mixture operation was chosen for the work reported in this chapter. Because the fully premixed charge does not form liquid fuel film, it is possible to eliminate the effect of liquid fuel film on soot formation.

The first part of this chapter is dedicated to defining the background PN emission—the minimum PN emission that can be obtained from engine experiments. The change of this background emission, which depends on engine operating conditions and fuel composition, are described in the second part. Theoretically, operation with a fully mixed charge with $\lambda = 1.00$ does not form soot. Therefore, this background emission can be considered soot generated from non-fuel sources. On the other hand, the last part of this chapter describes the experimental results from rich mixture of premixed gasoline, which are related to PN emissions caused by flame propagation in locally rich mixture.
4.1 Experiments description

The schematic diagram for the experimental set-up is shown in Figure 4-1. Compared to the experimental set-up in Chapter 3, the fuel delivery system was changed, but the other apparatus remained the same. Gasoline was delivered by an automotive port fuel injector, while gaseous fuels were controlled by a mechanical valve. The fuel injector was connected to the inlet of the premixing chamber, and the gaseous fuel line was connected to the upstream of the throttle valve. Only one of the two non-DI fuel systems (either gasoline or gaseous fuel) is used at a time.

The operating points were the same, but the MAP had to be 5-10% higher compared to direct injection cases due to the lack of charge cooling effect. Because the background PN emissions (either from premixed gasoline or methane) were very low, they were significantly influenced by uncontrollable ambient conditions, such as intake temperature and humidity. Therefore, some of the experimental results in this chapter have different absolute emission level. As a solution, the experimental data were normalized to a reference value (usually the PN concentration of 2 bar NIMEP operation), if necessary.
4.2 Experimental result

4.2.1 Background PN emission

The first part of the premixed charge experiments was done to obtain the minimum possible PN emission, which will be called the background PN emission in this study. It is believed that the background PN emission is derived from non-fuel sources, such as lubrication oil, abraded metal or particles floating in the air.

Theoretically, any fuel that is fully-mixed with lambda higher than a critical value (around 0.5) should not form any soot because carbon atoms will exist as CO rather than soot according to the law of thermodynamics. However, many experiments showed that soot is formed under premixed operation cases with the lambda higher than the critical value.

The best option is using hydrogen as fuel because it is possible to conclude that any PN emission from hydrogen combustion is not fuel-originated. However, hydrogen has very distinctive (and different) combustion characteristics compared to hydrocarbon fuels: it has very fast combustion speed, which results in a high pressure rise rate. Therefore, it is difficult to maintain $\lambda = 1.00$ combustion, unlike hydrocarbon fuels. Therefore it is hard to equate combustion conditions that may influence soot formation such as NIMEP, maximum pressure, MAP.

Figure 4-2: Pressure traces for methane and hydrogen combustion
Figure 4-2 and Table 4.1 show the pressure trace and corresponding characteristics of methane combustion under fast-idle condition and those of five different hydrogen combustion conditions at the same RPM.

Table 4.1: Comparison between methane and hydrogen combustion

<table>
<thead>
<tr>
<th>Fuel</th>
<th>MAP (bar)</th>
<th>NIMEP (bar)</th>
<th>lambda</th>
<th>Spark (ATDC)</th>
<th>P_{peak} (bar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Methane</td>
<td>0.465</td>
<td>2.00</td>
<td>1.00</td>
<td>-28</td>
<td>13.0</td>
</tr>
<tr>
<td>Hydrogen 1</td>
<td>1.14</td>
<td>1.50</td>
<td>-5</td>
<td>14.3</td>
<td></td>
</tr>
<tr>
<td>Hydrogen 2</td>
<td>1.04</td>
<td>1.50</td>
<td>-10</td>
<td>16.0</td>
<td></td>
</tr>
<tr>
<td>Hydrogen 3</td>
<td>0.75</td>
<td>1.50</td>
<td>-15</td>
<td>18.5</td>
<td></td>
</tr>
<tr>
<td>Hydrogen 4</td>
<td>0.73</td>
<td>1.50</td>
<td>-20</td>
<td>20.4</td>
<td></td>
</tr>
<tr>
<td>Hydrogen 5</td>
<td>0.819</td>
<td>1.99</td>
<td>2.50</td>
<td>-10</td>
<td>14.6</td>
</tr>
</tbody>
</table>

NIMEP from hydrogen combustion is approximately a half of that from methane combustion when MAP and maximum pressure were equated (Hydrogen 1). To obtain the maximum in-cylinder pressure (Hydrogen 5), MAP had to be almost doubled with very lean mixture. Similar peak pressure rather than NINEP was sought because the non-fuel PM sources such as lubricant oil particles carried out by the crevice out flow are more dependent on peak pressure than NIMEP. Figure 4-3 shows the corresponding accumulation mode PN emission and PN spectrum. As can be seen, the accumulation mode PN concentrations from methane and hydrogen combustion were similar, and no distinctive accumulation mode peak was observed for all cases.

Maier et al. performed a similar experiment. They compared PN emission from premixed hydrogen and methane to gasoline direct injection under various engine loads [58]. In that study, differences in the combustion characteristics between hydrogen and gasoline were compensated by nitrogen dilution of intake air, and the pressure or heat release traces were matched to those of gasoline operation. The study also showed that difference in PN emission between premixed methane and hydrogen was negligible; very small amount of particles larger than 30 nm could be measured regardless of operating condition for both fuels.

Therefore, methane was chosen as the fuel for the background PN emission ex-
experiment, instead of hydrogen. However, a few more experiments were performed to check whether there was any possibility to obtain a significantly lower level of the accumulation mode PN emission than that from stoichiometric premixed methane combustion.

**PN concentration of ambient air**

The accumulation mode PN concentration from ambient air (of the test cell) was measured and compared with that from methane combustion under fast-idle operation. To obtain the most accurate PN concentration, it was required to adjust the dilution ratio so that the total PN concentration fit the dynamic range of the DMS 500, as shown in Figure 4-4.

Considering the PN concentration, the dilution ratio 1 (undiluted gas) is suitable for ambient air, and this was the same for methane combustion under fast-idle condition. However, it was not possible to make the dilution ratio '1' because the high
temperature of the exhaust gas could damage the sampling system unless it were cooled by dilution.

The minimum dilution ratio required for the sampling engine exhaust is '5', therefore the ambient air was measured with two different ratios. The accumulation mode PN is shown in Figure 4-5a, and Figure 4-5b shows the corresponding PN spectrum.

![Figure 4-5: PN emission from ambient air and methane combustion](image)

Figure 4-5a shows that the accumulation mode PN of ambient air was approximately 1/3 of the accumulation emission from methane combustion, but they were approximately the same when the dilution ratio was equated to '5'. For methane combustion, the nucleation mode peak was higher compared to ambient air due to the combustion. However, the PN spectra were similar in the accumulation mode range, and did not show a noticeable accumulation mode peak.

**PN emission from motoring**

As a next step, PN emission from motoring was measured. Using an external supercharging system, the peak pressures were equated (the CAD where the peak pressure occurred was not the same). With the supercharging system, it was possible to obtain the peak pressure of 27 bar, which is similar to that of methane combustion with 4 bar NIMEP operation, as shown in Table 4.2. Corresponding accumulation mode PN emissions are shown in Figure 4-6, which confirms that PN concentrations were similar for both cases.

**The change of the accumulation mode PN after ignition-off** The decay of accumulation mode PN concentration was measured after ignition-off. The difference
Table 4.2: Peak pressure from methane combustion and motoring pressure

<table>
<thead>
<tr>
<th>NIMEP (bar)</th>
<th>P_{peak}(bar)</th>
<th>Required MAP (bar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.0</td>
<td>15-17</td>
<td>0.91-0.97</td>
</tr>
<tr>
<td>4.0</td>
<td>23-27</td>
<td>1.49-1.53</td>
</tr>
<tr>
<td>6.0</td>
<td>40-42</td>
<td>-</td>
</tr>
<tr>
<td>8.0</td>
<td>50-52</td>
<td>-</td>
</tr>
</tbody>
</table>

Figure 4-6: The accumulation mode from methane combustion (closed symbols) and motoring (filled symbols)

between the motoring experiment and this experiment is that the heat (from combustion) slowly dissipated over time in this case, while the engine remained cold in the motoring experiment. In addition, the peak pressure was equated in the motoring experiments, while the MAP was equated in this case.

In Figure 4-7, the engine was shutting off at time = 0. After shutting off, the engine speed was maintained by the motor. The throttle was fixed. Symbols on the left side of Figure 4-7 are the accumulation mode PN concentration during firing, and PN concentrations were measured at 1 s intervals after ignition off.

There was no change in the accumulation mode PN concentration in the cases of 2 bar and 4 bar NIMEP, the same as for the motoring test. At high load (6 bar and 8 bar of NIMEP), the accumulation mode PN concentrations were higher during firing,
but a similar order of PN emission was maintained for a while after shutting off the engine.

Throughout these experiments, it becomes possible to conclude that the accumulation mode PN emission from methane is background emissions. The accumulation mode PN concentration of the background was similar to that of ambient air, and there is no noticeable accumulation mode peak in the PN spectrum.

In terms of interpreting the accumulation mode PN concentration, one thing to notice is that there might be an error in the absolute number of particulates. Because the accumulation mode PN level from methane combustion is as low as ambient air, dilution should not be used for accurate results. However, the nitrogen dilution must be used to prevent damages from high temperature exhaust gas, and this off-set of the dilution ratio may generate inaccuracy in the sampling system. In addition, the fitting offered by the DMS 500 may not work well since no accumulation mode peak could be identified. Therefore, it is possible to say that the accumulation mode PN emission from the methane combustion is as low as ambient air, but it is impossible to define a background emission as a certain PN concentration.

Throughout this chapter, the accumulation mode PN concentration from methane under fast-idle condition will be frequently used as the reference value because it is the lowest accumulation PN emission.
4.2.2 Influence of RPM and NIMEP on PN emission from premixed mixture

In the previous section, it was shown that the accumulation PN emission from methane combustion under fast-idle condition was negligible. In this section, the accumulation mode PN concentrations were measured for various RPM and NIMEP.

Figure 4-8: Experimental condition for premixed methane combustion

Figure 4-8 shows the experimental condition. Four different engine RPM were chosen (same as for the other experiments), but NIMEP was swept at 0.5 bar intervals to investigate the change of the accumulation mode PN emission more precisely.

Figure 4-9: Accumulation mode PN emission from methane combustion

The accumulation mode PN concentration as a function of NIMEP and RPM is
shown in Figure 4-9. Unlike gasoline direct injection, the accumulation mode PN is not substantially affected by RPM. The change of the accumulation mode PN concentration as a function of NIMEP becomes more clear if the experimental regime is divided into two parts, as indicated in Figure 4-10.

As shown in Figure 4-11, the accumulation mode PN concentration remains constant around the $10^4$ particles/cc, regardless of NIMEP at lower loads. However, for high load operation, the PN emissions starts to exponentially increase as engine load increases, reaching $2 \times 10^5$ particles/cc at 8 bar NIMEP.

Figure 4-11 shows the PN spectrum for selected cases (1200/2 bar, 1400/4 bar, 1600/6 bar and 1800/8 bar of RPM/NIMEP). Corresponding the accumulation mode
concentrations were $9.5 \times 10^3$, $5.3 \times 10^3$, $1.6 \times 10^4$ and $1.8 \times 10^5$ particles/cc, respectively. No accumulation mode peaks were observed for all cases, although the accumulation mode PN concentration differed up to a factor of 30. Instead, the entire PN spectrum shifted upwards as the PN emission increased.

It is guessed that the increase in the accumulation mode PN emission at the higher load is due to other sources (e.g., crevice out-gassing) influenced by high pressure and temperature, than rather combustion. In the case of hydrogen combustion summarized in Table 4.1 and Figure 4-3a, the accumulation mode emission PN tends to increase as maximum cylinder pressure increases while other operating parameters are the same. To confirm this issue, premixed gasoline experiments were performed. If the combustion of fuel contributes to the background PN emission, the PN emission from premixed gasoline should be higher than that from methane. This is because gasoline has a higher C/H ratio, and more likely to condense due to its higher boiling point.

The experimental conditions were simplified to 16 operating points (same as for the gasoline direct injection cases), as shown in Figure 4-12. MAP was slightly lower (around 5%) in the cases with premixed gasoline, compared to the methane experiment, and the spark timing was set to MBT.

![Image of experimental conditions](image.png)

Figure 4-12: Experimental condition for premixed gasoline experiments

Figure 4-13 shows the accumulation mode PN concentration in premixed gasoline. The symbols are the accumulation mode PN concentration from premixed gasoline,
and the lines are those from methane combustion. The corresponding PN spectrum is shown in Figure 4-14.

Figure 4-13: Accumulation mode PN emission from premixed gasoline combustion (symbols) and premixed methane (lines)

Figure 4-14: PN spectrum of premixed gasoline (closed symbols) and methane (open symbols) combustion for selected operating conditions

Both the Figure 4-13 and 4-14 confirm that there is no fundamental difference between methane and premixed gasoline combustion regarding PN emission. The accumulation mode PN concentrations and PN spectra are similar when the engine operating condition is the same. Therefore, it is possible to conclude that the chemical composition of fuel is insignificant on premixed charge combustion with stoichiometric operating condition, as long as the fuel is well vaporized and fully mixed with air
before entering the combustion chamber.

Figure 4-15 shows the accumulation mode PN emission from three different fuel delivery methods. To see the relative magnitude of each case, all values were normalized by the accumulation mode PN concentration of methane combustion at 1200 RPM / 2 bar NIMEP operating condition. For gasoline direct injection, SOI timing was set to optimum timing, as obtained in Section 3.2.2.

![Figure 4-15: Comparison of accumulation mode PN emission depending on RPM, NIMEP and fuel delivery methods; methane(closed symbols), premixed gasoline(hollow symbols) and direct injection gasoline(half closed symbols)](image)

As previously mentioned, the accumulation mode PN emission from premixed gasoline combustion was almost the same as that of methane combustion. However, the accumulation mode PN emission with gasoline direct injection was 5–45 times higher compared to that of methane. Table 4.3 shows the normalized accumulation mode PN from gasoline direct injection. The accumulation mode PN from gasoline direct injection was divided by that from methane combustion for each operating point.

According to Table 4.3, the difference between gasoline direct injection and methane was more severe at low engine load. This is because the accumulation mode PN emission from methane combustion was very low at low engine load. In contrast, gasoline direct injection generates at least $10^5$ particles/cc. This trend is more clearly shown at low RPM because the accumulation mode PN emission from direct injection is
Table 4.3: Normalized accumulation mode PN emission of gasoline direct injection

<table>
<thead>
<tr>
<th>NIMEP</th>
<th>1200 RPM</th>
<th>1400 RPM</th>
<th>1600 RPM</th>
<th>1800 RPM</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 bar</td>
<td>34.7</td>
<td>45.0</td>
<td>29.4</td>
<td>12.9</td>
</tr>
<tr>
<td>4 bar</td>
<td>14.0</td>
<td>25.2</td>
<td>17.8</td>
<td>13.4</td>
</tr>
<tr>
<td>6 bar</td>
<td>11.6</td>
<td>18.8</td>
<td>8.6</td>
<td>8.5</td>
</tr>
<tr>
<td>8 bar</td>
<td>10.3</td>
<td>7.0</td>
<td>5.0</td>
<td>5.7</td>
</tr>
</tbody>
</table>

higher at low RPM while that from premixed charge is not influenced by RPM.

4.2.3 The accumulation mode PN emission from premixed, but fuel-rich charge experiments

In the previous two sections, the background PN emission, which is related to non-fuel contribution on soot emission, was investigated using stoichiometric premixed charge. This section is dedicated to describing the investigation of soot formation caused by flame propagation in locally fuel-rich mixture. Fully premixed, but fuel-rich mixture was used for this purpose. Although liquid fuel is considered the main source for soot emission from DISI engines, significant amounts of soot can be formed from a premixed mixture as long as it is rich enough. Abdul-Khalek and Kitterson [59] reported that a gasoline engine which showed PM level as low as air at stoichiometric operation generated more PM than did conventional diesel engines when it was operated a with very rich mixture (λ = 0.4). Because premixed mixture does not have liquid droplets, it is possible to investigate the soot formation without the contribution of liquid fuel film.

The lambda varied from 0.65 to 1.10, with an interval of 0.05. Within this operating range, COV was less than 7%. MAP was fixed with the value at lambda 1.00, and only fuel injection amounts were increased. Therefore, NIMEP decreases at very low lambda, but was less than 10% for all conditions.

Figure 4-16 shows the change of both the accumulation and nucleation mode PN emissions at 1200 RPM and under 2, 4, 6 and 8 bar NIMEP conditions. It can
be seen that both modes had thresholds of PN emission. The PN concentration remained approximately constant up to a certain point ($\lambda = 0.70-0.80$), then started to increased exponentially as lambda decreased.

![Graphs showing PN emission as a function of lambda at different bar pressures for (a) the accumulation mode and (b) the nucleation mode.](image)

Figure 4-16: PN emission as a function of lambda at 1200 RPM

This sudden increase is also shown in the PN spectrum (Figure 4-17). Although the threshold lambda was different, the behavior was similar. There was almost no change for lambda higher than this threshold, but the entire PN spectrum exponentially grew as lambda decreased for lambda lower than the threshold value.

![Graphs showing PN spectrum as a function of lambda at different bar pressures for (a) 2 bar NIMEP and (b) 8 bar NIMEP.](image)

Figure 4-17: Selected PN spectra at 1200 RPM
These threshold lambda values, approximately 0.70–0.80, are much higher than the theoretical lambda obtained from the basic principle of thermodynamics. Theoretically, soot should not be formed under C/O ratio higher than 1.0 (for test fuel, it is equivalent to λ = 0.45), because it is more energetically stable as CO molecules rather than soot particles under such circumstances.

Kettler [3] did the same experiments with a rapid compression machine (RCM) with gasoline and gasoline + toluene blended fuels. The pressure and temperature were set to 8 bar and 720 K, respectively, which corresponds to 1200 rpm and 2 bar gross indicated mean effective pressure (GIMEP) operating condition in the engine experiments. He obtained threshold values similar to theoretical values, as shown in Figure 4-18. It can be seen that the soot yield started to increase exponentially when the equivalence ratio = 2.5 for gasoline, which corresponds to λ = 0.4.

Figure 4-18: Soot Yield vs. Equivalence Ratio, [3]

However, many studies have shown that the threshold lambda for soot formation could be lower than the theoretical value. From the review paper about soot formation, Mansurov [60] argued that soot could be formed at low C/O ratio. In Bunsen burner experiments, many hydrocarbon fuels started to form soot at C/O ratio 0.4–0.5. This critical C/O ratio was similar to values obtained from the engine
experiment because lambda values of 0.70 and 0.75 are equivalent to the C/O ratio of 0.49 and 0.46, respectively, for the test fuel. Marciq et al. also reported a similar trend in soot formation with a vehicle test [40]. They measured the PN emission from a truck equipped with a port-fuel injection system. The truck was tested under the 50 mph cruise condition, and the lambda varied from 1.00 to 0.80. It was found that the PN increased exponentially as lambda decreased. Hageman et al. [61] also performed a similar experiment with a single cylinder DISI engine. They measured the onset of soot formation with a fully premixed and pre-vaporized fuel-air charge as a function of equivalence ratio at 2100 RPM and 334 kPa gross IMEP. The critical equivalence ratio was found to be between 1.345 and 1.349, which corresponds to 0.741 in terms of lambda.

A plausible explanation for the observed leaner threshold for PM formation in engines is that the theoretical limit and the observations from RCM experiments were for a homogeneous bulk reaction which was in equilibrium because of the high combustion temperature. For engine combustion, however, the flame itself is highly non-uniform and not in equilibrium. Thus, the flame condition may favor a leaner threshold based on the unburned fuel/air ratio.

To investigate this threshold behavior in detail, the same experiments were performed at different RPMs. Due to base line drift, the accumulation mode PN concentrations were normalized to PN concentration at $\lambda = 1.00$ for each case.

Figure 4-19 shows the experimental results for four different engine speeds. It is shown that the PN emission patterns were similar for all cases.

To remove the possibility of local condensation, premixed methane experiments were also performed in two different RPMs (1200 and 1800). However, due to the narrow flammability limit of methane, it was impossible to maintain stable combustion for lambda lower than 0.75 in both cases, as shown in Figure 4-20.

The methane experiments did not show threshold behavior, but it is hard to draw any conclusion from this result because the lambda limit was too high for methane to be compared to premixed gasoline.

To quantify the threshold behavior in detail, threshold lambda was computed from
all operating points. The "threshold lambda" was defined as the lambda value that showed a PN concentration 10 times higher than that of lambda = 1, and this value was obtained via interpolation using exponential fitting.

Figure 4-21 shows the threshold lambda for each case. The values were dispersed between 0.70 and 0.80, and neither were influenced by RPM or by NIMEP.

As a next step, the same experiments were performed at different dilution levels to investigate the influence of oxygen mole fraction. Two different dilution levels, 9:1 and 4:1 (corresponding to the oxygen mole fraction of 18.8 and 16.7%, respectively), were used while RPM was fixed at 1200. As in previous experiments, the accumulation mode PN concentrations were normalized by PN concentration at $\lambda = 1.00$. Normalized accumulation mode PN emissions are shown in Figure 4-22, and corresponding threshold lambdas are shown in Figure 4-23.
Figure 4-20: Normalized accumulation mode PN emission from methane combustion

Figure 4-21: Threshold lambda for different RPM and NIMEP

Figure 4-23: Threshold lambda for different dilution ratio at 1200 RPM
Unlike for RPM or NIMEP, there was a slight dependence on dilution ratio. The threshold lambdas were higher at higher dilution ratios. It was reported that the critical C/O ratio decreased as combustion temperature decreased [60]. However, it is hard to conclude that the lowered combustion temperature is the only reason for soot formation at less rich mixture, because the dilution also affects the flammability limit, which could increases quench rate.

Figure 4-22: Normalized accumulation mode PN concentrations with nitrogen dilution
4.3 Findings

In the work reported in this chapter, soot formation under the absence of liquid fuel film or droplets was studied using fully premixed homogeneous charge. A swirl-aided premixing chamber equipped with surface heaters was implemented to aid the mixing of the air-fuel mixture before entering the combustion chamber.

Soot formation from non-fuel contribution (background emission in this thesis), was measured using stoichiometric methane combustion. On the other hand, the soot generated from flame propagation in locally rich mixture was investigated using fuel-rich premixed gasoline mixture. One thing to notice is that the second experiments encompassed the soot formation caused by non-fuel sources, while the first experiment did not include the influence of flame propagation in locally rich mixture. The main results can be summarized as follows.

**Background PN emissions**

Stoichiometric premixed methane combustion with fast-idle condition shows the accumulation mode PN concentration as low as for ambient air and motoring condition. This means that it is the minimum PN emission can be obtained from the experiment. The accumulation mode PN concentration under this operating condition is approximately $10^4$/cc. This background emission is influenced by NIMEP, but not by RPM. For NIMEP lower than 5 bar, the accumulation mode PN emissions were similar to those from the fast-idle condition. However, PN emissions increased as engine load increases for the operating conditions when NIMEP was 5.5 bar or higher. Compared to 2 bar NIMEP cases, the accumulation mode PN concentration was approximately 15–20 times higher at 8 bar operating conditions. It is guessed that the increased PN emission at higher loads is due to other source, such as crevice out-gassing, than combustion of fuel. If well vaporized, the accumulation mode PN emission level of stoichiometric premixed gasoline is similar to that of methane. Compared to these background emissions, the minimum accumulation mode PN emission obtained from direct injection experiments was 5–45 times higher. This value (the accumulation
mode PN concentration from direct injection divided by that of premixed injection) is larger for low RPM and low engine load.

**PN emission due to flame propagation in locally rich premixed mixture**

Fuel-rich premixed gasoline experiments showed that there is a threshold at lambda between 0.70 and 0.80 where the accumulation mode PN concentration started to increase exponentially with the decrease of lambda. This threshold was slightly affected by the oxygen mole fraction, but was independent from engine RPM or NIMEP. Threshold lambda values obtained from the experiments were much higher than corresponding theoretical values. However, similar trends (soot formation under less fuel-rich condition) have also been found in other studies. It is guessed that the flame condition (non-uniform and not in equilibrium) favors a leaner threshold based on the unburned fuel/air ratio.
Chapter 5

Dual fuel experiments

Soot formation in the absence of liquid fuel film was investigated in the previous chapter. The work reported in this chapter was focused on the third mechanism of soot formation in DISI engines: the contribution from the liquid fuel film on the piston top. The other two mechanisms were studied in the absence of liquid fuel film by using premixed charge, but the same approach cannot be applied in this case because the most of liquid fuel evaporates before combustion regardless of injection methods. This makes it very difficult to test the influence of liquid fuel independently. Because the PN measurement device samples the gas at the downstream of the exhaust system, it is hard to determine whether the soot is generated due to the liquid fuel film or locally rich mixture.

As alternatives, a rapid compression machine (RCM) and an optical engine have been used. However, the experimental conditions of those devices are not exactly the same as real operating conditions.

In this study, dual fuel was implemented as a solution. The influence of liquid fuel on soot formation is indirectly investigated by using the combination of gaseous fuel (premixed methane) and liquid fuel (gasoline direct injection) under the same operating condition. Although this system is not completely free from the previously mentioned problem —evaporation of liquid fuel—this method can give a meaningful insight into the soot formation from liquid fuel film under realistic engine operating conditions.
5.1 Experiments description

Figure 5-1: Schematic diagram of the experimental apparatus

Figure 5-1 shows the schematic diagram of the experimental setup. The direct injection and premixed methane feeding system were used simultaneously. In addition to this, nitrogen dilution was also implemented.

The amount of liquid fuel injected was controlled by injection pulse, with intervals of 10% of mass. Due to the decreased charge cooling effect, MAP has to be increased as the fraction of liquid fuel decreases (up to 15%) to maintain the same NIMEP. Lambda and the spark timings were adjusted to 1.00 and MBT timing, respectively for each condition.

The fuel injector for direct injection is controlled by a pulse signal, and the amount of fuel injected is supposed to be proportional to the pulse width sent from the injector driver at a given pressure. Figure 5-2 shows the normalized amount of fuel that is actually injected as a function of injection pulse.

It can be seen that the amount of fuel injected and injection pulse can be correlated in linear function only if the injection pulse is longer than 600 $\mu$s. When the injection pulse is shorter than this length, correlation functions are still linear but the slope and y intersection are significantly changed.
This limit is not important for direct injection experiments because the smallest injection amount requires at least 950 $\mu$s of injection. However, it limits the operating regime in dual injection experiments because the amount of injected fuel was much smaller.

The minimum injection pulse width was set to 400 $\mu$s since it is the shortest pulse that can guarantee repeatability. Therefore, the operating regime was narrower for low NIMEP conditions.

5.2 Experimental result

5.2.1 Dual fuel combustion experiments

As shown in Figure 5-3, eight operating points were chosen for dual injection experiments with the combination of two RPM and four NIMEP. Fuel was injected at the optimum timing for the lowest accumulation mode PN emission under each condition.

Figure 5-4 shows the accumulation PN emissions as a function of the amount of liquid fuel. As can be seen, the PN emission patterns were different in two cases. At 1200 RPM, the accumulation mode PN emission exponentially decayed as the liquid fuel fraction decreased when the liquid fuel fraction was high. However, the PN concentration did not strongly depends on the liquid fuel fraction except for the case
Figure 5-3: Experimental conditions for dual injection experiments

Figure 5-4: Accumulation mode PN concentration as a function of a liquid fuel fraction; the disconnected symbols at liquid fuel fraction = 0 are data from the pure methane experiments.

of 2 bar NIMEP and 1800 RPM. This trend is more clearly seen if the accumulation mode PN concentration is plotted in a linear scale, as shown in Figure 5-5. The results imply that there is a certain threshold, which depends on RPM and engine load, for PN formation as a function of liquid fuel injected, as in the premixed charge experiments.

For 1200 RPM, the threshold of the liquid fuel fraction was 0.8 for 8 bar NIMEP, and 0.9 for other cases. The thresholds lasted a little longer in case with 8 bar because greater amounts of fuel were injected. In other cases, it is guessed that the increased amount of liquid was offset by the better evaporation caused by high temperature and Reynolds number, resulting in the same level of threshold regardless of NIMEP.
One possible explanation for why this behavior did not occur in 1800 RPM was that the mixing process is fast enough, therefore, only a small fraction of the injected fuel remained liquid by the time the flame arrived.

Figure 5-6 shows the PN spectrum for 2 bar and 8 bar NIMEP. The accumulation mode peak, approximately 40–100 nm diameter, declined as the fraction of liquid fuel decreased. As in Figure 5-5, this trend was more clearly observed in low-RPM condition.

To determine if there is an influence of fuel injection timing, the same experiments were performed using different SOI timings. For this, 1200 RPM and 8 bar NIMEP operating condition was chosen because the threshold occurs at the lower liquid fuel fraction at under these conditions.

Figure 5-7 shows the accumulation PN emission in various SOI timings. The values at the zero liquid fraction should be the same because the operating condition is the same (premixed methane only), but the PN emissions were higher when the liquid fuel was injected at SOI timings with high PN emission. The variation for the pure methane case may just be the run-to-run measurement variation at the low accumulation mode PN concentration. Another explanation is that, although it was a result from diesel engine combustion, it has been observed that a small portion of the soot generated, is absorbed by the lubricating oil during the early expansion stroke [62].
To eliminate this discrepancy, all values are normalized to the PN concentration from the premixed methane at the corresponding SOI timing, as shown in Figure 5-8. It can be seen that different SOI timing gave a different threshold for PN emission. For SOI timing with higher PN, the observed threshold had a small fraction of liquid fuel. Regardless of the SOI timing, the accumulation mode PN emissions at thresholds (the lowest PN emission under in the presence of liquid fuel) were 1.5–3 times higher than those from cases with premixed methane.

It was noticeable that the threshold behavior is also observed in case of late injection timings. Considering the fact that there is almost no contact between fuel spray and the piston top with late injection timing (e.g., SOI = 180 CAD ATDC), this result implies that piston impingement is not the only factor affecting soot formation during high load operation.
Figure 5-7: Accumulation mode PN concentration at 1200 RPM 8 bar with various SOI timings

![Graph showing accumulation mode PN concentration at different SOI timings](image-url)

The PN spectrum for different SOI timing, shown in Figure 5-9, shows the coincident result with the accumulation mode PN concentration. The entire spectrum shifted downward as the liquid fuel fraction decreases.

This change in the spectrum was more clearly observed when passing the threshold. For the case with SOI timing = 40 ATDC, the PN spectrum with liquid fuel fractions of 100, 80 and 50% were similar because the threshold occurred at 20% liquid fuel fraction. In contrast, the PN spectrum with 100% liquid fuel fraction was very different from the 80% liquid fuel fraction for 70 CAD ATDC SOI timing because the

Figure 5-9: PN spectrum as a function of the liquid fuel fraction at different SOI timing

(a) SOI timing = 40 CAD ATDC  
(b) SOI timing = 70 CAD ATDC
threshold occurred at 80% of liquid fuel fraction in this case.

Figure 5-8: Normalized accumulation mode PN concentration with various SOI timings; 1200 RPM and 8 bar NIMEP

Figure 5-10: Accumulation mode PN at 1800 RPM 8 bar with various SOI timings

The result from the same experiment, but at 1800 RPM and 8 bar NIMEP, is given in Figure 5-10. For 1800 RPM, the optimum SOI timing changed from 70 CAD ATDC to 65 CAD ATDC. As shown in Figure 5-4, threshold behavior was not observed at the optimum SOI. However, the threshold behavior appeared at other SOI timings. The same as in the 1200 RPM case, a threshold was observed with small fraction of liquid fuel for SOI timing with higher PN emission. Threshold liquid fractions were a little lower than for those in 1200 RPM cases.
5.2.2 Late gasoline injection experiment

In the previous experiment, the fuel injection timing was limited to cases in which the engine was able to run with direct injection only. Therefore, it was impossible to run the engine with late fuel injection timing, under which condition it would be more likely to have liquid fuel film on the piston top.

In this experiment, methane was the main fuel for combustion and only a very little gasoline was injected during the late compression stroke. Here, gasoline was not used as fuel, but as a source for soot. It is expected that most of gasoline injected remained in liquid form when the flame arrived because the time for evaporation was much shorter than for early injection. Figure 5-11 shows the conceptual diagram of this experiment. As in Chapter 3, nitrogen dilution was used to lower the combustion temperature.

![Figure 5-11: Schematic diagram for late gasoline injection experiment](image-url)
Compared to the dilution experiment in Chapter 3, this experiment focused more on the role of liquid fuel film on soot formation. This is because a larger portion of injected fuel remains in liquid form on the piston top during the expansion stroke. The 8 bar operating condition was chosen because the accumulation mode PN increased as the dilution ratio increased in previous dilution experiments. The dilution ratios are given in Table 5.1, and these were the same as in previous experiments.

Table 5.1: Dilution ratios and required MAP for 8 bar NIMEP

<table>
<thead>
<tr>
<th>Dilution ratio</th>
<th>Oxygen mole fraction (vol%)</th>
<th>Add nitrogen amount (vol%)</th>
<th>Required MAP (bar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No dilution</td>
<td>20.9</td>
<td>0.0</td>
<td>1.18-1.20</td>
</tr>
<tr>
<td>9:1</td>
<td>18.8</td>
<td>10.0</td>
<td>1.25-1.26</td>
</tr>
<tr>
<td>6:1</td>
<td>17.9</td>
<td>14.3</td>
<td>1.30-1.31</td>
</tr>
<tr>
<td>4:1</td>
<td>16.7</td>
<td>20.0</td>
<td>1.35-1.36</td>
</tr>
</tbody>
</table>

To impede vaporization, the fuel injection pressure was decreased to 30 bar. In this case, 400 μs of injection pulse with 30 bar injection pressure corresponds to 13% liquid fuel fraction under 8 bar NIMEP operating condition. Liquid fuel fraction varies from 13% to 20%, because 20% was the smallest liquid fraction in previous experiments. SOI timing was fixed to 40 BTDC compression for all cases. Injection timing was chosen as the latest possible fuel injection timing where the fuel injection event finishes before spark for all dilution levels. Lambda and spark timing were set to 1.00 and MBT for pure methane combustion; thereby the total metered lambda was less than 1.00 with gasoline injection. Table 5.2 summarizes the fuel injection parameters.

In summary, there were four variables in this experiment: amount of methane, amount of nitrogen dilution, amount of gasoline and the MAP. For pure methane operation with no dilution, the methane flow and MAP was set at $\lambda = 1.0$ and NIMEP = 8 bar. For each dilution level, the methane and MAP were readjusted to get $\lambda = 1$ and NIEMP = 8 bar. Then, at each dilution level, without changing the MAP and methane flow, a different amount of gasoline was injected, resulting in a
Table 5.2: Fuel injection parameters

<table>
<thead>
<tr>
<th>Injection duration(μs)</th>
<th>Mass fraction</th>
<th>Engine out lambda</th>
</tr>
</thead>
<tbody>
<tr>
<td>400</td>
<td>0.13</td>
<td>0.93-0.94</td>
</tr>
<tr>
<td>500</td>
<td>0.16</td>
<td>0.89-0.90</td>
</tr>
<tr>
<td>650</td>
<td>0.20</td>
<td>0.86-0.87</td>
</tr>
</tbody>
</table>

change of engine-out lambda and NIMEP.

Figure 5-12 shows the accumulation mode PN emission as a function of the oxygen mole fraction at the two different RPM. It is shown that PN emission decreased as dilution ratio increased with operation at 8 bar NIMEP. The decrease of PN emission was the greatest when the dilution ratio increases from 6:1 to 4:1. Even considering increased MAP at higher dilution ratio, it is possible to say that the accumulation mode PN emission actually decreased. This is because the accumulation mode PN concentration declined more than a factor of '2', while the change of MAP was less than 20%. Furthermore, the accumulation mode PN concentration increased in the dilution experiments in Chapter 3, under the same conditions.

![Figure 5-12](image_url)

(a) 1200 RPM

(b) 1800 RPM

Figure 5-12: The accumulation mode PN concentration with late fuel injection at 8 bar NIMEP

This result corresponds with the PN spectrum, shown in Figure 5-13. The accumulation mode peak decreased as dilution ratio increased when the same amount of fuel was injected at the same time. In the accumulation region, the spectrum
was diminished in both cases, but the degree of decrease was stronger at 1200 RPM. However, there was no consistent increase or decrease in the nucleation mode regime.

These results confirm the conceptual model offered by Ketteler and Cheng shown in Figure 5-14. In this experiment, pyrolysis (which triggers soot formation during the expansion stroke) was suppressed by nitrogen dilution, resulting in reduced PN emission; and the reduced combustion temperature caused by the dilution decreased the heat conduction required for pyrolysis.

Figure 5-13: PN spectrum with the liquid fuel fraction = 0.13

The lambda sweep experiments were performed without dilution at 1200 RPM and 8 bar NIMEP operating condition, with 400 μs of gasoline injection, as shown in Table 5.3. For the 'fix MAP' condition, the amount of methane injected was changed,
thereby the NIMEP varied as a function of lambda, while the MAP was adjusted to maintain constant NIMEP in the 'fix NIMEP' condition.

Table 5.3: Experimental conditions for lambda sweep experiments

<table>
<thead>
<tr>
<th>Methane lambda</th>
<th>Engine out lambda</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fix MAP</td>
</tr>
<tr>
<td>0.90</td>
<td>0.85-0.86</td>
</tr>
<tr>
<td>0.95</td>
<td>0.90-0.91</td>
</tr>
<tr>
<td>1.00</td>
<td>0.94-0.95</td>
</tr>
<tr>
<td>1.05</td>
<td>0.97-0.98</td>
</tr>
<tr>
<td>1.10</td>
<td>1.00-1.01</td>
</tr>
<tr>
<td>1.15</td>
<td>1.05-1.06</td>
</tr>
</tbody>
</table>

The purpose of the experiment was to see whether the excess oxygen had any chemical effect or just act as diluent to reduce the temperature of burned gas (which would affect pyrolysis). If the oxygen has no chemical effect, the accumulation mode PN emission should not change much as a function of lambda.

![Figure 5-15: Accumulation mode PN emission as a function of methane lambda](image)

However, as shown in Figure 5-15, the accumulation mode PN decreased as lambda increased for both cases. To see the effect of diluent in detail, case with 400 μs injection (Figure 5-12a) and the fuel-lean fixed NIMEP (Figure 5-15) were plotted.
together in Figure 5-16. Here, the x-axis (diluent mole fraction) means the sum of nitrogen and excess oxygen. However, the definition might be a bit tricky since the lambda is calculated based on the methane part of the mixture only when calculating diluent fractions, meaning that the extra gasoline injected is not considered. Figure 5-16 shows that oxygen is more effective than nitrogen at PN reduction for the same diluent fraction.

![Figure 5-16: Accumulation mode PN emission as a function of methane lambda](image)

As the final step, combined lambda sweep + nitrogen dilution experiments were performed under the condition of fixed NIMEP with methane lambda = 1.00, 1.05, 1.10 and 1.15 (plotted in Figure 5-17). As can be seen, regardless of the entire diluent fraction, the accumulation mode PN significantly dropped when the dilution ratio changed from 6:1 to 4:1. Especially for methane, lambda 1.10 and 1.15 showed almost no change of PN emission when the dilution ratio changed from 0 to 6:1.
These results imply that the excess oxygen has a chemical effect. However, it should be noticed that with the excess oxygen, the geometry of the soot producing plume also changes, because the equivalence ratio distribution of the gasoline vapor and premixed methane/air mixture has changed. This also can explain why the accumulation mode PN emission increased as methane lambda becomes fuel-rich.
5.3 Findings

The accumulation mode PN emission from the dual injection was measured to investigate the contribution of liquid fuel film on soot formation. Methane was chosen for the premixed injection fuel because methane always exists in gaseous state, and premixed methane showed the lowest soot emission under stoichiometric condition. Liquid fuel (gasoline) was injected via a high pressure direct injection system. With two different sets of experiments, the following things were revealed about the contribution of liquid fuel on soot formation.

Dual fuel combustion experiments
Both methane and gasoline were used as main fuel and engine-out lambda was fixed at 1.00. The PN emission was measured by changing the fraction of liquid fuel under given operating condition.

There is a threshold liquid fraction where PN emissions increase rapidly as a function of the liquid fuel fraction. This threshold liquid fraction is dependent on RPM and NIMEP. It is lower when air-fuel mixing is weak, and may not exist when air-fuel mixing is strong enough. For liquid fuel fractions higher than this threshold, the accumulation mode peak, located at 40–100 nm, is reduced as the fraction of liquid fuel decreases.

At the optimum injection timings, the lowest threshold liquid fraction was 0.8, which means that only 20% of the liquid fuel contributed significantly to soot formation. However, the threshold could be much lower for non-optimum SOI timings. In general, the threshold fraction is lower for SOI timings that resulted in higher PN emissions. Regardless of SOI timing, PN emissions at thresholds were 1.5–3 times higher than those from cases with premixed methane.

Late gasoline injection experiments
Methane was used as the primary fuel, and only a very small amount of gasoline is injected at the late compression stroke to generate soot. Nitrogen dilution was
implemented to affect pyrolysis of fuel-rich vapor above the piston top. Unlike dilution experiments done with purely direct injection, the accumulation mode PN emission declined as dilution ration increased under the 8 bar operating condition. With this experiment, it is shown that soot formation due to pyrolysis during the expansion stroke can be suppressed by nitrogen dilution. Reduced combustion temperature diminishes heat conduction from the burned gas into the rich vapor plume above the liquid fuel film, suppressing pyrolysis. However, lambda sweep experiments showed that the accumulation mode PN decreased much faster if the lambda increased.
Chapter 6

TEM imaging analysis

In previous chapters, PN concentration was measured to investigate the soot formation mechanism in DISI engines. As the final step of this thesis, this chapter addresses the morphology of accumulation mode particles, in relation to operating conditions, using TEM.

High-resolution transmission electron microscope (HRTEM) has widely been used for better understanding of in-cylinder soot formation processes [6, ?, 5, 63, 64]. Unlike particle detection devices that offer information based on particle size and concentration, TEM provides two-dimensional images of the accumulation mode particles in engine exhaust, enabling direct observation of nano-particles.

There are many options in the usages of TEM, such as sampling method, sample treatment, magnification rates and parameters to measure. In the case of sophisticated imaging analysis, very detailed geometry of particles was measured or calculated as shown in Figure 6-1. In this study, the general morphology of the particles was investigated to determine whether there is any fundamental difference between the soot generated by different mechanisms.
6.1 Experiments description

6.1.1 Sampling methods

The sampling system is one of the most important parts of TEM imaging analysis because TEM imaging requires controlled sampling on special analysis supports called TEM grids [65]. There are several options for driving the soot to the TEM grid: direct capture (using thermophoresis), filter sampling, electrostatic precipitation, soot-oil in oil centrifugation, and charge-space-guard modification. In this study, soot was collected by the direct capture and filter sampling methods.

With the direct capture methods, sampling grids are inserted into a sampling holder, and then directly exposed to the exhaust stream. Particles attached to the grid due to the thermophoretic force caused by the temperature gradient between the exhaust gas and TEM grids. The sampling time was between 30 seconds to 5 minutes depending on the accumulation mode PN concentration.

With the filter sampling method, soot was firstly collected on a 47-mm paper filter (PALL Fiberfilm filters). A vacuum pump was installed at the downstream of the filter to aid the sampling process. During sampling, the filter was held in a stainless steel sample holder. After the sampling process, the filter was dissembled from the holder. After this, the soot collected onto the filter was loaded on the TEM grids.
due to van der Waals force. The entire sampling process took approximately 15-20 minutes. Figure 6-2 shows the pictures of the sampling devices actually used for this study.

PELCO 200-mesh pure-carbon-film-coated copper grids were chosen for direct sampling in order to prevent thermal deflection caused by hot exhaust gas. For filter sampling, center-marked 200-mesh pure copper grids were used. In both cases, TEM grids were observed without any pre/post-treatment to prevent changes in the properties of the attached soot particles.

6.1.2 Experimental apparatus

Figure 6-3 shows the schematic diagram for the exhaust and sampling system. Other systems such as fuel delivery and intake systems were the same in previous experiments. The sampling ports were located at the downstream of the exhaust, 30-45 cm below the exhaust manifold.
6.1.3 Sampling condition

Table 6.1 shows the engine operating conditions that were chosen for sampling. All operating conditions were adopted from the experimental conditions in previous chapters. Similar to the PN concentration measurement, particles were sampled after the engine-out PN emission concentration was stabilized.

Each operation condition was selected to represent a single soot formation mechanism addressed in this study. Referring to Table 6.1, condition (1) and (4) are related to the background PN emission under low engine load, while (2) and (6) represent the background emission under high load operation. In addition, (3) was added to check whether there is any fundamental difference in particles generated by hydrogen combustion. Condition (5) was chosen to investigate the soot formed due to flame propagation in rich mixture. Conditions (7)–(10) were direct injection with 50 bar fuel pressure. (7) and (8) represents fast-idle condition with different SOI timings; while (9) shows high load operation. Conditions (10) and (11) were chosen to compare the particles generated with different fuel delivery methods, under the same operating condition. For (11), it was expected that a larger portion of particulates originated from the pyrolysis of liquid fuel films compared to condition (10). As for the experimental condition in Chapter 5, the fuel injection pressure was reduced to 30 bar for condition (11).
Table 6.1: Sampling conditions for TEM imaging

<table>
<thead>
<tr>
<th>No.</th>
<th>Fuel</th>
<th>Delivery method</th>
<th>NIMEP, lambda</th>
<th>RPM</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Methane</td>
<td>Premixed</td>
<td>4 bar, 1.0</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td></td>
<td>8 bar, 1.0</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Hydrogen</td>
<td>Premixed</td>
<td>2 bar, 2.5</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td>4 bar, 1.0</td>
<td>1200</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td>2 bar, 0.7</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td>8 bar, 1.0</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Gasoline</td>
<td>Direct (SOI = 60 ° ATDC)</td>
<td>2 bar, 1.0</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td>Direct (SOI = 110 ° ATDC)</td>
<td>2 bar, 1.0</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td></td>
<td>Direct (SOI = 60 ° ATDC)</td>
<td>8 bar, 1.0</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td>Direct (SOI = 60 ° ATDC)</td>
<td>8 bar, 1.0</td>
<td>1800</td>
</tr>
<tr>
<td>11</td>
<td>Dual-fuel</td>
<td>Direct (SOI = 320 ° ATDC)</td>
<td>8 bar, 0.94</td>
<td></td>
</tr>
</tbody>
</table>

With filter sampling, the sampling time was not directly related to the imaging process because the particles were collected onto the paper filter before loaded to TEM grids. Therefore, the sampling time was fixed at 20 min for all cases.

However, sampling time varied for direct sampling because it determines the quality of images. It is hard to detect enough particles when the sampling time was too short; while too long sampling times caused agglomeration of particles on the TEM grids. The sampling time was chosen based on trial-and-error. For conditions with low PN concentration such as (1) and (4), the sampling time was up to 3 min, and it was around 30 s in case of high PN emission cases like (5). Therefore, the number of particles on the image is not related to the actual PN concentration in the engine exhaust.
6.2 Experimental result

6.2.1 Filter sampling

Figure shows the TEM images with 100,000× magnification for cases (1), (3), (4) and (8). As can be seen, significant difference between samples is not observed. Particles had a layered micro structure, and primary particles were spherical with some irregularities, with mean diameter of 20-50 nm.

![TEM images](image)

(a) Methane, 1200 RPM, 2 bar NIMEP, $\lambda = 1.0$
(b) Hydrogen, 1200 RPM, 2 bar NIMEP, $\lambda = 2.5$
(c) Gasoline(premixed), 1200 RPM, 2 bar NIMEP, $\lambda = 1.0$
(d) Gasoline(direct, SOI = 60° ATDC), 1200 RPM, 2 bar NIMEP, $\lambda = 1.0$

Figure 6-4: TEM images of filter sampled soot particles with 100k magnification rate

To investigate the micro structure of primary particles more precisely, the magnification was increased to 300,000× for the same conditions, as shown in Figure 6-5. It is known that soot particles from DISI engines have less-ordered structures (ordered particles are characterized by smooth concentric fringe patterns surrounding the nucleus and a short interlayer distance between fringes), compared to carbon black and...
particles from diesel engines [6].

Several studies contain reports that the particles from ethanol-blended gasoline have a less ordered (amorphous) structure than those from pure gasoline. This is because particles from ethanol-blended gasoline could not experience the carbonization process as extensively as those from gasoline, due to reduced carbon availability in the fuel [6, 64]. In the same manner, the particles from hydrogen combustion may have a less ordered structure because the fuel does not contain carbon atoms. However, it is not clear from the pictures in Figure 6-4 or 6-5 if there are differences or not, because the small differences observed could be due to a focusing effect.

Compared to diesel soot images from other literature given in Figure 6-6, the differences between particles generated under the four operating conditions in this study were relatively small.
6.2.2 Direct sampling

Due to the agglomeration of particles, it was impossible to investigate the morphology of individual particles with filter sampling. As a consequence of this problem, the direct sampling method was implemented. Figure 6-7 shows the TEM grids used for filtered sampling. To capture particles in hot exhaust gas, thermal-resistive carbon-film-coated grids were used. Magnification rate was reduced to 25,000× to observe the entire shape of each particle.

Particles from background emission

Figure shows images of particles from methane and premixed gasoline combustion, with $\lambda = 1.00$ and 2 bar NIMEP conditions. In both cases, the particles are severely agglomerated, and have a diameter much larger than 100 nm. On the bottom of Figure 6-8a, a particle with long-chain structure is also observed.
(a) Methane, 1200 RPM, 2 bar NIMEP, \( \lambda = 1.0 \)
(b) Methane, 1200 RPM, 2 bar NIMEP, \( \lambda = 1.0 \)
(c) Gasoline (premixed), 1200 RPM, 2 bar NIMEP, \( \lambda = 1.0 \)
(d) Gasoline (premixed), 1200 RPM, 2 bar NIMEP, \( \lambda = 1.0 \)

Figure 6-8: Particles from premixed methane and gasoline (2 bar NIMEP) with 25k magnification rate

On the other hand, particles from hydrogen combustion show different morphology, as shown in Figure 6-9. The particles are not agglomerated, and the diameter of the primary particles is much smaller than for the previous two cases. It is worth noticing that Figure 6-8 and 6-9 show very different structures for the macro particles, (the particles in Figure 6-8 are larger than 100 nm, but those in Figure 6-9 have a mean diameter on the order of 10 nm). However, the elementary particles from Figure 6-5 are not that different. It is suspected that the particles could not grow well due to very low carbon availability.
Figure 6-9: Particles from hydrogen combustion (1200 RPM, 2 bar NIMEP, λ = 2.5) with 25k magnification rate

(a) Methane, 1200 RPM, 8 bar NIMEP, λ = 1.0
(b) Methane, 1200 RPM, 8 bar NIMEP, λ = 1.0
(c) Gasoline (premixed), 1200 RPM, 8 bar NIMEP, λ = 1.0
(d) Gasoline (premixed), 1200 RPM, 8 bar NIMEP, λ = 1.0

Figure 6-10: Particles from premixed methane and gasoline (8 bar NIMEP) with 25k magnification rate

These characteristics did not change at higher load. Figure 6-10 shows the images of particles from 8 bar NIMEP conditions with the same fuel delivery methods and
lambda as given in Figure 6-8. Particles have either agglomerated or chain shapes. However, the mean diameter and the entire length of chain-like particles (Figure 6-10b, and 6-10d) were larger than those for 2 bar NIMEP cases.

Those large agglomerates, which were not observed in other cases, might be considered ash or particulates from lubricants, because it is expected that the fuel barely contributes to soot formation under premixed and stoichiometric conditions. A study by Andrews et al. [66] revealed that the major portion of particulates from PFI engines is ash, with the second largest fraction being unburned lubricating oil. Carbonaceous particulates were found to be significant only at high load with mixture enrichment.

**Particles from fuel-rich premixed charge combustion**

Figure 6-11 shows the particles from fuel-rich premixed gasoline combustion. As can be seen, the diameter of both particles and primary particles are smaller than those from background emission cases. Most of them have a largely branched chain structure, but a few of them are agglomerated, like the particles from background emissions. There was a large fluctuation in the mean diameter of particles of both shapes.

![Particulate Images](a) Gasoline(premixed), 1200 RPM, 2 bar NIMEP, \( \lambda = 0.7 \)

![Particulate Images](b) Gasoline(premixed), 1200 RPM, 2 bar NIMEP, \( \lambda = 0.7 \)

Figure 6-11: Particles from fuel-rich premixed gasoline combustion with 25k magnification rate

Because the accumulation mode PN concentration under this operating condi-
tion is approximately 1000-times higher than that of background emission with the same NIMEP, most of the particles taken in 6-11 were generated due to the fuel-rich mixture. Therefore, it is believed that the images in Figure 6-11 show the geometrical characteristics of particles generated due to flame propagation in fuel-rich mixture.

**Particles from gasoline direct injection and dual-fuel combustion**

Figure 6-12 shows the particle images from gasoline direct injection with 2 bar NIMEP operating condition at two different fuel injection timings.

![Images of particle images from gasoline direct injection combustion (2 bar NIMEP) with 25k magnification rate.](image)

(a) Gasoline(direct, SOI = 60° ATDC), 1200 RPM, 2 bar NIMEP, $\lambda = 1.0$

(b) Gasoline(direct, SOI = 110° ATDC), 1200 RPM, 2 bar NIMEP, $\lambda = 1.0$

Figure 6-12: Particles from gasoline direct injection combustion (2 bar NIMEP) with 25k magnification rate

As can be seen, the diameter of the primary particles is slightly larger than those of the fuel-rich mixture, but smaller than those of background emission cases for both SOI timings. Particles were more nearly shape fuel injection timing was early (Figure 6-12a). When the injection timing was retarded, particles becomes chain-like shape as shown in Figure 6-12b, but did not have the complex shapes seen in fuel-rich premixed cases. This is consistent with the morphology study performed by Symonds et al.[67], who argued that the accumulation mode particles from DISI engines are less likely to have a chain-like structure and more likely to have a spherical structure than those from diesel engines.

However, these morphological characteristics are changed at high load operation. Figure 6-13 shows the images of particulates from 8 bar NIMEP with different RPM.
Noticeable differences between two RPM are not observed, but particles under the same operating condition had different morphology. Particles shown in Figure 6-13a and 6-13c have shapes similar to Figure 6-12a, but some particles shown in the other figures have shapes similar to shown in Figure 6-11 or 6-10d. This is because all three soot formation mechanisms are possible with direct injection. Compared to the 2 bar NIMEP condition (Figure 6-12), the primary particles were generally bigger, which increased the size of agglomerated particles as well.

(a) Gasoline (direct, SOI = 60° ATDC), 1200 RPM, 8 bar NIMEP, $\lambda = 1.0$
(b) Gasoline (direct, SOI = 60° ATDC), 1200 RPM, 8 bar NIMEP, $\lambda = 1.0$

(c) Gasoline (direct, SOI = 60° ATDC), 1800 RPM, 8 bar NIMEP, $\lambda = 1.0$
(d) Gasoline (direct, SOI = 60° ATDC), 1800 RPM, 8 bar NIMEP, $\lambda = 1.0$

Figure 6-13: Particles from direct injection (8 bar NIMEP) with 25k magnification rate

The diversity of morphology in particles from DISI engines was also observed in a study done by Miyashita et al. [5], as shown in Figure 6-14.
The experimental conditions for Figure 6-14 were very similar to that of Figure 6-13. The engine was operated at 1200 RPM with 70% load, and particles sampling time varied from 10 s to 5 min, depending on the accumulation PN concentration. It is possible to see particles with different morphologies because the liquid fuel film is not the only source of soot formation. The experiments in a previously mentioned study (by Symonds et. al.), in which it was suggested that the particles from DISI engines are more likely to have a spherical structure, were performed under low RPM and low load condition (1000 RPM and 3.27 bar BMEP). Therefore, it might be possible to say that the particles generated by liquid fuel film are more likely to have a sphere-like structure. This is because the experimental results in Section 3.2.2 showed that the liquid fuel film was a dominant source of PM emission in low load, while air-fuel mixing became important during high load operation.

Figure 6-15 shows the particles from two different fuel delivery methods under the 8 bar NIMEP operating condition. Compared to the same operating condition, but with direct injection as shown above, particles are more likely to have the morphology typical of idle condition with early injection timing, but some particles (shown in Figure 6-15d) have a chain-like structure. It is also shown that the size of primary particles varies a great deal, regardless of the particle morphology.
From the particle images from the direct injection, it is guessed that the particles generated due to the pyrolysis of liquid fuel film have (or at least, are more likely to have) a sphere-like structure with diameter ranging from 50–100 nm, this is because this shape is only observed in particles from direct injection. However, this hypothesis is not completely confirmed because particles with other shapes were observed as well, even in the dual-fuel combustion with late gasoline injection experiments. It is possible to argue that late injection could not eliminate the second soot formation mechanism, but there is no clear evidence for the argument.

Therefore, statistics based quantification will be required for further investigation on the morphological differences of the particles from different mechanisms. Alternatively soot formation under more controlled conditions, such as RCM, could also be considered.
6.3 Findings

For this chapter, TEM imaging analysis was performed to investigate the morphology of particulates generated by different mechanisms. Particles from various operating conditions were sampled using two different methods. The filtered sampling method showed that there is no fundamental difference in micro structure. However, TEM imaging result from direct sampling shows that particulates from different fuel delivery method have different size and shape, which can be summarized as follows.

Common characteristics
Engine load has a strong impact on particle morphology. The mean diameter of primary particles increases as engine operation load increases. (For direct injection, the primary particles from high load operation can be as small as those from low load operation) As a result, the average particle size becomes larger under high load operating condition.

Particles from background emissions
Except for particles from hydrogen combustion, which have sphere like structures composed of very small primary particles, the particles from background emissions are either severely agglomerated or form a long chain structure.

Agglomerated particles are distinguished from the particles generated by other mechanisms because their mean diameter is much larger. The morphology itself does not change as engine load increases, but the size of the primary particles grows. At low loads, most particles have a mean diameter around 100 nm, but particles with mean diameter > 300 nm are observed in high load operation.

Particles from fuel-rich premixed mixtures
The majority of particles have a chain structure with many branches. In any structure, primary particles are smaller than those from background emission, and the diameter of primary particles does not differ a great deal. On the other hand, the
mean diameter of agglomerated particles can be very differ, depending on how many primary particles are attached together. They range from 40 to 200 nm, although most are smaller than 100 nm.

**Particles from gasoline direct injection**

The size of primary particles is between that from fuel-rich mixtures and background emissions under the same NIMEP condition. Particles are more likely to have a spherical shape during idle operation, and particles with a chain-like structure do not have as many branches as do the particles from fuel-rich mixture. On the other hand, particles can have the shapes shown in background emission or premixed fuel-rich combustion under high-load operation because all soot-formation mechanisms are possible for direct injection with high NIMEP operating condition. It is guessed that these are particles generated from the liquid fuel film because this sphere-like structure is only observed in direct injection cases. However, further studies based on statistical methods will be required to confirm this hypothesis.
Chapter 7

Conclusion

This chapter provides a summary of the experiments performed in this study, the results of the experiments and their significance. The PN emission problem of DISI engines, especially under cold-start condition, was well known. However many questions about the in-cylinder soot formation process have been unanswered due to its complexity. The purpose of the work presented in this thesis was to reveal the soot formation process in DISI engines by decoupling various contributing factors, thereby finding potential soot emission mitigation strategies.

The following sections provide an overview of the experiments, present the key findings of this study and their contributions for future research on cold-start emissions of DISI engines.

7.1 Research overview

This study aimed to investigate PN emission under cold-start condition. From the vehicle test data, a set of operating points that best represent the cold-start condition were chosen. A series of steady-state experiments were performed at these points. The operating regime lies between 1200–1800 RPM and 2–8 bar NIMEP.

In the first step of the thesis, the PN emission of a DISI engine with a conventional gasoline direct injection system was measured with fixed fuel injection pressure (50 bar). In addition, nitrogen dilution experiments were performed to investigate the
influence of the combustion temperature.

It was found that the fuel injection timing, which determines the geometrical relationship between the fuel spray and piston crown/cylinder wall, is the most important factor for the accumulation mode PN emission. The nucleation mode PN emission was not strongly influenced by fuel injection timing, but there was a slight trade-off relationship between the nucleation mode and the accumulation mode PN emissions. The result implies that the nucleation mode particles are generated where the air/fuel ratio is near stoichiometric, while the accumulation mode particles are formed in much fuel-richer regions, such as where the fuel vapor is derived from liquid fuel films. The SOI timing where records the lowest PN emission was advanced as the RPM increased and engine load increased. The nitrogen dilution, which lowers the combustion temperature, did not change the PN emission pattern determined by the SOI timings. However, nitrogen dilution influenced the absolute amount of PN emission for early SOI timings. For 60 ATDC intake or earlier SOI timings, dilution reduces PN emission at lower NIMEP, but increased PN emission for higher NIMEP. For SOI timing 90 ATDC intake or later, where only a small amount of liquid fuel contacts the piston or cylinder wall, dilution did not substantially affect the PN emission.

As a next step, three primary soot formation mechanisms: (1) soot formation from non-fuel sources (e.g., lubricants), (2) flame propagation in locally rich mixture, and (3) pyrolysis of fuel-rich vapor from liquid fuel film on the piston top, were independently investigated along with various fuel delivery methods. In addition to the measurement of the PN concentration, the particle morphology was also investigated with TEM imaging. It was found that each mechanism has its own characteristics which can be summarized as follows.

1. **Non-fuel contribution**

   The PN emission from non-fuel contribution could be measured with a stoichiometric methane mixture. The PN level was independent of load up to 5 bar NIMEP. From 5.5 to 8 bar, the PN emission started to increase as the engine load increased. The
PN emissions were the roughly the same for premixed gasoline/air and methane/air mixtures at \( \lambda = 1 \), implying that the PN emission was not the result of the combustion of the fuel. One explanation is that the PN level when operating under low load, (approximately \( 10^4 / \text{cc} \)), is within the noise level of the instrument because a similar level of PN emission was recorded for ambient air. The increase in PN at the higher load is due to other sources such as crevice out-gassing, rather than combustion of fuel. For all cases, the accumulation mode peak, which appears between 50 and 100 nm in the PN spectrum for other mechanisms, was not observed. As the PN emission increased, the entire spectrum was shifted upwards without a noticeable peak. The particles from background emissions were either severely agglomerated or formed a long chain structure. Agglomerated particles are distinguishable from particles generated by other mechanisms because their mean diameter is much larger when NIMEP was the same.

2. Flame propagation in fuel-rich mixture
Fuel-rich premixed gasoline was used to investigate the soot formation generated from flame propagation in locally rich mixture. For both nucleation mode and accumulation mode particles, PN remained approximately constant up to a certain point (\( \lambda = 0.70 - 0.80 \)), then started to increase exponentially as \( \lambda \) decreases. This threshold was not influenced by RPM or engine load, but was slightly affected by oxygen concentration; the threshold occurs at higher \( \lambda \) when oxygen mole fraction was lower. Threshold lambda values obtained from the experiments are much higher than theoretical values (0.40-0.45). Similar results (soot formation under high \( \lambda \)) have also been found in other studies. The majority of particles have a chain structure with many branches. The primary particles are smaller compared to particles from other mechanisms, but the size of agglomerated particles varies a great deal.

3. Pyrolysis of the vapor from liquid fuel films
A dual injection system (premixed methane + gasoline direct injection) was used to investigate the soot formation by pyrolysis of the vapor from liquid fuel films. Dual
combustion (both methane and gasoline were used as the main fuel) experiments showed that there was a threshold gasoline fraction level beyond which the PN emissions increase rapidly. The threshold gasoline fraction was strongly influenced by the SOI timing. The threshold fraction was lower for SOI timings that resulted in higher PN emissions. However, the accumulation mode PN emission at the threshold was 1.5–3 times higher than those from premixed methane regardless of SOI timing. In addition, late gasoline injection (methane was used as fuel, gasoline was injected at the late compression stroke to generate soot) was performed to see the contribution of liquid fuel films on the accumulation mode PN emission. With nitrogen dilution experiments, it was shown that soot formation during the expansion stroke, caused by pyrolysis of rich vapor above the liquid fuel film, can be suppressed by lowering the combustion temperature. It is guessed that particles generated from the liquid fuel film are more likely to have a spherical shape.

7.2 Contribution on the PN emission reduction

This section includes the major contributions of the thesis. The experimental results from Chapter 3 to Chapter 6 are integrated here to explain the PN emission behaviors of DISI engines under cold-start condition. From this, possible PN abatement operating strategies.

1. Three primary soot formation mechanisms have different characteristics, resulting in irregular trends of PN emission in DISI engines

Soot formation in the fuel-rich premixed gasoline experiments showed that the threshold lambda increases as oxygen mole fraction decreases, which means that soot formation becomes more violent at low combustion temperature. However, the late gasoline injection experiments in Chapter 5 showed that soot formation from the pyrolysis of the vapor from liquid fuel films is suppressed as combustion temperature decreases. On the other hand, non-fuel originated soot formation is influenced by the maximum in-cylinder pressure. All mechanisms are not strongly influenced by engine speed, but
the accumulation mode PN emission decreases as RPM increases in gasoline direct injection experiments. This is because increased piston speed aids air-fuel mixing, reducing locally fuel-rich regions.

2. The relative contribution of the three soot formation mechanisms varies depending on the engine operating condition

The difference between the accumulation mode PN emission from gasoline direct injection and that from background emission under the same condition varies much, depending on engine operating condition. Normalized PN emission for gasoline direct injection (the accumulation mode PN from direct injection / that from background emission) was 34.0 at 1200 RPM and 2 bar, 10.7 at 1200 RPM and 8 bar, and 5.7 at 1800 RPM and 8 bar (NIMEP conditions). This large fluctuation means that the relative contribution of each mechanism on soot formation changed. In addition, for SOI timing = 60 CAD ATDC, the nitrogen dilution increases the PN emission at 8 bar NIMEP condition, but reduces at 2 bar NIMEP condition. These inconsistencies imply that the relative portions of the three mechanisms vary when operating conditions change.

Because of points 1 and 2, the PN abatement operating strategy should be varied as a function of engine operating condition. At low load, when the liquid fuel film is a major source of soot formation, the piston impingement should be minimized by retarding the SOI timing. In addition, introducing cold EGR could also be considered to reduce the combustion temperature. However, at high load, the fuel injection timing should be advanced to maximize air-fuel mixing, and combustion temperature should be elevated.

3. Transient operation strategies are very important for reducing PM

For this research, the engine operations in this thesis are designed to represent the first acceleration of the FTP-75 cycle which shows a very steep peak during the actual vehicle test. However, the experimental results showed that the accumulation mode PN emission from steady state operation was not as severe as expected. The PN emission from direct injection at 8 bar NIMEP was approximately 5–10 times higher
than that from background emission. This results means that the PN emission during transient emission is much higher than the summation of the PN emission from steady states.

During transient operating conditions, the overall air/fuel ratio is not constant around '1'. Dynamic changes in MAP and engine speed may cause deviations from the targeted air/fuel ratio, especially at the beginning of acceleration. In addition, as shown in Section 3.2.2., a slight deviation from the optimum fuel injection timing (the injection timing shows the lowest PN emission) significantly increase PN emission because even a slight deviation in injection timing may cause severe wall impingement or inhomogeneous mixture preparation when the amount of fuel injected is large. To reduce PN emission during transient operation, all combustion parameters, such as fuel injection timing, spark timing and cam phasing angles, should be changed smoothly from low load/speed to high load/speed operating conditions by a proper control strategy. Some preliminary works about the transient PN emission is introduced in Chapter 8.

7.3 Outlook

The scope of this thesis was limited to PN emission under cold-start condition, and all experiments were performed with a specific engine and fuel. In addition, explanations for some results are not clear yet. Exploring the following ideas and topics would expand understanding of PN emissions of DISI engines.

Topics that could not be addressed in this thesis

- Quantification of the contribution of each mechanism depending on engine operating conditions.

- Correlation of results of the engine experiments to TEM imaging results, more clearly explaining why the particles have different sizes and morphologies.

- Break-down of the non-fuel contribution, including oil consumption past the
rings, oil introduced via the intake ports and oil from the positive crankcase ventilation.

- Investigation of the fuel sensitivity. Different fuels will likely show a different response between homogeneous and wall film/evaporation/diffusion flame mechanism.

- Documentation of enrichment versus PN relationship on the different fuel types under fully-premixed operation.

**Investigation using experimental conditions closer to real driving**

- Expansion of experimental regimes to high load operation (BMEP higher than 20 bar) and severe transient operation where shows high PN emission even after an engine is warmed-up.

- Investigation of solid particle PN emission (using the heated oxidation catalyst).

### 7.4 Closing remarks

It has been widely known that PM negatively affects both public health and the public welfare, and internal combustion engines are one of the main sources of anthropogenic PM. Therefore, the study of the cold-start PN emission of DISI engines will continue to be a relevant research topic due its high-share of the overall PN emissions, and corresponding environmental and health impacts.

It is expected that knowledge and insights from this thesis about the cold-start PN emission of DISI engines will be useful for reducing the PN emissions of modern DISI engines; thereby reducing the harmful impacts of internal combustion engines on people and environment.
Chapter 8

Suggestions and preliminary works for future study

Although all the experiments done in this thesis were steady state experiments, the results revealed the importance of transient operation in the PN emissions. It has been known that PN emission from DISI engines peaks during periods of transient operation such as rapid accelerations and cranking-start [68, 69, 70]. In several studies, the soot formation during transient operations has already been already investigated, and the impact of control and calibration during transient cold start is already well understood by automobile manufacturers. However, most of them address on qualitative implications because it is hard to define the characteristics of transient operation with a limited number of operating parameters. This chapter provides preliminary test results for the emission during transient engine operation.

In addition, other recommended research topics except PN emission during transient operation are included at the end of the chapter.

8.1 Experiment description

Because the single-cylinder engine operation could not provide enough power to achieve fast-enough transient operation, multi-cylinder engine experiments were performed for transient operation. The LNF engine, the predecessor of the LTG engine
was chosen. The LNF engine has almost the same configuration as the LTG engine, except for a slightly lower compression ratio (9.5:1 vs 9.2:1). The engine was controlled and monitored by ECU(INCA); therefore operating parameters such as fuel injection timing, spark timing and cam opening time were automatically controlled. Figure 8-1 shows the schematic diagram of the experimental set-up.

![Figure 8-1: Schematic diagram of the experimental setup](image)

Engine operating points are given in Figure 8-2. The lower limit of NIMEP was determined by the combustion stability, and the upper limit was determined by knock or dynamometer capacity. To avoid abnormal engine operating strategies under cold-start condition (e.g., retarded spark timing for faster catalyst light-off), experiments were performed under warmed up condition. Coolant and lubricant temperature were maintained at 80 °C.

Under the operating regime given in Figure 8-2, PN concentration was measured with at intervals of 100 RPM and 1 bar NIMEP. Lambda was adjusted to 1.0 with feedback control during the experiments.

RPM was controlled using a dyno controller, and NIMEP was controlled by changing throttle position. For transient operation, NIMEP was chosen as a variable (i.e., NIMEP was varied at fixed RPM). Changing RPM with fixed NIMEP was more challenging because the throttle position needs to be changed as RPM the changes...
to maintain the same NIMEP. To simulate more realistic operation, a control system which can adjust both RPM and NIMEP simultaneously will be required.

Figure 8-3 describes a transient operation. The operation starts from idle NIMEP (2.0–2.5 bar), reaches maximum NIMEP at given RPM within 15 s and then is reduced to idle NIMEP in the next 15 s. The time scale is slow enough so that the engine calibration would be quasi-static, but the engine temperature would not be at the steady state temperature of the load and speed operating point that is ramped through. Because the throttle position was controlled with a linear profile, air flow and corresponding NIMEP was not linear.
8.2 Results

Both the nucleation and accumulation mode PN concentration were measured during steady state operation, then results were compared to those from transient operation. During transient operation, PN emission was measured with 0.5 s interval.

8.2.1 Steady state PN emission MAP

Figure 8-4 shows the PN contour for steady state condition, and corresponding geometric mean diameters are given in Figure 8-5. As can be seen, the PN emission from both modes was lowest with the operating condition of 1400-1600 RPM and 4-6 bar NIMEP. It is guessed that PN emission increases at lower RPM/NIMEP due to lower combustion temperature which causes incomplete oxidation. On the other hand, PN emission increases at higher RPM/NIMEP because the amount of fuel injected increases.

The change of the accumulation mode PN concentration is consistent with that in Chapter 3 in terms of RPM, but not NIMEP. In Figure 3-11, the accumulation mode PN decreases as RPM increases from 1200 to 1800 RPM, and increases as NIMEP increases from 2 to 8 bar NIMEP. However, In Figure 8-4, the accumulation mode decreases under the same NIMEP condition for RPM lower than 1700. One possible explanation is that the warmed-up cylinder block enhances the vaporization of fuel, so that the oxidation process, which is more violent at high temperature, becomes more important than under the cold-start case.

The mean diameter of the nucleation mode particles shows a similar trend with the nucleation mode PN concentration. However, the mean diameter of the accumulation mode particles is strongly influenced by NIMEP, rather than RPM. The mean diameter of the accumulation mode particle was 35-40 nm for low load, but increased to 70-80 nm as the engine load increased.
8.2.2 PN emission under transient operation

The transient PN emission was measured at 1200, 1600 and 2000 RPM, and then compared to the PN emission map given in Figure 8-4. In Figure 8-6, the transient PN emission is shown in the straight line with filled symbols, and that from steady state measurement at the corresponding load and speed is shown by the dotted line with hollow symbols.

The general trend of PN emission as a function of NIMEP was similar for both steady state and transient operation cases, but there were a few differences for both the nucleation and accumulation mode PN emissions: (1) the fluctuation of PN con-
centration was severe in transient case, and (2) the transient PN emission during high load operation (from 10 to 20 s) was higher than steady state PN emission.

The first difference was rather obvious because the fuel injection and spark timings were changing during transient operation. For the second one, it is suspected that higher equilibrium temperature for high-load operation aided vaporization of fuel during the steady state experiment, resulting in lower PN emission.

### 8.2.3 Conclusion

The PN emission from the transient operation with 15 s of ramp-time was similar to that from the steady state operation, but showed severe fluctuations and higher PN emission during high-load operation. This is suspected to be due to difference in the equilibrium temperature. Because the operating strategies for transient operation varies a great deal, it is impossible to characterize the PN emission during transient operation with this one experiment. Various experiments with different operating profiles (ramp-time, targeted air/fuel ratio and variation in RPM/load) will be required to investigate the soot formation behavior during transient operation. For further research on soot formation during transient operation, an experimental system that enables simultaneous control of engine load and engine speed will be required.
The main reason for measuring PN during transient operation is to assess whether the soot process is approximately quasi-steady or not. If the process can be considered as quasi-steady, it becomes possible to set-up PN mitigation strategies with steady state data.

As the time step for transient operation keeps reduced, the measurement would essentially become a cycle-to-cycle measurement, which is related to assessment of the engine calibration and residual fuel effect.

This experiment will be more challenging because the time resolution of the DMS 500 (and most of the fast PM analyzers available today) is not short enough detect the changes of PN concentration at this time scale, as shown in Figure 8-7.

![Figure 8-7: Transient operating condition](image)

In order to enable cycle-to-cycle PN measurement, an exhaust low-pass filter has to be developed to average out the pulsating flow, but still maintain the resolution of the longer-time-averaged values.

### 8.3 Other suggestions for future work

#### Optical experiments
In this thesis, liquid fuel film on the piston top was identified as one of the three mechanisms involved in soot formation. The idea that liquid fuel films contribute to soot emission, especially under cold start condition, is not new. However, there
have been several different explanations about the role of liquid fuel films on soot formation. This thesis utilized the hypothesis that pyrolytic reactions are involved in soot formation, which was conceptualized by Ketteler and Cheng[3]; however, sometimes the soot formation process due to liquid fuel films are described as pool-fire or diffusion flame.

To confirm this hypothesis, an optical experiment that could visualize the growth of vapor plumes might be required. Laser aided optical experiments such as laser induced fluorescence (LIF) may be used to measure equivalence ratios, to determine what reactions are possible under the given fuel and air concentrations.

**Fuel sensitivity on soot formation**

Another interesting topic about the PN emission in DISI engines that cannot be addressed in this thesis is fuel sensitivity. Many studies have shown that fuel properties are closely related to soot formation [56, 57]. In terms of fuel composition, it is also known that ethanol-blended gasoline is effective for soot-mitigation. It is expected that this effect is driven by both a chemical factor (ethanol has a lower C/H ratio than gasoline and contains an oxygen atom, resulting in a lower stoichiometric air-fuel ratio) or a physical factor (vapor pressure of ethanol is higher than that of gasoline, making it more likely to evaporate), but the relative magnitudes of the two factors have not been clearly investigated.

The research aimed at investigating the effect of fuel sensitivity on soot emission has this problem; it is impossible to independently control physical properties and chemical compositions. Therefore, if it becomes possible to decouple those two factors with a well-designed mixture of fuels, this would be very helpful to expand understanding on in-cylinder soot formation process.
Bibliography


Nomenclature

λ  Relative air fuel ratio

Φ  Fuel equivalence ratio

ATDC  After Top Dead Center

BMEP  Brake Mean Effective Pressure

BTDC  Before Top Dead Center

CA_{50}  Crank angle position where 50% of the heat is released

CAD  Crank Angle Degree

DAQ  Data Acquisition

DISI  Direct Injection Spark Ignition

ECU  Electronic Control Unit

EPA  Environmental Protection Agency

FTP  Federal Test Procedure

GIMEP  Gross Indicated Mean Effective Pressure

GMD  Geometric Mean Diameter

GSD  Geometric standard deviation

HEPA  High Efficiency Particulate Arrestance
LIF Laser Induced Fluorescence
LII Laser Induced Incandescence
MAP Manifold Absolute Pressure
MBT Maximum Brake Torque
NIMEP Net Indicated Mean Effective Pressure
$P_{\text{peak}}$ Maximum in-cylinder pressure
PFI Port Fuel Injection
PM Particulate Matter
PN Particulate Number
SOI Start Of Injection
TEM Transmission Electron Microscope