Influence of negative corona discharge on the Zeta potential of diesel particles

He Huang1, Xiao Zhang2, Xue Xiao3 and Song Ye4
1School of Traffic Engineering, Nanjing Institute of Industry Technology, Nanjing, China
2Zhenjiang Campus, Army Military Transportation University of PLA, Zhenjiang, China
3School of Automotive and Traffic Engineering, Jiangsu University, Zhenjiang, China
4SAIC Volkswagen Automotive Co., Ltd., Shanghai, China

Abstract
Electrical agglomeration as a pretreatment means can reduce the exhaust particle number concentration of diesel engine. The charge of particle is an important factor affecting the coagulation process. Therefore, an experiment was carried out to study the charging characteristic of diesel particles. Zeta potential for diesel particle was used to represent the charged state and the charge of particles could be calculated according to the value of Zeta potential. Influences of various factors on the charge of particle were investigated by changing the charged voltage, internal temperature of charging zone, and the load of engine. Experimental results show that the increase of charged voltage can improve the charge and the absolute value of diesel particles. With increase of charging zone temperature, corona inception voltage declines and the charge of particle increases. The load of engine has a positive effect on the charge of particles which reaches its peak at full load.

Keywords
Diesel engine, corona discharge, particulate matter, Zeta potential, charging characteristics

Introduction
Modern diesel engine has the characteristics of strong power, low fuel consumption, and high reliability. However, the emission of micro-nanosized particles poses great harm to the atmospheric environment and human health. How to reduce the emission of particles in diesel engine exhaust is an urgent problem to be solved.1–5

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Diesel particles are associated with a large amount and tiny particle size, most of which are less than 100 nm and can enter the human body through the respiratory tract. Research indicates that particles < 100 nm can penetrate the cell wall of the human body, causing numerous diseases such as blood disease, and can even result in some genetic diseases. Moreover, diesel particles are not easy to subside and suspend in the atmosphere, which has a certain impact on the environment. In recent years, countries around the world have paid more and more attention to the diesel particle emissions. The Euro V and Euro VI emission regulations have added the limit requirements of particle number emissions. Therefore, controlling diesel particle emissions, especially for small particles, is of great significance.

Electrical agglomeration (EA) is a relatively mature technique, which has been extensively applied in numerous fields such as chemical industry, building materials, and metallurgy. EA technique can enhance the agglomeration effect between particles through electrical charging of particulate matter (PM), thus promoting particle diameter enlargement and reducing the number of micro-nanoparticles. Recently, scholars at home and abroad have applied such technique to control and purification of the diesel particles, opening up a new research field for diesel particles purification.

Electrical charging of PM is the key to EA technique, which is mainly realized through gas discharge. The discharge modes mainly include corona discharge, glow discharge, and dielectric barrier discharge. Of them, corona discharge can generate a large amount of free electrons under high temperature and atmospheric pressure. The electrons would be driven the space far away from the electrodes after they have induced impact ionization and the negative ions are formed. The generated ions can collide with the particles, so that the particles are charged. Such technique has displayed promising industrial application prospect. Ke et al. used the wire-type charging device to charge the diesel particles by corona discharge technique. The research results suggested that the Bosch smoke value of the charged particles was remarkably reduced under different working conditions. Wang discovered that the charge-to-mass ratio of diesel particles was in direct proportion to interelectrode voltage under corona discharge condition, and load had more significant influence on the charge-to-mass ratio than rotational speed. Du et al. explored the relationship between the charged effect of diesel particles and the exhaust flow rate through experiment. Okubo et al. had collected and measured the diesel particles after corona charging using Faraday cage and voltammeter, and their results suggested that the charge-to-mass ratio of particles would reach a maximum of $\frac{126}{m} \mu C/g$ at the gas flow rate of 0.28 m/s. The above studies suggest that corona discharge technology can effectively enhance the diesel particles charge, and then control the particulate emissions to a certain extent.

The particle charge will directly affect their motion and agglomeration when diesel particles get charged in the electric field. Some scholars at present have carried out related research on the charging process of diesel particles under corona discharge condition, but research on the particle charge is rarely reported. It is necessary to explore the charged characteristics of diesel particles under corona
discharge condition, so as to further carry out dynamic analysis on the charged diesel particles, and thus intensively examine their motion and agglomeration processes. Therefore, this study had constructed the diesel particle charged test bench under corona discharge condition for pre-charge treatment of diesel particles, characterizing the particle charged status through analyzing their Zeta potentials, and explored the influence of different factors on the particle charge. Findings in this study are of important guiding significance to enhance the charged effect of diesel particles and establishing the mathematical model of EA.

**Experimental equipment and method**

**Experimental equipment**

The test engine was a four-cylinder, turbocharged, intercooled, electronic control common-rail diesel engine, and its main technical parameters are shown in Table 1. The control parameters such as fuel injection timing and fuel injection pressure remained unchanged during the experiments.

The experimental system are shown in Figure 1, which is mainly composed of diesel, temperature control system, charged reactor, power supply device, and particle collector. First, the condenser was cooled by the centrifugal fan to remove the water vapor in the emissions, and the emissions were then heated by the ceramic tube heater, so that emissions at a certain temperature were provided to the charged device. In addition, the internal temperature in the discharge region of the charged reactor was monitored using the PT100 thermal resistance sensor. Its technical parameters are as follows. The collection temperature range is $-200^\circ C$ to $+200^\circ C$, the display precision is $0.1^\circ C$, and the comprehensive precision is $0.3^\circ C$. The charged reactor had coaxial bobbin structure, the charged region was a stainless steel tube 52 mm in diameter, the insulating medium was a ceramic tube 3 mm in inner diameter and 1.5 mm in wall thickness, and the discharge electrode was located in the axis of the stainless steel tube. The experimental power supply was the TE4020 negative high-voltage direct current (DC) electrical source. Its technical parameters are shown in Table 2. Such high-voltage power supply could be

| Item                                | Specification                                      |
|-------------------------------------|----------------------------------------------------|
| Type                                | Four-cylinder, in-line, turbocharged, and intercooled|
| Bore × stroke (mm)                  | 105 × 118                                          |
| Max. injection pressure (MPa)       | 160                                                |
| Working volume/L                    | 4.09                                               |
| Compression ratio                   | 17.5                                               |
| Rated power/speed (kW/r/min)        | 95/2600                                            |
| Type of combustion chamber          | Direct injection                                    |
| Fuel injection system               | Electronically controlled high pressure common-rail |

Table 1. Specifications of testing engine.
continuously adjusted at 0–50 kV, with the maximum output current of 1 mA. A gas washing bottle filled with deionized water was equipped in the rear end of the charged reactor to collect the exhaust particles, and the sampling gas flow was controlled using the flowmeter and vacuum pump.

**Table 2.** The parameters of high-voltage power supply.

| Item                          | Parameter                               |
|-------------------------------|-----------------------------------------|
| Rated input voltage           | 220 V ± 10% AC                          |
| Rated output voltage          | −50 kV                                  |
| Voltage adjustable range      | 0–50 kV                                 |
| Dimensions (L×W×H)            | 243 × 155.6 × 123 (mm)                  |
| Relative load                 | ±0.1%                                   |
| Input adjustment rate         | Standard value 0.01%                    |

**Measuring principles and methods of the particle charge**

**Measuring principles of charge.** The charged state of particles was characterized through measuring the Zeta potential of the particles. As shown in Figure 2, Zeta potential referred to the potential difference (PD) between the internal solution and the sliding interface between the diffusion layer in the double electric layer and the stern layer on solid surface. The surface charge density and polarity could be speculated through the Zeta potential value of particles in the suspension. In addition, the Zeta potential of particles dispersed in the deionized water could be measured using the Malvern Zetasizer Nano ZS90 potentiometer. The potentiometer can accurately measure the Zeta potential of water dispersed and non-dispersed systems. The detection range of particle diameter is 3.8 nm–100 m, and...
there is no practical limit on the Zeta potential range of the sample. The experimental solution was 0.01 mol/L NaCl solution (pH 6), the temperature was controlled at 24.5°C ± 1.5°C, and the experimental electric field intensity was 16 V/cm. All data were measured for 3 times to take the average. The charge density on particle surface could be calculated according to the Gouy–Chapman formula

$$\sigma = \sqrt{8\varepsilon_w \varepsilon_0 n k_B T \sin \left( \frac{e\xi}{2k_BT} \right)}$$  \hspace{1cm} (1)$$

where $\xi$ was the Zeta potential value of particles, $\varepsilon_w = 78.3$ was the relative dielectric constant of water at 298 K, $\varepsilon_0 = 8.86 \times 10^{-12}$ C/(V m) was the vacuum dielectric constant, $n$ was the number concentration of NaCl solution, $e = 1.6 \times 10^{-19}$ C was the elementary charge, $k_B = 1.38 \times 10^{-23}$ J/K was the Boltzmann constant, and $T$ was the experimental temperature. Assumed that the diesel particles were spherical, and the particle charges could be obtained according to the following formula based on the charge density on particle surface $\sigma$

$$q = 4\pi \left( \frac{d_p}{2} \right)^2 \sigma$$  \hspace{1cm} (2)$$

Figure 2. Schematic diagram of particle distribution around the charged particles.
where \( d_p \) was the particle size. The Zeta potentials of the uncharged and charged particles measured using the potentiometer were \( \zeta_0 \) and \( \zeta_1 \), respectively, which were then substituted into formulas (1) and (2) to calculate the electric quantity \( q_0 \) and \( q_1 \). Thus, the electric quantity \( q \) obtained due to corona discharge of particles was shown as follows

\[
q = q_1 - q_0
\]

\( q \) shows the electric quantity obtained due to corona discharge of particles.

**Measurement of charge.** Corona discharge can be divided into positive corona discharge and negative corona discharge according to the corona electrode polarity. At the time of positive corona discharge, the electron avalanche is produced by photo ionization, the electrons will be neutralized as long as they arrive at the positive corona line, and the positive ions will slowly move in the space electric field. In the case of negative corona discharge, the electron avalanche is mainly produced by impact ionization, and the negative ions produced in the corona region will continue to disperse outwards.\(^{24}\) Under the same discharge parameters, the negative corona discharge process is more stable, along with higher produced ion concentration.\(^{25}\) Consequently, this study was carried out under negative corona discharge condition.

The experiments were carried out under the diesel rated speed of 2600 r/min, and the working conditions of 25%, 50%, and 100% load. The diesel working conditions were adjusted until steady running, and later, the internal temperatures within the discharge region were adjusted using the ceramic tube heater to 300°C, 400°C, and 500°C, respectively. Subsequently, the high-voltage power supply was connected, and the voltage were adjusted to 0, −5, −10, −15, and −20 kV, respectively. The current in the electrode ranged from 0 to 0.42 mA under the voltage of 0–20 kV, while the power consumption range of the charging device was only 0–8 W. After 5 min, the voltage of charging device became steady, the vacuum pump was opened, the sampling gas flow was controlled at 10 L/min, and the sampling time was 5 min.

**Results and discussion**

**Influence of charged voltage on the particles Zeta potential**

The charged modes of particles can be classified into electric field charging and diffusion charging modes under corona discharge condition, and the former is dominant for particles with the particle size of >500 nm, while the latter is predominant for those with the particle size of <200 nm, and both these two modes exist for particles with the particle size of 200–500 nm.\(^{26}\) Diesel particles are tiny, with the particle size of <100 nm, and are dominated by the diffusion charging mode.

At the conditions of 2600 r/min full load and charged region temperature of 500°C, the Zeta potential of diesel particles was changed with the change in charged voltage, as shown in Figure 3(a), and the calculated particle charge was presented in Figure 3(b). In the power-off status, the Zeta potential of diesel
particles was −0.60 mA, due to the presence of negatively charged functional groups (such as −OH). With the increase of voltage, the absolute value of Zeta potential and particle charge increases. When the voltage increases from −5 to −10 kV, the Zeta potential changes obviously, its absolute value reaches 0.80 mV, and the charge of a single particle also elevates remarkably. At this moment, electron avalanche begin to take place, and the particles begin to be infiltrated in the free electrons and negative ions. With the increase of voltage, the energy injected into the charged region elevates, the air ionization degree aggravates, the average space field intensity and charge density increases, and the particle charge also increases. According to the extended Derjaguin–Landau–Verwey–Overbeek (EDLVO) theory, the enhancement of particle electronegativity leads to an increase in electrostatic repulsion between particles, along with an increase in absolute value of particle Zeta potential.27

Influence of internal temperature within the charged region on the particles Zeta potential

Figure 4 displays the influence of temperature on the Zeta potential and charge of diesel particles at 50% load. It could be seen from Figure 4(a) that, with the increase of temperature, Zeta potential could respond to the charged voltage at a faster speed. The increase of absolute Zeta potential is only 0.02 mV when the charged region temperature is 300°C and the charged voltage increases from 0 to −10 kV. However, under the same charged voltage, such value could reach 0.11 mV at the charged region temperature of 400°C. It could be mainly ascribed to the fact that high temperature makes the gas molecules gain greater kinetic energy and reduces the molecular ionization energy. At the same time, the free path of electrons becomes greater, the acceleration distance of electrons in the electric field becomes larger, and the electron energy also increases. Therefore, the electrons are
more likely to open the peripheral electrons of gas molecules at high temperature to form the new free electrons, thus giving rise to corona discharge.\textsuperscript{28} As a result, high temperature could effectively reduce the critical coronal voltage at the time of corona discharge, so that the diesel particles are rapidly charged. Figure 4(b) shows the calculated charge of a single particle under different temperatures at 50\% load and the charged voltage of $-15$ kV. As could be observed from the figure, when the temperature increases from 300$^\circ$C to 400$^\circ$C, the charge of diesel particles increases markedly, which is consistent with the variation rule of the particle Zeta potential with temperature. It could also be seen from Figure 4(b) that, when the charged region temperature increases from 400$^\circ$C to 500$^\circ$C, the particle charge increases slightly, which could be mainly ascribed to the elevated migration rate of negative ions with the increase of temperature. Thus, the ion velocity is faster under the same electric field intensity, along with the collision probability of particles and ions increases, and the particle diffusion charge increases slightly.

**Influence of engine load on the particles Zeta potential**

Figure 5(a) shows the changes in particle Zeta potential at the diesel loads of 25\%, 50\%, and 100\% at the time of corona discharge. It could be figured out from the figure that, the absolute value of particle Zeta potential is greater under lower load in power-off status, which is 0.65 mV at 25\% load. It is mainly because that, the air–fuel ratio was relatively high under low load, which can increase the probability of the produced soot to contact with oxygen, thus contributing to more carbon layer edge atoms combine with the free hydroxyl. Moreover, the temperature is lower at lower load, which would lead to weakened oxidation of hydroxyls attached onto soot surface. Therefore, there are more hydroxyl functional group (C–OH) on the surface of particles formed under low load, along with stronger electronegativity on particle surface and greater absolute value of Zeta potential. When charging
the discharge electrode with negative high voltage, the particles absolute value of Zeta potential under each load shows an increasing trend with the increase of voltage. Typically, the particles absolute value of Zeta potential increases with the increase of load when the charged voltage reaches \(2 \times 10^5 \) kV. These findings indicate that, with the proceeding of corona discharge, the diesel particles has carried more charges under high load, with electronegativity is greatly enhanced. Figure 5(b) shows the calculated charge under various loads at the charged voltage of 15 kV. It could be seen from the figure that, the particle charge increases slightly with the increase of load, and the best charged effect could be achieved at full load, which is consistent with the variation rule of particle Zeta potential. It is mainly because that there is lower fuel delivery per cycle under low load working condition, the area of lean-burn region within the combustion chamber increases, and the low temperature in the cylinder increases the fuel incomplete combustion, forming more unburned hydrocarbon (HC). Moreover, the mass fraction of soluble organic fraction (SOF) absorbed by particles increases, thus aggravating particle agglomeration and adhesion. Therefore, the diesel particles are tightly distributed under low load working condition. However, the particles are mostly the disperse crotch-shape or chain structure under full-load working condition.\(^{29}\) According to the theory proposed by Shin et al. 2010 the particle charge is in direct proportion to their surface area and capacitance under unipolar diffusion charged condition.\(^{30}\) When the particle migration diameters are the same, particles with disperse chain structure has greater surface area and capacitance. Consequently, diesel particles could obtain more electric quantity through diffusion charge.

**Conclusion**

In this study, the negative corona discharge method is employed to pre-charge the diesel particles. The Zeta potential of diesel particles are analyzed to characterize the particle charged state. Later, factors, including charged voltage, temperature in
charged region, and engine load, are taken into comprehensive consideration, so as to explore the influence of different parameters on the Zeta potential of diesel particles. The following conclusions can be drawn:

First, under negative corona discharge condition, the charged voltage is increased, the space ion density within the charged region is elevated, the impact probability of diesel particles with negative ions is enhanced, and the amount of charge is markedly improved. Particles carrying more charges have stronger electronegativity, which results in enhanced electrostatic repulsion between particles and manifests as the increase in the absolute Zeta potential of particles.

Second, high temperature can effectively reduce the gas corona voltage at the time of corona discharge of diesel particles. After corona discharge, the increased temperature is to the benefit of the increased mobility of negative ions, which can partially improve the amount of particle charge.

Third, under the same discharge parameters, increase in load can greatly improve the surface area and capacitance of particles, thus enhancing the amount of diffusion charge of particles.

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ORCID iD
He Huang https://orcid.org/0000-0003-3386-9426

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**Author biographies**

**He Huang** contributed to the conception of the study. Her research area is diesel engine electrical agglomeration technology.

**Xiao Zhang** contributed significantly to analysis and manuscript preparation. His research area is diesel emission control technology.

**Xue Xiao** performed the data analyses and wrote the manuscript. Her research area is diesel combustion technology.

**Song Ye** helped perform the analysis with constructive discussions. His research area is diesel emission control technology.