On the charged aerosols generated by atmospheric pressure non-equilibrium plasma

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Abstract
Recently, charged aerosols are used widely in many applications, such as fog elimination, inactivation of airborne viruses and so on. Atmospheric pressure non-equilibrium plasma (APNP) have become an efficient way to generate charged aerosols. Except the traditional pin-plate or wire-plate APNP sources, the new APNP sources such as large scale corona discharge system and air plasma jet array are introduced. These sources can increase the ion density in open air and generate adjustable large plasma plume, which are helpful in increasing the charging efficiency on aerosols. Although the interactions between plasma and aerosols is quite complicated, the preliminary study suggests that the diffusion charging dominates aerosols below 0.1 μm, field charging dominates aerosols larger than 1 μm, and the photo charging also contributes to aerosols charging. These reactive charged aerosols have shown their potential in the artificial rain enhancement, biomedicine, and material processing. Finally, challenges and opportunities for theoretical, experimental, and application research in related cross-disciplinary areas are presented to stimulate critical discussions and collaborations in the future.

1 | INTRODUCTION

Aerosols, also known as airborne particles, or particulate matter (PM), are usually referred to the particles suspend in air, which can be as ultrafine particles, fine particles (PM2.5) and respirable suspended particles (RSP, PM10). These airborne particles can be generated by evaporation of liquid droplets leading to micrometre powders through spray drying and pyrolysis [1, 2], or gas-to-particle conversion leading to nanometre to sub-micrometre particles [3]. Aerosols are made up of different compositions such as sulphates, nitrates, elemental and organic carbon, metals, biological compounds, and polycyclic aromatic hydrocarbons (PAHs) [4, 5], which have side effects on human health and climate [6].

Aerosols have been widely studied in different fields, and the charging process plays an important role in many applications. Charged aerosols are used widely in aerosols processes and environment protection, and have the advantage of low cost, simple operating condition, adjustable reactive and ionizing properties [1, 7]. Aerosols measurement by electrical low pressure impactors need particles to be charged by a corona discharge system, and the consequent faradic current generated by charged particles along each flow channel can be measured. Charging aerosols is also technical foundation for gas purification facilities such as air filtration equipment and electrostatic precipitators [8, 9]. Because the unipolar charging has higher charging efficiency, and recombination of charged particles with opposite polarities can be avoided, it receives more attentions than bipolar charging for these applications. In addition to that, besides the traditional low pressure coating processes by ion etching and sputtering, and laser, rf and micro-wave plasma is employed in the gas to particle conversions [3]. Recently, highly charged aerosol particles are used in many applications such as electrostatic painting, separation technology or surface coating [10], fog elimination [11], and inactivation of airborne viruses [12].

Therefore, this review address charged aerosols generation and application based on atmospheric pressure non-equilibrium plasma (APNP), especially, the potential applications of charged aerosols in rain enhancement and biomedical field are...
introduced. Figure 1 shows the interactions between aerosols, APNP sources, and plasma characteristics decide the generation efficiency and application direction of charged aerosols.

The first part introduces eight kinds of typical APNP sources used widely in charged aerosols generation. The pre-requisites about aerosol (definitions, formation and evolution kinetics) are firstly presented. Discharge characteristics, such as ion density, plasma volume, and generation mechanism of charged species, closely related to charged aerosols generation are depicted.

The second part addresses the charging mechanism of aerosols in plasma. The charging process is actually affected by particle properties, electric field, and discharging conditions. Besides the diffusion and field charging mechanism, because of strong UV optical emission generated by air plasma, the photo charging mechanism is also discussed. Diagnostics of charged aerosols are introduced in view of applications.

The third part deals with the potential applications of charged aerosols in the field of the enhanced droplet condensation, biomedical and chemical processing. The special designed APNP sources, charging efficiency, and interaction mechanism between charging aerosols and application object are systematically discussed.

2 | APNP SOURCES TO GENERATE CHARGED AEROSOLS

2.1 | Aerosols formation and kinetics

Aerosols are gaseous dispersion systems composed of solid or liquid particles suspended in gaseous media [13, 14]. Aerosol particles generally range in size from 0.01~10 μm, but due to a wide range of sources and causes of formation, such as pollen and other plant aerosols with particle sizes of 5~100 μm, and wood and tobacco burning aerosols with particle sizes of 0.01~1000 μm [15]. The particles come in a variety of shapes, from nearly spherical, such as liquid fog beads, to flaky, spice-like, and other irregular shapes. From the point of view of hydrodynamics, aerosol is essentially a multiphase fluid with gas as continuous phase and solid and liquid as dispersed phase [16, 17].

The fragmentation of liquid jets (spraying), solids (millling), and gas-to-particle conversion (nucleation) can generate suspension of liquid/solid particles in gases [18, 19]. The formation of nucleated nanoparticles from the gas phase requires supersaturation that can be achieved by physical and chemical methods. The physical routes of nucleation include the cooling of hot vapours through adiabatic expansion or collisional heat transfer with a colder gas by non-isothermal diffusion or turbulent mixing [18, 20–22]. The chemical routes of nucleation imply gas-phase reactions generating condensable products and nanoparticles with different compositions than the precursors [18, 21, 23, 24].

The ions generated by plasma provide additional electric field forces on the condensable particles, and facilitate the nucleation starts for lower saturation than homogeneous nucleation. The ion-induced nucleation also decreases the activation energy for the formation of stable clusters substantially [18]. Therefore, APNP is also used to enhance the nucleation process through plasma-induced gas phase reactions and the consequent gas-to-particle conversions, and plasma etching [18, 25].

After the generation by the physical or chemical route of nucleation, nanoparticles grow by coagulation with eventual simultaneous condensation, depending on the concentration of condensable species remaining after the nucleation phase [19, 26]. Coagulation occurs by ballistic collisions of particles smaller than the mean free path of gas molecules (60 nm at normal atmosphere). For the small particles (dp < 10 nm), the collision frequency is in the free molecular regime according to the kinetic gas theory. For particles bigger than the mean free path, the particle collision frequency depends on Brownian motion of particles related to their diffusion coefficient [27]. The Brownian coagulation rate is affected by temperature, primary particle and agglomerate sizes, and the concentration squared. Therefore, coagulation mainly concerns highly diffusive sub-micrometre particles with concentration more than 10^6–10^7 cm^{-3} [28].

The condensation on pre-existing nuclei also facilitates the growth of nanoparticles when condensable species still remain after the homogeneous nucleation phase [27]. The condensation is homogeneous nucleation indeed, which happens when the temperature decreases around the vapour source and the consequent supersaturation required for condensation is reached further, the supersaturation for nucleation is decreased by hetero-molecular nucleation, or condensable species are produced during the transition process in the reactor [18].

2.2 | Typical APNP sources

As APNP are employed to generate charged aerosols, the goal here is to define categories of discharge sources based on the structure and materials of electrode, and excitation mode.
The wire-plate or wire-cylinder corona discharge source usually consists of a corona discharge wire placed between two parallel plate electrodes, or in the axis of cylinder electrode, respectively (Figure 2a) [29]. The wire electrode is connected to the positive or negative power supply through a high resistance of ~10 MΩ to prevent corona-spark discharge transition. The gap between the plate electrode and the wire is ~3 cm. The applied voltage can be ~ −15 kV or +9−15 kV. Numerous discharge streamers flow to the collecting electrode plate, collide with particles transported by the gas with high velocity, and lots of charged aerosols are generated after collision. In order to decrease the re-entrainment of sub-micrometre particles deposited on the electrodes, the pulsed-DC power supply was employed to drive the discharges [30], besides that, the length of plate or cylinder electrode was shortened to avoid the precipitation of charged particles within the corona discharge sources [31, 32].

Counter-flow corona discharge source consists of sharp needle electrodes mounted on rods or rings each row of electrodes facing a grounded mesh electrode (Figure 2b). The corona discharges happen between sharp needle electrodes and opposite grid, and the plasma plume generated by the strong corona discharges collides with aerosols moving at a certain velocity to produce charged aerosols [33, 34]. Depending on the propagation direction of plasma plume is in the same or opposite moving direction of aerosols, the counter- or co-flow direction to the gas flow can be distinguished. Many electrode pairs can be arranged in series to increase the charging efficiency of aerosols. The high voltage power supply such as 15−30 kV DC or AC 20 kHz (RMS 20 kV) can be used to drive the discharge system.

The dual corona discharge source consists of two multipoint discharge electrodes, facing each other, and two parallel grids between these two discharge electrodes (Figure 2c). The grid electrode can not only control the discharge intensity emitted from the multipoint discharge electrodes, but also work as the counter electrode for the other oppositely located discharge grid electrode. The charging zone is the region between grids electrode. Two AC high voltage sources running in the anti-phase are used to drive the discharge electrodes and grids electrode. Aerosols can be charged by the plasma plume emitted from the discharge electrodes and the subsequent flows between the grids perpendicularly to the gas flow. The charge amount of charged aerosols through this charging zone can be close to the Pauthenier limit [29]. The strong alternating electric field between the grids due to the anti-phase AC high voltage make the aerosols flow in sine-wave trajectories, and the subsequent penetration of these aerosols through the discharge source is more than 80%, which is higher than that of DC corona discharge sources. Besides that, the energy consumption and ozone generation of dual corona discharge sources are less than the traditional DC corona discharge sources [35].

The Masuda boxer discharge source bases on the high frequency surface discharge and low frequency electric field discharge (Figure 2d). The surface discharge generated by a strip electrode embedded in the ceramic wall can form a charger channel and facilitate the charging process of aerosols. The low frequency electric field realized by the external electrode pair can induce the oscillation movement of charged aerosols, which prevents the precipitation of aerosols on the strip electrode and metal electrode. This discharge source is especially suitable to charge the aerosols with high resistivity, and its charging efficiency on aerosols is also higher than the DC corona discharge source [36, 37].

The key characteristics of spiked-corona discharge source is the flat rods or plates with spikes, placed between two parallel plates (Figure 2e) [34, 38]. The spike electrode is power
electron, and its direction is usually perpendicular to the gas flow. The flat belt with spiked edges resembling a saw-tooth is another common structure of spiked electrode [39]. The spike edges on the adjacent two plates can generate plasma and numerous positive and negative ions, which facilitates the bipolar charging of aerosols and agglomeration, therefore, the higher deposition of aerosols on both plates especially on the edges is still the area needs improving. The advantage of spiked-corona discharge source is that it can provide larger and more stable discharge volume than the wire-type corona discharge sources [34].

The dielectric barrier discharge (DBD) source consists of two parallel plate metal electrodes covered with a dielectric barrier (Figure 2f). The kHz AC or RF power supply is used to energize the discharge. More uniform and larger plasma volume between the two dielectric barriers promotes the bipolar charging of aerosols and the collection efficiency of aerosols. Besides the application such as removal of NOx, hydrocarbons, and particulate matter [40, 41], the large volume of DBD source can charge and react with mist droplet (micrometre in diameter), and also generate plasma activated vapour [31, 42, 43].

The large scale corona discharge source is an efficient way to increase the local ion density and generate charged aerosols in the open air. Figure 2g shows the setup of large scale corona discharge wires with length of several kilometre. The wire electrode is usually five strands of stainless steel stranded wire with the diameter of 1–2 mm to withstand the elements. The pole and crosspieces can be used to avoid the interference between corona discharge wires. According to the reference [44], this large scale corona discharge source has the corona wire with length of 7.2 km and the applied voltage of −90 kV, can generate a high ion density volume of approximately 30 (long) × 23 (height) × 90 m (width). Aerosols can be charged in this volume and work as more efficient condensation nucleus. More details of this source can be found in Section 4.1.

The air plasma jet can generate the plasma plume with adjustable length and direction. The insert of air plasma jet array into the flow channels of aerosols can provide a new way to generate the charged aerosols (Figure 2h). The plasma plume of the plasma jet array can cover the whole cross section of the flow channels of aerosols. Although the generation of plasma jet is in the pulse mode with repetition frequency of 20–50 kHz, more plasma jet arrays along the delivery channel can promote the bipolar charging of aerosols substantially, and its application in enhanced water vapour condensation in cloud chamber indicates its potential in fog dispersal.

Changes on aerosol also affect the coagulation of particles significantly. The coagulation process is suppressed by electrostatic repulsion force for the unipolar charged aerosols. However, for the bipolar charged aerosols, the coagulation process is enhanced because of electrical forces such as ion-induced Van der Waal’s force and image force, and Columbo attraction force [45]. The recent experiment result suggests that charged sorbent nano-powder attract and collect on their surface fine opposite-charged or electroneutral particulates (drops) from the air or room surface [46–48], which indicates the coagulation of charged aerosol can provide a new way to sorption of harmful aerosol particles.

2.3 | Discharge characteristic

The geometry (wire, point, wire-to-plane, gas gap) and nature (insulating or conductive materials) of electrode, the excitation mode (DC, AC, pulsed) and the working gas directly affect the discharge regimes of APNP sources. The common discharge regimes include DC corona discharge, filamentary streamer or sparks discharges in asymmetric gap, homogenous and filamentary DBD, plasma jet and so on. The classification of discharge characteristics of APNP sources help understanding of aerosols charging process.

2.3.1 | Corona discharges

The positive corona discharge occurs mainly in two different forms: burst corona discharge and streamer corona discharge [49]. The first stage of corona has been thought of being made up with imperfectly resolved current impulses of extremely high frequency by Trichel. The photoionization and impact ionization around the pin electrode tip as shown in Figure 3a generate the intermittent corona with a small discharge current. The second stage of streamer discharge is attributed to numerous individual current pulses distributed over the surface of pin electrode tip with sufficient electric field. Figure 3b suggests that the positive space charge concentrates in a region where the electric field is strong enough to sustain the Townsend discharge mechanism by electron avalanche. The large streamer pulses are observed at the last stage of the discharge (Figure 3c). The discharge streamers penetrate into the gas gap in the strong electric field, which is believed to be incited by energetic α-rays or properly timed ions [50–52].

The negative corona discharge can be triggered when the electric field around the pin electrode tip exceeds the ionization electric field of ~3 kV/mm. The generation of negative charges around the pin electrode tip and the subsequent removal leads to the repetitive discharges. The attachment of electrons to O2 and electron detachment through excitation and photoionization compete in the negative corona discharge.

\[
\begin{align*}
N_2 + e & \rightarrow N_2^+ + 2e, \Delta H = 104 \text{ kJ/mol} & \text{(Reaction 1)} \\
O_2 + e & \rightarrow O_2^+ + 2e, \Delta H = -125 \text{ kJ/mol} & \text{(Reaction 2)} \\
O_2 + N_2 + e & \rightarrow O_2^- + N_2 & \text{(Reaction 3)} \\
O_2 & + h\nu \rightarrow O_2^+ + e & \text{(Reaction 4)} \\
O_2 & + h\nu \rightarrow 2O, \Delta H = 498 \text{ kJ/mol} & \text{(Reaction 5)} \\
O_2 + O & \rightarrow O_3 & \text{(Reaction 6)}
\end{align*}
\]
The ionization of N\(_2\) (Reaction (1)) and photoionization (Reaction (4)) dominate the ionization in the strong electric field region adjacent to the pin electrode tip. The photoionization also strips O\(_2\) to generate O and O\(_3\). The ionization of O\(_2\) by electron (Reaction (2)) dominates the ionization reaction in the lower field because its energy for ionization is less than that for nitrogen ionization (Reaction (1)). Electrons attach to a neutral O\(_2\) by three-body reaction (Reaction (3)) create a negative space charge at a distance from the electrode tip [52].

The increased rate of removal of negative charges and the reduced rate of generation of negative ions due to detachment effect in high field is responsible for the corona discharge in the pulsed mode [53].

### 2.3.2 Dielectric barrier discharges

The gas electrical breakdown firstly leads to the charging of the dielectric barrier surface in the case of kHz AC operated DBD. The charge accumulated on the dielectric surface induces an electric field opposite to the applied voltage. Therefore, the total electric field decreases and the discharges extinct. The dielectric barrier works as a load which not only limits the amount of charge and the discharge current density in the gas, but also avoid the spark or arc discharge transition. For the DBD driven by AC high voltage, a breakdown happens during the rising phase of applied voltage (i.e. twice per cycle), and the discharge stops at the voltage peak. For the DBD driven by unipolar pulsed DC high voltage, the surface charges generates an electric field at the falling edge of high voltage pulse, which causes the second breakdown during the same high voltage period [54–56].

Gas discharge usually constrict at atmospheric pressure because of the streamer breakdown mechanism [57]. Electron avalanches create a space charge and thus an additional electric field which enhances the growth of secondary electron avalanches locally. The ionization region and the perturbation of the electric field grows rapidly, and numerous distinct plasma channels form in the end. These so-called micro-discharges appear as thin discharge channels because of the repetitive generation. DBDs in air are typical examples of such filamentary discharges. The micro-discharge channels can spread on the dielectric barrier surface and cover a much larger area than the original filamentary spot. The increasing applied voltage can increase the number of micro-discharges per active phase, but will not change the charge amount of a single micro-discharge. The charge amount depends on the gas gap and the specific capacitance of the barrier [58].

It is interesting to note that a filamentary DBD can be uniform to naked eyes when the number density of micro-discharges is high enough. There can also be transition regimes, where uniform and filamentary mode exists within the same half period or stagger between two successive half periods. DBDs in the atmospheric pressure glow discharges or Townsend discharge mode are also diffuse discharges. The diffuse DBDs have been generated in pure gas (helium, nitrogen) or gas mixtures (argon with oxygen, air with helium) [59, 60].

### 2.3.3 Plasma jet

Plasma jet is an efficient way to generate an adjustable plasma volume with high charge density. The adjustable plasma plume can increase the charging efficiency of aerosols. Although the noble gas can generate plasma jet easily, the air plasma jet is obviously more suitable to generate charged aerosols at lower cost. Actually, because of the presence of electronegative O\(_2\), it is difficult to sustain the air plasma jet. Nevertheless, several different air plasma jets have been developed [61].

The micro-plasma jet shown in Figure 4a can operate in several gases including air. The anode and cathode is separated by a dielectric barrier with a thickness of 0.2–0.5 mm thickness and a centre hole of 0.2–0.8 mm in diameter. The centre hole is also the development channel for the plasma jet. The ballast resistor between the power and ground electrode is 51 k\(\Omega\). A relatively low temperature air plasma jet is generated in the open air with the length of \(~1\) cm, when the DC applied voltage is 1 kV and air flows through the centre hole. The gas
The temperature of plasma jet is about 1000 K within the micro-gap, but decreases quickly to about 50°C at 5 mm away from the nozzle. The corresponding air flow rate is 200 SCCM, and the discharge current is 19 mA.

Another typical air plasma jet shown in Figure 4b has the notable characteristics that a porous anodic alumina dielectric barrier is used to separate the inner HV stainless steel electrode and the outer ground electrode [62]. The porous anodic alumina has 30 vol% porosity and average pore diameter of 0.1 mm. The cantered hole with diameter of 1 mm in the ground electrode facilitates the injection of plasma jet into the open air. An air plasma jet with length of ~2 cm is generated, when 60 Hz HV power is applied and several standard liter per minute air flow through the hole. The gas temperature of plasma is ~60°C at 1 cm from the nozzle.

The air plasma jet array shown in Figure 4c can generate a much larger plasma plume than single plasma plume. The single plasma jet of the jet array has a stainless steel needle electrode within the dielectric tube. The radius of needle electrode is ~50 μm. The resistor of 50 MΩ is used to limit the discharge current. The applied voltage of the air plasma jet array is 18 kV. The discharge of each plasma jet appears as a periodic sequence of nanosecond repetitive pulses with a pulse repetition rate of ~20 kHz [63]. The air plasma plume can be touched by human hand directly. Because of the interaction between plasma plumes, and diffusion and transportation of charged particles [64, 65], the effective plasma volume of the plasma jet array is expected to be larger than the sum of plasma volume of three single plasma jet.

The plasma plume appears continuous to naked eyes, is actually made up of fast propagating plasma bullet, which is proved by using the fast imaging technology. The plasma bullet travels at a velocity of $1.5 \times 10^5$ m/s, higher than drift velocity of electrons $1.1 \times 10^4$ m/s at the corresponding electric field [66–68]. The streamer propagation based on the photoionization theory is used to explain the plasma bullet propagation. Different from the cathode directed streamer generated by positive corona discharge, the plasma bullet behaviour is mostly repeatable [61]. In most cases, the plasma bullet propagation behaves in a repeatable fashion: they consistently propagate the same distance after the same period. The plasma bullet propagation can be in a chaotic mode when the applied voltage is low or immediately following breakdown [69, 70].

3 | CHARGING MECHANISM AND DIAGNOSTICS OF AEROSOLS

APNP sources produce clouds of electrons and bipolar ions, which can be used for generation of charged aerosols, neutralization of particles or separation of unipolar charged aerosols [71, 72]. Actually, based on particle properties ($d_p$, dielectric constant, $C_p$ capacitance), electric field, and charging conditions $N_i \times t$ ($N_i$, the ion density, and $t$, the transit time of aerosols in cloud), the unipolar diffusion and field charging laws can predict the number of charges. Figure 5 outlines the charging mechanism depends on the size of aerosols: the diffusion charging dominates aerosols below 0.1 μm, field charging dominates aerosols larger than 1 μm, and both mechanism are active for intermediate sizes, additionally, strong UV optical emission generated by air plasma affects the charging dynamics of aerosols. The critical points to perform the charge number, size and concentration measurements on charged aerosols are also discussed.

3.1 | Diffusion charging of aerosols

Brownian motion of ions, independently of the applied electric field, induces collision with aerosols and the consequent charging. The probability of ion-aerosols collision depends on particle radius ($r_p$), the ion density ($N_i$) and the aerosol's residence time ($\tau$) in the ion cloud. The resulting electric field around the charged aerosols will repel
ions and decrease the charging rate. For the aerosols with $0.1 \mu m < d_p < 2 \mu m$, the charge number of per aerosol ($n_p$) is defined as

$$n_p(t) = \frac{4\pi\epsilon_0 r_p k_b T}{e^2} \ln\left(1 + \frac{e^2 c_i r_p N_i \tau}{4\pi\epsilon_0 k_b T}\right)$$

where $\epsilon_0$ is absolute permittivity of the void, $r_p$ is the radius of the particle, $k_b$ is the Boltzmann constant, $T$ is the environment temperature, $e$ is the charge of electron, $c_i$ is the mean thermal speed of ion, $N_i$ is the ion concentration, $\tau$ is the aerosol’s residence time. Because there are no theoretical maximum of the charge imparted to aerosols by diffusion charging, characteristic charging time cannot be used [3], actually, the diffusion charging of 0.1 $\mu m$ aerosols to 3$e$ usually takes up 1 ms [3]. Based on the charging condition and aerosols properties, the charge distribution is Boltzmann’s distribution with a mean charge shifted from zero [73].

The diffusion charging method have been used to develop the particles surface area monitor (NSAM) [74, 75]. The NSAM mix particles with positive ions generated by corona discharges in a mixing chamber. A conductive filter can collect the positively charged particles, and then an electrometer connected to a sensitive amplifier measure the current. Excess ions can be absorbed by an ion trap before they get into the conductive filter. The relations between the lung deposited surface area (LDSA) concentration of particles and the electrometer current can be expressed as $S_A = k \times I$, where $S_A$ is LDSA concentration, $k$ is the calibration coefficient, and $I$ is the electrometer current (FA). This relation is consistent for the aerosols diameter between tens of nm and 400 nm. The surface area concentration of PM2.5 aerosols can be approximately measured by this method [76].

The bipolar diffusion charging can make aerosols acquire both positive and negative charges after collision with positive and negative ions [77]. If the product of ion concentration and residence time is high enough, aerosols reach stationary charge states. The bipolar diffusion charging has been also important in aerosol sizing and measurement systems [78]. The corona discharge can easily generate the high density positive and negative ions (up to $10^9$/cm$^3$) and therefore neutralize aerosols at higher flow rates and concentrations, even including large particles of micrometre and super-micrometre size [79]. The Coulomb screening effect of charged aerosols can be relatively strong, when high ion density in the ionizer is achieved ($n_i \approx 10^9$–$10^{10}$ cm$^{-3}$). The corresponding Debye radius $R_D \approx 100 \mu m$ at such ion density, therefore the screening effect might be neglected for small particles of micrometre and sub-micrometre size, $R_p << R_D$. The traditional theory of bipolar charging of aerosols particles in the continuum regime, based on the solution of the diffusion equation for ions in the Coulomb electrostatic field of charged particles, can explain the charging mechanism of these small particles, reasonably. However, for the large particles ($R_p \geq 10 \mu m$), the Coulomb field screening effect becomes important, and its theory of bipolar charging should base on the diffusion equations for positively and negatively charged ions migrating to the central particle in the collective electrostatic field with the potential obeying the Poisson equation [78].

### 3.2 Field charging of aerosols

The classified field charging mechanism describes the charging rate of a spherical aerosol, within an environment of highly gaseous ion density ($\sim 2 \times 10^{10}$/cm$^3$) with an applied electric field [80]. Ions travel along the electric field lines and collide with particles when the field lines intersect particles. The number of field lines intersect the particle diminishes when the particle is charged, and the subsequent charging rate also decreases, until the charge on particles is saturated, afterwards, charges can only be added to particles through diffusion charging mechanism. The saturation charge $n_s$ of a particle in an electric field $E_0$ is developed based on Pauthenier’s equation as [80].

$$n_s = \left(1 + \frac{2(e-1)}{(e+2)}\right) \frac{4\pi\epsilon_0 E_0 r_p^2}{e}$$

where $r_p$ is the particle radius, and $e$ is the dielectric constant.

The charging rate $dn_p/dt$ is defined as

$$\frac{dn_p}{dt} = \frac{eN_i \mu_i n_s}{4\epsilon_0} \left(1 - \frac{n_p}{n_s}\right)^2$$

where $N_i$ is the ion density, and $\mu_i$ is the electrical mobility of ions.

For the initially neutral particle, the charge of particle after a time $\tau$ spent in the ion density is defined as

$$n_p = n_s \times \frac{e \mu_i N_i \tau}{(1 + e \mu_i N_i \tau)}$$
This charging law is valid if the following assumptions are met.

i. The Knudsen number is < 1 for the continuum regime. Because the mean free path of gas molecules (≈60 nm in standard temperature and pressure) is smaller than particle diameter, the gas can be considered as a continuous medium.

ii. Particles are not deformed by repulsion forces (charge number much less the Rayleigh charge limit) and spherical.

iii. The diffusion charging can be neglected when the electric field is uniform (spatially constant) and \( \geq 10^8 \) V/m [80]. For example, the electric field in the gas gap of asymmetric dc APNP is \( \approx 10^5 \) V/m and very heterogeneous only in the ionization region, about 100 μm around the wire. The electric field surrounding a particle in the drift region, can be uniform [3].

iv. The ion density is several orders of magnitude higher than the particle density. The corona discharge has the ion density about \( 10^7 \) cm\(^{-3} \), however, the aerosol particle density is between \( 10^8 \) and \( 10^9 \) cm\(^{-3} \) [80]. The excess of ion facilitates the efficient charging of aerosols.

The comparison between theoretical Pauthenier and measured mean number of charges suggests the Pauthenier's law fits well with experimental measurement for \( N_i t \) product \( \approx 10^6 \) scm\(^{-3} \) when aerosols in the range from 0.44 to 1.33 μm are injected into asymmetric dc corona discharge region [3]. The further comparison of the mean charge per particle versus particle diameter suggests Pauthenier's law is valid for charges acquired by diffusion and field charging [80].

### 3.3 Photo charging of aerosols

Atmospheric pressure air discharges can generate UV radiation in the \( \lambda = 250–300 \) nm range [81]. The particle charging based on energetic UV photon usually happens in two different steps. Particles are exposed to UV-irradiation generated by air plasma directly in the first step. UV photons absorbed by particles can lead with certain probability to generate free electrons, when the photon energy is higher than the particle material's work function [82]. Gas molecules immediately capture these electrons and form ions, which are in turn absorbed by particles as the result of charge diffusion and charge desorption of charged particles.

The particle charge distribution is dominantly influenced by diffusion discharge in the second step. Aerosols leaving strong UV emission region contains positively charged aerosols and negative ions. Therefore, the charge level of aerosols can be only influenced by diffusion charging mechanism in the UV irradiation free region. The dynamic behaviour of the photo charging process for an aerosol containing chemically dissimilar particles of different sizes can be described as.

\[
\frac{dN_{q,R_p,C_m}}{dt} = \left[ a_{R_p,C_m}^{q \rightarrow q+1} N_{q-1,R_p,C_m} - a_{R_p,C_m}^{q \rightarrow q+1} N_{q,R_p,C_m} \right] \downarrow \text{photo - charging}
\]

\[
\frac{dn_i}{dt} = \sum_{C_m} \sum_{R_p=R_{p_{\text{max}}}}^{R_{p_{\text{min}}}} \left[ a_{q,R_p,C_m}^{q \rightarrow q+1} N_{q,R_p,C_m} \right] \downarrow \text{production by photo - charging}
\]

\[
d_{q,R_p,C_m}^{q \rightarrow q+1} = \Delta_i \downarrow \text{diffusion to particles}
\]

\[
d_{q,R_p,C_m}^{q \rightarrow q+1} = \beta_{q+1,R_p,C_m} N_{q+1,R_p,C_m} - \beta_{q,R_p,C_m} N_{q,R_p,C_m} \downarrow \text{diffusion charging}
\]

Where \( R_p \) is particle radius, \( C_m \) is chemical composition, \( q \) is charge, \( N_{q,R_p,C_m} \) is representative of a set of balance equations for each charge fraction, \( a_{q \rightarrow q+1} \) is the combination coefficient for the photo charging of a particle from charge level \( q \) to \( q+1 \).

Although the photo charging production of charged particles have not been studied in non-thermal APNP, the experimental study based on UV irradiation system suggests that the photo charging process and particle concentration significantly affect the particle charge distribution. The increasing particle concentration obviously decreases the mean particle charge number at the constant irradiation intensity. Simultaneously, the bipolar charge distribution of charged aerosols is obtained, which is similar to particle neutralization by radioactive source at higher temperatures. This allows consideration of the photo-charger to be the possible replacement of neutralizer [82, 83].

### 3.4 Diagnostics of aerosols

The real time diagnostics of aerosols can be realized by Mie ellipsometry for the detection of nano-particles, by laser Doppler method for the size distribution of particles larger than 0.5 μm, and by laser-induced light emission for chemical analysis [3]. However, before detection or collection for classical chemical analysis, the aerosol's diagnostic tools often
The more economic method, holographic deflection imaging, is also used to measure electric charge on individual aerosol particles. This method uses common optical hardware and a novel particle sizing algorithm for poly-dispersed size distributions [87]. A charged particle sample is introduced into a vertical channel air flow and a horizontal electric field, simultaneously, in-line digital holography is used to simultaneously track particle position and decode size information. Finally, from velocity and size information above, particle charge is calculated using a force balance, with the assumption of Stokes drag on spherical particles [87].

Size distribution can be defined by coupling a size-selection device with a particle counter. Size selection in differential mobility analyser bases on the monotonous decreasing electrical mobility function of singly charged particle with their size [3]. Actually, the bipolar Boltzmann charge distribution includes a defined proportion of singly charged particles, generated by bipolar ion clouds with radioactive sources or bipolar corona neutralizers [90]. Plasma sources are also employed for unipolar charging of aerosols. Afterwards, the collection of charged particles selected in electric field can obtain size distribution information through current measurement [91]. Besides that, the sampling strategies with their related specific limits should be emphasized when dealing with particle measurements. For example, the mean diameter are very different when the y-axis label of the distribution is in number (cm⁻³), surface (cm²cm⁻³) or volume (cm³cm⁻³). Number and surface size distribution are mainly used in physical process discussion, while volume size distributions are used in industrials for filter efficiencies and mass transfer [1–3].

4 | APPLICATIONS OF CHARGED AEROSOLS

Charged aerosols are used widely in aerosols processes and environment protection, and have the advantage of low cost, simple operating condition, adjustable reactive and ionizing properties. Recently, highly charged aerosol particles are used in many industrial applications such as electrostatic painting, separation technology or surface coating. Here, we concentrate on the research progress on the potential applications of charged aerosols in the artificial rain enhancement, biomedicine and chemical processing.

4.1 | Droplet condensation by charged aerosols

Charged particles have long been demonstrated to be able to induce water droplet coalescence. Wilson firstly found that ions generated by radioactive materials acted as condensation nuclei in super-saturated water vapour in small cloud chamber in 1895 [92]. But the question that still exist: can charged aerosols induce macroscopic precipitation in relatively large space?
The rain and snow formation in air induced by corona discharges. (a) The schematic of the 15,000 m$^3$ cloud chamber experiment. (b) The schematic of droplet cross section in the case of neutral condition and charged condition. The cross section of collecting droplets increases with charge amount. (c) Temporal evolution of the particle size distribution in a 15,000 m$^3$ atmospheric cloud chamber (particle radius range: 0.25–10 μm). (d) Generation of snow in the cloud chamber. The macroscopically visible snow induced by corona discharge. Filament-like ice crystals on the corona wire after 30 min operation [89].

Figure 7a shows the schematic of 15,000 m$^3$ cloud chamber to study precipitation phenomenon in larger space. The diameter and height of the cylindrical cloud chamber was 25 and 27.5 m, respectively. For the rain formation experiment, the chamber was first cooled down to 10°C. Superheated water vapour generated by a pressurized boiler was injected into the cloud chamber. Afterwards, the temperature increased to 20 ± 1.5°C. For the snow formation experiment, the chamber was first cooled to −20°C. The temperature increased to −15 ± 1.5°C after the water vapour injection. The stainless wire with the diameter of 0.1 mm and length of 20 m was used as the corona discharge electrode. The applied voltage was −60 kV and the measured current was 10 mA.

The enhanced precipitation by charged particles can be explained by Figure 7b. In the case of pure gravity collisions, the collision efficiency between collector droplet and smaller background droplets is usually much smaller than the unity because of hydrodynamic effect as the repulsion force prevents the droplet collision. However, when the droplet is charged, the attractive force between the collector charge and induced dipole moment of the smaller droplets may dominate at smaller distance and exceed the force of hydrodynamic interactions. Therefore, the collision cross section and the collision frequency increase significantly [92].

The temporal evolution of the particle size distribution in the cloud chamber after injection of water vapour at 20°C is shown in Figure 7c. The particle distribution of aerosols in cloud chamber at $t = 0$ s show that the radius of most particles was less than 1 μm, and no particles larger than 3.75 μm were detected. After the corona discharge was switched on, the growth of larger particles was relatively fast. A lot more particles with radius larger than 2 μm arose after $t = 60$ s, and particle radius even increased to 10 μm after $t = 120$ s.

Figure 7d shows the formation of snow induced by corona discharge in the cloud chamber at −15°C. After the corona discharge was turned on for only 30 s, the snow/ice particles
with hundreds of micrometre to several millimetre in size were generated. Some of the ice particles adhere to the corona wire and formed the filamentary like ice on the wire.

The study on the growth dynamics of single charged droplet in micrometre scale can help understand on the interaction mechanism between charged nuclei and aerosols. Figure 8a shows the cloud chamber to study the enhanced growth of single droplet by controlling equivalent charge on droplet [93]. The cloud chamber is 3 m in height and has a cross section of $2 \times 2$ m$^2$. A 1.5-m-diameter cylindrical stainless steel can which controls temperature, humidity and purity is inserted in the cloud chamber.

A DC power supply is used to produce electric field at the tip of a stainless steel needle so that the droplet can form at the tip. The growth dynamics of the charged droplet is shown in Figure 8b. A 5-μm diameter droplet is used as the starting point. The applied voltage on the needle electrode is 300 V, and the electric field on the edge of droplet is $4.9 \times 10^7$ V/m (Figure 8c). The equivalent charge on the surface of droplet is $2.15 \times 10^6$ e (e is electron charge). The diameter of the charged droplet increases to 10.5 and 19 μm at the 72nd and 180th second, respectively. Afterwards, the stable droplet means the evaporation and collection of water molecules reach balance. The electric field at the edge of 19 μm charged droplet is $1.8 \times 10^7$ V/m.

The increasing charge (realized by higher applied voltage) enhances the collection of water molecules and small aerosols, so the stable diameter of the charged droplet increases to 50 μm when the equivalent charge on 5 μm droplet increases to $5 \times 10^6$ e (applied voltage 700 V, Figure 8b). The collision rate coefficient between charged droplet (5 μm) and aerosols shown in Figure 8d indicates that for 0.1 μm radius aerosol, the collision efficiency coefficient increases from increases from $2.1 \times 10^{-11}$ to $8.3 \times 10^{-11}$ m$^3$ s$^{-1}$ as the equivalent charge of droplet increases from $1.43 \times 10^5$ e to $5 \times 10^6$ e. The collision efficiency coefficient of charged droplet is three orders of magnitude higher than that of uncharged droplet for the aerosols smaller than 0.03 μm. The further calculation indicates that the contribution of electric force to the growth of charged droplet is at least three orders of magnitude higher than thermophoretic and diffusophoretic forces [93, 94].

The measurement of charge amount of charged aerosols at humidity supersaturation condition is a challenge for the traditional measurement device. Figure 9a shows the system to measure the charge amount at the humidity supersaturation of 130 ± 10%. The air temperature inside the chamber was 2°C ± 1°C [45]. The negative DC high voltage coupled air plasma jet array (introduced in Section 2.3.3) can generate plasma with volume more than 2 cm$^3$, which can charge the aerosols efficiently.
Figure 9b shows the braking experiment (the deceleration of aerosols in horizontal movement) of the charged aerosols to measure the charge amount. The negative electrostatic field can stop the propagation of the negative charged aerosols, and the horizontal movement of charged aerosols is inversely proportional to the charge amount. Therefore, the uncharged aerosols were not affected by the negative electrostatic field. However, the most of charged aerosols generated by negative air plasma with the driving voltage of −10 kV stop at the position of 0.027 m from the electrode. The travelling distance of these charged aerosols is 0.35 m. The averaged charge of charged aerosols calculated is 3740 e. As the discharge voltage decreased from −10 to −15 kV, the travelling distance of charged aerosols decreases from 0.35 to 0.25 m. The averaged charge of charged aerosols increased to 5818 e, which was ~20% of the maximum charge (31250 e) that the aerosols with diameter of 2 μm [98].

These highly charged aerosols can obviously accelerate the settling process of droplet. Figure 9c shows droplets mainly distributed in the range 5~40 μm for the natural settling case. More frequent collisions happened between charged aerosols and uncharged water molecules and aerosols [96–102] therefore, the droplets distributed in the range of 40~70 μm for the case of charged aerosols settling case, while, the uncharged aerosols can only cause limited increase of the particle size distribution compared with natural settling case.

The indoor experiment above lay the foundation for the setup of large scale corona discharge system to generate charged aerosols in the open air. The large scale corona discharge system (Figure 10a,b) has the floor area ~11304
FIGURE 10 Large-scale installation for ion generation for precipitation of atmospheric aerosols, using over 300,000 corona plasma discharges. (a) The large-scale corona discharge system consists of six poles and 7.2 km electric wires. (b) The design diagram of the hexagonal discharge arrays. (c) The satellite image of the test zone (taken from https://map.baidu.com, last access: 29 December 2019). The hexagon shows the corona plasma discharge system. The red points show the locations of ion density measurements. (d) The hydrogen balloon carried the ion density measurement device. (e) The ion density distribution in vertical direction and downwind direction (experimental measurement, applied voltage – 90 kV).

m². Six poles each with the height of 20 m supporting the 7.2 km long wire electrode are erected vertically and arranged in a regular hexagon array. The wire electrodes are divided into two layers, at the height of 20 and 15 m, that is, with 5 m separation between the layers. There are 10 wire electrodes in each layer, and the horizontal distance between the wires is 50 cm to avoid the mutual interference. The stainless-steel wire has six strands and a diameter of 1 mm [44].

The satellite image of test zone is shown in Figure 10c. The hexagon shows the large scale corona discharge system. The red points show the location of the ion density measurements. The ion counter carried by the hydrogen balloon is used to measure the vertical (5 ~ 50 m) and horizontal (20 ~ 50 m) ion density in the downwind as shown in Figure 10d.

Figure 10e shows the coverage range of negative ions generated by the large corona discharge system shown in Figure 10a. The ion density decreases from 5 x 10⁵/cm³ to 8 x 10³/cm³ as the distance from the wire electrode increases from 20 to 30 m. Afterwards, the ion density decreases to the background value as the horizontal distance increases to 40 m. The relatively high density 10⁴/cm³ at the horizontal position of 50 m can be attributed to the random enhanced transport by gusts. The width of the coverage area in radial direction is 90 m. Therefore, the whole coverage volume was approximately 30 (long) x 23 (height) x 90 m (width). Aerosols can combine with the ions generated in this volume, thus creating a large number of charged nuclei within the volume [103–107].

4.2 Biomedical application of charged aerosols

The interactions between liquid vapour in the micrometre range (aerosols) and plasma not only couple ions or electrons on the aerosols, but also make the gaseous reactive oxygen and nitrogen species (RONS) dissolve in the small liquid aerosols. These aerosols with high concentration of dissolved RONS are called as plasma activated mist (PAM). The plasma jet can be used to generate PAM. The plasma jet is produced along the noble gas channel because the noble gas can extend the length of plasm plume and increase the active species concentration. The liquid vapour is delivered through another channel. The plasma and the aerosol mix in the chamber behind the two channels [108]. The air DBD similar to the device introduced in the Section 2.2 can also be used to generate air plasma perpendicular to the aerosols flow. Different from the plasma jet system, the longer dielectric barrier covered electrode tube is needed to ensure the well mixing between plasma and liquid aerosols. The air flow of ultrasonic nebulizer transports directly aerosols to the air DBD tube, which simplifies the gas supply [91, 92].

The ultrasonic nebulizer provides a humidity over-saturation environment in the plasma chamber. The humid air promotes the production of OH and H₂O₂ by the plasma. The air flow lead aerosols through the plasma, and the aerosol with high surface-volume ratio absorb the reactive plasma species including OH, H₂O₂, O₃, NO₃⁻ and so on efficiently (Figure 11) [66]. The interaction between aerosols and plasma also produce lots of charged aerosols. The charged aerosols
absorb the small uncharged aerosols through the image charge force (a short range attractive force proportional to $q^2$).

The size of wet aerosol produced by ultrasonic nebulizer are mainly distributed in 2~3 μm and 4~6 μm. Figure 8 shows the diameter of single charged aerosol (equivalent charge $\sim 2.15 \times 10^4 e$) increases from 5 to 19 μm during 180 s under the humidity supersaturated conditions [109]. Although the time of aerosol stay in the plasma chamber is less than ~1 s, the density of aerosol produced by ultrasonic nebulizer is about $10^5$/cm$^3$, so the plasma with charge density of $10^{12}$~$10^{14}$/cm$^3$ can produce highly charged aerosol, which can make up the insufficient time to absorb the small uncharged aerosols. Eventually, the highly reactive, growing aerosol deposits continuously onto the object surface to achieve optimal therapeutic outcomes.

Aerosol propagation is an important way of disease transmission such as COVID19, SARS and MERS. The charging and electrostatic adsorption of aerosols provides a new way to inactive the airborne viruses [12]. The packed bed DBD reactor can decrease the viral aerosol concentration by 2.3 log. The reactor consists of a larger Plexiglas tube and two smaller tube. The smaller tubes slide freely relative to the larger tube. Over 500 inert borosilicate glass beads are inserted in the space between the larger and smaller tube.

Lots of filamentary discharges are generated in the narrow spaces between the beads. Ions generated by these filamentary discharges can charge viral aerosols, afterwards, the AC electric field can possibly make the charged aerosols oscillate in the space between the beads and extend the transition time of aerosols through the reactor, therefore, RONS and UV generated by plasma has enough time to inactivate these airborne viruses [12]. When the reactor is powered by 30 kV AC high voltage, the reactor causes a decrease of infected virus concentration of more than 2.3 log infective MS2 [12]. In contrast, the nearly constant reduction in gene copy concentrations with and without plasma suggests that the virus is inactivated, and not removed physically.

### 4.3 Chemical processing by charged aerosols

Droplets dispersed in gas can transform into solid particles by precipitation during spray drying. This process is also a micro-sized reactors for the synthesis of multi-metal oxides s with adjustable size, structure and stoichiometry indeed [85]. In addition to that, liquid aerosol can transport reactants to the substrate for film synthesis easily through aerosol based CVD in atmospheric pressure thermal plasmas [110].

Atomised spray plasma deposition (ASPD) based on perfluorotributylamine–nanoparticle slurry mixtures can generate super-hydrophobic nanocomposite layers in a single solventless step [111]. ASPD is carried out in an electrodeless, cylindrical, T-shape glass reactor enclose in a Faraday cage. The copper coil driven by RF power supply can generate large volume plasma which can charge 20 μm diameter median droplet efficiently. The optimal ASPD rate based on these charged droplets for the perfluorotributylamine precursor is about $49 \pm 4$ nm min$^{-1}$ at a liquid flow rate of $16 \pm 4 \times 10^{-4}$ ml s$^{-1}$, while the atomized spray deposition without plasma is below $0.1 \pm 0.1$ nm min$^{-1}$ [111]. Besides that, the charged drops of monomer produced by DBD electro-spray coating in air is also used for polymer coatings. The thickness of film can be adjusted from a few tens of nanometre to micrometre [3]. The polymer film deposited by these charged droplets, can resist hot water washing required for bio-functionalization of surfaces [3].

Electro spray of charged droplets is also used emulsion and suspension preparation. For example, when two immiscible solutes are mixed by spraying 15% dextran aqueous solution in 8% PEG aqueous solution [112], cytompatible capsules suspended in water can be achieved by water in water emulsions. This method is also used for the generation of composite particles suspended in liquids, for example, the electro spray of polystyrene beads in a aqueous solutions is used for surface functionalization and immunoassay bio-diagnostics [113], and the electro spray of chitosan core in a calcium phosphate solution is used for encapsulation of liquids up to living yeast cells in hydroxyapatite shell [114].

### 5 CONCLUSION

APNP have become a very effective new way to generate charged aerosols. Except the traditional pin-plate or wire-plate APNP sources, the large scale corona discharge system and air plasma jet array can increase the ion density in open air and generate adjustable large plasma plume, respectively. These new plasma sources are helpful in increasing the charging efficiency. Although the interactions between plasma and aerosols is quite
complicated, the preliminary study suggests that the diffusion charging dominates aerosols below 0.1 μm, field charging dominates aerosols larger than 1 μm, and the photo charging also contributes to aerosols charging. These reactive charged aerosols have shown their potential in the artificial rain enhancement, biomedicine, and material processing.

Fundamental, technical and economical contexts are favourable, regarding the following:

i. The charging, coagulation and movement of aerosols in the discharge region can be analysed by finite element simulation tools from aerosols transportation and coagulation, to discharge dynamics

ii. Charged aerosol generation and diagnostic tools for studies and online monitoring down to sub-micrometre scale as well as low cost plasma sources are available

iii. The low cost and environment friendly charged aerosols generation system will be widely applied in biomedicine and material processing area

Some basic processes related with charged aerosols generation and processing by APNP still need further investigation. The nucleation induced by ions and the effect of photo charging have not been studied systematically. Moreover, the surface reactions between plasma and aerosols not only charge aerosols and change the functionality of aerosols, but also affect the discharge stability. In that respect, the study on reaction chain between plasma, suspended particles and gaseous reactants can speed up the charging process.

In the end, quite new APNP sources have been developed during the past decade. The portable, low cost and environment friendly air plasma sources with adjustable discharge intensity can be employed to develop the charged aerosols sources used in daily life. The gliding arc and propeller arc, with the high reactivity and selectivity of plasma, can be used to control size and structure of charged aerosols, so to increase the material surface processing efficiency.

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