

CHAPTER 2

THEORY

In this Chapter, some of the basic fundamentals of aerosol particle properties and theories needed to characterize the performance of the electrical mobility spectrometer (EMS) are presented briefly. The first Section of the Chapter covers some fundamental properties of aerosol particles. The second Section describes briefly a theory of the particle motion in a gas. In the third Section, a brief review of the particle charging mechanisms, and the unipolar diffusion and unipolar field chargings are described. The flow field and electric field modelings are described in the fourth and last Sections of the Chapter, respectively.

2.1 Aerosol Properties

Aerosol is defined as a suspension of any solid or liquid particles (or a combination of both) in a gas with diameter in the range between 1 nm and 100 μm (Hinds 1999). The most important parameter for characterizing the behavior of aerosols is the particle size. The behavior, chemical composition and physical properties of aerosol particles vary strongly as a function of the particle size. Most aerosols consist of a number of different particle sizes. Monodisperse aerosols by definition consist of particles that have all the same size and can be produced in the laboratory for use as test aerosols. However, most aerosols are polydisperse, with a wide range of particle sizes, and statistical measures should be used to characterize their particle size. The distribution of particle sizes within the aerosol is referred to as the aerosol size distribution. Usually, polydisperse aerosol size distributions are well described by a lognormal distribution function. There is no theoretical basis for the application of the lognormal distribution function; its particular suitability has been arrived at through a great deal of empirical experience. The lognormal distribution function is given by the following equation (Hinds 1999):

$$df = \frac{1}{\sqrt{2\pi} \ln \sigma_g} \exp\left(-\frac{(\ln d_p - \ln \bar{d})^2}{2(\ln \sigma_g)^2}\right) d \ln d_p \quad (2.1)$$

where d_p is the particle diameter, \bar{d} is the count median diameter, and σ_g is the geometric standard deviation given by the equation

$$\sigma_g = \exp\left(\frac{\sum n_i (\ln d_i - \ln d_g)^2}{N-1}\right)^{1/2} \quad (2.2)$$

where d_g is the number weighted geometric mean diameter, or the count mean diameter, is given by (Hinds 1999).

$$d_g = \exp\left(\frac{\sum n_i \ln d_i}{N}\right) \quad (2.3)$$

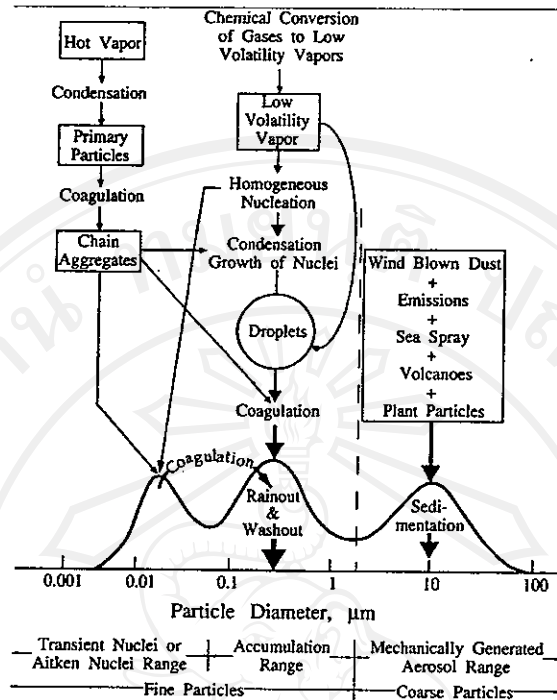


Figure 2.1 Atmospheric aerosol size distributions (Flagan 1998).

where n_i is the number of particles in group i , having a midpoint of size d_i , and N is the total number of particles (Hinds 1999).

The most common way to present the particle size distribution of aerosol is in terms of the three modes, referred to as the nuclei, accumulation, and coarse particle modes, shown in Figure 2.1. The nuclei mode is composed of particles which have equivalent diameters of less than about 100 nm. Nuclei mode particles are produced from recent source emissions and particles formed in the atmosphere by gas-to-particle conversion and condensation processes. They are present in great quantity and thus dominate the number weighted aerosol size distribution. However, since the nuclei mode particles are so small, they have very little mass associated with them and consequently contribute very little to the mass weighted size distribution. The accumulation mode is composed of particles which have equivalent diameters in the size range between approximately 100 nm and 1 μm , primarily sulphuric acid, ammonium sulphate and ammonium nitrate, organic products, smog particles, combustion particles, and nuclei mode particles that have coagulated with accumulation mode particles. This mode is where most of the mass of atmospheric aerosol is, and it is this size range that acts as the cloud condensation nuclei. Because accumulation mode particles are formed primarily through the agglomeration of the smaller nuclei mode particles, they are present in much smaller (though still highly significant) numbers. Because of their greater size, accumulation mode particles carry significant mass with them, allowing them to dominate the mass weighted particle size distribution. The coarse mode is composed of particles which have equivalent diameters greater than approximately 1 μm , such as windblown dust, crustal material, large sea-salt particles from sea spray, and mechanically generated anthropogenic particles such as those from agriculture and surface mining. Even though coarse mode particles are highly massive (when compared with nuclei and accumulation mode particles), they are present in negligible number compared to nuclei and accumulation mode particles; consequently, they do not contribute significantly to either the number or mass weighted size distributions.

The shape of aerosol particles is another factor that varies considerably. Liquid aerosol particles are nearly always spherical, while solid aerosol particles usually have complex shapes; most other types of particles are nonspherical. Some have regular geometric shapes, such as cubic, cylindrical, single crystals, or clusters of spheres. The shape of a particle affects its drag force and settling velocity. For the most application, an aerosol is usually assumed to be spherical in nature, leading to the concept of the equivalent diameter, which is the diameter of the sphere that has the same value of a particular physical property as that of an irregular particle. For approximate analysis, shape can usually be ignored. Particle with extreme shapes, such as long, thin fibers, are treated as simplified nonspherical shapes in different orientations. A correction factor called the dynamic shape factor is applied to Stokes's law to account for the effect of shape on particle motion. It is defined as the ratio of the actual resistance force of the nonspherical particle to the resistance force of a sphere having the same volume and velocity as the nonspherical particle, and is given by

$$\chi = \frac{F_D}{3\pi\eta V d_e} \quad (2.4)$$

where F_D is the actual resistance force, η is the gas viscosity, V is the particle velocity, and d_e is the equivalent diameter. The shape of aerosol particles having a complex structure, such as an agglomerated metal fume or soot particle, can be characterized in terms of a fractal dimension. The fractal dimension relates a property such as the perimeter or surface area of an object to the scale of the measurement (Hinds 1999).

2.2 Particle Motion in Gases

The following paragraphs describe the theory of the particle motion in a gas. These include the gas viscosity, the Reynolds number and annular flow velocity profile, the molecular and particle velocity, the mean free path and Knudsen number, the particle Reynolds number, the settling velocity and mechanical mobility, the Stokes number and inertial impaction, the electrical mobility, the Brownian motion and thermal diffusion and the particle losses.

2.2.1 Gas Viscosity

Viscosity is the constant of proportionality for relationship between shear stress and velocity gradient for a Newtonian fluid:

$$F \propto \frac{du}{dy} \quad (2.5)$$

The dynamic viscosity for air at 20°C is 1.81×10^{-5} Pa·s. Viscosity can be related to a reference viscosity, η_r , at a reference temperature, T_r , as following equation (Willeke and Baron 1993):

$$\eta = \eta_r \left(\frac{T_r + S}{T + S} \right) \left(\frac{T}{T_r} \right) \quad (2.6)$$

where S is the Sutherland interpolation constant (Willeke and Baron 1993). It should be noted that viscosity is independent of pressure.

2.2.2 Reynolds Number and the Annular Flow Velocity Profile

The fluid flow inside the electrical mobility classifier is operated at laminar flow conditions. To obtain that, the Reynolds number has to be kept within the laminar regime. The Reynolds number is a dimensionless number that characterizes a fluid flow through a pipe. The most commonly used classifier employ annular flows and the Reynolds number in the annular flows geometries is defined as (Janna 1993):

$$\text{Re} = \frac{2r_2(1-\kappa)\rho\bar{U}}{\eta} \quad (2.7)$$

where r_2 is the radius of the outer cylinder of the annulus, κ is the diameter ratio of the inner over the outer cylinder, \bar{U} is the mean (axial) flow velocity, and ρ is the gas density. The velocity profile in the annular flow geometries is given by

$$u(r) = \frac{r_2^2}{4\eta} \left(-\frac{dp}{dz} \right) \left(1 - \frac{r^2}{r_2^2} - \frac{1-\kappa^2}{\ln(\kappa)} \ln\left(\frac{r}{r_2}\right) \right) \quad (2.8)$$

where dp/dz is the constant pressure gradient (Janna 1993).

2.2.3 Molecular and Particle Velocity

Derived from the ideal gas law, the square root of the mean square velocity of every particle or molecule of the gas is given by

$$c_{rms} = \sqrt{\frac{3RT}{M}} \quad \text{for molecules} \quad (2.9)$$

$$c_{rms} = \sqrt{\frac{3kT}{m}} \quad \text{for particles} \quad (2.10)$$

where R is the gas constant, M is the molecular weight of the gas, k is Boltzmann's constant, m is the molecular mass of the gas and T is the absolute temperature. Because aerosol particles exchange energy with the surrounding gas molecules they have the same kinetic energy. The kinetic energy of one molecule of particle within the gas is given by the following equation

$$KE = \frac{3RT}{2} \quad (2.11)$$

However, all molecules or particles do not have the same velocity at an instant time in the gas. Assuming a Maxwell-Boltzmann velocity distribution for the molecular velocity along any direction is given by

$$f(c)dc = 4\pi c^2 \left(\frac{m}{2\pi kT} \right)^{3/2} \exp\left(-\frac{mc^2}{2kT}\right) dc \quad (2.12)$$

where $f(c)dc$ is the fraction of velocities between c and $c + dc$ (Hinds 1999). The average mean molecular or particle velocity is given by integrating Equation 2.12 over all possible speeds

$$\bar{c} = \int_0^{\infty} cf(c)dc = \left(\frac{8kT}{\pi m}\right)^{1/2} = \left(\frac{8RT}{\pi M}\right)^{1/2} \quad (2.13)$$

2.2.4 Mean Free Path and Knudsen Number

The mean free path of a gas is defined as the average distance a molecule travels before colliding with another molecule. The mean free path, λ , of a gas can be determined from the average number of collisions a particular molecule undergoes in one second, n_z , and the average distance it has traveled in that second, is given by

$$\lambda = \frac{\bar{c}}{n_z} \quad (2.14)$$

where n_z is the average number of collisions between molecules is given by

$$n_z = \sqrt{2}n\pi d_m^2 \bar{c} \quad (2.15)$$

where n is the concentration of molecules, the number per unit volume, d_m is the collision diameter of the particle, defined as the distance between the centers of two molecules at the instant of collision. Combining Equation 2.14 and 2.15 gives

$$\lambda = \frac{1}{\sqrt{2}n\pi d_m^2} \quad (2.16)$$

For air at 20°C and atmospheric pressure, the mean free path of air is 0.066 μm (Hinds 1999). Willeke (1976) showed that the mean free path of a certain aerosol as a function of pressure, P , and temperature, T , is given by the following equation

$$\lambda = \lambda_r \left(\frac{P_r}{P}\right) \left(\frac{T}{T_r}\right) \left(\frac{1+(S/T_r)}{1+(S/T)}\right) \quad (2.17)$$

where λ_r is the mean free path at the reference conditions, P_r and T_r are the pressure and temperature in the reference condition, respectively, and S is the Sutherland constant.

The aerosol transport regime as defined by the Knudsen number, Kn . This defines how the fluid suspending the particle views the particle. It is defined as the ratio of the gas mean free path to the diameter of the particle is given by

$$\text{Kn} = \frac{2\lambda}{d_p} \quad (2.18)$$

where d_p is the diameter of the particle and λ is the gas mean free path. For aerosol particles, this is taken as the mean free path of air. The Knudsen number is used to define which the

transport regime and associated equations should be used when dealing with aerosols. There are three cases: the free molecular regime ($d_p \ll \lambda$), the continuum regime ($d_p \gg \lambda$) and the transition regime ($d_p \approx \lambda$).

2.2.5 Particle Reynolds Number

Particle Reynolds number is an important parameter used in the classification of aerosol dynamics, which describes the aerodynamic properties of the particle. It is defined as the ratio of the inertial forces to the frictional forces acting on the particle, and is given by (Willeke and Baron 1993):

$$\text{Re} = \frac{\rho V d_p}{\eta} \quad (2.19)$$

where ρ is the gas density, V is the relative velocity between the fluid and particle, d_p is the particle diameter and η is the gas viscosity. Generally, if $\text{Re} < 1$, the flow around the particle can be described as laminar flow, which will be a requirement for many practical applications.

2.2.6 Settling Velocity and Mechanical Mobility

When an aerosol particle moving with a net velocity corresponding to the carrier gas which it is suspended in, the particle is subject to the gas resistance force or drag force, F_D . This provides the following Stoke's law formulation for drag as

$$F_D = 3\pi\eta V_p d_p \quad (2.20)$$

where V_p is the velocity of the particle and d_p is the particle diameter. Stoke's law is a specific solution to the generally insoluble Navier-Stokes equations. The Navier-Stokes equations are the general differential equations describing fluid motion and are used to describe the drag force acting on a particle, with the assumptions that inertial force are small when compared with viscous force ($\text{Re} < 1$, i.e. particle Reynolds number is small), motion is constant, the particle is a rigid sphere with no walls or other particles nearby, the fluid is incompressible, and the fluid velocity is zero at the particle surface. For the size of particles and flow conditions being dealt with here, all of the above assumptions are valid, except for the assumption of zero fluid velocity at the particle surface. As particle size approaches the gas mean free path, the particles experience "slip" at their surface (i.e. the velocity of the surrounding fluid at the particle surface is not zero). In order to account for this surface slip, the empirical correction factor, C_c , the Cunningham slip correction factor, is therefore introduced as a correction factor to allow for this, giving Stokes' law as

$$F_D = \frac{3\pi\eta V_p d_p}{C_c} \quad (2.21)$$

The value of C_c has been reevaluated over the year. It was first derived by Cunningham (1910) and later by Allen and Raabe (1982) who reanalyzed the earlier data of Millikan from experiments carried out between 1909 and 1923, giving C_c as

$$C_c = 1 + \frac{\lambda}{d_p} \left(2.34 + 1.05 \exp \left(-0.39 \frac{d_p}{\lambda} \right) \right) \quad (2.22)$$

For air at 20°C and atmospheric pressure, C_c has a value of 224.332 for a 1 nm particle and 1.155 for a 1 μm particle (Hinds 1999). The terminal settling velocity, V_{TS} , of a particle suspended in a still gas can be determined by equating the Stokes drag force to the gravitational force (i.e. $F_D = F_G$). Neglecting buoyancy force, this terminal settling velocity becomes

$$V_{TS} = \frac{\rho_p d_p^2 g C_c}{18\eta} \quad (2.23)$$

where ρ_p is the particle density and g is gravitational acceleration. It should be noted that the settling velocity is not significant for particles smaller than 1.0 μm in diameter (Hinds 1999).

The ability of a particle to move in the suspending gas and a specific force field is referred to as the particle mobility. The particle mobility, B , is defined as the ratio of the terminal velocity of particle (with respect to carrier gas) to the magnitude of the drag force exerted on the particle, and for small particles is given by

$$B = \frac{V}{F_D} = \frac{C_c}{3\pi\eta d_p} \quad (2.24)$$

where the definition of diameter being used is that of aerodynamic diameter. The particle mobility has unit of m/Ns and is often called mechanical mobility. Knowing the mobility of any particle in the gas its terminal velocity can be calculated given the acting force.

2.2.7 Stokes Number and Inertial Impaction

If the particle is accelerated to some initial velocity V_0 by a force which is then removed, the aerodynamic drag force will tend to decelerate the particle until it is traveling at the same velocity as the fluid, V_f . The distance which the particle travels as it decelerates from V_0 to V_f in the absence of external forces is given by the stopping distance, S :

$$S = B m_p V_0 \quad (2.25)$$

where B is the particle mobility and m_p is the particle mass. This stopping distance is typically quite small; for a particle with an initial velocity of 1000 cm/sec in a stationary fluid, the stopping distance is 3.6×10^{-3} cm for a 1000 nm particle and 6.8×10^{-6} cm for a 10 nm particle.

When the aerosol flow comes to an obstruction (e.g. a bend in a tube), the fluid flow must change its direction. However, because the aerosol particles have a greater momentum than the gas molecules, they will not be able to change their direction of travel as quickly as the gas. If particle momentum is sufficiently high, it will not be able to follow the path of the gas flow; the particle will then cross the gas streamlines and impact on the obstruction (e.g., the tube wall). The tendency of a particle to either follow gas streamlines or

impact inertially is measured by the particle Stokes number, Stk . The Stokes number is defined as the ratio of particle stopping distance to the characteristic dimension, d_c , of the obstruction is given by the following equation

$$Stk = \frac{S}{d_c} = \frac{\rho_p d_p^2 V_f C_c}{18\eta d_c} \quad (2.26)$$

where the Stokes definition of particle diameter is used. It has been experimentally determined that significant impaction losses occur for particles with Stokes numbers greater than 0.6.

2.2.8 Electrical Mobility

When a charged particle enters the presence of an electrostatic field, a charged particle will experience a net electrostatic force, F_E , is given by

$$F_E = neE \quad (2.27)$$

where n is the net number of elementary charges on the particle and e is the value of elementary charge on an electron and E is the electric field strength. In the absence of any other external forces (generally $F_E \gg F_G$), the electrostatic force will be balanced by the Stokes drag force (i.e. $F_E = F_D$), resulting in a terminal electrostatic velocity of a charged particle, V_{TE} , is given by

$$V_{TE} = \frac{neEC_c}{3\pi\eta d_p} = neEB \quad (2.28)$$

The ability of a charged particle to move in the presence of an electrostatic field is referred to as the electrical mobility of particles, Z_p , the velocity of a charged particle in an electric field of unit strength is given by

$$Z_p = \frac{V_{TE}}{E} = \frac{neC_c}{3\pi\eta d_p} \quad \text{for } Re < 1 \quad (2.29)$$

and is directly related in terms of the particle mechanical mobility as

$$Z_p = qB = neB \quad (2.30)$$

2.2.9 Brownian Motion and Thermal Diffusion

The random motion of particles due to collision with gas molecules is the Brownian motion. This random motion causes particle diffusion in a concentration gradient. Hence, there is a net transport from higher to lower concentrations with the gas. The net flux of particles, J , is in the direction of lower concentration and characterized by the particle diffusion coefficient, D . The larger the value of D , results in the more vigorous the Brownian motion, and the more rapid the mass transfer in a concentration gradient. This motion, in the absence of external forces, is described by Fick's first law of diffusion is given by

$$J = -D \frac{dn}{dx} \quad (2.31)$$

where dn/dx is the particle concentration gradient. The diffusion coefficient, D , given by the Stokes-Einstein equation (Hinds 1999)

$$D = kTB = \frac{kTC_c}{3\pi\eta d_p} \quad (2.32)$$

The diffusion coefficient has unit of m^2/s . Given the diffusivity of a particle, the root mean square net displacement due to the particle Brownian motion is given by

$$dx_{rms} = \sqrt{2Ddt} \quad (2.33)$$

where dt is the traveling time.

Some effects occur when a temperature gradient is established for the surrounding gas molecules. The force experienced by an aerosol molecule is proportional to this temperature gradient and in the direction of decreasing temperature. This is the case of hot aerosol gas the blows over a hot surface. Particle are transported and deposited on the cold surface. The thermal force, F_T , on a particle with diameter, d_p , is given by

$$F_T = -\frac{p\lambda d_p^2 \nabla T}{T} \quad \text{for } d_p < \lambda \quad (2.34)$$

$$F_T = -\frac{9\pi\eta^2 d_p H \nabla T}{2\rho_g T} \quad \text{for } d_p > \lambda \quad (2.35)$$

where p is the gas pressure, ∇T is the temperature gradient, ρ_g is the gas density and H is a function of the thermal conductivities of gas and particle, the particle size and the gas mean free path. The negative sign indicates that the force is always directed in the opposite direction of the gradient (i.e. from hot to cold)

2.2.10 Particle Losses

(a) Sampling Losses

An isokinetic sampling system is generally desirable to obtain a sample of particles from a flowing gas stream. In an isokinetic sampling system, the sampling probe is isoaxial with the flow (i.e. it is aligned in parallel with the gas streamlines) and the sample velocity entering the probe is identical to the free stream gas velocity, the isokinetic condition for a properly aligned probe is

$$U = U_0 \quad (2.36)$$

where U is the free stream gas velocity and U_0 is the sample velocity in the probe. Any deviation from isokinetic conditions may result in under-sampling or over-sampling of particle in certain size ranges due to inertial effects. In addition, further losses may occur due

to inertial impaction of particle at the sample tube inlet. Fortunately, under normal conditions, sampling losses due to inertial impaction and anisokinetic sampling generally become significant only for particle larger than 1 μm . Consequently, for particle smaller than 1 μm , sampling losses are not a significant issue (Graskow 2001).

(b) Transport Losses

Particle should be transported from the sample tube inlet to analyzer without bias. However, particles of different sizes penetrate sampling tubes with different efficiencies. Two of the most common mechanisms for particle losses during aerosol transport are due to diffusion and thermophoresis, as described above. Diffusion loss results in the selective loss of smaller particles as they diffuse to and are deposited on tubing walls. In the tube flow, the transport efficiency with diffusive particle loss, η_{diff} , can be expressed as (Willeke and Baron 1993)

$$\eta_{\text{diff}} = \exp\left(-\frac{\pi d L V_{\text{diff}}}{Q}\right) \quad (2.37)$$

where d is the inside diameter of tube, L is the length of the tube, V_{diff} is the deposition velocity for particle diffusion loss to the wall, and Q is the volumetric flow rate of gas through the tube. The most effective method for reducing diffusion losses is simply to minimize the physical length of the transport tubing. Thermophoretic deposition is only a cause for concern for aerosols that are hot (as is often the case with combustion aerosols). Thermophoresis can be reduced by heating and insulation of sample lines as well as by reduction of aerosol temperature through dilution.

Precipitation of small particles by electrostatic effects due to image force and space charge effects can occur in the sampling lines during transport. Because it is not always possible to know the distribution of charge on aerosol particles or the electric field in a sampling line that is subject to static charge, electrostatic deposition of particles in sampling lines is most difficult to characterize (Willeke and Baron 1993). This problem is largely avoided through the exclusive use of metal or electrically conductive lines. Having no electrically isolated section will obviate the problem of electrostatic deposition. If metal lines cannot be used, TygonTM is an acceptable substitution. Materials to be avoided for aerosol transport are TeflonTM and PolyfloTM (Willeke and Baron 1993). Losses of large particles can occur due to sedimentation, gravitational settling, and inertial impaction. Generally speaking, such losses only become significant for particles larger than 1 μm , and consequently are usually negligible.

2.3 Particle Charging Mechanisms

Particle charging is the first basic step in the aerosol measurement based on electrical mobility technique. The aim of the charging mechanism for a mobility analyzer is to impose a known charge distribution on the aerosol particles. Because particle size distribution is commonly determined through the electrical mobility classification, prediction of particle size requires the knowledge of the charge distribution for every particle size. There are many mechanisms by which aerosol particles acquire net charge distributions. These are flame charging, static electrification, diffusion charging and field charging. The most commonly used method for charging particles in electrical measurement instruments is diffusion charging. Generally speaking, particles are allowed to collide with ions and the charge carried by these ions is transferred to the particles. This method is so called due to the mechanism

ions travel in the gas and collide with the particles. Diffusion charging of particles can be that unipolar or bipolar depending on the polarity of the ions colliding to the particles. Bipolar diffusion charging leads to a charge equilibrium which has low charging efficiencies, e.g. 3.3 % for positively charged particles and 5.7 % for negatively charged particles (Wiedensohler 1988). Unipolar diffusion charging has advantages over bipolar diffusion charging as it does not reach an equilibrium charge distribution, therefore potentially enabling the attainment of a higher charging efficiency. There are two conventional ways to produce ions for unipolar diffusion charging in a gas; by corona discharge and by ionizing radiation from α -ray or β -ray sources such as ^{85}Kr or ^{241}Am .

Corona discharges are among the most common technique to produce high ion concentrations and there have been numerous extensive studies in the past. The phenomenon is used in many industrial applications such as electrostatic coating and precipitation (Lawless and Sparks 1988). Electrostatic charging by the corona dischargers is also commonly used in determining aerosol size distribution by electrical mobility analysis. Corona discharge is produced by a nonuniform electrostatic field such as that between a needle and plate or a concentric wire and a tube. Air and other gases can undergo electrical breakdown when the electric field strength is high. For the case of the wire and the tube the only place this breakdown can occur is in a very thin layer at the wire surface. In this corona region, electrons have sufficient energy to knock an electron from gas molecules creating positive ions and free electrons. During this process, aerosol flow is directed across the corona discharge field and is charged by random collisions between the ions and particles due to Brownian motion of ions in space. The amount of ion deposition on the particle surface depends on resident time, particle radius and shape, electric field, etc. The technique has been applied successfully and several designs of wire-cylinder corona charger are employed and described in the published literature, both wire-cylinder corona (Hewitt 1957; Liu *et al.* 1967; Liu and Pui 1974; Buscher *et al.* 1994; Unger *et al.* 2004; Biskos *et al.* 2005) and needle chargers (Whitby 1961; Medved *et al.* 2000; Marquard *et al.* 2003 Hernandez-Sierra *et al.* 2003). A number of particle sizing instruments employ unipolar corona chargers (Liu and Pui 1975; Keskinen *et al.* 1992; Kulon *et al.* 2001; Tammiet *et al.* 2002; Reavell *et al.* 2002; Biskos 2004) as important upstream component to impart known charge to the aerosol system. The charger performance of these instruments depends on the stable operation of their chargers. Aerosol charging is a function of the ion concentration, N_i , and the mean residence time of the particles to the ions, t . For this reason, a well-designed corona charger should provide a stable $N_i t$ product that can be accurately determined for any given operating conditions.

2.3.1 Unipolar Diffusion Charging

When an aerosol particle is exposed to gaseous ions, capture of ions by the particle occurs leading to the appearance of an electrical charge on the particle. The magnitude of the charge depends upon the size of the particle, the unipolar ion density encountered, and the time that particle spends within this region. In the absence of any appreciable electric field, this particle will be diffusively charged by the Brownian random motion of the ions with respect to the particle. This diffusion charging, first characterized by White (1963) and more recently modified by Pui (1976), can be expressed in a convenient analytic form. For an initially neutral particle immersed in a unipolar ion cloud, the flux of ions, J , impinging on the particle surface area is given by

$$J = 4\pi a^2 \left(\bar{c}_i \frac{N_i}{4} \right) \quad (2.38)$$

where a is the particle radius, N_s is the concentration of ions above the surface and \bar{c}_i is the mean thermal speed of the ions. The spatial distribution of ions is given by the classical Boltzmann distribution for the equilibrium state. Neglecting the image force attraction between the ions and the particle, the Boltzmann distribution at the particle surface is given by

$$N_s = N_i \exp\left(-K_E \frac{n_p e^2}{akT}\right) \quad (2.39)$$

where N_i is the ion concentration at infinity, n_p is the particle charge, e is the elementary unit of charge, $K_E = 1 / 4\pi\epsilon_0$ with the vacuum permittivity, k is the Boltzmann's constant (1.380658×10^{-23} J/K), and T is the operating temperature of the system. Substituting Equation 2.39 into Equation 2.38 gives

$$J = \pi a^2 \bar{c}_i N_i \exp\left(-K_E \frac{n_p e^2}{akT}\right) \quad (2.40)$$

The above equation was originally derived by White (1963). This is valid for the kinetic theory (Free molecular regime, $Kn \gg 1$). The charging rate expression can be described by the system of differential equation as

$$\frac{dn_p}{dt} = \pi a^2 \bar{c}_i N_i \exp\left(-K_E \frac{n_p e^2}{akT}\right) \quad (2.41)$$

With the initial condition that $n_p = 0$ at $t = 0$ for the charging of an aerosol (initially neutral), the average charge of particle can be integrated analytically to give

$$\int_0^{n_p} \frac{dn_p}{\exp\left(-K_E \frac{n_p e^2}{akT}\right)} = \int_0^t \pi a^2 \bar{c}_i N_i dt \quad (2.42)$$

Thus, the average charge, n_{diff} , caused by the diffusion charging in a time period, t , by a particle diameter d_p , can be found from

$$n_{\text{diff}} = \frac{d_p kT}{2K_E e^2} \ln\left(1 + \frac{\pi K_E d_p \bar{c}_i e^2 N_i t}{2kT}\right) \quad (2.43)$$

where d_p is the particle diameter.

2.3.2 Unipolar Field Charging

In the case where an electric field exist in the charger, the effect of the finite electric field used in the charging region can be estimated by the classical field charging

equation derived by White (1963), the saturation charge, n_s , of a particle (diameter, d_p and dielectric constant, ε) in an electric field E is given by

$$n_s = \left(1 + 2 \frac{\varepsilon - 1}{\varepsilon + 2}\right) \left(\frac{Ed_p^2}{4K_E e}\right) \quad (2.44)$$

The charging rate expression, dn_p/dt , is given by the following differential equation

$$\frac{dn_p}{dt} = n_s K_E e Z_i N_i \left(1 - \frac{n_p}{n_s}\right)^2 \quad (2.45)$$

If the particle is initially neutral, the average number of charge, n_{field} , acquired in an average electric field E is given by

$$n_{\text{field}} = \left(1 + 2 \frac{\varepsilon - 1}{\varepsilon + 2}\right) \left(\frac{Ed_p^2}{4K_E e}\right) \left(\frac{\pi K_E e Z_i N_i t}{1 + \pi K_E e Z_i N_i t}\right) \quad (2.46)$$

where ε is the particle dielectric constant. In the present study, $\varepsilon = 3.0$ is arbitrarily assumed for the dielectric constant of the particle.

2.4 Flow Field Modeling

For flow field modeling, flow conditions inside the charger and classifier column of the EMS are assumed to be steady, incompressible and laminar. Based on the principle of momentum conservation, the incompressible Navier-Stokes equations (N-S equation) can be applied in this case. In these axisymmetric geometries, the continuity and N-S equations used in this model can be written in the 2-D cylindrical coordinates is given as follows:

Continuity equation:

$$\frac{1}{r} \frac{\partial}{\partial r}(ru_r) + \frac{\partial}{\partial z}(u_z) = 0 \quad (2.47)$$

N-S equation:

For the radial component (in r -direction),

$$u_r \frac{\partial u_r}{\partial r} + u_z \frac{\partial u_r}{\partial z} - \frac{u_\theta^2}{r} = -\frac{1}{\rho} \frac{\partial p}{\partial r} + \mu \left(\frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial}{\partial r}(ru_r) \right) + \frac{\partial^2 u_r}{\partial z^2} \right) - \mu \frac{1}{r^2} u_r \quad (2.48)$$

For the axial component (in z -direction),

$$u_r \frac{\partial u_r}{\partial r} + u_z \frac{\partial u_z}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial z} + \mu \left(\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_z}{\partial r} \right) + \frac{\partial^2 u_z}{\partial z^2} \right) \quad (2.49)$$

For the circumferential component (in θ -direction)

$$u_r \frac{\partial u_\theta}{\partial r} + u_z \frac{\partial u_\theta}{\partial z} = \mu \left(\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_\theta}{\partial r} \right) + \frac{\partial^2 u_\theta}{\partial z^2} \right) - \frac{2\mu}{r} \left(\frac{\partial u_\theta}{\partial r} \right) \quad (2.50)$$

where u_r is the velocity component in the r -direction, u_z is the velocity component in the z -direction, u_θ is the velocity component in the θ -direction, p is the pressure and μ is the kinematic viscosity of air.

2.5 Electric Field Modeling

For electrostatic field modeling, the Poisson's equation for the electric potential can be used. However, the space-charge effect on the electric field can be neglected ($\rho = 0$) for low aerosol concentration, typically less than 10^{12} particles/m³, and with low particle charge level (Chen and Pui 1997; Camata *et al.* 2001). Thus, the Laplace's equation is used in the present formulation:

$$\nabla^2 V = 0 \quad (2.51)$$

where V is the applied voltage. To further simplify the equation, axisymmetric assumption is used due to the charger and classifier configurations. The resulting equation in the 2-D cylindrical coordinates is given as follows:

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial V}{\partial r} \right) + \frac{\partial^2 V}{\partial z^2} = 0 \quad (2.52)$$

Once the electric potential is obtained, the electric field strength in the r - and z - directions can be calculated by the following equations

$$E_r = -\frac{\partial V}{\partial r}, \quad E_z = -\frac{\partial V}{\partial z} \quad (2.53)$$

In the simplified geometry, the electric field strength are expressed by

$$E_r(r, z) = E_r(r) = -\frac{V}{r \ln(r_2/r_1)}, \quad (2.54)$$

$$E_z(r, z) = 0 \quad (2.55)$$