Turbulent Particle Deposition in a Rectangular Chamber: Study of the Effect of Particle Size and Ventilation Regimes

by

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April 1996

MASTER

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CLARKSON UNIVERSITY

**TURBULENT PARTICLE DEPOSITION IN A RECTANGULAR CHAMBER: THE
STUDY OF EFFECT OF PARTICLE SIZE AND VENTILATION REGIMES**

A Thesis

by

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Department of Chemistry

Submitted in Partial Fulfillment of the Requirements

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The undersigned have examined the thesis entitled: **"Turbulent Particle Deposition in a Rectangular Chamber: the Study of Effect of Particle Size and Ventilation Regimes"**, presented by Mr. Yoshio Nomura, a candidate for the degree of Master of Science, and hereby that it is worth of acceptance.

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Abstract

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The interaction of aerosol particles with wall surfaces is important in modeling their behavior. This interaction is usually represented in theoretical models as a loss term. The loss rate is the rate at which particles deposit or react with the surfaces. This loss term is important in many branches of aerosol science including human health and indoor air quality. Increased surface deposition usually means lower concentrations of airborne particles and hence, lower exposure to the inhabitants. If the efficiency of the particle deposition is influenced by factors other than the particle size, such as a natural convection of the air, this has to be taken into account to evaluate the results.

In this research, test aerosol sized from 15 nm to 3 μm are produced by several different aerosol generators; the gas burner, the Collison nebulizer, the condensation aerosol generator, the orifice atomizer and the Vibrating Orifice Aerosol Generator (VOAG). Those generators are reasonably stable and able to produce high concentrations enough to measure the particle concentration decay in a chamber.

A rectangular chamber whose dimensions are $75 \times 75 \times 180 \text{ cm}^3$ was used in this study. The particles were injected into the chamber, with a known ventilation and the concentration decay was monitored by the Ultrafine Condensation Particle Counter (UCPC) and Optical Particle Counter (OPC). During the measurement, the air inside the chamber is moved by natural convection and ventilation effect. The ventilation rate (air exchange rate) used in this research is

0.8 h⁻¹, 1.7 h⁻¹ and 2.4 h⁻¹. Those values were measured by monitoring the radon gas concentration decay.

The results shows that the particle loss rate under the higher air exchange rate is larger and this is not only due to air exchange itself but also the wall deposition. The theoretical model presented by Benes and Holub (1996) agree with the experimental data better than the Crump and Seinfeld (1981) model with the hypothesis of Plandtl's mixing length.

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CHAPTER 1 Introduction

Fine aerosol particles can be defined as relatively stable suspensions in a gaseous medium of liquid droplets or solid particles with a gravitational settling velocity less than the settling velocity of aerosol particles. Such aerosols are encountered in many scientific phenomena and there is great interest in fine particles not only because of their use in industry, their deposition in the respiratory system during inhalation and exhalation, but also because of their involvement in the nature and perturbed atmosphere.

1.1. Health Effects of Fine Particles

Aerosols, especially those in the indoor environment, may become a critical issue for people in the view of the potential health effects of indoor airborne particles. If aerosol particles deposit inside of a human body, they may cause disease (Lieberman, 1969; Brian, 1977; Thor, 1977). The inhaled particles deposit in different parts of the respiratory system according to their size. The particles can be cleared from the respiratory tract by mucous clearance. Soluble particles are absorbed directly from the respiratory system or the digestive system and accumulate in organs according to their biological affinity. As a result, they can damage the organ's function because of their chemical toxicity or radioactivity. Insoluble or partially soluble particles deposit in the respiratory system. This deposition can lower a respiratory function and lead to chronic lung disease.

The respiratory system can be divided into three parts which are the head region (nose, mouth, pharynx, and larynx), the tracheobronchial region, and the alveolar region. Because each part of the respiratory system has a different function and clearance mechanisms in order to explain the

aerosol effect on the human body, it is important to know how particles deposit and in which part of respiratory system particle deposit.

Particles deposit in the various regions of the respiratory tract by several physical mechanisms. Deposition mechanisms depend on the aerodynamic properties of the particles, the anatomy of the airways, and the airflow through them. Lippmann (Lippmann *et al.*, 1980; Lippmann, 1983) describes a number of deposition mechanisms. These are impaction, sedimentation, diffusion, interception, and electrostatic interception. However, the predominant mechanisms are impaction, sedimentation, and diffusion. Interception and electrostatic interception are not significant. In larger airways, impaction is more significant. Inhaled air follows a tortuous path through the nose or mouth and the branching airways in the lung. Every time the air change directions, the inertia of particles tends to keep them moving in their prior directions, which can carry them onto airway surfaces. Impaction probability depends on the stopping distance.

The location of the particle within the airway must also be considered. In a bifurcated airway, the deposition probability during the inhalation is higher for a particle traveling along the center line of the parent tube than for particles nearer to the walls, and the highest density of deposition is at or near the carina of the bifurcation. Computational fluid dynamics has been used to estimate deposition.

Gravitational sedimentation is an important mechanism for deposition in the smaller bronchi, the bronchiolus, and the alveolar spaces where the airways are small and the air velocity is low. Sedimentation has the greatest effect when the airway is horizontal. Sedimentation becomes less effective than diffusion when the terminal settling velocity of the particles falls below 0.001 cm s^{-1} , which corresponds to a unit density sphere of about $0.5 \text{ }\mu\text{m}$ diameter.

When the particle size is smaller, Brownian motion increases and becomes a more effective mechanism of particle deposition in the lung as the root-mean-square displacement approaches the size of the air sacs in the alveoli. Diffusive deposition is important in small airways, alveoli and at airway bifurcations for particle diameters below about $0.5 \text{ }\mu\text{m}$. For radon and its decay products

attached to particles that are nearly molecular in size, the diffusive deposition efficiency will be high in the nasopharynx passage and in large airways such as the trachea and early generations of the bronchia.

The total deposition inside the respiratory system is usually measured experimentally. The experiments are done by measurement of monodisperse aerosols during inspiration and expiration.

In many cases, the size or chemical composition of deposited particles has changed from original composition (Martonen *et al*, 1982; Anselm *et al*, 1986) because the size of the particle increases due to the absorption of water vapor onto the particle especially when the particle contains water soluble compounds. This phenomena is called hygroscopic effects. Ferron (1977) presented the mathematical equations for calculating particle growth during respiration.

Deposition in the head region or tracheobronchial regions prevents particles' alveolar deposition. In case of inspiration through a mouth (30 Lpm), 20% of particles whose aerodynamic diameter is 5 μm and 70% of particles whose aerodynamic diameter 10 μm deposit before the larynx. From the nose, 70% of 5 μm and 100% of 10 μm particles deposit before the larynx. However, smaller particles such as submicron particles are able to go deep enough for alveolar deposition.

Therefore particles sized in the submicron range are the major hazard to public health.

Besides the possible impact a human health, the damages by the accumulation of particles on cultural artifacts and electronic equipment have been reported. Nazaroff (1990) observed that airborne particles can deposit on cultural artifacts causing soiling and material damage. The fine particles can also deposit on electronic equipment resulting in equipment failures (Weschler, 1991). If the deposition velocities of particles in several size ranges are known, one can calculate the rate at which particles accumulate on surfaces. Such information has been used to develop predictions of the benefits and costs derived from improving air quality in many offices. Also this information can be used to estimate the potential risks of artifacts and electronic equipment under different ventilation rates or air conditioning.

1.2. Indoor Radon

It has been over one hundred years since radon was recognized as a hazardous material by Harting and Hesse (1879). They studied about toxicity from the high rate of lung cancer in miners. Through substantial research efforts (Aurand *et al*, 1955; Bale and Shapiro, 1956), it became obvious that the source of lung cancer was not radon but its short lived radon decay products. Exposure to alpha particle radiation from the short lived daughters of ^{222}Rn (^{218}Po half-life=3.04min, ^{214}Pb half-life=26.8min, ^{214}Bi half-life=19.8min, ^{214}Po half-life=162 μsec) can cause bronchogenic cancer or lung cancer. Because radon is a gas at room temperature and has comparatively long half-life (3.85 days), it is able to enter indoors from soil, building materials or from ground water. Since the late 70's high levels of radon and its decay products have been detected in ordinary residences in the U.S., Canada and Sweden. Although ^{222}Rn itself is inert, when the radon decay products are formed, they tend to attach to airborne particles immediately and are then transported by the particles. When the radon decay products attach to submicron particles, these radioactive materials can be carried into the tracheobronchial and pulmonary region of the respiratory system during inhalation process. Because the submicron particles are too small for immediate sedimentation but small enough for high diffusivity, these particles are very difficult to be removed from the air. With the presence of trace of volatile organic compounds (VOC) or semi volatile compounds (SVOC), radon decay products change in their physical and chemical state (Wilson, 1987; Berglund *et al*, 1986) and this can change their aerodynamic behavior (Castleman, 1991). It should be borne in mind that not only uranium miners and other underground miners but also normal houses face this problem because of indoor radon and its decay products and according to Bricard *et al* (1968) even in thoroughly filtered air, aerosols will spontaneously form from trace gases under the influence of radiant energy. Mechanisms for removal of submicron airborne particles will help to reduce the radiation dose from submicron particles including radioactive materials.

1.3. Indoor Ozone and Nitrogen Oxides

The concentration of indoor ozone is normally lower than those of outdoors. However, the large amount of time spent indoors will increase indoor ozone exposures (concentration $\times$ time). Sometimes the exposure is greater than outdoors. The contribution of indoor exposure to total exposure is especially important when one considers possible chronic health effects resulting from long term ozone exposure (Absil, 1992).

In most cases, the indoor ozone has infiltrated from outdoors. In some cases, it is generated indoors by sources such as electrostatic air cleaners and photocopying machines (Yocum, 1982, Allen, 1978). In the absence of indoor emission source, the concentration of indoor ozone can be estimated from the outdoor ozone concentration, the air exchange rate, and the rate of removal on indoor surfaces. The last factor is the effect of deposition on the concentration change in a room. The contribution of other factors can be measured or calculated easily and knowledge of the deposition velocity allows one to estimate indoor ozone levels.

Because NO₂ is a criterion pollutant, the presence of nitrogen oxides in indoor air should be paid attention. Moreover, indoor concentrations are sometimes much higher than those outside due to the emissions from combustion such as gas stoves, gas ovens and kerosene-fired space heaters. Excluding the effect of ventilation, the nitrogen oxide loss rate is observed to be a first order process. The surface loss rate of nitrogen oxide is significantly affected by humidity (Yamanaka, 1984). In chamber studies of the uptake of NO₂ of a variety of indoor materials, Miyazaki (1984) found moderate increase in deposition velocity with increasing humidity. Spicer *et al* (1989) reported no effect of increased humidity on NO₂ losses to walls and decreasing loss rates to cement blocks and increasing loss rates to wool carpets and furnace filters. This resulted mainly from the increase of the surface area available for deposition.

1.4. Deposition Study

When gaseous molecules in motion hit a wall, the molecules experience a reaction force and

rebound as shown in Figure 1.1. This force is called the gas pressure. However, in case of aerosol particles, when they hit a wall, they can adhere to the surface of the wall. This phenomenon is called deposition. Because of the particle deposition, the concentrations very close to the wall surface are zero. According to Fick's first law of diffusion, the particles in the higher concentration area move to lower concentration area by diffusion. By continuous diffusive

Figure 1.1 Particle Deposition and Gas Molecule Bounce off

deposition, the particle concentration of the aerosol keep decreasing. Diffusive deposition is predominant for submicron particles in the air. Diffusion is given by

$$\frac{\partial c}{\partial t} = D \nabla^2 c - \text{div} (BcF) \quad (1.1)$$

where c is the concentration of particles, F is the external force on particles, B is the mobility, t is the time, and D is the diffusion coefficient. When an external force, F , does not exist, the square-

mean particle displacement during the time t is $2Dt$. D and B are related by Einstein's relationship.

When the air has a flow or experiences turbulence (non-steady state), the deposition mechanism becomes more complicated. The general equations were given by Levich (1962). In a turbulent flow system, a very thin laminar sublayer is formed in the area which is very close to the walls. In this layer, the behavior of airflow is very similar to the one of a laminar boundary layer. Fuchs (1964) presented models of deposition to vertical walls in turbulent flow.

Friedlander and Johnston (1957) took the inertia of particles into consideration. In this model, turbulent transport carries particles to outside the laminar boundary layer, and then particles maintain their direction and velocity towards the walls. If the stopping distance of a particle is larger than the thickness of the layer, the particles will deposit onto the walls. This kind of model is called the free flight model. From this model, they calculated the deposition rate with the assumption that diffusion coefficient of a particle is the same as the one of a fluid.

Davis (1966) used the sum of the diffusion coefficient of a fluid and the Brownian motion for particle diffusion coefficient. Because Davis derived a smaller value by calculation than the experimental value, Beal (1970) improved the idea of particle velocity near the walls. Yoshioka (1972) applied Friedlander's method to a diffusion tube. Sehmel (1970) and Liu (1974) adopted Owen's equation (1960) for a fluid diffusion coefficient. Although Hutchinson (1971) and Cleaver (1975) estimated the effect of turbulent flow to the laminar sublayer, the free flight model is not complete. In this model, diffusion by Brownian motion is ignored in the turbulent flow. Moreover, particle behavior near the walls or in the laminar boundary layer is not fully understood.

The first research which was concerned with turbulent deposition and gravitational sedimentation in a vessel was performed by Langstroth and Gillespie (1947). They observed the change of the number and the size of ammonium chloride particles in a chamber. They found that the mixing rate was closely related to the particle loss. Corner and Pendlebury (1951) derived a

formula for the aerosol deposition rate in a rectangular box. In the same manner, the aerosol deposition rate in a tube also can be derived. Bigu (1985) used radon decay products as aerosol particles. By the α -counting method, he analyzed the concentration of radon decay products and determined deposition coefficients. Van de Vate (1972) used monodisperse polystyrene latex particles with diameters ranging from 0.09 to 1.3 μm and obtained a diffusion boundary layer thickness 0.085 cm. This result agreed reasonably with equation (1.2) which many investigator suggested

$$\beta = \frac{v_{ts}}{h} + \frac{S D}{V \delta_d} \quad (1.2)$$

where v_{ts} is particle terminal settling velocity, h is vessel height, S is vessel surface area, D is the particle diffusion coefficient, V is the vessel volume, and δ_d is a diffusion boundary layer thickness. Harrison (1979) also used polystyrene latex to estimate the boundary layer thickness. He showed that the gravitational settling and convective diffusion losses of spherical latex aerosol particles to the inside walls of the aerosol container are dependent on the nature of the surface.

Crump and Seinfeld (1981) derived an equation for arbitrary shape vessel. They assumed that turbulent diffusivity near the wall is proportional to 2nd power of the distance from the wall surface. This theoretical result agreed with experimental data (Crump *et al*, 1983). Okuyama (1986) used monodisperse NaCl, DEHS (diethylhexyl sebacate), and polystyrene latex particles. In this research, the deposition rate showed clear dependance on both the particle size and turbulence intensity. The results also agreed with the theory of Crump and Seinfeld. Byrne *et al* (1995) used neutron-activation analysis (NAA) to detect rare elements. Dysprosium and indium were used as aerosol labels in his study. This sensitive tracer aerosol analysis technique yielded good agreement with Corner and Pendlebury's predictions. This study also showed the very high sensitivity of neutron activation analysis for quantitative aerosol particle detection. By analysis of

a small number of air and surface samples, it is possible to predict the aerosol mass deposited on a body, especially the part which cannot be destructively analyzed. Eklund (1995) created turbulence by changing the ventilation rate in a rectangular chamber and observed that the dependence of particle losses by surface deposition on ventilation rate was weak. His model did not agree with the theory of Crump and Seinfeld. Previous studies on particle loss rate in a vessel are summarized in Table 1.1. The detail theories are described in Chapter 2 and Chapter 3.

The purpose of this research is to investigate the deposition of particles as a function of size. In order to do so, it was necessary to construct aerosol generators using several methods such as evaporation and condensation or vibrating orifice aerosol generators for different size ranges. These aerosol generator systems are described in Chapter 4. The deposition experiments and the conclusions are given in Chapter 5 and 6, respectively.

Table 1.1 Summary of Previous Studies of Particle Loss in a Vessel

Particle Used	Chamber Shape and Volume (m ³)	Analysis Method	Conclusion
Langstroth and Gillespie (1947)	Ammonium Chloride Rectangular, 1.12 (m ³)	Thermal Precipitation, Microscopy	Observed particle loss depends on mixing rate
Van de Vate (1972)	Polystyrene Latex Spherical, 1 (m ³)	Electrostatic Precipitation, Electron Microscopy	Showed porosity of polystyrene latex from deposition study
Harrison (1979)	Polystyrene Latex Cubic, 0.195 (m ³)	Light Scattering	Showed relationship between boundary layer thickness and the particle size
Crump <i>et al</i> (1983)	Sodium Chloride, Polystyrene Latex Spherical, 0.118 (m ³)	Electrical Aerosol Analysis, Light Scattering	Presented the validity of theory for arbitrary shape vessels
Okuyama <i>et al</i> (1986)	Sodium Chloride, Diethylhexyl Sebacate Spherical, 0.0026 (m ³)	Light Scattering	Suggested the eddy diffusivity is proportional to 2.7th power of the distance from the wall
Holub <i>et al</i> (1988)	Radon Decay Products Spherical 0.23 (m ³), Cylindrical 4 (m ³)	Condensation Nucleus Counting	
Byrne <i>et al</i> (1995)	Silica Labeled with Dysprosium Cubic, 8 (m ³)	Neutron Activation Analysis	Showed neutron activation analysis on deposition study

CHAPTER 2 Particle Loss in Steady State

2.1. Basic Aerosol Behavior

2.1.1. Aerodynamic Drag and Stokes' Law

In order to understand the aerodynamic properties, the Reynolds number is a very useful parameter. The Reynolds number is the ratio of inertial to frictional force on the particles and is given by $\rho_g v_p d_p / \eta$, where ρ_g is the density of the fluid, v_p is the relative velocity between the fluid and an object such as a particle, d_p the particle diameter, and η is the viscosity of the fluid. When the Reynolds number of an aerosol particle moving in a gas is greater than 3 the viscous forces on the particle are outweighed by the inertia effect. Above 3, Newtonian drag comes into effect.

An externally applied force on an aerosol particle is opposed and rapidly balanced by the force induced by its aerodynamic drag force. A particle's drag force, F_{drag} , is determined by the relative motion between the particle and the surrounding gas. Under the condition that the particle size is much larger than the distance among the gas molecules because the surrounding gas can be considered as a continuous fluid, the drag force is given by

$$F_{\text{drag}} = \frac{\pi}{8} C_d \rho_g v_p^2 d_p^2 \quad (2.1)$$

where C_d is the drag coefficient. It should be borne in mind that the aerodynamic drag is related to the gas density not the particle density. The drag coefficient, C_d , relates the drag force to the velocity pressure and expressed in terms of gas flow parameters as

$$C_d = \frac{24}{\text{Re}} \quad \text{Re} < 0.1 \quad (2.2)$$

where Re is the particle Reynolds number. From equation (2.2) and the definition of Reynolds number, the equation of the drag force can be derived as

$$F_{\text{drag}} = 3 \pi \eta v_p d_p \quad (2.3)$$

This equation is known as the Stokes' law. According to this equation, the drag force of spherical particles depends on the gas viscosity, η , and the relative particle velocity, v_p , and the particle diameter, d_p . When the shape of particles are not spherical and a dynamic shape factor, χ , is known, this parameter can be applied to predict the exact value of their drag force. χ 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.

2.1.2. Slip Correction Factor

The assumptions of Stokes' law are

1. Surrounding air is incompressible. (The velocity of the particle is much smaller than the speed of sound.)
2. Particles are rigid spheres.
3. There is no slip condition at particle surface.

However, when a particle is small enough to be the same order of size as the mean free path of ambient air, the relative velocity of the gas at the particle surface is not zero and the third assumption cannot hold. Under standard conditions, this error can not be ignored for particles less than $1 \mu\text{m}$ in diameter. In order to correct this problem, Cunningham (1910) derived a correction factor for Stokes' law. This factor is called slip correction factor or Cunningham correction factor and is always greater than one. This original factor, which is applicable for particles more than $0.1 \mu\text{m}$ in diameter, is

$$C_c = 1 + 2.52 \frac{\lambda}{d_p} \quad (2.4)$$

where λ is the mean free path of the gas (0.646×10^{-5} cm for air at 20°C). Experimental results (Davis, 1945) have developed an equation that extends the range of the particle diameter from 0.002 to 1.00 μm by the equation below.

$$C_c = 1 + \frac{\lambda}{d_p} \left[2.514 + 0.800 \exp \left(-0.55 \frac{d_p}{\lambda} \right) \right] \quad (2.5)$$

For the particles less than 0.002 μm , Ramamurthi *et al* (1989) developed the above equation by substituting

$$d_p^* = d_p \left(1 + 3 e^{-2.20 \times 10^{-7} d_p} \right) \quad (2.6)$$

for d_p in the equation (2.5). Note that d_p in equation (2.6) is in centimeters. Rader (1990) reported that equation (2.5) is very accurate both in molecular regime ($2\lambda/d_p > 20$) and transition regime.

2.1.3. Brownian Motion

If the particles are sufficiently small and suspended in a gas, they will experience an agitation from the randomly and swiftly moving gas molecules bombarding the particle surface. This will create a thermal motion analogous to that of the surrounding gas molecules. Even though there is a net displacement with time of any given particle, this motion appears almost as a vibration of the ensemble of particles. The agitation and migration of small colloidal particles has been known since the work of Robert Brown (1828), and the phenomenon thus has been called Brownian motion or Brownian movement. Bodaszewski (1883) linked the Brownian motion of smoke particles to the movement of the gas molecules as postulated by the kinetic theory. By the work

of Einstein (1905), Svedberg (1909), Sedding (1908) and others in the early 1900s, the applicability of Fick's law for the diffusion of the particles in a fluid has been widely accepted. Hidy and Brock (1970) reviewed Brownian diffusion in detail.

2.1.4. Molecular Diffusion in Steady State

Aerosol particles move from higher concentration regions to lower concentration regions along the concentration gradients. This phenomena is explained by Fick's law of diffusion. Fick's first law of diffusion states that the concentration of particles crossing a unit area in a unit time which is called the flux, J , is proportional to the concentration gradient normal to the unit area dc/dx . The constant of proportionality, D , is known as the diffusion coefficient. Symbolically, for the current through a plane at right angles to the x direction,

$$J = -D \frac{dc}{dx} \quad (2.7)$$

In general, the diffusion coefficient is a variable depending on the particle size, ambient temperature, and concentration. However, its concentration dependance can often be neglected. If the properties of the fluid are the same in all directions, it is said to be isotropic. This is the usual case, and D then has the same value for diffusion in any direction.

Fick's second law represents the time dependent case in which the change in concentration of an aerosol, with respect to time at a point in space, is proportional to the divergence of the concentration gradient at that point, the constant of proportionality again being D , the diffusion coefficient (Jost, 1952).

$$\frac{\partial c}{\partial t} = -D \nabla^2 c \quad (2.8)$$

The term ∇^2 is the Laplacian operator, which in Cartesian coordinates is

$$\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \quad (2.9)$$

in spherical coordinates ($x=r\sin\theta\cdot\cos\phi$, $y=r\sin\theta\cdot\sin\phi$, $z=r\cos\theta$)

$$\nabla^2 = \frac{\partial^2}{\partial r^2} + \frac{2}{r} \frac{\partial}{\partial r} + \frac{1}{r^2 \sin^2 \theta} \frac{\partial^2}{\partial \phi^2} + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} + \frac{1}{r^2} \cot \theta \frac{\partial}{\partial \theta} \quad (2.10)$$

and in cylindrical coordinates ($x=r\cos\theta$, $y=r\sin\theta$, $z=z$)

$$\nabla^2 = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial z^2} \quad (2.11)$$

Thus in Cartesian coordinates, Fick's second law of diffusion is written

$$\frac{\partial c}{\partial t} = D \left[\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} + \frac{\partial^2 c}{\partial z^2} \right] \quad (2.12)$$

The particle diffusion by Brownian motion is analogous to the molecular diffusion of gas molecules and its diffusion coefficient is given by

$$D = k T B = \frac{kT}{3 \pi \eta d_p} C_c \quad (2.13)$$

where k is Boltzmann's constant, T is the absolute temperature, B is the mechanical mobility, η is the viscosity, d_p is the diameter of particle and C_c is the Cunningham correction defined by equation (2.5).

The mobility is defined as

$$B = \frac{v_p}{F} \quad (2.14)$$

where F is the external force on the particle.

2.2. Particle Loss Mechanisms from Stagnant Air

There are mainly two mechanisms which remove aerosol particles from stagnant air. One is deposition and the other is sedimentation. In Figure 2.1, the diffusion coefficient and the terminal settling velocity are shown as functions of the particle diameter. A diffusion coefficient is very large if the particles are less than 100 nm in diameter. This indicates that the diffusive deposition is significant for smaller particles. A diffusion coefficient increases with temperature as would be expected from the similar relationship for gas. For large particles, a diffusion coefficient is inversely proportional to particle size. For small particles, with large slip correction factors, diffusion coefficient is proportional to d^{-2} (Hinds, 1982). On the other hand a sedimentation becomes more significant when particles are larger. The aerosol particles which are moving by Brownian motion have possibility to collide one another and this result in coagulation. The coagulated particles are larger than the original particles and then, they are tend to undergo the gravity effect which is called a terminal settling. In this section, several processes of particle losses and transport properties are introduced and analyzed.

2.2.1. Diffusive Deposition

In the stagnant air, if aerosol particles are transported by diffusion as expressed by Fick's law and deposit on a wall, this phenomenon is called diffusive deposition. The deposition velocity is defined as follows, when particles move in the y direction and the deposition surface is x - z plane,

$$v_d = \frac{\phi_{y=y_1}}{n(x, y_1, z, t)} = \frac{\text{particle number deposited/cm}^2 \text{sec}}{\text{particle number/cm}^3} = \text{cm/sec} \quad (2.15)$$

where v_d is the deposition velocity. $\phi_{y=y_1}$ is the deposition flux. ϕ has the unit of mass per area per time or particle number per area per time, of the particles at the point of y_1 . n is the concentration at the point (x, y_1, z, t) . For three dimensional particle deposition which includes the particle velocity of x , y , and z component, the deposition flux can be expressed with the sum of each component. If the deposition flux is measured in units of mass per volume and the concentration is measured in units of mass per volume, then the ratio, v_d has units of length per time which is the unit of the velocity. However, it should be noted that the deposition velocity does not represent any “velocity” of the particles or the flow but instead represents an efficiency of deposition.

When the concentration of aerosol particles decreases by deposition in a given finite region, the rate of the average concentration change with time in the finite area is expressed as follows.

$$\frac{d\bar{n}}{dt} = -\beta\bar{n} \quad (2.16)$$

where β is deposition coefficient which has a unit of mass or particle number per time. Equation (2.16) indicates that the particle deposition progresses as a first order process. In a steady state, the deposition coefficient is v_d/h , in a rectangular chamber with height h . According to Fick's law of diffusion, deposition flux at the surface of deposition wall is

$$\phi_{y=0} = D \left(\frac{\partial n}{\partial y} \right)_{y=0} \quad (2.17)$$

2.2.2. Error Function

In many cases, the integrals of diffusion equation use the error function as part of the solution.

Figure 2.1 (a) Particle Diffusion Coefficient and Particle Diameter, (b) Particle Terminal Settling Velocity and Particle Diameter (a) is calculated with equation (2.13) and (b) is from

$$V_{ts} = \rho d^2 g / 18 \eta. \quad (20^\circ\text{C}, \text{ in air at standard pressure})$$

The error function $\text{erf}(q)$ can be written as

$$\text{erf}(q) = \frac{2}{\sqrt{\pi}} \int_0^q e^{-t^2} dt \quad (2.18)$$

The first two derivatives of the error function are

$$\frac{d}{dq} \text{erf}(q) = \frac{2}{\sqrt{\pi}} \exp(-q^2) \quad (2.19)$$

$$\frac{d^2}{dq^2} \text{erf}(q) = -\frac{4}{\sqrt{\pi}} q \exp(-q^2) \quad (2.20)$$

2.2.3. Deposition on a Vertical Wall

In the case of deposition on a vertical wall of the infinite area, the effect of gravity can be ignored. The solution of the diffusion equation gives the change in particle concentration. The direction of the y axis is perpendicular to the deposition surface of the vertical wall.

$$\frac{\partial n(y, t)}{\partial t} = D \frac{\partial^2 n(y, t)}{\partial^2 y} ; y > 0 \quad (2.21)$$

With the boundary conditions of

$$n_{t=0} = n_0 \quad (\text{Condition 1})$$

$$n_{y=0} = 0 \quad (\text{Condition 2})$$

the solution is given as follows.

$$n(y, t) = n_0 \operatorname{erf} \left(\frac{y}{2\sqrt{Dt}} \right) \quad (2.22)$$

The deposition flux and deposition velocity are given, respectively, as

$$\phi = D \left(\frac{\partial n}{\partial y} \right)_{y=0} = n_0 \sqrt{\frac{D}{\pi t}} \quad (2.23)$$

$$v_d = \sqrt{\frac{D}{\pi t}} \quad (2.24)$$

2.2.4. Gravitational Deposition and Diffusive Deposition on a Horizontal Wall

When the direction of the y axis is taken perpendicular to the horizontal deposition wall, the diffusion equation becomes

$$\frac{\partial n(y, t)}{\partial t} = v_{ts} \frac{\partial n(y, t)}{\partial y} + D \frac{\partial^2 n(y, t)}{\partial y^2} ; y > 0 \quad (2.25)$$

where v_{ts} is terminal settling velocity. With the condition of

$$n_{t=0} = n_0 \quad (\text{Condition 1})$$

$$n_{y=0} = 0 \quad (\text{Condition 2})$$

the solution is given (Takahashi, 1982)

$$n(y, t) = \frac{n_0}{2} \left[1 + \operatorname{erf} \left(\frac{y + v_{ts} t}{2\sqrt{Dt}} \right) - \exp \left(-\frac{v_{ts} y}{D} \right) \operatorname{erfc} \left(\frac{y - v_{ts} t}{2\sqrt{Dt}} \right) \right] \quad (2.26)$$

where $\operatorname{erfc}(y)$ is $1 - \operatorname{erf}(y)$. The deposition flux is

$$\phi = n_0 \left[\sqrt{\frac{D}{\pi t}} \exp\left(-\frac{v_{ts}^2 t}{4D}\right) + \frac{v_{ts}}{2} \left[1 + \operatorname{erf}\left(\frac{v_{ts} t}{2\sqrt{Dt}}\right) \right] \right] \quad (2.27)$$

when $t \ll 4D/v_{ts}^2$,

$$\phi = n_0 \left(\sqrt{\frac{D}{\pi t}} + \frac{v_{ts}}{2} \right) \quad (2.28)$$

In equation (2.28), the first and second term of the right hand side shows the effects of diffusion and gravitational sedimentation, respectively. Although the first term is equal to equation (2.14), the second term appears to be half the gravitational sedimentation flux, $n_0 v_{ts}$.

2.2.5. Deposition of Particles Between Two Parallel Vertical Walls

If the distance between two walls is h and y axis is perpendicular to the walls, then

$$\frac{\partial n(y,t)}{\partial t} = D \frac{\partial^2 n(y,t)}{\partial y^2}; \quad 0 < y < h \quad (2.29)$$

With the condition of

$$n_{t=0} = n_0 \quad (\text{Condition 1})$$

$$n_{y=0} = n_{y=h} = 0 \quad (\text{Condition 2})$$

the solution is (Kaneoka, 1973)

$$n(y,t) = \frac{4n_0}{\pi} \sum_{m=1}^{\infty} \frac{1}{(2m-1)} \exp\left[-\frac{(2m-1)^2 \pi^2 D t}{h^2}\right] \sin\left[\frac{(2m-1) \pi y}{h}\right] \quad (2.30)$$

The deposition flux for one wall is given as follows.

$$\phi = \frac{4 n_0 D}{h} \sum_{m=1}^{\infty} \exp \left[-\frac{(2m-1)^2 \pi^2 D t}{h^2} \right] \quad (2.31)$$

The average time that the particles need to deposit on either wall, *i. e.*, the average time that the particle can exist between the walls without deposition, is given by equation (2.32) bellow

$$\bar{t} = \int_0^{\infty} t \frac{2\phi}{n_0 h} dt = \frac{h^2}{12D} \quad (2.32)$$

If h is very small, it is possible to observe the average residence time and the diffusion coefficient can be obtained from equation (2.32).

2.2.6. Diffusive Deposition on the Inside a Spherical Vessel

If the radius of vessel is R and spherical coordinates are used, the diffusion equation is given by

$$\frac{\partial n(r,t)}{\partial t} = D \frac{1}{r} \frac{\partial^2 [r n(r,t)]}{\partial r^2} ; 0 < r < R \quad (2.33)$$

With the boundary conditions of

$$n_{t=0} = n_0 \quad (\text{Condition 1})$$

$$n_{r=R} = 0 \quad (\text{Condition 2})$$

the solution is (Takahashi, 1982)

$$n(r,t) = \frac{2 n_0 R}{\pi r} \sum_{m=1}^{\infty} \frac{(-1)^{m+1}}{m} \exp \left(-\frac{m^2 \pi^2 D t}{R^2} \right) \sin \left(\frac{m \pi r}{R} \right) \quad (2.34)$$

The average number concentration of particles in the vessel is

$$\bar{n}(t) = \frac{3}{4\pi R^3} \int_0^R 4\pi r^2 n(r,t) dr = \frac{6n^2}{\pi^2} \sum_{m=1}^{\infty} \frac{1}{m^2} \exp\left(-\frac{m^2 \pi^2 D t}{R^2}\right) \quad (2.35)$$

Thus, the deposition flux is

$$\phi = \frac{1}{4\pi R^2} \frac{4}{3} \pi R^3 \frac{d}{dt} (n_0 - \bar{n}) = \frac{2n_0 D}{R} \sum_{m=1}^{\infty} \exp\left(-\frac{m^2 \pi^2 D t}{R^2}\right) \quad (2.36)$$

From equation (2.35), the half-life ($n/n_0=1/2$), of the particle is

$$t_{1/2} \approx \frac{0.03055 R^2}{D} \quad (2.37)$$

The average residence time for the particles is

$$t_R = \int_0^{\infty} \frac{n(r,t)}{n_0} dt = \frac{R^2}{15D} \approx 2t_{1/2} \quad (2.38)$$

As can be seen, in many cases for stagnant aerosol gas, both the deposition flux and deposition velocity can be calculated using Fick's law of diffusion with boundary conditions.

2.2.7. Particle Coagulation in Steady State (Thermal Coagulation)

Another important mechanism to remove particles from air is coagulation, which is also known as aggregation or agglomeration. Coagulation of aerosol particle is a process where aerosol particle collide with each other due to the relative motion between them and adhere to form larger particles. This results in the continuous decrease in the number concentration and an increase in the particle size. This increase size and mass encourages particle sedimentation. However, in the absence of any loss or removal mechanisms there is no change in mass concentration. If the relative motion between particles is due to Brownian motion, the process is called thermal

coagulation which is easily observed. This phenomenon is significant at high number concentration of aerosol particles. Because during the coagulation process, both particle number concentration and particle sizes change with time, numerical models which predict these variables as a function of time are complex and have to be simplified to use them. The fundamental theory of particle coagulation was given by Smoluchowski (1917). He derived that dn/dt is proportional to $-n^2$ and the constant of proportionality is called coagulation constant, K . Several coagulation constants are tabulated in Table 2.1.

Table 2.1 Coagulation Constant, K , for Several Particle Diameter at 20°C

Particle Diameter (μm)	Coagulation Coefficients ($\text{cm}^3 \text{s}^{-1}$)
0.05	15×10^{-10}
0.1	8.6×10^{-10}
0.5	4.0×10^{-10}
1	3.5×10^{-10}
5	3.1×10^{-10}

2.3. The Other Transport Properties

2.3.1. Thermophoresis

Thermophoresis is a complex phenomena and a comprehensive mathematical theory is not yet available. An aerosol particle is continuously struck from all direction by gas molecules. In a temperature gradient, these molecules in the higher temperature side of the particle will have a higher momentum than those in the low temperature. Due to this difference, the momentum impacted from either side the net momentum delivered from the gas to the particle is not zero and

so the particle will obtain a momentum in the direction from high to low temperature. In order to formulate a precise mathematical description of the momentum transfer, precise knowledge of the velocity distribution of the gas molecules around the particle is necessary. This is the major complicating factor in developing a comprehensive theory.

Waldmann and Schmitt (1966) suggested approximate formula for calculating this net force for a particle with a diameter d

$$F_{th} = \frac{-p\lambda d^2 \nabla T}{T} \quad \text{for } d < \lambda \quad (2.39)$$

where p is the gas pressure, λ the gas mean free path, ∇T the temperature gradient, and T the absolute temperature of the particle. The velocity of thermophoresis is given as

$$V_{th} = \frac{-0.55\eta \nabla T}{\rho_g T} \quad \text{for } d < \lambda \quad (2.40)$$

Equation (2.40), above indicates that for the particles whose diameters are smaller than fluid molecule mean free path, the velocity resulting from thermophoresis is not dependent on the particle size. This velocity under standard condition per unit temperature gradient is

$$\frac{V_{th}}{\nabla T} = -2.8 \times 10^{-4} \quad \text{for } d < 0.1 \mu\text{m} \quad (2.41)$$

For larger particles, the particles are described as being in the continuum regime. Those particle are affected by several phenomenon. Because the size of the particle is large, gas molecules rebounding from the particle are very likely to hit the particle again. Thus, a velocity distribution derived on for isolated system is no longer applicable. Moreover, a temperature gradient can be

established in the particle. This internal temperature gradient affects the temperature of the surrounding gas. The net effect results in the total thermophoretic velocity. Brock (1962) suggested the equation for particles with $d > \lambda$.

$$V_{th} = \frac{-3 \eta C_c H \nabla T}{2 \rho_g T} \quad \text{for } d > \lambda \quad (2.42)$$

Where the coefficient H includes the effect of the internal temperature gradient of the particle. Because the particle moves through the temperature gradient, every time the particle changes its position, the surrounding air temperature changes. In this case the ratio of thermal conductivity have to be taken into consideration. The coefficient, H , adjust this effect and is defined by

$$H \approx \left[\frac{1}{1 + 6\lambda/d} \right] \left[\frac{\kappa_a/\kappa_p + 4.4\lambda/d}{1 + 2\kappa_a/\kappa_p + 8.8\lambda/d} \right] \quad (2.43)$$

where κ_a and κ_p are thermal conductivity of the air and the particle, respectively. The thermal conductivity of air is $5.6 \times 10^{-5} \text{ cal cm}^{-1} \text{ s}^{-1} \text{ K}^{-1}$, while that for particle ranges from 0.16 for a iron metal to $1.9 \times 10^{-5} \text{ cal cm}^{-1} \text{ s}^{-1} \text{ K}^{-1}$ for an insulator (asbestos) (Mercer, 1973). Thermophoresis is relatively independent of particle size over a wide range. Thermal precipitators have been used for collecting small samples for applications such as electron microscope observations. Although this phenomenon can be considered as a pollutant cleaner of indoor air, this is not a effective way in the normal rooms.

2.3.2. The Effect of Electric Charge

The minimum charge on an aerosol particle is zero. However, this condition is rarely achieved because of the random charging of aerosol particles by omnipresent air ions. There are approximately 10^3 ions per cubic centimeter and when neutral aerosol particles are hit by ions

which are moving in random thermal motion, these particles are charged, and then, these particles start attracting ions of opposite sign. This offsetting process results in the loss of charge from aerosol particles and leads to an equilibrium state called the Boltzmann equilibrium or residual charge distribution.

In the atmosphere, ions can be formed by the decay of radioactive materials. Most of the naturally occurring radioactive isotopes are part of the uranium or thorium decay chains. These are present in the soil or water near the surface of the earth. Because of that, the strength of the radiation depends on the location, time, and altitude. For example, one of the most important radioactive material, radon and its decay products are formed by $^{238}\text{U} \rightarrow ^{222}\text{Rn} \rightarrow ^{218}\text{Po} \rightarrow ^{214}\text{Po} \rightarrow ^{214}\text{Bi} \rightarrow ^{214}\text{Po}$.

When a charged particle is placed in an electric field, it experience the electric force. The velocity of motion resulting from the force can be determined in a manner similar to the terminal settling velocity. In order to understand the charged particle behavior, it is convenient to express the ability of a particle to move in an electric field in terms of electrical mobility, Z . This is the velocity of a particle with a given charge in an electric field of unit strength.

The electrical mobility, Z , is given by

$$Z = \frac{q C_e}{3 \pi \eta d_p} \quad (2.44)$$

where η is viscosity of the gas in poise, d_p is particle diameter in cm and q is the number of electrical charge of the particle in e.s.u.. The electrical mobility, Z , here has a unit of $\text{cm}^2\text{s}^{-1}\text{V}^{-1}$.

Particles can be charged in several ways. The basic processes which give rise to a charge on a particle are direct ionization, static electrification, collisions with ions or ion clusters (either with or without an external electric field present), or ionization of the particle by electromagnetic radiation such as ultraviolet light, visible light, x or gamma radiation. These processes can be considered separately.

One of the important charging mechanisms is electrolyte effects. In this mechanism, solutions of liquids of high dielectric constant exchange ions with metals or solid surfaces. When a droplet of a high dielectric constant is swept from a metal surface, it can develop and can carry away a high charge. For a given surface and liquid, droplets will all have a net charge of the same sign, so that the droplets will repel each other. This is an important mechanism in this field because the surface of Vibrating Orifice Aerosol Generator jet or Collision Nebulizer jet is made from aluminum or the other metal and droplets formed from liquid with a high dielectric constant are charged immediately. Table 2.2 lists dielectric constants for various materials.

In case of atomizing sodium chloride solution, droplets of the solution will be charged at the surface of the nebulizer because of the large value of dielectric constant. During the evaporation process, the volume of the droplets decrease but the charge remains, so the solid sodium chloride particles formed will retain some or all the original charge. Two effects of such electric charges on the particle behavior must be taken into consideration. Attractive electrical forces between aerosol particles encourage coagulation, but simultaneously repulsion forces will reduce coagulation. The net result is a negligible change in coagulation for aerosols with the Boltzmann equilibrium charge distribution. However, there can be a significant increase in coagulation for aerosols with strong bipolar charge distribution. Charged aerosols are more likely than uncharged aerosols to form chainlike agglomerates. (Hinds, 1982)

Aerosol particles with a unipolar charge distribution are repelled from one another, which decelerates the coagulation. The repelling force leads to electrostatic dispersion or the spreading of the aerosol cloud, which also results in a reduction in number concentration because those charged particles are attracted by the electric field around the aerosol system. When the number concentration of the particle is n , the sign of the electrical charge of all the particles is the same and the number of the charge, q , is also the same, the strength of the electric field is given by

$$\text{div} E = 4 \pi n q \quad (2.45)$$

Table 2.2 Dielectric Constants of Several Materials (20°C)

Material	Dielectric Constant
Oil	2 -2.2
Methyl alcohol	31
Ethyl alcohol	24.3
Sodium chloride	5.9
Water	78
Magnesium oxide	9.65
Grass	5 - 10
Air	1
PVC	3.3 - 4.5

On the other hand, by definition $v_{TE}=BqE=ZE$, substituting E gives

$$\text{div } v_{TE} = 4 \pi n q Z \quad (2.46)$$

where v_{TE} is electrostatic terminal velocity. This equation shows that the aerosol particles transport to circumference of the aerosol system with their terminal electrostatic velocity, v_{TE} .

This phenomena is called electrical diffusion. Furthermore, because $\text{div } v_{TE}$ can be written as

$$\text{div } v_{TE} = -\frac{1}{n} \frac{dn}{dt} \quad (2.47)$$

From the solution of equation (2.47), the number concentration of aerosol particle decreases by

$$\frac{1}{n} - \frac{1}{n_0} = 4 \pi q Z t \quad (2.48)$$

where n_0 is the initial concentration. Figure 2.2 shows the number concentration decay by the electrical diffusion with time for aerosol particles of several size range.

Figure 2.2 Concentration Decay of the Unipolar Charged Particle. $n_0=10^9 \text{ cm}^{-3}$ and $n_0=10^5 \text{ cm}^{-3}$.

Calculated from equation (2.48)

2.4. Particle Deposition in the Laminar Flow

Laminar flow is the streamline flow of an incompressible, viscous Newtonian fluid in which all parts of the fluid move in distinct and separate lines.

For larger particles that are more affected by gravity than by diffusion, the rate of deposition on a horizontal surface is proportional to the density of the particle because the rate of deposition is equal to the products of the particle concentration and the falling speed.

For smaller particles, whose size is less than 1 μm , the particles can cross the streamline by inertia or by molecular diffusion. From equation (2.7), the flux of particle is proportional to the concentration gradient and it is very important to understand this concentration gradient. In order to evaluate this gradient, it is essential to know how the concentration is maintained at a distance from the deposition wall. If the wall works as a perfect absorber of the aerosol particles, the concentration on the surface of the deposition wall is zero and the concentration increases outwards. With the hypothetical condition where the concentration is uniform beyond a certain distance from the deposition surface (within a certain distance, this region is called diffusion boundary layer), the deposition flux can be calculated by (Davies, 1964)

$$\phi = \frac{D n_0}{\delta_d} \quad (2.49)$$

where δ_d is the thickness of the diffusion boundary layer. In this layer, from the surface the entire concentration gradient should be linear. n_0 is the concentration outside the boundary layer. With equation (2.15), the deposition velocity is expressed by

$$v_d = \frac{\phi}{n_0} = \frac{D}{\delta_d} \quad (2.50)$$

Figure 2.3 shows the deposition velocity with various diffusion boundary layer thickness. The larger particles, more than 1 μm in size, have an invariably small deposition velocity. For

the larger particles, diffusive deposition is not significant except when the boundary layer is very thin. Most molecules have a size of between 10^{-4} μm and 10^{-3} μm . For small particles whose size is less than 1 μm , the deposition velocity is large especially when the thickness of the diffusion boundary layer is thin. This indicates that small particles have an ability to penetrate the streamline of the main laminar flow by diffusion while larger particle can reach the deposition wall by the inertia or the gravitational settling. The thickness of the diffusion boundary layer is determined by the characteristics of main flow.

2.4.1. Laminar Boundary Layer and Diffusive Deposition over a Flat Plate

Laminar boundary layer is also called laminar velocity layer because this layer develops the velocity gradient between the main flow and the flow very close to the flat plate surface. Because the flow near the flat plate surface is affected by the viscosity of the fluid, the flow rate becomes smaller than free stream. Figure 2.4 (a) shows the uniform flow over a flat plate. If the fluid is inviscid, the velocity profile appears as in Figure 2.4 (b). The velocity at the wall is not zero, as it would be for a real fluid. A better depiction of a real flow is given in Figure 2.4 (c). Far from the surface, the fluid is not affected by the presence of the plate; consequently, the effect of viscosity in this region can be neglected. Except the boundary layer, both the velocity and the pressure are equal to the free stream value. Inside the laminar boundary layer, the velocity varies from zero at the wall to the free stream value at the edge of the laminar boundary layer. The thickness of the laminar boundary layer increases with length downstream. In spite of this thickness, its effect is extremely important in such problems as the computing resistance to motion of a body through a fluid, in determining heat transfer characteristics at a body surface or calculating the aerosol particle deposition velocities.

2.4.2. The Thickness of the Laminar Boundary Layer

With sufficiently large velocity, almost all the velocity gradient is confined to the laminar

Figure 2.3 Deposition Velocity by Diffusion through a Diffusion Boundary Layer of Various Thickness (Calculated from equation (2.50))

(a) Uniform flow

(b) Inviscid fluid

(c) Real fluid

Figure 2.4 Uniform Flow over the Flat Plate

boundary layer and the thickness of the boundary layer influences the deposition velocity of the aerosol particles from the main stream towards the surface. Takahashi (1982) estimated the thickness of this boundary layer and a deposition velocity by using differential equations of fundamental fluid dynamics in the following way.

Suppose that the flow is two-dimensional and steady. Furthermore, the fluid is taken to be Newtonian and incompressible with constant properties. The boundary layer equations that are not applicable to a region of backflow. Because the laminar boundary layer is very thin, the pressure does not vary in the direction along the body surface. The y axis extends upward normal to the surface. The continuity equation in differential form is

$$\frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} = 0 \quad (2.51)$$

The Bernoulli equation applies to the region outside of the laminar boundary layer in differential form.

$$\frac{1}{\rho} \frac{\partial p}{\partial x} + u_0 \frac{\partial u_0}{\partial x} = 0 \quad (2.52)$$

where u_0 is a main stream velocity in the positive direction of x axis. The x component of the Navier-Stokes equation, neglecting gravity, is

$$u_x \frac{\partial u_x}{\partial x} + u_y \frac{\partial u_x}{\partial y} = -\frac{1}{\rho} \frac{\partial p}{\partial x} + \nu \frac{\partial^2 u_x}{\partial y^2} \quad (2.53)$$

where p is the pressure and ν is the kinematic viscosity ($\nu = \eta / \rho$). For laminar boundary layer flow over a flat plate, equations (2.51), (2.52) and (2.53) should be solved simultaneously for u_x , u_y , and p . Assuming uniform flow ($\partial u / \partial x = 0$), because equation (2.52) shows that there is no pressure variation with x , equation (2.53) becomes

$$u_x \frac{\partial u_x}{\partial x} + u_y \frac{\partial u_x}{\partial y} = \nu \frac{\partial^2 u_x}{\partial y^2} \quad (2.54)$$

The boundary conditions are

$$x = 0, y = \infty \quad u_x = u_0 \text{ (condition 1)}$$

$$y = 0 \quad u_x = 0, u_y = 0 \text{ (condition 2)}$$

From equation (2.51) and (2.54), the solution of this differential equations is (Levich, 1962)

$$u_x = \frac{1}{\sqrt{2}} u_0^{3/4} \alpha^{1/2} \varphi^{1/2} \nu^{-1/4} x^{-1/4} \approx \frac{1.33}{4} u_0 \sqrt{\frac{u_0 y}{\nu x}} \quad (2.55)$$

$$u_y \approx \frac{1.33}{16} \frac{u_0^{3/2} y^2}{\nu^{1/2} x^{3/2}} \quad (2.56)$$

where φ is the function of flow

$$\varphi = \frac{\alpha}{8} \frac{u_0^{3/2} y^2}{\nu^{1/2} x^{1/2}} \quad (\alpha = 1.33) \quad (2.57)$$

Because the diffusive transportation of aerosol particles can be expressed in the same manner as momentum transfer of fluid, differential equation about aerosol concentration can be written as follows

$$u_x \frac{\partial n(x,y)}{\partial x} + u_y \frac{\partial n(x,y)}{\partial y} = D \frac{\partial^2 n(x,y)}{\partial y^2} \quad (2.58)$$

Equation (2.58) should be expressed as a function of x and φ in order to use equations (2.55) -

(2.57). After arrangement of the formula, the following equation is obtained.

$$\frac{\partial^2 n}{\partial \zeta^2} + \frac{v}{D} \frac{\alpha}{2} \zeta^2 \frac{\partial n}{\partial \zeta} = 0 \quad (2.59)$$

where $\zeta = \frac{1}{2} \sqrt{\frac{u_0}{v x}} y$. Equation (2.59) can be solved as ordinary differential equation with following conditions

$$n_{y=0} = 0 \quad n_{\zeta=0} = 0 \text{ (condition 1)}$$

$$n_{y=\infty} = 0 \quad n_{\zeta=\infty} = 0 \text{ (condition 2)}$$

The solution is

$$n(\zeta) = C_1 \int_0^\zeta \exp(-0.22 S_c \zeta^3) d\zeta + C_2 \quad (2.60)$$

or

$$n(x, y) = \frac{(0.22 S_c)^{1/3}}{0.89} n_0 \int_0^{1/2 \sqrt{\frac{u_0 y^2}{v x}}} \exp(-0.22 S_c \zeta^3) d\zeta \quad (2.61)$$

where S_c is Schmidt number which is the ratio of a kinematic viscosity to diffusion coefficient ($=v/D$). As the Schmidt number increases, convective mass transfer increases relative to Brownian diffusion of particles. S_c is relatively independent of temperature and pressure near standard conditions. Because the deposition flux is given by equation (2.17), after expansion and differentiation of equation (2.61), the deposition flux can be obtained.

$$\phi = 0.34 n_0 D S_c^{1/3} \sqrt{\frac{u_0}{v x}} = 0.34 n_0 D^{2/3} v^{-1/6} u_0^{1/2} x^{-1/2} \quad (2.62)$$

Figure 2.5 shows the deposition velocity through the laminar boundary layer which is calculated from equation (2.62). The particle diameter is 10 nm and u_0 is 500 cm s⁻¹.

The thickness of the diffusion layer δ_d , in which the particle concentration gradient is constant,

Figure 2.5 Deposition Velocity through a Boundary Layer (Particle diameter is 10 nm and u_0 is 50 cm s⁻¹)

is obtained from the equation (2.49).

$$\delta_d = \frac{D n_0}{\phi} \approx 3 Sc^{-1/3} \sqrt{\frac{v x}{u_0}} \quad (2.63)$$

In the same manner, when the velocity gradient is constant in the laminar boundary layer, the thickness of the laminar boundary layer is

$$\delta_L \approx 5 \sqrt{\frac{v x}{u_0}} \quad (2.64)$$

The term δ_L represents distance from the surface of the wall to the point where 99 percent of u_0 is reached. At the distance to the point where 85 percent of u_0 is reached, the numerical coefficient becomes 3 instead of 5. If u_0 is 50 cm s^{-1} , δ_L becomes 0.61 cm ($x=5 \text{ cm}$) or 0.87 cm ($x=10 \text{ cm}$).

Laminar boundary layer velocity profile for flow past a flat plate is tabulated in Table 2.3.

When the Reynolds number is defined as $u_0 x / v$, the formula above can be changed as

$$\delta_L = \frac{5x}{\sqrt{Re_x}} \quad (2.65)$$

The relationship between the distribution of flow u_x and the distribution of aerosol particle concentration is shown in Figure 2.6. From equation (2.63) and (2.64), eliminating x and u_0 , the ratio of δ_d and δ_L is expressed by

$$\frac{\delta_d}{\delta_L} = 0.6 \sqrt[3]{\frac{D}{v}} \quad (2.66)$$

The kinematic viscosity of the air, v , is about $0.15 \text{ cm}^2 \text{ s}^{-1}$ for the room condition. The value of D is less than this for practically everything except for a few light gases such as water vapor, while for heavy gases and particles, it is very much smaller. Hence, only for a few gases of low

Figure 2.6 Velocity and Particle Concentration Profile in Laminar Flow

molecular weight have the thicker diffusion boundary layer than the laminar boundary layer.

Table 2.3 Laminar Boundary Layer Velocity Profile for Flow past a Flat Plate: The Blasius solution (Reist, 1984)

Coefficient	u_x / u_0	Coefficient	u_x / u_0
0.0	0.0	2.8	0.81152
0.2	0.06641	3.0	0.84605
0.4	0.13277	3.2	0.87609
0.6	0.19894	3.4	0.90177
0.8	0.26471	3.6	0.92333
1.0	0.32979	3.8	0.94112
1.2	0.39378	4.0	0.95552
1.4	0.45627	4.2	0.96696
1.6	0.51676	4.4	0.97587
1.8	0.57477	4.6	0.98269
2.0	0.62977	4.8	0.98779
2.2	0.68132	5.0	0.99155
2.4	0.72899	∞	1.00000
2.6	0.77246		

CHAPTER 3 Deposition of Aerosol Particles in Turbulent Flow

3.1.1. Laminar and Turbulent Flow

When a fluid is undergoing laminar flow, a particle in one layer generally stays in that layer unless it is highly diffusive. The layers of fluid slide by one another without apparent eddies or swirls. In turbulent flow, eddies and vortices mix the fluid by moving particles about the cross section.

In both laminar and turbulent flow, the flow velocity at the wall is zero. This phenomenon, called the nonslip condition, is due to viscosity. In laminar flow, in which inertia or momentum of the fluid is small, the viscous effect is able to penetrate farther in to cross section from the wall than it can in turbulent flow. In turbulent flow, in which the viscous effects are small, the momentum or inertia of the flow is able to penetrate farther outward toward the wall from the centerline than it can in laminar flow. This penetration of momentum or inertia is called the momentum transport phenomenon. However, although the effect of viscosity is very small in the turbulent flow, there is a region that exists very close to the wall where the flow is not turbulent. This region is called a laminar sublayer.

Because there are local fluctuations in time and space in the turbulent flow, the readily measured experimental properties of the turbulent flow are the time average velocity and the intensity and spectrum of the turbulence. For example, for flow velocity, two velocities can be identified: the mean velocity which is an average or mean value, and the instantaneous velocity, which fluctuates randomly about the mean. The instantaneous velocity can be average over a finite time to yield the mean value.

$$\bar{u} = \frac{1}{t} \int_0^t u \, dt \quad (3.1)$$

where t is the time period for the measurement. A similar time-smoothing operation can be performed for pressure at a point that also fluctuates in the turbulent flow.

3.1.2. Time-smoothed Equation of Motion

In general, the forces acting on a fluid are gravitational, viscous/frictional and pressure forces. If the continuity and momentum equations can be written for all three principal directions, and the fluid is Newtonian with constant properties of density and viscosity, a set of differential equations can be obtained. The momentum equation written for each principal direction gives what are called the Navier-Stokes equations. The other equations are called continuity equations. The continuity equations are statement of the conservation of mass.

In order to apply these equations, new parameters have to be used to express the fluctuation about the mean which are u' for stream velocity and p' for pressure. The results for steady incompressible turbulent flow in Cartesian coordinates becomes the following continuity equation

$$\frac{\partial \bar{u}_x}{\partial x} + \frac{\partial \bar{u}_y}{\partial y} + \frac{\partial \bar{u}_z}{\partial z} = 0 \quad (3.2)$$

Equation of motion for the x component is

$$\begin{aligned} & \rho \left(\bar{u}_x \frac{\partial \bar{u}_x}{\partial x} + \bar{u}_y \frac{\partial \bar{u}_x}{\partial y} + \bar{u}_z \frac{\partial \bar{u}_x}{\partial z} \right) \\ &= -\frac{\partial \bar{p}}{\partial x} + \eta \left(\frac{\partial^2 \bar{u}_x}{\partial x^2} + \frac{\partial^2 \bar{u}_x}{\partial y^2} + \frac{\partial^2 \bar{u}_x}{\partial z^2} \right) - \frac{\partial}{\partial x} (\rho \overline{u'_x u'_x}) - \frac{\partial}{\partial y} (\rho \overline{u'_x u'_y}) - \frac{\partial}{\partial z} (\rho \overline{u'_x u'_z}) + \rho g_x \end{aligned} \quad (3.3)$$

flow, an analogous equation can be used

$$\bar{\tau}_{yx} = -\eta_e \frac{d\bar{u}_x}{dy} \quad (3.7)$$

where η_e is the eddy viscosity. The total mean shear stress is then

$$\bar{\tau}_{yx} = -\eta \frac{d\bar{u}_x}{dy} - \eta_e \frac{d\bar{u}_x}{dy} \quad (3.8)$$

where η is a viscosity that result from molecular motions, whereas η_e is the eddy viscosity that result from eddying motions in turbulent flow.

Boussinesq (1877) introduced the concept of eddy viscosity in order to find the model which explained Reynolds shear stress. He assumed that the molecular action that is responsible for viscous shear is analogous to the eddies that cause turbulent stress. This assumption implies that the turbulent stresses are proportional to the velocity gradient which is shown in equation (3.7). The eddy viscosity, η_e , assumed to be the product of a velocity and a length like the kinematic viscosity which is the ratio of absolute viscosity to density (η / ρ). According to the kinetic theory of gasses, the kinematic viscosity, ν , is given by (Kennard, 1938)

$$\nu = 0.499 \bar{c} \lambda \quad (3.9)$$

where $\bar{c}$ is the mean molecular speed and λ is the mean free path. The basic idea in equation (3.7) is that the eddies in turbulent flow are moving in a fluid and thus in their motion approximate the motion of free molecules of a gas. The eddies disappear in short time and new ones are continuously fed into the fluid system. The effective distance which they travel in the transverse direction is called the mixing length, l , and the velocity with which they move in relation to the surrounding fluid is of the order of, or at least proportional to the velocity fluctuation. This idea

Figure 3.1 Schematic Diagram of the Mixing Length

is illustrated in Figure 3.1. Two points, y_1 and y_2 , are selected from the boundary such that they extend well into the turbulent portion of the flow. These points are separated by a distance l that has the average size of eddies. The time-smoothed velocities at the point of y_1 and y_2 are $\bar{u}_{x1}$ and $\bar{u}_{x2}$. It is theorized that a positive turbulent fluctuation in the vertical direction at y_2 causes a corresponding reduction in the x -directed component of u_{x1} , the instantaneous velocity at y_1 (Cebeci and Smuth, 1974). The reduction u_{x1}' is approximately equal to the differences in mean velocities. So at y_1 ,

$$u_{x1}' \approx \bar{u}_{x1} - \bar{u}_{x2} \approx l \frac{d\bar{u}_x}{dy} \quad (3.10)$$

and the shear stress becomes

$$\bar{\tau}_{yx} = \rho \overline{u_x' u_y'} \approx \rho u_y' l \frac{d\bar{u}_x}{dy} \quad (3.11)$$

Figure 3.2 Simplified Diagram of Momentum Transfer

If it is further assumed that the turbulent fluctuation, u_y' , is approximately equal to the horizontal fluctuation, u_x' , then equation (3.11) becomes

$$\bar{\tau}_{yx} = \rho \overline{u_x' u_y'} \approx \rho l^2 \frac{d\bar{u}_x}{dy} \left| \frac{d\bar{u}_x}{dy} \right| \quad (3.12)$$

where l is a length of the same order as the root mean square of the random length l' and absolute value signs are placed to ensure that the sign on the shear stress properly reflects the sign of the gradient.

In Figure 3.2, particles of fluid are assumed to migrate as a result of the turbulence. A particle at point 1 may migrate to level y_0 . By that time, it will be deficient in velocity by some amount that should be proportional to $\partial \bar{u}_x / \partial y$. Since the particle has a velocity different from the mean, the deficiency is u_x' and, to first order, $u_x' = l \times (\partial \bar{u}_x / \partial y)$, where l is an unknown length. In kinetic theory, l is the mean free path, the average distance between collisions. In Prandtl's theory (1926), it is reasonable to assume that there is some effective, characteristic length. The

momentum deficiency is ρu_x or $\rho l(\partial u_x / \partial y)$. The fluid elements will have a vertical velocity proportional to u_y , and the rate of transport of excess momentum to the line y_0 will be proportional to u_y' . Because of continuity, u_y' can hardly be other than proportional to u_x' . For instance, if a particle from point 2 overtakes a particle coming from point 1, fluid is forced out from between them at a rate proportional to their velocity difference, which is the same as saying that $u_y' \propto u_x'$. Therefore

$$u_y' \approx l \left(\frac{\partial u_x}{\partial y} \right) \quad (3.13)$$

and the eddy viscosity can be written by

$$\eta_e = l^2 \left| \frac{d\bar{u}_x}{dy} \right| \quad (14)$$

where l is a length which generally depends on the coordinates and characterizes the scale of turbulence (the mean dimension of the turbulent fluctuations) at a given point.

Figure 3.3 Mixing Length Distribution across the Turbulent Boundary Layer (according to the data of Klebanoff, 1954)

Equation (3.12) requires some empirical data if it is to be used for quantitative calculations. It is necessary to make assumptions for the distribution of eddy viscosity. Figure 3.3 show the mixing length distributions on a flat plate according to the measurement of Klebanoff (1954). The y axis is taken perpendicular to the wall surface and δ is the thickness of the turbulent boundary layer. The graph shows that in the region of $y/\delta < 0.15$, the mixing length changes linearly with the distance from the wall surface. In this region, the mixing length can be approximated by

$$l = \kappa y \quad (3.15)$$

where κ is a proportional constant known as Kàrmàn's constant. For y/δ greater than about 0.3, the mixing length remains approximately constant.

In the same manner of the development of equation (3.14), the eddy diffusion coefficient can be obtained.

$$D_e = l_2^2 \frac{d\bar{u}_x}{dy} \quad (3.16)$$

Where l_2 is another mixing length. The relationship between l_2 and l determines the value of the turbulent Prandtl number, Pr_t , which is the ratio of η_e to D_e . The momentum transfer theory supports $Pr_t = 1$.

3.1.3. Particle Deposition over a Flat Plate through a Turbulent Boundary Layer

In laminar flow, the laminar boundary layer on the smooth surface grows in thickness downstream. However, the thicker the boundary layer grows, the smaller the velocity gradient across the laminar boundary layer becomes. Therefore, the momentum towards the surface becomes too large to overcome viscous drag force of the fluid near the surface. In this very thick boundary layer, near the surface, an unstable velocity profile develops because of the influence of a reversed pressure gradient downstream. This flow becomes turbulent although the main stream

flow may be laminar. Generally, when the Reynolds number exceeds 10,000, break up of the laminar flow occurs.

The velocity gradient at the surface in a turbulent boundary layer is steeper than in a laminar boundary layer, and the rate of diffusion is enhanced because the thinner diffusion layer is maintained near the bulk concentration region. In this case, the eddy diffusion coefficient is used to estimate the diffusive deposition.

In order to analyze the particle deposition through the turbulent boundary layer, the tangential shear stress, τ_s , on the surface over which the boundary layer is flowing is used. The shear stress is also the rate of transfer of momentum perpendicular to the surface which depends on the amount of turbulence. In laminar flow, the shear stress due to the viscosity of the fluid. However, in turbulent flow, in addition to the effect of viscosity, the momentum carried across the main stream has to be taken into consideration.

Kármán (1930) showed how mean velocities in a turbulent boundary layer could be rendered dimensionless by defining the friction velocity, u_* , which is

$$u_* = \sqrt{\frac{\tau_s}{\rho}} \quad (3.17)$$

In flows the tangential stress is greatest at the wall and falls off linearly. Outside the sublayer, it is equal to the Reynolds stress, $\rho \overline{u_x u_y}$. Within the sublayer, the turbulence decreases and the shearing stress becomes greater. This is due to the effect of viscosity as the surface is approached. The friction velocity is therefore similar in value to the mean turbulent velocity amplitude at a distance from the surface where turbulence is greatest.

Using the friction velocity, the turbulent boundary layer can be generalized with the dimensionless variables of the distance, y^+ , and the velocity, u_x^+ .

$$u_x^+ = u_x / u_* \quad (3.19)$$

$$y^+ = y u_* / \nu \quad (3.18)$$

When the particle has small inertia, Fuchs (1964) introduced a relatively simple model. The entire system consists of the turbulent region and very thin laminar sublayer (thickness = δ_L) region which is very similar to the laminar boundary layer. In the laminar sublayer, the turbulent effects can be ignored. Suppose a plane is placed in the turbulent flow. When the x axis aligned in direction of main flow and the y axis perpendicular to the wall surface, the velocity profile of each region can be written as

$$\frac{u_x(y)}{u_*} = 2.5 \ln y^+ + 5.5 \quad ; y > \delta_L \quad (3.20)$$

$$\frac{u_x(y)}{u_*} = y^+ \quad ; y < \delta_L \quad (3.21)$$

Generally speaking, equations above are valid only when $y^+ > 20$, $y^+ < 5$. On the other hand, turbulent kinetic viscosity coefficient, ν_t , can be expressed as $\kappa u_* z$ (κ is Kármán's constant and ≈ 0.4). The momentum transfer in the turbulent flow is written as

$$\tau_s = \nu_t \frac{\partial(\rho u_x)}{\partial z} \quad (3.22)$$

and mass transfer is

$$\phi = D_e \frac{\partial n(x, y)}{\partial y} \quad (3.23)$$

From equation (3.22) and (3.23), the turbulent kinematic viscosity, ν_t , and the eddy diffusion coefficient, D_e , are physically similar and their values are considered as being close to each other. The ratio of ν_t / D_e was determined by the experiment as being between 1.4 and 2.0. Thus, $D_e = 1.4 \sim 2.0 \kappa u_* y = 0.6 \sim 0.8 u_* y$; $y > \delta_L$.

Because diffusive deposition by Brownian motion is dominant when inside the laminar sublayer, the deposition flux in that region can be written using equation (2.17). If the particle concentration is zero at the surface of the wall, the concentration at height y , $n(y)$ becomes $\phi y / D$. On the other hand, in the vicinity of $y > \delta_L$ the deposition flux is

$$\phi' = D_e \frac{\partial n'(y)}{\partial y} = \alpha u_* y \frac{\partial n'(y)}{\partial y} \quad (3.24)$$

where $\alpha = 0.6 \sim 0.8$ and prime means its deviation component is caused by turbulence. Hence, if $n' = n_0$ at $y = h$, n' can be written as the function of y

$$n'(y) = n_0 + \frac{\phi'}{\alpha u_*} \ln \frac{y}{h} \quad (3.25)$$

Because at the point $\delta_L = y$, ϕ and n can be considered as being ϕ' and n' respectively, the deposition flux is

$$\phi = \frac{n_0 D}{\delta_L - \frac{D}{\alpha u_*} \ln \frac{\delta_L}{h}} = \frac{n_0 D}{\delta_L \left(1 - \frac{1}{10 \alpha Sc} \ln \frac{\delta_L}{h} \right)} \quad (3.26)$$

Generally, in case of aerosol particles, Sc is much larger than 1

$$\phi \approx \frac{n_0 D}{\delta_L} \quad (3.27)$$

However the velocity distribution in a turbulent flow is not simple. There is a complex transition region between the deposition wall and the turbulent flow region. In this transition region, both v_t and v effect the momentum transfer. Moreover, to be exact, the value of v_t can not be expressed as $\kappa u_* y$. Levich (1962) divided this transit region into four sections shown in Figure 3.4 and assumed the eddy diffusion coefficient took effect even very close to the wall.

Figure 3.4 The Structure of the Turbulent Boundary Layer

Region[1] $z > d$; *turbulent region* $d = u_* x / u_0$ $n_1 = n_0$

Region[2] $\delta_{vs} < z < d$; *buffer region*, $\delta_{vs} \approx 10\nu / u_*$ and $D_e \gg D$. In this region D_e is expressed as

$$D_e = \beta_0 y^2 \frac{\partial u_x}{\partial y} \quad (3.28)$$

From equation above, the deposition flux is

$$\phi = D_e \frac{\partial n}{\partial y} = \beta_0 y^2 \frac{\partial u_x}{\partial y} \frac{\partial n}{\partial y} \quad (3.29)$$

If u_x can be expressed like equation (3.20) in this region, the deposition flux becomes

$$\phi = \beta u_* z \frac{\partial n}{\partial z} \quad (3.30)$$

This equation is equivalent to equation (3.23) substituting D_e for $\beta n_* z$ ($\beta = 0.5 \sim 0.8$). Hence, if $n_2 = n_1 = n_0$ at $y = d$, the number concentration of this area is

$$n_2 = \frac{\phi}{\beta u_*} \ln \frac{y}{d} + n_0 \quad (3.31)$$

Region[3] $\delta_{ds} < y < \delta_{vs}$; *viscous sublayer region*

In this region, he assumed D_e is much larger than D as well. If D_e can be written as

$$D_e = \frac{\gamma u_* y^4}{\delta_{vs}^3} \quad (3.32)$$

where γ is a constant. The deposition flux is

$$\phi = \gamma u_* \frac{y^4}{\delta_{vs}^3} \frac{\partial n}{\partial y} \quad (3.33)$$

Hence, with the condition of $n_3=n_2$ at $y=\delta_{vs}$, the solution of equation above is

$$n_3 = n_0 + \frac{\phi}{u_*} \left(\frac{1}{\beta} \ln \frac{\delta_0}{d} + \frac{1}{3\gamma} - \frac{\delta_0^3}{3\gamma z^3} \right) \quad (3.34)$$

Region[4] $y < \delta_{ds}$: diffusion sublayer region

In this region, D is the dominant factor and $\phi=D(\partial n/\partial y)$. The solution of this equation with the condition of $n=0$ at $z=0$ and $n_4=n_3$ at $y=\delta_{ds}$ becomes as

$$\phi = \frac{D n_0}{-\frac{D}{\beta u_*} \ln \frac{\delta_{vs}}{d} + \frac{\delta_{vs}^3 D}{3 \gamma u_*} \left(\frac{1}{\delta_{ds}^3} - \frac{1}{\delta_{vs}^3} \right) + \delta_{ds}} \quad (3.35)$$

If $D_e=D$ at $z=\delta_{ds}$, $D=\gamma u_* \delta^4/\delta_{vs}^3$, and if $\delta_{vs} \approx \delta_L \approx 10\nu/u_*$,

$$\delta_{ds} \approx \frac{10^{3/4} \nu}{u_* (\gamma Sc)^{1/4}} \quad (3.36)$$

From equation (3.35) and (3.36),

$$\phi = \frac{D n_0}{\delta_{ds} \left[1 + \frac{1}{3} \left[1 - (10 \gamma Sc) Sc^{-3/4} \right] - \frac{\gamma^{1/4}}{10^{3/4} \beta Sc^{3/4}} \ln \frac{\delta_{vs}}{d} \right]} \quad (3.37)$$

Because generally Sc is much larger than 1 in case of an aerosol particle, equation (3.37) can be rewritten as

$$\phi \approx \frac{D n_0}{\delta_{ds} \left(\frac{4}{3} - \frac{\delta_{ds}^{1/4}}{10^{3/4} \beta Sc^{3/4}} \ln \frac{\delta_{vs}}{d} \right)} \quad (3.38)$$

With $\beta \approx \gamma \approx 1$, and equation (3.36), equation (3.38) can be further simplified to

$$\phi \approx \frac{D n_0}{4/3 \delta_{ds}} = \frac{n_0 u_*}{7.5 \gamma^{-1/4} Sc^{3/4}} \quad (3.39)$$

In order to evaluate the deposition velocity, it is necessary to know u_* which is dependent on the degree of turbulence and the roughness of the surface along which the air is flowing. The deposition velocity is shown in Figure 3.5. The deposition velocity is calculated from equation (3.39) with $\nu = 0.15 \text{ cm}^2\text{s}^{-1}$ and is shown as a fraction of the friction velocity, $v_d^+ = v_d/u_*$.

3.1.4. Inertial Deposition of Particles

For small particles, the diffusive deposition is the dominant process but when the inertia of the particles is sufficient for them to travel some distance independent of the stream line of medium fluid, another model should be discussed.

The relaxation time characterizes the time required for a particle to adjust its velocity to a new condition. The relaxation time, τ , is defined as

$$\tau = mB = \frac{\rho_p d^2 C_c}{18 \eta} \quad (3.40)$$

where m is the mass of the particle, B is the mechanical mobility, ρ_p is the density of the particle and d is the particle diameter. Several values in standard air condition are shown in Table 3.1.

Figure 3.5 Diffusive Deposition Velocity through the Turbulent Boundary Layer. The Deposition Velocity Expressed as a Fraction of the Friction Velocity according to Equation (3.39). (In air, with standard condition)

Table 3.1 Relaxation Time at Standard Condition ($\rho_p=1 \text{ g cm}^{-3}$)

Particle Diameter (μm)	Relaxation Time (s)
0.01	6.8×10^{-9}
0.1	8.8×10^{-8}
1.0	3.6×10^{-6}
10.0	3.1×10^{-4}
100	3.1×10^{-2}

The relaxation time increases with the particle size because larger particles in motion have larger inertia which tends to keep the particles moving along their current trajectory.

When there is no force except the drag force against a particle in stagnant air, the particle travels until its relaxation time elapses. If the particle had an initial velocity V_0 , the distance the particle is transported is given by

$$d_{\text{stp}} = V_0 \tau \quad (3.41)$$

where d_{stp} is the total distance traveled by the particle, called the stopping distance or inertial range. The stopping distance represents the effective momentum as well. From the definition of the relaxation time (equation (3.40)), the stopping distance can be expressed in terms of the product of the particle's mobility and its initial momentum

$$d_{\text{stp}} = B m V_0 \quad (3.42)$$

In most cases, particles reach their stopping distance very rapidly. In a time equal to 3τ , particles travel 95% of their stopping distance. When a particle is moving with the air flow and

Figure 3.6 Stopping Distance of Particles calculated from equations (3.41) and (3.42). Initial velocity, V_0 , is 1000 cm s^{-1} and the particle density, ρ_p , is 1.0 g cm^{-3} .

the flow is turned through 90° , the stopping distance represents the distance the particle continues to travel in its original direction and thus can be thought of as a measure of the persistence of a particle to continue along its original path.

For larger particles with high initial velocity, those particles are not in the Stokes region. Because in this case, the drag force and the velocity on the particles are changing proportionally, it is very difficult to estimate the stopping distance numerically. Mercer (1973) derived the

empirical formula for a particle having an initial Reynolds number of between 1 and 400.

$$d_{sp} = \frac{\rho_p d}{\rho_g} \left(Re_0^{\frac{1}{3}} - \sqrt{6} \arctan \left(\frac{Re_0^{\frac{1}{3}}}{\sqrt{6}} \right) \right) \quad (3.43)$$

Where $Re_0 = \rho_g V_0 d / \eta$, and the arctan is expressed in radians. Figure 3.6 shows the stopping distance of several different sized particles.

Smaller particles are transported to the border between the turbulent region and the laminar sublayer by the fluid turbulence and then they deposit on the wall surface by the molecular diffusion. In this case, the interposol of even a thin layer of very low turbulence forms an effective hindrance to deposition. However, for larger particles, because of their long stopping distance, the mechanism of turbulent deposition is different that of small particles. Because the inertia of larger particles is sufficient for them to travel some distance independently, they can reach the wall surface by eddy impingement. According to the size of the particles and strength of turbulence, d_{sp} can be of any of a wide range of magnitude. Therefore, it is not convenient to break down the boundary layer into laminar and buffer regions. When the d_{sp} is longer than the thickness of the sublayer, the particles can be reached the deposition surface, across the intervening regions of lesser turbulence.

Based on this model Friedlander and Johnstone (1957) calculated the deposition velocity in a cylindrical tube with a smooth surface. They assumed the diffusion coefficient of particle is equal to that of the fluid and the deposition velocity is given by

$$\begin{aligned}
\frac{1}{v_d^+} &= \frac{1524.3}{d_{stp}^{+2}} - 50.52 + \frac{1}{\sqrt{f/2}} ; d_{stp}^+ < 5 \\
\frac{1}{v_d^+} &= 5 \ln \left(\frac{25.2}{s^+ - 4.795} \right) - 13.73 + \frac{1}{\sqrt{f/2}} ; 5 < d_{stp}^+ < 30 \\
\frac{1}{v_d^+} &= \frac{1}{\sqrt{f/2}} ; d_{stp}^+ > 30
\end{aligned} \tag{3.44}$$

where f is the Fanning's friction coefficient (Schlichting, 1968) which is given by

$$f = 0.0791 \text{Re}^{-1/4} \tag{3.45}$$

The dimensionless parameter is given by

$$\begin{aligned}
(\text{velocity})^+ &= (\text{velocity}) / u_* \\
(\text{length})^+ &= (\text{length}) \times u_* / \nu \\
(\text{diffusion coefficient})^+ &= \text{diffusion coefficient} / \nu \\
(\text{concentration})^+ &= (\text{concentration}) / \bar{n} \\
(\text{time})^+ &= (\text{time}) \times u_*^2 / \nu
\end{aligned} \tag{3.46}$$

Figure 3.7 shows the results of Friedlander's equation (equation 3.44). In Friedlander's model, the explanation of the initial velocity or the eddy diffusion coefficient of particle is not complete. Moreover, the particle's behavior very close to the deposition surface is not theoretically explained. Because of this, many models of turbulent deposition have been proposed.

Davies (1966) assumed that the eddy diffusion coefficient of particle is equal to the sum of the molecular and eddy diffusion coefficient of the fluid and the initial velocity is the instantaneous velocity, u' , of the point on the border between the turbulent region and the turbulent boundary layer region. From the experimental data (Laufer, 1954), the instantaneous velocity at the border is given by

Figure 3.7 Dimensionless Plot of Deposition Velocity against Particle Relaxation Time
(Calculated from Equation (3.44) with a condition of $Re=5000$, $\bar{u}_x = 1000 \text{ cm s}^{-1}$.)

$$u'_{y=\delta}^+ = \frac{\delta^+}{\delta^+ + 10} \quad (3.47)$$

Yoshioka *et al* (1972) took the gravitational sedimentation into consideration and calculated the deposition velocity based on Friedlander's model. Owen *et al* (1960), Hutchinson *et al* (1971) and Cleaver *et al* (1975) analyzed the model that the turbulent boundary layer is affected by turbulent bursting phenomena. Kaneoka (1973) considered the effect of the lifting force on particles in the turbulent boundary layer. Caporaloni (1975) introduced turbophoresis model which contained the transport velocity by non-isotropic turbulence.

3.1.5. Particle Loss in a Vessel

Langstroth and Gillespie (1947) observed the change of ammonium chloride particle size and number by coagulation and deposition in a turbulent flow in a rectangular smoke chamber. They showed (Langstroth *et al*, 1949; Gillespie, 1953) the reproducibility of the losses of particles by the coagulation in the cloud and deposition on the chamber wall surface. They also observed that the loss rate was strongly dependant on the intensity of stirring.

Twomey (1976) suggested a very simple model. In this model, it is assumed that the aerosol is enclosed in some vessel within which a uniform aerosol concentration is maintained throughout the main volume by convective mixing. Particles whose sizes are microns or submicrons are transported to the vicinity of the deposition walls by convection, and then reach the deposition walls through the boundary layer by the diffusion. Diffusion is the dominant factor inside the boundary layer and negligible at a distance greater than the boundary layer thickness, δ . Thus, at the border of boundary layer, the convective diffusion coefficient, D_c , and molecular diffusion coefficient, D , have the same value. Those particles are also affected by gravity. The particles are fall to the bottom of the vessel with a settling velocity, v_{ts} . If the vessel has a height, h , volume, V , and total surface area, s , then the net rate of aerosol concentration decay is

$$-\frac{dn}{dt} = \frac{\lambda_{ts}}{h} + \lambda_d \frac{s}{V} \quad (3.48)$$

where λ_{ts} is the rate of deposition by gravitational settling and λ_d is the rate of deposition by molecular diffusion. The solution of this equation (3.48) is

$$n = n_0 \exp \left(\frac{v_{ts}}{h} + \frac{s}{V} \frac{D}{\delta} \right) \quad (3.49)$$

where n_0 is the initial aerosol concentration. This equation is valid only for a cubic vessel.

Figure 3.8 Schematic Diagram Illustrating Convective Mixing and the Boundary Layer in a Rectangular Vessel

Fuchs (1964) concluded that when only convective diffusion and gravitational sedimentation are important in the closed vessel, the aerosol particle losses with time, t , is

$$n = n_0 \exp(-\beta t) \quad (3.50)$$

From the comparison of equation(3.49) and (3.50), β is given as

$$\beta = \frac{v_{ts}}{h} + \frac{s}{V} \frac{D}{\delta} \quad (3.51)$$

Figure 3.9 shows the plot of equation (3.51).

Figure 3.9 Deposition Coefficient, β , as a Function of Particle Size Calculated from Equation (3.51) . (In a rectangular vessel which is $75 \times 75 \times 180$ cm, in air at room temperature)

In Figure 3.9, there is a minimum value of β where the particle diameter is about $1 \mu m$. The particles larger than this size are removed by gravitational settling because of their high terminal settling velocity. On the other hand, the smaller particles than this diameter have a large diffusion coefficient and they are removed by the diffusive deposition. These two counteracting

phenomena result in a minimum value of the deposition coefficient.

This simple equation does not explain the eddy diffusion due to the turbulent flow generated by the natural convection. Assuming that the air inside the chamber is mixed well and the concentration inside of the chamber, except in the very thin layer region which is located very close to the chamber wall, is constant, Corner and Pendlebury (1951) developed a theory of a deposition and coagulation in a vessel. They also assumed that in the very thin layer, the flow rate distribution and the particle concentration distribution can be considered to be linear. The deposition flux of particles is

$$\phi = v_{ts} n + D_{\text{Eff}} \left(\frac{dn}{dy} \right)_{y=0} \approx v_{ts} \bar{n} + D \frac{\bar{n}}{\delta} \quad (3.52)$$

where v_{ts} is terminal settling velocity and D_{Eff} is effective diffusion coefficient in the very thin layer. Equation (3.52) indicates that the deposition flux is the sum of the sedimentation flux and the diffusive deposition flux. In equation (3.52), δ correspond to the diffusion layer in equation (2.49). Corner and Pendlebury included more details in order to generalize the solution of this equation. Because in the boundary layer, they assumed that both the turbulent diffusion and Brownian motion diffusion exist, the effective diffusion coefficient, D_{Eff} , is given as $(D_e + D)$. From the Prandtl's mixing length concept which is shown in equation (3.18), the turbulent diffusion coefficient of fluid, D_e , is

$$D_e \approx \kappa u_* y = \kappa y^2 \frac{\partial \bar{u}}{\partial y} = k_e y^2 \quad (3.53)$$

where k_e is the turbulent intensity ($= \kappa^2 (\partial \bar{u} / \partial y)$), κ is Kármán's constant and u_* is friction velocity. The concentration distribution in the boundary layer for the entire deposition wall can be expressed with the turbulent intensity for the effective diffusion coefficient. When the y axis is taken perpendicular to the deposition wall, the particle losses on the side walls are

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial y} \left[(k_e y^2 + D) \frac{\partial n}{\partial y} \right] \quad (3.54)$$

The particle losses on the top and the bottom of the chamber are, respectively

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial y} \left[(k_e y^2 + D) \frac{\partial n}{\partial y} \right] + v_{ts} \frac{\partial n}{\partial y} \quad (3.55)$$

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial y} \left[(k_e y^2 + D) \frac{\partial n}{\partial y} \right] - v_{ts} \frac{\partial n}{\partial y} \quad (3.56)$$

Corner and Pendlebury simplified these equations and obtained the solution to equations (3.55) and (3.56). The number of deposited particles on the entire walls of the chamber per unit time is

$$\Phi = n_0 \left[\frac{8}{\pi} \sqrt{k_e D} + v_{ts} \coth \left(\frac{\pi v_{ts}}{4 \sqrt{k_e D}} \right) \right] \quad (3.57)$$

With the average number concentration of the vessel, $\bar{n}$, equation (3.57) can be rewritten as

$$\beta = \frac{1}{L} \left[\frac{8}{\pi} \sqrt{k_e D} + v_{ts} \coth \left(\frac{\pi v_{ts}}{4 \sqrt{k_e D}} \right) \right] \quad (3.58)$$

where L is the side length of the cubic chamber. To use equation (3.17) the value of k_e is needed.

In many cases, the value of k_e are obtained experimentally. Takahashi and Kasahara (1968)

derived equations for a cylindrical vessel in the same manner as Corner and Pendlebury.

Crump and Seinfeld (1981) developed Corner and Pendlebury's model for wall loss

coefficient, β , for a vessel of arbitrary shape. For instance, for the spherical vessel, let R denote the radius of the sphere, θ the polar angle and suppose the turbulent boundary layer has thickness δ . In the boundary layer, they assumed that there exist three types of transport; turbulent diffusion, Brownian diffusion, and gravitational sedimentation. The flux due to molecular diffusion is given by $-D\nabla c$. Turbulent diffusion is assumed to be represented by $-D_e\nabla c$. They also assumed the average gas velocity very near the walls is negligible. Then the effect of gravitational sedimentation is accounted for by the term $v_{ts}\mathbf{k}\nabla c$, where $\mathbf{k}$ is the unit normal vector in the vertical direction. They also assumed that the concentration in the turbulent core to be uniform as is the models mentioned before. Under these assumptions the aerosol concentration in the boundary layer is governed by

$$\nabla[(D+D_e)\nabla n] - v_{ts}\mathbf{k}\nabla n = 0 \quad (3.59)$$

for $R-\delta < r < R$. The boundary condition on equation (3.59) are

$n = 0$ when $r = R$ (condition 1)

$n = n_0$ when $r = R-\delta$ (condition 2)

where n_0 is the bulk concentration. For the eddy diffusivity, they used Prandtl's mixing length expression, $D_e = k_e y^2$, near a wall. Where y denotes the distance from the wall and k_e may be evaluated from the turbulent energy dissipation rate. Hinze (1959) and Tchen (1947) had solved the equation of motion for a particle moving in a homogeneous turbulent flow field and had shown that in the long time limit, particle and fluid eddy diffusivities are equal.

With the assumption that $D \ll k_e \delta^2$, in polar coordinates, equation (3.59) becomes

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 (D+D_e) \frac{\partial c}{\partial r} \right) + \frac{(D+D_e)}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial c}{\partial \theta} \right) + v_{ts} \cos \theta \frac{\partial c}{\partial r} - \frac{v_{ts} \sin \theta}{r} \frac{\partial c}{\partial \theta} = 0 \quad (3.60)$$

With simplification and application of the Debye function, the deposition coefficient per unit volume can be calculated as

$$\beta = \frac{6\sqrt{k_e D}}{R} D_1 \left(\frac{\pi v_{ts}}{2\sqrt{k_e D}} \right) + \frac{v_{ts}}{4R/3} \quad (3.61)$$

where D_1 denotes the Debye function (Abramowitz and Stegun, 1964)

$$D_1(x) = \frac{1}{x} \int_0^x \frac{t dt}{e^t - 1} \quad (3.62)$$

where x is set as $\pi v_{ts} / 2(k_e D)^{1/2}$.

The second term of equation (3.61) corresponds to the gravitational sedimentation term since for a sphere $4R/3$ is the average height. However, first term of equation (3.61) also depends on v_{ts} , so this is not a purely diffusive contribution. Because the spherical vessel does not have a completely vertical deposition surface, the particles are always affected by gravitational sedimentation.

When x is much smaller than 1, because $D_1(x)$ is nearly equal to $\pi^2/6x$ for small x , β can be simplified.

$$\beta \approx \frac{2k_e D}{r v_{ts}} + \frac{v_{ts}}{4R/3} ; x \gg 1 \quad (3.63)$$

In the same manner, by choosing the x coordinates to lie along the inward unit normal to the surface, and the other two coordinates to be at a tangent to the surface, the deposition coefficient for arbitrary shape is given by

$$\beta = \frac{1}{V} \int_{\Sigma} \frac{v_{ts} \mathbf{n}(y) \cdot \mathbf{k} dA(y)}{\exp \left(\frac{\pi v_{ts} \mathbf{n}(y) \cdot \mathbf{k}}{2\sqrt{k_e D}} \right) - 1} \quad (3.64)$$

where Σ is the surface area, V is the volume of the vessel, $dA(x)$ is the differential area element on

Σ , k is the unit vector in the vertical direction and $n(y)$ is the unit outward normal to the surface. When equation (3.64) is applied to a cubic vessel with the side L , the Corner and Pendlebury's equation can be obtained.

The particular form of D_e is needed because Lin *et al* (1953) and Friedlander (1977) suggested that D_e is proportional to the 3rd power of the distance from the deposition surface in the viscous sublayer. In Crump and Seinfeld's model, when D_e is assumed to be proportional to the n th power of distance, the deposition coefficient is expressed by

$$\beta = \frac{1}{V} \int_{\Sigma} \frac{v_{ts} n(y) k dA(y)}{\exp \left(\frac{\pi v_{ts} n(y) k}{n \sin(\pi/n) \sqrt{k_e D^{n-1}}} \right) - 1} \quad (3.65)$$

Figure 3.10 shows equation (3.65) applied to the rectangular chamber with several different k_e . Crump *et al* (1983) used a spherical vessel and obtained an agreement between the theoretical formula of Crump and Seinfeld and the experimental results with $n = 2$.

In order to obtain the value of unknown parameter, k_e , Okuyama *et al* (1977) suggested that the average value of the energy dissipation rate, ϵ , is taken to be equal to the power consumption rate per unit mass of mixing fluid. In this approximation, k_e is considered to be proportional to the velocity gradient of fluid as $\kappa(du_x/dy)$ where κ is the Kármán's constant (≈ 0.4). The value of du_x/dy is evaluated from the energy dissipation rate per unit mass of fluid by Saffman and Turner (1956) as

$$\frac{du_x}{dy} = (2\epsilon/15\nu)^{0.5} \quad (3.66)$$

where ν is kinematic viscosity of fluid. Accordingly, turbulent diffusive deposition of particles depends on the velocity gradient evaluated from the energy dissipation rate per mass of fluid. In the case of a turbulent flow in a vessel, the value of the energy dissipation rate is approximated

Figure 3.10 Deposition Coefficient as a Function of Particle Size for Various Values of Turbulence Intensities, k_e . (Calculated from equation (3.65) with $n = 2$, and under the same condition as in Figure 3.9)

by the power consumption rate per unit mass of mixing fluid. If a fan is used to convect the air inside the chamber, ϵ is given by (Holub *et al*, 1988)

$$k_e = 0.4 \left(\frac{E}{8v} \right)^{0.5} ; E = 4 \times \text{RPS}^3 \times D_B^5 / V \quad (3.67)$$

where RPS is the revolution per second of fan and D_B is the diameter of the fan.

With equation (3.65), Okuyama *et al* (1986) found from their empirical data that the D_e is proportional to the 2.7th power of the distance ($n = 2.7$). Holub *et al* (1988) showed the agreement between experimental and theoretical values of deposition rates with using 2.6th power. According to the theoretical arguments, n should be an integer number either 2 or 3. Also the Crump and Seinfeld model is only valid when calculation is done with data in cgs unit. In order to remove the dimensional problem of the Crump and Seinfeld model, Benes and Holub (1996) proposed a new expression for the eddy diffusion coefficient based on the rules of dimensional analysis (Szűcs, 1980) which is

$$D_e = k_e \delta^2 \left(\frac{y}{\delta} \right)^n \quad (3.68)$$

where δ is the thickness of the turbulent boundary layer. With the eddy diffusion coefficient of equation (3.68), the deposition coefficient, β , is given by

$$\beta = \frac{S}{V} \frac{n}{\pi} \sin \left(\frac{n}{\pi} \right)^n \sqrt{k_e \delta^{2-n} D d_p^{n-1}} \quad (3.69)$$

However, this equation does not include the gravitational sedimentation factor and is good for only very small particles. When the sedimentation term from Crump and Seinfeld model is added to this formula, the applicable size range can be extended to micrometers. Figure 3.11 shows the plot of equation (3.69) with $n=2$.

Figure 3.11 Deposition Coefficient as a Function of Particle Size for Various Values of Turbulence Intensities, k_e . (Calculated from equation (3.69) with $n = 2$, and under the same condition as in Figure 3.9)

CHAPTER 4 Aerosol Generator

4.1. Introduction

Generation of aerosol particles is required for experimental applications. These applications may vary widely in the aerosol characteristics required and therefore, means of aerosol production that are consistent with the scientific investigation are needed. Because most of the properties of aerosol particles are strongly dependant on the particle size or particle size distribution, to generate aerosols with a specified size range is an important task. The characteristics of a good aerosol particle generator are that it is reproducible and provides constant output of monodisperse, stable, uncharged aerosol particles whose size and concentration can be easily controlled.

In this chapter, review of monodispersity and several aerosol generators, an orifice generator, a Vibration Orifice Aerosol Generator (VOAG) and a condensation aerosol generator which are used in this research work, are described.

4.2. Monodisperse Aerosol

The characteristics of the ultimate form of the aerosol particles produced by various means depend not only on the generation method, but also upon the physical and chemical nature of the source material and the temperature and humidity conditions or treatment received. On this basis, aerosols can be produced that are heterodisperse, polydisperse or monodisperse. The term heterodisperse is used to describe aerosols of particles which vary widely not only in size but also in physical and chemical characteristics. Polydisperse usually implies homogeneity of basic physical and chemical characteristics but with widely different particle sizes and differences in

those aerodynamic properties which depend in part upon particle size. Monodisperse is a term which implies both homogeneity of basic chemical and physical characteristics including composition and physical density and relative uniformity of particle size and aerodynamic properties. Generally the term “monodisperse” is relative term. A distribution of particles which is satisfactory monodisperse in one application may be considered as polydisperse in another. Fuchs and Sutugin (1966) suggested practical monodisperse aerosol size distribution which uses the degree of dispersity. The degree of dispersity can be characterized by the value of the relative standard deviation, α which is the ratio of the standard deviation of the particle radii, σ , to the mean radius r

$$\alpha = \frac{\sigma}{\bar{r}} = \left[\frac{\sum_i^N (r_i - \bar{r})^2}{N} \right]^{1/2} \times \frac{1}{\bar{r}} \quad (4.1)$$

Since the particle size distribution in aerosols usually does not follow the normal (Gaussian) distribution but approaches log-normal distribution, it is reasonable to express the degree of dispersity by the value of the standard deviation of the logarithms of the radii. This is given by

$$\ln \sigma_g = \left[\frac{\sum_i^N \left(\ln \frac{r_i}{r_g} \right)^2}{N} \right]^{1/2} \quad (4.2)$$

where r_g is the geometric mean of the particle radius. For small values of α ; $\ln \sigma_g \approx \alpha$. Their definition of monodisperse particle size distribution is one having a value of α less than 0.2. Nevertheless, a distribution of particles which extends from 1 to 5 μm and has a coefficient of variation much bigger than 0.2 may be clearly polydisperse to most, but a distribution that extends from 0.01 to 0.05 μm may be considered as practically monodisperse even though the degree of dispersity may be greater than 0.2.

In addition, the phenomenon being studied may affect the definition. Even though aerosol particles are numerically monodisperse with respect to geometric size, because of their variation of density, they could be polydisperse aerosol particles with respect to aerodynamic properties such as terminal settling velocity.

The extensive use of aerosols has led to the design of many types of aerosol generators which fall into basic groups which are compressed air atomizers, ultrasonic nebulizers, spray nozzles and dust blowers. In addition to these basic groups, there are specially designed aerosol generators in order to produce uniform droplets or particles such as condensation aerosol generators and photochemical aerosol generators.

4.3. Atomization

4.3.1. Basic Background of Atomization

Basically the atomization process is converting a liquid into small droplets. The generated droplets tend to be spherical because this shape has the smallest surface energy. Aerodynamic forces acting on the bulk liquid surface may promote the disruption process by applying disruptive force to the bulk liquid. A breakup occurs when the magnitude of the disruptive force just exceeds the consolidating surface tension force. Many of the larger droplets produced in the initial atomization process are unstable and undergo disintegration into smaller droplets. Thus, the final range of drop sizes produced in a spray depends not only on the drop sizes produced in primary atomization but also on the extent to which these droplets disintegrate during secondary atomization.

The most elementary form of atomization is the quasi-static case of the hanging or pendant drop. This is exemplified by the slow discharge of a liquid from the end of a burette or a dripping faucet. When the force of gravity on the liquid exceeds the force the droplet to the burette end, the surface tension, the liquid detaches and a free droplet forms. The mass of the droplet formed is determined by equating the gravitational and surface tension force on the hanging droplet. For

the slow emission of liquid from a thin circular tube of diameter d_0 , the mass of the droplet, m_d , formed is

$$m_d = \frac{\pi d_0 \sigma}{g} \quad (4.3)$$

where σ is the surface tension of the liquid. The size of the spherical droplet corresponding to this mass is given by

$$d_p = \left(\frac{6 d_0 \sigma}{\rho_L g} \right)^{1/3} \quad (4.4)$$

For a sharp edged opening 1 mm in diameter, equation (4) would predict droplet sizes of 3.6 mm for water. If the hole size were reduced to 10 μm in diameter, the predicted size for water would be 784 μm .

When atomization occurs as a result of interaction between a liquid and the surrounding air, the overall atomization process involves several interacting mechanisms, among which is the splitting up of the larger drops during the final stages of disintegration. It is of interest, therefore, to examine the various ways in which a single drop of liquid can break up under the action of aerodynamic forces. In order to obtain a strict formula for the breakup of a droplet, exact knowledge of the distribution of aerodynamic forces on the droplet is needed. However, every time droplet changes its shape by under those forces, the pressure distribution around it also changes. Klusener (1933) examined the influence of the air pressure distribution change around the droplet. Under equilibrium conditions, the internal pressure at any point on the droplet surface, p_i , is just sufficient to balance the external aerodynamic pressure, p_A , and the surface tension, p_σ , so that

$$p_i = p_A + p_\sigma = \text{constant} \quad (4.5)$$

for a spherical droplet

$$p_{\sigma} = \frac{4\sigma}{d_p} \quad (4.6)$$

Equations (4.5) and (4.6) indicate that a droplet can remain stable as long as a change in air pressure at any point on its surface can be compensated by a corresponding change in p_{σ} such that p_i remains constant. However, if p_A is large compared with p_{σ} , then any appreciable change in p_A can not be compensated by a corresponding change in p_{σ} to maintain p_i constant. In this situation, the external pressure p_A may deform the drop to an extent that leads to further reduction in p_{σ} and finally to the disruption of the droplets into smaller ones (Griffen and Muraszew, 1953). For these smaller droplets, the higher value of p_{σ} as indicated by equation (4.6) may be large enough to accommodate the variations in p_A . If not, further subdivision may occur until p_{σ} is large enough to maintain a constant value of p_i at all points on the drop surface. When this stage is reached, the drop is stable and no further breakup can occur.

4.3.2. Factors Influencing Atomization

The performance of any given type of atomizer depends on its size and geometry and on the physical properties of the dispersed phase which is the liquid being atomized and the continuous phase which is the gaseous medium into which the droplets are dispersed.

The flow and spray characteristics of most atomizers are strongly influenced by liquid properties such as density, viscosity, and surface tension. A spray can be characterized by the surface area change resulting from atomization. The surface area before a breakup is very simple. However, after atomization, the area is the sum of the surface areas of all the individual droplets. This multiplication factor provides a direct indication of the level of atomization achieved and is useful in applications that emphasize surface phenomena such as evaporation and absorption. Surface tension is also important in atomization because it represents the force that resists the

formation of new surface area. The minimum energy required for atomization is equal to the surface tension multiplied by the increase in liquid surface area. Whenever atomization occurs under conditions where surface tension forces are important, the Weber number, which is the ratio of the inertial force to the surface tension force, is a useful parameter for correlating droplet size data.

Viscosity is also an important factor to influence atomization. Even though its influence on atomization is not as significant as that of surface tension, it affects not only on the droplet size distribution but also the nozzle flow rate and the spray pattern. An increase in viscosity lowers the Reynolds number and also hinders the disintegration and produce a significant increase in the size of the droplets.

4.4. Liquid Aerosol Generators

Liquid aerosol generators can be classified by the method of particle formation which is achieved either by mechanical dispersion of the liquid into droplets, or through condensation of a vapor into droplets. In all condensation aerosol generators, the basic principal is the same: the liquid to be aerosolized is first heated to form a vapor, then the vapor is cooled and condensed into droplets. However, there are several different techniques which have been used to mechanically disperse liquids to form aerosols. Some of the more common methods, including air nebulizers, spinning-disk atomizers, ultrasonic nebulizers, and vibrating orifice generators, are introduced in this section.

4.4.1. Simple Air Nebulizers

The Devilbiss nebulizer (Kerker, 1975) is one example of this type of nebulizers. Air passes through a narrow tube. The high velocity air emerging from this tube creates a low pressure region at the tube exit, drawing liquid from the reservoir upward through a liquid feed tube. The emitted liquid is atomized by the high velocity air into small droplets.

Another common aerosol nebulizer is the Collison nebulizer (Fig 4.1) which was invented by W. E. Collison (1935). This type of nebulizer has been developed and used mainly in medical usage (Rosebury 1947, Henderson 1952, Druett 1959, 1969).

Figure 4.1 Collison nebulizer (a) and the magnification of Nebulizing head (b)

Details of its design and operating characteristics were given, including air and liquid consumption, aerosol output rate and droplet size distribution by May (1973).

The original design had a baffle in front of the nebulizing jet in order to remove the larger

droplets from the generated spray by impaction. The removed liquid flows back to the bulk liquid and is resprayed. The spray fluid is pulled up through a feed tube to a nozzle by a pressure drop created by a high velocity air jet and is atomized by the air jet. The diameter of the jet is 0.35 mm and three jets models are often used. In this nebulizer, during the time between formation of drops and their escape from the aerosol outlet at the top of the nebulizer, a period which averages about 3 seconds in the three jet model, the droplets come to equilibrium with the air and their evaporation cools it. For pure water, aqueous suspensions or dilute aqueous solutions, saturation of the air by evaporation from the droplets is complete and the fall in temperature is almost entirely due to this evaporation. The temperature fall from the adiabatic expansion of the compressed air is largely absorbed by the thermal energy released by breakup and impact of the liquid. The temperature in the vicinity of a head spraying water is about 6°C below ambient but by the time the aerosol emerges its temperature initially rises by exchange with the ambient air via the hardware of the nebulizer to about 3°C below ambient. Because of the heat capacity of the system further temperature changes are slow and after about 30 minutes of operation the aerosol and hardware reach an equilibrium at about 5°C below the ambient. When solutions of any substance which lowers the vapor pressure are sprayed the relative humidity and temperature of the emerging aerosol will depend on the nature of the solute and its concentration. With volatile liquids, no significant change in temperature can be expected.

In order to obtain monodisperse aerosols of small and uniform particle size, a very dilute solution should be used. In many cases, polystyrene latex spheres which can be obtained from carefully graded monodisperse suspensions or bacterial cells are used for laboratory preparation of monodisperse particles. The dilution must be such that the chance of any droplet containing more than one particle is small. Henderson (1952) suggested that the number concentration of cell suspension should be kept below 10^9 ml^{-1} , when 95 per cent of the resulting dried aerosol particles consist of single cells. This concentration is equivalent to an average of 1 cell per $12.5 \mu\text{m}$ drop and is consistent with the experimental data (May, 1973). The upper limit of droplet size is

relatively larger than is generally realized, around $43\mu\text{m}$ over the pressure range of 15 and 50 psi. However, those large droplets are infrequent in comparison with the number of average sized droplets.

4.4.2. Spinning disk atomizers

In this type of aerosol generators, the liquid is atomized by being thrown from the edge of a spinning disk (Liu, 1967). The relationship between the size of droplets atomized from this generator and density of the liquid, ρ , the diameter of spinning disk, D , angular velocity, ω , is given by

$$d\omega(D\rho/\gamma)^{1/2} = K \quad (4.7)$$

Walton and Prewett (1949) obtained the $K=3.8$ empirically.

4.4.3. Ultrasonic Nebulizers

The liquid to be atomized is fed through or over a transducer and horn, which vibrates at ultrasonic frequencies to produce the short wavelengths necessary for fine atomization. The system requires a high frequency electrical input. Two piezoelectric transducers, and a stepped horn. The concept is well suited for applications that require very fine atomization and a low spray velocity. At present, an important application of ultrasonic atomizers is for medical inhalation therapy, where very fine sprays and the absence of gas to affect atomization are important attributes.

4.4.4. Vibrating Orifice Aerosol Generator

Vibrating Orifice Aerosol Generator (VOAG) was introduced by Berglund and Liu (1973) and Wedding and Stuckel (1973). This type of generator is normally used to generate

monodisperse aerosols in the size range of 0.5 to 50 μm . In the vibrating orifice aerosol generator, the liquid is forced through an orifice (5-20 μm in diameter) at a constant flow rate with a pump, as shown Figure 4.2. The liquid stream passing through such an orifice becomes unstable and breaks up into droplets. By vibrating the orifice, the stream is broken into uniform droplets, with one droplet formed for each cycle. Thus, if the flow rate and cycle frequency are kept constant, the droplets should all be of the same size, with a very small geometric standard

Figure 4.2 Vibrating Orifice Aerosol Generator

deviation. The size of droplets generated by VOAG can be calculated directly from two parameters which are liquid flow and oscillating frequency

$$d_p = \left(\frac{6Q_L}{\pi f} \right)^{1/3} \quad (4.8)$$

where Q_L is a liquid flow rate in $\text{cm}^3 \text{s}^{-1}$ and f the frequency in Hz. Although the generated particles are monodisperse, the coagulation of particles immediately after dispersion can lead to doublets and triplets resulting in lowered monodispersity of the generated particles. To reduce this problem, dispersion air is normally added to the aerosol stream immediately after generation. The dispersion air flow rate is sufficiently high to produce turbulence near the aerosol stream, thus efficiency diluting and dispersing the particle before doublets can be formed. Any kind of low viscosity liquid which does not contain impurities that might clog the vibrating orifice can be used. In order to generate smaller solid monodisperse particles, volatile solvents containing dissolved solutes are used for VOAG. In this case, immediately after the generation of droplets, they flow through a dryer and the volatile solution is removed from the droplets.

If a liquid with high dielectric constant is used, droplets may be charged by electrolytic charging when the liquid is separated from the VOAG. During atomization, these droplets strip charge from the surface of the VOAG and produce moderately charged droplets as they separated from these surfaces. Pure water is a high dielectric liquid that can become charged during atomization. Because of that, in many cases, a neutralizer is used directly after the droplets' formation.

4.4.5. Condensation Generators

In condensation generators, the liquid or solid to be generated is first heated to form a vapor, and then the vapor is cooled and condensed into droplets. Upon cooling, the vapor condenses

homogeneously if the supersaturation is sufficiently high, or the vapor condenses onto nucleation centers introduced into vapor. The aerosol particles generated by this method are normally fairly monodisperse. One of the earliest practical generators of this type was the Sinclair-LaMer condensation generator (Sinclair and LaMer 1941, 1949; LaMer 1951, 1952; LaMer and Hochberg, 1949). Later that further research had been done by Matijevic *et al* (1962), Muir (1966), Swift (1967), Okuda *et al* (1969), Prodi (1970, 1972), and Prodi and Moelter (1986). In the earlier development of these generators, the liquid was vaporized in a boiler, and the nuclei were generated by an electric arc. The nuclei and the liquid vapor were then passed into a second chamber where the mixture was reheated to ensure that any residual droplet from the boiler would be evaporated. The nuclei centers and the reheated vapor were then passed through a cooling tower where the vapor condensed upon the nucleation centers.

In Matijevic system, they heated AgCl to between 404°C and 645°C and used it as the condensation nuclei. Helium was used as the carrier gas. In their experiment with linolenic acid and octanoic acid, the particles sized from 0.6 μm to 2.0 μm in diameter (geometric standard deviation $\sigma_g = 1.26 - 1.58$) and were generated with good stability. In this system, the flow rate of the bubbler affects both the concentration and size range of the particles. The particle size becomes smaller and monodispersity becomes lower as flow rate increases.

Prodi's method is to atomize the liquid, evaporate the droplets and then recondense the vapor onto the nonvolatile nuclei of the evaporated droplets. A schematic of one such generator is shown in Figure 3. Here, liquid atomization is achieved through a Collison atomizer fed with a solution or suspension, followed by a silica gel diffusion dryer. The carrier gas is nitrogen because the oxygen impurities in particular must be low in order to avoid the thermal degradation of the particle materials, carnauba wax, caused by the presence of oxygen in the gas stream. The aerosol is subsequently vaporized as it passes through a glass tube which is wrapped with electric heating tape. The vapor and the residue of the evaporated droplets then move downward into the unheated portion of the glass tube, where the vapor recondenses onto the residue particles. In this

Figure 4.3 Condensation Aerosol Generator

system, the generated particles were sized between $0.3\ \mu\text{m}$ and $2.2\ \mu\text{m}$ with geometric standard deviation of less than 1.2. This system is a very effective aerosol generator for submicron particles.

The upper 25 cm of the reheater column is heated by a heating tape and the lower part is used for the cooling process. Down flow is used in the reheater to improve thermal stability and avoid reevaporation and eddy formation which results in condensation onto the glass wall (Liu *et al*, 1966). When the vapor-nuclei mixture enters the reheating column from the bubbling system, the vapor is unsaturated. At the bottom part of heating tape, vapor-nuclei mixture is gradually cooled down under the condition of laminar flow so that the gas temperature at the center of the reheating column is higher than at the wall. At a certain point, the temperature at the wall reaches the dew point and the vapor begins to condense on them. Because of this, the vapor concentration near the wall becomes lower than in the center.

If the concentration of condensation nuclei is too low or the flow rate inside the reheating column is too low, most of the vapor condenses on the wall surface because at this point, the degree of supersaturation is not high enough for spontaneous condensation in bulk space. If the concentration of the condensation nuclei increases, the ratio of the amounts of vapor which condenses in bulk space to the one which condenses on the wall rises dramatically. Therefore, even though the mean volume of the droplets formed decreases, the product of (concentration of nuclei) $\times$ (the mean volume of droplets) increases. Moreover, the size of the droplets increases with increasing vapor concentration if the concentration of nuclei remains the same. This process is equivalent to raising temperature of the bubbling system. In other words, this allows the particle size to be controlled within certain limits by changing the temperature of the oil bath which governs the bubbling temperature. However, it should be noted that the control of particle size by varying the number concentration of nuclei is possible only within certain limits because very high concentration can not be attained due to their coagulation, whereas too low a concentration leads to homogeneous nucleation. The temperature of the reheater has little effect

on the particle size.

For the generation of small aerosol particles, high boiling substances with a large heat of evaporation should be used in order to obtain a very high degree of supersaturation.

High monodispersity requires a number of condition be attained. As mentioned before, the size of generated particles is very sensitive to the bubbling temperature because it affects the amount of vapor inside the reheating system. The temperature of bubbler should be kept constant. Otherwise, the aerosol particle size will continually change. At least several hours are needed to stabilize the bubbling system temperature.

The purity of the evaporating liquid is of great importance. Volatile impurities should be removed from the bubbling system before it is used because with their gradual removal from the bubbling system, the particle size decreases. If the generator is working for a long time, because the bubbled liquid is continuously heated, the liquid gradually decomposes, especially when air is bubbled through it. This results in the walls of bubbler and the reheater becoming covered with a film of the decomposition products and the monodispersity of the aerosol is impaired (LaMer, 1952). In an atmosphere of inert gas, decomposition can be effectively prevented.

CHAPTER 5 Experimental Results

5.1. Aerosol Generator

For this research, several types of aerosol generators were used because of their different characteristics. Figure 5.1 shows the generated particle size and aerosol generators used.

- A: Gas Burner Particles
- B: Sodium Chloride Particles
- C: Carnauba Wax Particles
- D: Oil Droplets
- E: Fluorescein Particles

Figure 5.1 Particle Materials Used in This Research

5.1.1. Gas Burner Particles

Natural gas and propane gas are the major source of the very fine particles (< 30 nm) in

normal residences (Li and Hopke, 1993). Olawoyin (1995) generated the fine particles from the gas burner in a barrel. Similarly in this research, a gas burner was placed on the bottom of a 55 gallon drum. Generated aerosol particles are subsequently sampled and sized with a Scanning Mobility Particle Spectrometer (SMPS) which is the combination of a Differential Mobility Analyzer (DMA) and an Ultrafine Condensation Particle Counter (UCPC).

Figure 5.2 Gas Burner Particle Generator

The DMA used in this research is TSI Model 3071. From equation (2.54), the velocity of the charged particles can be determined when the particle diameter, the number of charges and the strength of the electric field is known. The velocity of the particles can be controlled by changing the flow rate of the sheath air in the DMA. The power supply controls the strength of the electric field inside the tube. Therefore, if the number of charge on the particles is known, the particle size can be calculated from the electrical mobility.

This analyzer was originally designed by Knutson and Whitby (1975). Figure 5.3 shows the schematic diagram of the DMA used.

Figure 5.3 Cross -sectional View of DMA

Polydisperse aerosol particles are passed through a neutralizer. In the neutralizer, particles are exposed to bipolar ions. The particles and ions undergo continual collisions due to the random thermal motion of the ions. The particle reaches an equilibrium state in accordance with Boltzmann's law. The charged particles enter the outer layer of the column, while the particle-free sheath air forms the inner layer of the column. The aerosol and sheath air are laminar and do not mix. The inner cylinder has an electrical potential controlled by the power supply. The outer cylinder is electrically grounded. The trajectory of the particles is bent by their charge and electrical field. Only particles with electrical mobilities within a narrow range are able to follow the trajectory which leads to an output slit of inner cylinder. If the particles have an electrical mobility larger than the range, those particles hit the inner cylinder before the output slit. Particles with smaller electrical mobilities will tend to keep their original direction and pass through the output slit.

The DMA can classify the particles in the range of 0.005 to 1.0 μm . When the particles are smaller than 0.01 μm , the concentration of monodisperse particle obtained may be unstable because the ratio of the uncharged to charged particle becomes large.

The Condensation Particle Counter (CPC) was originally invented by Pollak and Nolan (1946). The equipment used in this research is TSI Model 3025. This type is called a continuous flow condensation nuclei counter. It was developed by Bricard *et al* (1976) and Agarwal and Sem (1980). A schematic diagram is shown in Figure 5.4. A supersaturation of *n*-butyl alcohol is achieved by saturating the gas stream in the saturator which is maintained at 35°C. The condenser tube is kept at 10°C. The aerosol particles are passed through the saturator first and then cooled in the condenser tube. In the condenser tube, particles grow by heterogeneous nucleation and are measured by light scattering. The use of alcohol condensation favors the detection of very small oil and combustion particles that are hydrophobic to water. The instruments has a flow rate of 5 $\text{cm}^3 \text{s}^{-1}$ and a concentration range of 0.01 to 9.99×10^{-4} particles cm^{-3} .

Figure 5.4 Schematic Diagram of UCPC

The measured size distribution of the particles produced by the gas burner is shown in Figure 5.5. The distribution was measured with the SMPS operated at 3 LPM for sheath and excess air and 0.3 LPM for monodisperse aerosol flow. In Figure 5.5, the number size distribution is given in terms of fractional number per unit $\ln(d_p)$. It is log-normal with a geometric mean of 14 nm and geometric standard deviation, σ_g , of 1.57.

Figure 5.5 The Size Distribution of the Gas Burner Particles

5.1.2. Sodium Chloride Particles

The Collison Atomizer (Fig 4.1) was used for generating sodium chloride particles. Filtered nitrogen was used as a carrier gas with the air pressure of 738 Pa. The generated particles were passed through a heating tube which was kept 40°C and they passed through a diffusion dryer. After removal of the water, the size distribution was measured by the SMPS which was described

in the previous section. The setups of the SMPS was the same as the gas burner particle measurement. Figure 5.6 shows the size distribution of the sodium chloride particles from several deferent concentration of NaCl/H₂O solution.

Figure 5.6 Size Distributions of Sodium Chloride Particles generated by the Collison Atomizer.

(a) 10 mg L⁻¹, (b) 20 mg L⁻¹, (c) 50 mg L⁻¹, and (d) 100 mg L⁻¹.

Because the concentration of sodium chloride solved in each droplet from the Collison atomizer is the same, when the droplet size is uniform, the formula relating the solution concentration and the particle size can be written as

$$d_p = A \times c^{1/3} \quad (5.1)$$

where A is the constant of proportionality and c is the concentration of the solution. From the peak diameter of each size distribution in Figure 5.6, Figure 5.7 can be obtained.

Figure 5.7 Sodium Chloride Particle Size Plot against the Solution Concentration. The line shows the regression of the four points.

The sodium chloride particle sizes generated by the Collison atomizer were very stable and reproducible. The geometric standard deviation was between 1.37 and 1.54. In this experiment,

even though the Collison atomizer was immersed in ice water in order to prevent evaporation of the solvent, the lack of monodispersity can still be considered caused by a change in concentration during the atomization.

5.1.3 Carnauba Wax Particles

Carnauba wax is a natural wax. The main components are $\text{CH}_3(\text{CH}_2)_{24}\text{COO}(\text{CH}_2)_{29}\text{CH}_3$, several fatty acids and alcohols. Its melting point is $82 \sim 85.5^\circ\text{C}$. For generation of carnauba wax particles, the condensation type generator (Figure 4.3) was used.

Figure 5.8 shows a schematic diagram of the bubbling system and reheating system.

Figure 5.8 Illustration of Bubbler (a) and Reheater (b)

The temperature of the bubbler was maintained by the temperature controller (OMEGA, Model CN2002) which monitors the temperature of the bubbler through the RTD (OMEGA

PRX-NP 100) and controls the output voltage for the oil bath in which the bubbler is immersed. The reheating temperature is controlled by a similar RTD switching the power to heating tape.

As the condensation nuclei, sodium chloride particles are used. The concentration of the solution was 20 mg L^{-1} . The droplets leaving the atomizer section were dried by passing them through a diffusion drier tube containing silica gel desiccant. The gas flow was subsequently passed into the bubbler. The depth of the bubbling tube in the wax was maintained at $5.0 \text{ cm} \pm 1.0 \text{ cm}$ because Horton *et al* (1991) suggested that the ability of the gas stream entering the bubbler to collect wax vapor depends on the residence time of the gas in contact with the liquid wax, and this parameter was controlled by the mass of the liquid in the bubbler. The amount of bubbling flow was maintained at 24 L h^{-1} . The resulting gas-nuclei-wax vapor mixture was transferred via a glass tube at room temperature to the reheater. The inner diameter and the length of the reheater is about 2.5 cm and 240 cm, respectively. The first 25 cm of reheater is covered by the heating tape. In order to vaporize the wax completely, the reheater temperature was kept over 200°C . To measure the size distribution, the SMPS system which is mentioned in the section 5.1.1, was used.

Prodi (1972) and Scheuch and Heyder (1986) produced di-2-ethyl hexyl sebacate (DEHS) particles and showed the importance of the reheater temperature. Hence, a series of experiments was carried out to explore the effect of the reheater temperature on the quality of the carnauba wax aerosols. Aerosols were produced at bubbler temperature of 170°C . Figure 5.9 and 5.10 shows the size distribution of the wax particles and geometric standard deviation, σ_g , with different reheating temperatures. The reheater temperature was measured by locating the RTD axially inside the tube so that the tip was positioned in the hottest region. σ_g was obtained after stable conditions were reached at each reheater temperature. From Figure 5.9, the most important property is the remarkable decrease in σ_g as the reheater temperature was raised above 300°C . The poor monodispersity at low reheater temperature results from the problem of premature condensation along with the incomplete vaporization of the carnauba wax liquid. This result

Figure 5.9 Size Distribution of Carnauba Wax Particles with Different Reheater Temperature.

(a) 200°C, (b) 250°C, (c) 300°C, (d) 350°C, (e) 400°C, (f) 500°C.

Figure 5.10 Effect of the Reheater Temperature on the Geometric Standard Deviation, σ_g , of the Carnauba Wax Particles

agrees with previous research (Horton *et al*, 1991) very well. They used DEHS particles instead of the carnauba wax and obtained monodisperse particles above a reheater temperature of 200°C.

In this study, the bubbling flow was kept at 24 L h⁻¹ and this is equivalent to the constant concentration of the condensation nuclei in the system. While the concentration of the nuclei remains the same, the size of the carnauba wax particles increases with increasing vapor concentration. Because the temperature of the bubbler governs the vapor concentration, by changing the temperature the final size of the carnauba wax particles can be controlled.

The carnauba wax particles generated at a range of different bubbler temperature were

Figure 5.11 Effect of Bubbler Temperature on the Final Wax Particle Size

measured. A reheater temperature was maintained at 350°C through out a series of experiments. Figure 5.11 gives the result. The dotted line in the graph is the third order regression with a correlation coefficient of 0.998. The particle diameter increased with increasing bubbler temperature, as more carnauba wax vapor was produced from the liquid phase. This results are consistent with the previous research (Swift, 1967; Prodi, 1972; Prodi and Moelter, 1986; Horton *et al*, 1991). For carnauba wax particle size; the dependance on the bubbling temperature can be well approximated by a third order regression in the temperature range from 110 to 170 °C.

Figure 5.12 The Bubbling Temperature Fluctuation When the Temperature Controller is Set at 155°C. The Line in the Middle Shows the Average Temperature. Upper and Lower Line Indicates the Average $\pm$ Standard Deviation.

During the aerosol generation the bubbler temperature should be kept constant because of the dramatic effect on the final particle size. In this research, a temperature fluctuation was noticed even though temperature was carefully maintained. The temperature fluctuation was shown in Figure 5.11. This measurement started about 75 minutes after when the reading of the temperature reached its designed stable state. The temperature controller was set at 155°C but the average temperature for 100 minutes test was 155.33°C and the highest and lowest temperature during this test were 157.1°C and 153°C. This temperature fluctuation is large enough to result in the poor monodispersity and unstable particle output.

5.1.4 Oil Particles

For oil particles, vegetable oil was used because of its non-toxicity and easy preparation. Figure 5.13 is the cross sectional view of the orifice atomizer used for oil particles. The oil is pulled up through the tube to the liquid chamber because of the pressure difference and the liquid surface is disturbed by a high velocity air stream coming through the orifice. This

Figure 5.13 Cross-sectional View of the Orifice Atomizer

disturbance makes droplets from the bulk liquid. The carrier gas was filtered dry air. The atomizing pressure was maintained at 1291 Pa by the pressure regulator placed just before the atomizer orifice. Large droplets hit the wall on the opposite side of the orifice because of impaction and flow back down to the liquid chamber. Excess oil in the liquid chamber flows back to the oil jar, ready to be resprayed. Because the generated particles were polydisperse, those droplets are classified into the desired size range by the DMA. This system was operated at room

temperature.

5.1.5. Fluorescein Particles

In this study, in order to generate the larger particles, fluorescein (Aldrich Chemical) was used. Figure 5.14 is the structure of the fluorescein.

Fluorescein particles were generated by the Vibrating Orifice Aerosol Generator (VOAG, Figure 4.2). Figure 5.15 shows the experimental setup for sampling the fluorescein particles. Fluorescein powder was dissolved into the mixture of equal amounts of water and ethanol and stirred overnight in the room temperature. All the air applied to this experiment was filtered

Figure 5.14 Structure of the Fluorescein Molecule

Figure 5.15 Experimental Setup for the Sampling of Fluorescein Particles

by molecular sieve before entering the experimental system. The dispersion and the dilution air flows are 24 L h^{-1} and 48 L min^{-1} , respectively. The compressed air pressure was maintained with the pressure regulator and the compressed air pressure kept the liquid flow rate at 0.1 ml min^{-1} . The frequency of the vibration orifice was 0.55 MHz . Generated particles were passed through

Figure 5.16 Size Distribution of the Fluorescein Particles from the Solutions Concentration 0.125 g L^{-1} (Left) and 1.25 g L^{-1} (Right). For each Experiment, 200 Particles' Sizes were Measured with the Optical Microscope.

a neutralizer and the diffusion dryer and were captured on the filter. The captured particles were examined with the optical microscope. Because of their yellow-orange color, those particles can be easily observed and counted.

Figure 5.16 is the size distribution of the fluorescein particles from 0.125 g L^{-1} and 1.25 g L^{-1} solution. Sampling time was 5 minutes. From the equation (4.8), the expected particle diameter is $0.89 \mu\text{m}$ for 0.125 g L^{-1} solution and $1.92 \mu\text{m}$ for 1.25 g L^{-1} . The empirical data, from the Figure 5.16, shows that the particles sized between 0.5 and $0.75 \mu\text{m}$ for 0.125 g L^{-1} and 1.0 and $1.5 \mu\text{m}$ for 1.25 g L^{-1} were most observed. This is relatively good agreement of theoretical and experimental data.

5.2. Deposition Experiment

Figure 5.17 shows the experimental setup.

Figure 5.17 Experimental Setup for Deposition Study

The dimensions of the rectangular chamber used in this research was $75 \times 75 \times 180 \text{ cm}^3$. The surface volume ratio, S/V , was $1.125 \times 10^4 \text{ cm}^{-1}$. This chamber had an air inlet and a sampling outlet on the ceiling. There were two outlets. One was the main exhaust which is connected to a flow meter and a vacuum pump. The other outlet was used in order to keep the inside pressure constant. An electric fan and a sampling probe were inside the chamber.

5.2.1. Air Exchange Rate of the Chamber

In order to measure the air exchange rate, radon gas was used in this research. At first, the chamber was filled with radon gas. The radon concentration was measured with a diffusion cell which was placed in the middle of the chamber, and then, the dry air line and the exhaust line were opened. The flow rate of the dry air kept higher than the one of the exhaust to keep inside the chamber slightly over pressure. The dry air flow and the exhaust flow were controlled by a mass flow controller and the orifice valve, respectively. An electric fan was running during this procedure. Figure 5.18 shows the results of the concentration decay of radon inside the chamber.

Figure 5.18 Concentration Decay of Radon inside the Chamber

From the slope of the regression of the Figure 5.18, the air exchange rate can be obtained. The results and flow rate of the dry air and the exhaust air are shown in Table 5.1.

Table 5.1 Flow Rate of the Dry Air and Exhaust

Dry Air Flow	Exhaust Air	Air Exchange
(LPM)	Flow (LPM)	Rate (h⁻¹)
32.0	19.2	0.8
78.0	62.0	1.7
100.0	78.5	2.4

5.2.2 Particle Loss Rate Measurement

In this study, the particle loss rate was measured by knowing the concentration decay of the particles inside the chamber. At first, from the aerosol generating system, the aerosol particles were injected into the chamber until the particle concentration reached high enough to be measured the concentration decay inside the chamber. The particle concentration was monitored by the CPC (for the particles $< 1 \mu\text{m}$) or an Optical Particle Counter (OPC, for the particles $> 1 \mu\text{m}$). During this procedure, the electric fan was running to ensure the complete mixing. When the desired particle concentration was achieved, the line from the aerosol generating system was closed and the electric fan was turned off. Subsequently, the vacuum line and the dry air line were opened. The particles were sampled and the concentration was recorded in the computer system. The concentration change during the entire operation is shown in Figure 5.19. When the aerosol generator started, the elapsed time was set to zero. The aerosol generator line was closed as soon as the particle number exceed 20,000 and simultaneously, the dry air line and the vacuum line were opened. The concentration declined logarithmically due to the air ventilation and the wall deposition.

Figure 5.19 Concentration Change during the Entire Operation. (80 nm Carnauba Wax Particles were used. The Air Exchange Rate = 0.8 h^{-1})

5.2.3. Method of Determination Particle Loss Rate

The mass balance expression for the particle concentration as a function of time is

$$\frac{dn}{dt} = -\lambda_v n - \lambda_d n \quad (5.2)$$

where n is the number concentration, t is the elapsed time, λ_v is the particle loss rate due to the

air exchange and λ_d is the particle loss rate due to the deposition. It is assumed that the wall deposition is a first-order process and there was no homogeneous production or decay of the particles. If the variables other than n are constant for a given situation, the solution to Equation (5.2) is

$$n(t) = n_0 \exp[-(\lambda_v + \lambda_d)t] \quad (5.3)$$

where n_0 is the initial concentration. Because the air exchange rate is known, the particle loss rate resulted from deposition can be calculated from the concentration decay with equation (5.3).

5.2.4. Experimental Results and Discussion of Particle Deposition in the Chamber

Figure 5.19 shows the particle concentration decay of each materials. The triangle symbols, ($\blacktriangle$), square symbols, ($\blacksquare$), and circle symbols, ($\bullet$) indicate the air exchange rate of 0.8 h^{-1} , 1.7 h^{-1} and 2.4 h^{-1} , respectively.

The gas burner particles were directly injected from the oil drum (Figure 5.2). The flow rate of the compressed air was 1 LPM and this air was pre-filtered by the molecular sieves and an air filter. The temperature of the air from the oil drum was higher than the room temperature but the temperature inside the deposition chamber did not change.

For the sodium chloride particles, the Collison atomizer was used (Figure 4.1). The NaCl/ H_2O solution concentration was 50 mg L^{-1} for 30 nm particles in diameter and 400 mg L^{-1} for 60 nm particles. The atomizing pressure was 738 Pa and the carrier gas was nitrogen. The operating conditions were described in section 5.1.2. After the sodium chloride particles were passed through the diffusion dryer, these particles were classified into desired size by the DMA and injected in the chamber.

The condensation aerosol generator (Figure 4.3 and 5.8) was used for carnauba wax particles. The particles were injected into the chamber directly after they passed through the reheating

Figure 5.20 Concentration Decay of the Particles of Each Material. (a) gas burner, (b) Sodium Chloride particle (30 nm), (c) Carnuba Wax Particle (80 nm), (d) Oil Particle (300 nm), (e) Fluorescein Particle (2.0 μm). Air Exchange Rate; $\blacktriangle = 0.8\text{h}^{-1}$, $\blacksquare = 1.7\text{h}^{-1}$, and $\bullet = 2.4\text{h}^{-1}$

column. The bubbling flow rate was 24 L h^{-1} . The bubbling temperatures were 155°C for 80 nm particles and 168°C for 103 nm particles. During the particle generation, the temperature fluctuation was $\pm 0.7^\circ\text{C}$. The reheater temperature was 350°C .

After the generation of the oil particles by the orifice atomizer (Figure 5.13), they were sorted into several different sizes by the DMA. The operating condition was described in section 5.1.4. For this research, the oil particles with 200, 300 and 400 nm in diameter were produced by arranging the voltage of the DMA.

Fluorescein particles were generated by the VOAG (Figure 4.2). The operating condition was the same as in the size distribution measurement as described in 5.1.5 except the frequency of the vibrating orifice and the concentration of the fluorescein solution. The frequency and the solution concentration used in the particle loss experiment were 5.0 MHz and 0.16 g L^{-1} . The size distribution of the particles were measured with the optical microscope.

The concentrations of the gas burner particles, the sodium chloride particles, the carnauba wax particles and the oil particles were measured by the UCPC. The fluorescein particle concentration was examined by the OPC.

From Figure 5.20, the plots fit well to a linear regression line for all size ranges. These results indicate the validity of the assumption that the particle loss is the first-order process and there is no redispersion of particles into the bulk region of the chamber. For all graphs in Figure 5.20, the slope increases with increasing air exchange rate. Because the slope determines the particle loss rate from equation (5.2), this means that when the air exchange rate is larger, the more particles are removed from the chamber. The deposition coefficients, the particle loss rate due to the wall deposition, for several particle sizes are given in Figure 5.21. The deposition coefficient also increases with the air exchange rate of the chamber. The particles are more efficiently removed with higher air exchange rate because of the effect of not only the ventilation but also the increase of the contribution of the wall deposition inside the chamber. In both Crump and Seinfeld model (Figure 3.10) and Benes and Holub model (Figure 3.11), the increase of the deposition coefficient

Figure 5.21 Deposition Coefficient and Air Exchange Rate of the Chamber.

is equivalent to the increase of the turbulent intensity, k_e , which determines the eddy diffusion coefficient. From the curve fit of these two models the value of k_e can be approximated. Figure 5.22 shows the comparison of the experimental data and the both the Crump and Seinfeld and the

Figure 5.22 Comparison of Experimental Data and the Models (a) Air Exchange Rate = 0.8 h^{-1} ,
(b) Air Exchange Rate = 1.7 h^{-1} , (c) Air Exchange Rate = 2.4 h^{-1}

Benes and Holub models. According to the prior research, two values for n in equation (3.65) are suggested by Okuyama *et al* (1986) and Holub *et al* (1988) for the Crump and Seinfeld model. In Figure 5.22 $n = 2.6$ is used.

As can be seen in Figure 5.22, Benes and Holub model fits the experimental data at least as well as the Crump and Seinfeld model. The submicron particles have larger deposition coefficients and this phenomena was observed in charged particles (McMurry and Rader, 1985). Best fits to k_d were obtained by the regression. Those parameters are shown in Table 5.2 and Figure 5.22.

Table 5.2 The Turbulent Intensity Value for the Theoretical Models

Air Exchange Rate (h^{-1})	Crump and Seinfeld (S^{-1})	Benes and Holub (S^{-1})
0.8	4.5	1.66
1.7	18.32	8.32
2.4	37.1	13.75

Figure 5.23 The Change of k_d Value with the Air Exchange Rate

From Figure 5.21, k_e changes linearly for Benes and Holub model. In the Crump and Seinfeld model, the slope is steeper and not linear. Crump *et al* (1983) suggested that in the tube, value of k_e is proportional to the $2/3$ power of the flow rate. In this rectangular chamber, k_e is proportional to the air exchange rate.

CHAPTER 6 Conclusion

In order to study particle deposition, monodisperse, stable, and uncharged particles are required in high concentration. The aerosol generators used in this research were simple to be used and relatively dependable. Even though the size cannot be controlled, the gas burner generated the high concentration fine particles with reasonable monodispersity. The widely used Collison Atomizer was stable and easy-operating. The three jet model used in this study can produce higher concentration of aerosol than the single jet model and if the concentration of the solution is carefully maintained, this generator can be used in various applications. The condensation type aerosol generator is a very useful system. The combination of carnauba wax and this system provided nearly monodisperse particles with 50 nm to 130 nm. The key to operate this type of generator is keeping the generator stable, especially the temperature of the bubbler and the reheater. The concentration of the wax vapor is strongly dependent on the bubbler temperature and heterogeneous nucleation is affected by the temperature of the reheater. A smaller geometric standard deviation, σ_g , was obtained when the reheater temperature is above 300°C. If the bubbler temperature fluctuation can be prevented, more satisfactory results might be obtained. The bubbler temperature was influenced from not only the ambient temperature but also the bubbling flow rate and the amount of wax inside the bubbling flask. These values are constantly varying and the temperature controller used could not follow this effect sufficiently.

The particle loss rate result showed that the losses due to surface deposition are relatively small compared to losses due to ventilation, but the dependence of particle loss rate on the air exchange rate was not weak. When the air exchange rate increases, the loss rate due to the surface deposition rises. Two model were investigated in this research and they fit to the

empirical data reasonably well. There still were differences between the empirical data and the model predictions of the particle loss rate. The possible reason is that the assumption of uniform mixing throughout the bulk region may fail especially for the larger particles because of their low diffusion coefficient. Moreover, the transportation processes within the boundary layers may need to be considered in theoretical investigations because the deposition rate through boundary layers may depend on near-surface air flow conditions. Thus, some assumption about k_e and δ are necessary and further work is required to parameterize the eddy diffusion coefficient.

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Figure 5.21 Comparison of Experimental Data and the Models (a) Air Exchange Rate = 0.8 h^{-1} , (b) Air Exchange Rate = 1.7 h^{-1} , (c) Air Exchange Rate = 2.4 h^{-1}

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Benes and Holub models. According to the prior research, two values for n in equation (3.65) are suggested by Okuyama *et al* (1986) and Holub *et al* (1988) for the Crump and Seinfeld model. In Figure 5.21, $n = 2.6$ is used.

As can be seen in Figure 5.21, Benes and Holub model fits the experimental data at least as well as the Crump and Seinfeld model. The submicron particles have larger deposition coefficients and this phenomena was observed in charged particles (McMurry and Rader, 1985). Best fits to k_e were obtained by the regression. Those parameters are shown in Table 5.2 and Figure 5.22.

Table 5.2 The Turbulent Intensity Value for the Theoretical Models

Air Exchange Rate (h^{-1})	Crump and Seinfeld (S^{-1})	Benes and Holub (S^{-1})
0.8	4.5	1.66
1	18.32	8.32
2.4	37.1	13.75

Figure 5.22 The Change of k_e Value with the Air Exchange Rate

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