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PARTICLE DEPOSITION IN HUMAN
AND CANINE TRACHEOBRONCHIAL CASTS

Annual Progress Report

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Particle Deposition in Human and Canine Tracheobronchial Casts

ABSTRACT

The aim of this work is to measure deposition patterns and efficiencies of aerosol particles within realistic, single-pathway physical models of the tracheobronchial airways of humans and experimental animals over a range of particle sizes for a variety of respiratory modes and rates. This will provide data needed to assess the dose to the bronchial epithelium from inhaled radon progeny.

In prior grant years an empirical expression for diffusional deposition efficiency of particles in the upper airways was obtained based on experimental data collected in central airway casts. The work also provided new quantitative data of airflow distribution in a realistic central airway cast for two species for both steady and pulsatile inspiratory flow and for expiratory flow.

In the current year theoretical studies based on a developing flow model were extended. We concluded that although the developing flow model is a better predictor of the data than assumption of parabolic flow, the predicted deposition is significantly lower than that predicted by our best fit equation.

The mass size dispersion of particles classified according to their electrical mobilities was also studied. For the conditions tested, doubly charged particles constituted 9-13% of the total particle number concentration and 24-48% of the total mass concentration. We demonstrated that the multiply charged particles are effectively removed, and

nearly monodisperse particles produced, if an impactor is placed in the aerosol generation system.

A condensation aerosol generator was constructed and tested for the production of high concentrations of radioactively tagged aerosol particles with diameters from 0.2 to 10 μm .

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I. SCOPE

The research program addresses Goal # 2 of the DOE/ER Plan for Radon Exposure and Dose to the Critical Cells of the Respiratory Tract. It responds to the indicated need for data on the effect of particle size and charge, breathing pattern and respiratory tract morphology on the dose to the critical cells of the bronchial epithelium.

The aim of this work is to measure deposition patterns and efficiencies of aerosols within realistic, single-pathway, physical models of the tracheobronchial airways of humans and experimental animals over a range of particle sizes from 0.01 to 1.0 μm , for a variety of respiratory modes and rates. The pathways are a major branch path, a minor branch path and one which consists of alternating major and minor branches. The particle size range has been extended to include aerosols with $d < 0.02 \mu\text{m}$ to represent the unattached fraction of Po-218. Substantial deposition is expected for these particles. Additional studies will compare the deposition of singly charged with neutral ultrafine particles. The measurements will permit the development and critical evaluation of quantitative predictive models for the deposition efficiency of the inhaled radioactive particles.

II. SIGNIFICANT RESULTS

Full morphometric and inhalation flow distribution measurements on the human and canine airway casts were completed in the prior grant periods. The work provided new quantitative data of airflow distribution in a realistic central airway cast for two species and for two different airflow patterns; steady inspiratory flow and pulsatile

inspiratory flow. Inertance (inertia of the airflow) was demonstrated to carry more of the flow to lower lobes at higher inspiratory flowrate. The effect was substantially greater in the canine cast. The inertial effects seen for constant flow appear to be enhanced by pulsatile flow. The difference is more apparent for the dog cast which is consistent with morphometric differences between the species. We further demonstrated that inertial effects are not nearly as pronounced during exhalation as for inhalation. Air flow resistance in these casts is considerably greater during exhalation than inhalation.

An empirical expression for the diffusional deposition efficiency of particles in the upper airways was obtained based on experimental data collected in central airway casts (Cohen, B.S. and Asgharian, B., J. Aerosol Sci., 1990). In the uppermost airways of the lung the deposition predicted using our empirical equation is about twice the value obtained using Ingham's equation for particle deposition by diffusion from parabolic flow. Further theoretical investigation was carried out to more completely evaluate the effect of skin friction and Schmidt number on deposition efficiency. We concluded that although the developing flow model is a better predictor of the data than assumption of parabolic flow, the predicted deposition is significantly lower than that predicted by our best fit equation. A detailed report of this work is included here as Appendix A.

III. CURRENT TECHNICAL STATUS OF THE PROJECT

As part of our ongoing efforts to improve the quality of our measurements we undertook an investigation of the mass size dispersion of particles classified according to their electrical mobilities. This is of primary concern in our experiments in which the radioactivity is a concentration dependent property of the classified particles. Initially, the mass size distribution of particles produced by 3-jet Collison, 6-jet Collison and Misty-Ox nebulizers was measured. A 0-2 stage Mercer impactor was placed after the nebulizers and the size mass distribution measured again. The polydisperse particle stream was then used to generate "size classified" aerosol and the mass size distribution of the equal mobility particles was calculated and measured. It was found that without an impactor in line, the aerosol stream contained a significant fraction of multiply charged particles. For the conditions tested, the doublets constituted 9-13% of the total particle number concentration and 24-48% of the total mass concentration. When an impactor was inserted directly after the nebulizers the multiply charged particles were effectively removed and the particles were nearly monodisperse. A report of this work has been submitted to the journal Aerosol Science and Technology for publication. The manuscript is included in this report as Appendix B.

The aerosol generator we use for ultrafine particles, $d < 0.2 \mu\text{m}$, is not suitable for studies with particles larger than $0.2 \mu\text{m}$. A condensation aerosol generator was constructed and tested to produce monodisperse radioactively tagged aerosol particles with diameters from 0.2 to $10 \mu\text{m}$. A detailed report of this laminar flow aerosol generator is

included in this report as Appendix C.

We are continuing to carry out cast deposition experiments in order to increase the data base. Current experiments for particles with $d < 0.2 \mu\text{m}$ are done with an impactor in the aerosol generation system. A few experiments have been completed using the condensation aerosol generator.

IV. PUBLICATIONS

Cohen, B.S., Deposition of Ultrafine Particles in the Human Tracheobronchial Tree: A Determinant of the Dose from Radon Daughters, In: Radon and Its Decay Products: Occurrence, Properties and Health Effects (Ed., P. Hopke), American Chemical Society, Washington, DC 20036, pp. 475-486, 1987.

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Cohen, B.S., and B. Asgharian, Deposition of Ultrafine Particles in the Upper Airways: An Empirical Analysis, J. Aerosol Sci. 21:789-797, 1990.

V. REPORT OF EFFORT - P.I.

The PI has devoted 2 person months to the project in the current calendar period (7/1/90-1/15/91). She will devote 1 more person month to the project prior to the end of the current grant year (4/30/91).

VI: COLLECTION EFFICIENCY OF PARTICLES BY DIFFUSION IN DEVELOPING FLOWS

In the upper airways of the respiratory system, due to deviation of flow from parabolic flow, one can not use the expression

$$\eta_D = 1 - 0.819e^{-14.63\Delta} - 0.0976e^{-89.22\Delta} - 0.0325e^{-228\Delta} - 0.0509e^{-125.9\Delta^{2/3}}, \quad (1)$$

where

$$\Delta = \frac{\pi LD}{4Q}, \quad (2)$$

in which L is the airway length, Q is the flow rate through the airway and D is the diffusion coefficient (Ingham, 1975). Equation (1) is derived under the condition of fully developed laminar (parabolic) flow in a tube. When the flow enters a tube, it requires a distance before becoming fully developed. The transitional region is defined by a non dimensional entrance length, ξ , as

$$\xi = \frac{z}{RRe}, \quad (3)$$

where z is the entrance length, R is the tube radius, and $Re = 2Q/\pi Rv$, with v being the air kinematic viscosity, is the flow Reynolds number. The developing flow becomes fully developed when $\xi = 0.15$. In the upper region of the lung the flow profile departs considerably from parabolic flow due in part to high flow velocity.

In developing flows the velocity gradient near the wall is larger than that in parabolic flows. Therefore, more particles are carried near the walls and consequently there is a higher probability of particle deposition in developing flows.

To obtain an expression for collection efficiency by diffusion in the upper airways we assumed a developing flow model. Although this simplified model ignores other possible deposition mechanisms such as that caused by the secondary flows, it is a better representation of the physical phenomenon and improves the current commonly-used results (equation 1). The solution for particle deposition in developing flows have

been obtained by several investigators. Since the Peclet number, Pe is large in aerosol flow in lung airways, the mass transfer in the axial direction is negligible as compared to that in the radial direction. Using the Integral Method, Chen and Comparin (1976) showed that when the Schmidt number, $Sc=v/D$, approaches zero, deposition in developing flows can be approximated by the expression derived for uniform (slug) flow (see also Ingham, 1975) and when Sc approaches infinity, deposition can be calculated from equation (1). In these limiting cases deposition efficiency is only a function of Δ described by equation (2). For intermediate values of Sc , however, one can not assume such a relationship and a more detailed study becomes necessary.

For aerosol flow in the lung airways both the Peclet number and the Schmidt number are large. In developing flows, Ingham (1983) found that η_D depends on ξ and Sc , as following

$$\eta_D = 1.709 I(\xi) Sc^{-\frac{2}{3}}, \quad (4)$$

where,

$$I(\xi) = \frac{\int_0^\xi \sqrt{\Phi} d\xi}{\left(\int_0^\xi \int_0^\xi \sqrt{\Phi} d\xi d\xi \right)^{1/3}}, \quad (5)$$

in which $\Phi(\xi)$ is the wall skin friction. An analytical solution for the integral of equation (4) in developing flow has not been derived. However, the numerical values of the wall skin friction, $\Phi(\xi)$ in developing flow are available in the literature (Mohanty and Asthana, 1978). To obtain an expression for $I(\xi)$ in equation (4), we fitted these data with polynomials of various degrees to maximize the correlation coefficient for the fit, $r^2=1$.

The flow in the entrance region of the tube was divided into 3 sub-

regions; one, immediately at the entrance where the flow is nearly uniform ($\xi < 0.004$), two, the inlet region where the centerline velocity in the tube is the same as velocity at the inlet ($0.004 < \xi < 0.04$), and three, the filled region where the flow boundary layers meet at the centerline but the flow is not fully developed ($0.04 < \xi < 0.15$). The inlet and filled regions are defined by Shingo (1966). The fitted expressions for these sub-regions are as following: for $0 < \xi < 0.004$,

$$\Phi(\xi) = 7.132 - \frac{4430}{3}(\xi - 0.004), \quad (5)$$

for $0.004 < \xi < 0.04$,

$$\begin{aligned} \Phi(\xi) = & 9.499 - 796.7\xi + 5.956 \times 10^4 \xi^2 - 2.343 \times 10^6 \xi^3 + 4.551 \times 10^7 \xi^4 \\ & - 3.434 \times 10^8 \xi^5, \end{aligned} \quad (6)$$

and for $0.04 < \xi < 0.15$,

$$\begin{aligned} \Phi(\xi) = & 5.633 - 58.926\xi + 914.4\xi^2 - 7426.9\xi^3 + 3.088 \times 10^4 \xi^4 \\ & - 5.178 \times 10^4 \xi^5. \end{aligned} \quad (7)$$

Expressions for $\Phi(\xi)$ are substituted in (4) and the values of $I(\xi)$ are calculated. A Trapezoidal numerical integration technique was employed to calculate the results. If the integrand in equation (4) is denoted by $f(\xi)$,

$I = \int_a^b f(\xi) d\xi = \frac{\Delta\xi}{2} \left(f_a + f_b + 2 \sum_{j=1}^{N-1} f_j \right)$, where $\Delta\xi$ is the interval width, N is the total number of intervals selected, f_a and f_b are the integrand calculated at the limits of the integral, and f_j is the value of $f(\xi)$ at the j^{th} interval. $I(\xi)$ is calculated for different values of ξ . The numerical results are then fitted with a 5-degree polynomial with $r^2 = 1$.

$$\begin{aligned} I(\xi) = & 0.0209 + 12.711\xi - 216.38\xi^2 + 2748.9\xi^3 - 17058\xi^4 \\ & + 40191\xi^5 \end{aligned} \quad (8)$$

The calculated values along with the fitted line are shown in Fig.1.

Upon substitution of equation (8) into (3), η_D in developing flows can be computed. Figure 2 shows the deposition efficiency of particles by

diffusion (η_D) in developing and parabolic flows plotted against ξ for $Sc=3.48 \times 10^4$ (0.05 μm particle diameter). For $\xi < 0.005$, η_D from developing flow is larger than that of parabolic flow, but, as ξ increases the two results converge. When $\xi > 0.005$, η_D for developing flow shows also a log-linear relationship with ξ . Figure 3 is the same plot for $Sc=6.76 \times 10^4$ (0.2 μm diameter). The same behavior is observed. From Figs. 2 and 3 it is concluded that for $\xi > 0.005$, η_D and ξ are log-linear.

Deposition of particles by diffusion in an airway with parabolic and uniform flow is shown by Ingham (1975) to depend on the non-dimensional parameter Δ given by equation (2). If the values obtained from these results are fitted into the expression

$$\eta_D = a_0 \Delta^{a_1}, \quad 10^{-9} < \Delta < 10^{-4} \quad (9)$$

the correlation coefficient, $r^2=1$. Coefficients a_0 and a_1 are found to be 7.388 and 0.674 for parabolic flow and 4.42 and 0.499 for uniform flow respectively. therefore, the plot of $\log_{10}(\eta_D)$ against $\log_{10} \Delta$ is essentially a straight line.

Values of ξ calculated for the experimental data points obtained in earlier measurements in central airway casts, showed that in all cases $\xi > 0.005$. Therefore, in the absence of an analytical expression for η_D in this region, we formulated an expression by curve fit of the experimental data with Δ . This will simplify the calculations. Due to the scattered nature of the data, little will be gained by a more detailed analysis such as might be obtained by mass transfer analogy. For an expression of the form of equation (9), we calculated $a_0=2.965$ and $a_1= 0.568$ by fitting the experimental data of Cohen (1987). The correlation coefficient for the fit, was 0.57.

Deposition fraction of particles calculated from the best fits and the experimental data are plotted in Fig. 4. As shown, parabolic flow results is

the lower limit and the uniform flow results show the upper limits. The best fit of the experimental data falls in between, simulating a developing flow phenomenon. In developing flow, however, η_D is related to both Sc and ξ and only in the limits the two are combined to give

$$\Delta = \frac{\xi}{2Sc}. \quad (10)$$

In accordance with the developing flow analogy, we formulated η_D as

$$\eta_D = a_2 \xi^{a_3} Sc^{a_4}, \quad (11)$$

by fitting with the experimental data. Coefficients a_2 , a_3 , and a_4 were found to be 1.73, 0.541, and -0.565 respectively. A plot of different fits along with the limiting lines are shown in Fig. 5. The results demonstrate that both fits are very similar and that equation (9) is sufficient to predict the deposition efficiency in the upper airway based on developing flow model.

Deviation of flow from parabolic flow in the airways is not merely the results of high airflow in the airways which delays the flow development. Other parameters such as secondary flow caused by the bifurcating airways may also be significant. In addition, the flow in the experiment simulating actual inspiratory respiratory breathing was pulsatile rather than constant which also may have caused enhanced deposition particularly at the bifurcation. A comparison of the current fit with the theoretical results of developing flow is made in Fig. 6. It shows that although the developing flow model is a better prediction of the data than the parabolic flow results (lower limit), it is significantly lower than the best fit. Therefore, developing flow will explain the difference partially. A more general expression must include the flow rotational and pulsatile effects. In other words, equation (11) must also include the Deans number and the Wormersly parameter. This will be explored in the future.

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Fig 4

APPENDIX B

**VII. GENERATION OF SUBMICRON MASS/VOLUME-MONODISPERSE
AEROSOLS WITH A NEBULIZER-IMPACTOR-ELECTROSTATIC
CLASSIFIER SYSTEM**

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ABSTRACT

A study was undertaken to investigate the mass size dispersion of particles classified according to their electrical mobilities because this is of primary concern in experiments that measure a concentration dependent property of the classified particles. Initially, the mass size distribution of particles produced by 3-jet Collison, 6-jet Collison and Misty-Ox nebulizers was measured. A 0-2 stage Mercer impactor was placed after the nebulizers and the size mass distribution measured again. The polydisperse particle stream was then used to generate "size classified" aerosol and the mass size distribution of the equal mobility particles was calculated and measured. It was found that without an impactor in line, the aerosol stream contained a significant fraction of multiply charged particles. When an impactor was inserted directly after the nebulizers the multiply charged particles were effectively removed and the particles were nearly monodisperse.

INTRODUCTION

Monodisperse aerosol particles are essential for measurement of the behavior of airborne particles in a complex flow field such as that in the respiratory airways. Monodisperse airborne particles with diameters smaller than $0.2\ \mu\text{m}$ can be produced from a polydisperse aerosol by charging the particles, then separating a narrow range of particle sizes by electrical mobility with an electrostatic classifier (ESC, model 3701, TSI inc., St. Paul, MN) (Liu and Pui, 1974). The number of ultrafine particles produced by this method will be very low because few of these particles become charged in the bipolar diffusion charger, and particles of only one sign are deflected into the resultant aerosol stream. The particle stream selected for a given electrical mobility will contain multiply charged as well as singly charged particles. The former carry substantially more mass than the latter and will distort analytical results if the particle detection method measures a concentration dependent property of the classified particles. For example, in experimental studies of particle deposition that measure radioactivity or fluorescence, the deposited mass/volume of particles is of primary concern. The goal of the study reported here was to produce a highly monodisperse ultrafine aerosol with a sufficiently high particle number concentration to study particle deposition in full hollow airway casts.

EXPERIMENTAL METHODS

Salt particles were generated from a 0.1% saline solution with 3 different types of atomizers: 3-jet Collison, 6-jet Collison, and Misty-Ox nebulizers. These three nebulizers were selected for testing because we had found them convenient for use with a highly radioactive $\text{Tc}^{99\text{m}}$ solution; each can be sealed quickly, shielded conveniently, and has a sufficiently large reservoir for the duration of the experiments. Conditions were selected to obtain the highest possible particle number concentration while reducing

the mass fraction from multiply charged particles. Figure 1 shows the schematic diagram of the particle generation system. Atomized droplets from each nebulizer were put through a Mercer impactor containing zero, one, or two stages to remove the large droplets from the aerosol stream. The performance characteristics of the impactor are provided in Table I. Insertion of the impactor immediately after the nebulizer avoids some common problems associated with using an impactor. Because of the large droplet size, initial stages of the impactor are sufficient to remove the larger droplets, consequently, the pressure drop caused by the addition of the impactor will be minimal. In addition, captured droplets drip off the impactor plate and will not cause an overload which may affect the impactor characteristics (Pui et al., 1990). Furthermore, the captured droplets cannot bounce back into the aerosol stream as is always a possibility with the solid particles. It is also essential to remove the large droplets not only to produce a finer size distribution of the particles, but because after evaporation they increase the relative humidity of the flow significantly and consequently will quickly saturate the silica gel driers. The remaining droplets in the flow pass through a heater to evaporate the water from the salt particles. The water vapor is removed by channeling the flow through a silica gel diffusion dryer. A Dew Point Transmitter (Model 655, General Eastern, Watertown, MA) was placed in the system after the diffusion drier to monitor the relative humidity of the aerosol stream. The silica gel was replaced when relative humidity exceeded 40%, since dry salt particles are achieved and maintained only below this relative humidity level (Orre et al., 1985; Tang and Murkelwitz, 1984). The dry salt particles are charge neutralized (^{85}Kr) to eliminate excess charge and produce a Boltzmann charge distribution of the particles. The particles were size classified by an ESC connected in series to a Condensation Nucleus Counter (CNC, model 3020, TSI inc., St. Paul, MN). Particle size analysis was controlled by a personal computer connected to both the ESC and the CNC. An automated system varied the voltage on the ESC electrode and recorded the measurements of the particle

concentration from the CNC; the software program then calculated the particle count and mass size fractions.

To produce a monodisperse aerosol (MDA), the dry, polydisperse aerosol (PDA) from the nebulizer was charged neutralized with the ^{85}Kr and passed through an independent ESC to select particles of a given mobility. Number concentration of the MDA particles is directly related to the flow rate of the sheath air and PDA. The sheath air flow rate was kept at 20 lpm. The PDA stream for the Misty-Ox and the 6-jet Collison nebulizers was at approximately 5 lpm. For the 3-jet Collison nebulizer, the maximum flow rate was 3 lpm. The equal mobility particles were charge neutralized and particle size analysis was performed utilizing the computer controlled ESC - CNC system described above (Figure 2).

If the concentration and size dispersion of the PDA particles are known, the concentration of singly and doubly charged aerosols, c , in the MDA stream separated by the ESC can be determined from

$$c = \delta_1 \delta_2 C, \quad (1)$$

where δ_1 is the number or volume fraction of the PDA particles at size interval $\Delta \ln d$, C is the total number or volume concentration of the PDA particles entering the ESC for size classification and δ_2 is the fraction of particles carrying n elementary charge units.

$$\delta_2 = \frac{\exp(-\frac{n^2}{2\sigma^2})}{\sum_{n=-\infty}^{+\infty} \exp(-\frac{n^2}{2\sigma^2})}, \quad (2)$$

where σ is the characteristic Boltzmann's charge.

RESULTS AND DISCUSSIONS

Particle mass size distributions of the PDA particles generated by different nebulizers and bypassing the impactor are shown in Fig. 3. The resulting mass size distributions, $dc/Cd(lnd)$, are calculated from the measured volume fraction data ($\Delta c/C$) at different particle size intervals (Δlnd). As shown, the mass size distribution functions are not log-normal. The distribution is not monotonic, but exhibits multi-modal peaks. All 3 nebulizers exhibit similar distributions and modal peaks. Bimodal peaks were also observed in mass size distribution measurements of Rodes et al. (1990) for wet and dry salt particles generated by an ultrasonic room humidifier. This phenomenon may be attributed to the kinematic coagulation of the particles in the nebulizers after bouncing off the walls.

The corresponding plot of number distribution of the PDA particles for the 3 nebulizers is shown in Fig. 4. The 3-jet Collison nebulizer produced a lower particle concentration than did the 6-jet Collison or Misty-Ox nebulizer because of the lower flow rate. The modal peak for the 3-jet Collison nebulizer occurred at a larger particle diameter than that for the other two nebulizers. Difference in mass size distributions and in number concentrations of the PDA particles will result in a different size dispersion and concentration of MDA particles when they are classified by an ESC.

Figure 5 shows the distribution of equal mobility MDA particles separated by the ESC when the PDA particles are generated by a 3-jet Collison nebulizer. The impactor was not included in the system. The resulting particle mass size distribution are for the ESC voltages of 0.93 kV, 3.26 kV, and 8.57 kV. These voltages correspond to the count median diameter (CMD) of singly charged particles of 0.05 μm , 0.1 μm , and 0.18 μm respectively. As shown, doublets are present even for particles as small as 0.05 μm . The sizes of the doublets corresponding to these single particles are 0.073 μm , 0.15 μm , and 0.29 μm respectively. The relative number of doublets increases as the size of the singly charged particles increases. For 0.05 μm particles, the peak of the doubly charged

particles is near that of singly charged ones and the doublets and the triplets make up only a few percent of the total distribution, therefore they are negligible. For $0.1\ \mu\text{m}$ and $0.18\ \mu\text{m}$ particles, there are significant numbers of multiply charged particles and the particles cannot be described as of uniform size. The multiply charged particles should be removed from the aerosol stream, if this particle stream is to be used as a source of monodisperse aerosol, (Romey-Novas and Pui, 1988).

Figure 6 shows the mass size distribution of the MDA particles classified by the ESC when the PDA particles are generated by a 6-jet Collison nebulizer. No impactor was used in the system after the nebulizer. Again, the particles generated are nominally $0.05\ \mu\text{m}$, $0.1\ \mu\text{m}$, and $0.18\ \mu\text{m}$. For $0.05\ \mu\text{m}$ particles, although no apparent peak due to multiply charged particles is noticed, the wide distribution is evidence that they are still present. For $0.1\ \mu\text{m}$ and $0.18\ \mu\text{m}$ particles, the doublet peaks are obvious and some triplets are also evident. The results show that as the ESC voltage increases (i.e., as the size of the singly charged particles is increased), the fraction of singly charged particles reduces and the particle mass size distribution spreads. This implies that the larger the particle size, the more the contribution of multiply charged particles.

The mass size distributions of the MDA particles separated by the ESC when the PDA particles are generated by the Misty-Ox nebulizer, are shown in Fig. 7. No impactor was used in the system after the nebulizer. The same particle sizes as the previous cases are considered. As seen, the doublets and triplets are present for all particle sizes and significant even for $0.05\ \mu\text{m}$ particles. By comparing Figs. 6 and 7, it appears that, although, Misty-Ox and 6-jet Collison nebulizers generate similar PDA size distributions, after size classification by the ESC, more doublets for $0.05\ \mu\text{m}$ particles and fewer doublets for $0.1\ \mu\text{m}$ and $0.18\ \mu\text{m}$ are obtained when the PDA particles are generated by the Misty-Ox nebulizer.

Values of particle concentration of MDA particles for various voltages of the ESC were calculated from equations (1) and (2) utilizing the measured size fractions

and the total concentrations of the PDA particles. Results are listed in Table II. In addition, the concentration ratio of singly charged to the doubly charged particles were calculated. From Table II, it can be seen that for singly charged particles, as the size of the particles is increased, the number concentration tends to decrease, but the mass concentration will increase. Furthermore, the number concentration ratio ranges from 7 to 10 while the mass concentration ratio ranges from 1.1 to 3.2. Therefore, the doublets constitute 9-13% of the the total particle number concentration and 24-48% of the total mass concentration. When the size of the classified singly charged particles increases, the mass ratio decreases again demonstrating that doublets are more significant for larger particles. Based on the results, it can be concluded that the contribution of doublets to total mass fraction is significant as was observed in Figs 5-7.

The mass size distribution functions of the PDA particles generated by different nebulizers when an impactor is added to the system after the nebulizer to remove the large droplets, are shown in Figs. 8-10. Figure 8 depicts the results for a 3-jet Collison nebulizer. The 3-jet Collison nebulizer could only produce a maximum flow rate of 3 lpm. This flow rate was not sufficient for the impactor to efficiently remove the large particles. As shown in Fig. 8, there is a small decrease in mass fraction for large particles when a 0-stage or a 1-stage impactor is employed. There is a noticeable reduction of the larger particles when a 2-stage impactor is employed.

The mass size distribution function of the PDA particles generated by the 6-jet Collison nebulizer with the impactor in line is shown in Fig. 9. The 6-jet Collison nebulizer has less constriction than the 3-jet Collison nebulizer and is thus capable of transporting a higher flow rate. As a result, when an impactor is used, a significant reduction in the mass of large particles is observed. Figure 9 depicts the results for a flow rate of 5 lpm. When no impactor is used, as for the 3-jet Collison nebulizer, there is a significant fraction of large particles and particle mass size distribution is wide. When the impactor is added to the system, the fraction of large particles decreases as the

number of included impactor stages is increased. The size distribution looks more log-normal. It is also interesting to note that for 0-stage impactor in Fig. 9, the right tail has a sharp cut-off. Thus, this initial droplet distribution can be used to produce monodisperse particles as large as $0.4\text{ }\mu\text{m}$.

Figure 10 shows the results of particle mass size distribution of the PDA particles from a Misty-Ox nebulizer when a flow rate of 5 lpm through the system is employed. Comparing Figs. 9 and 10, striking similarities between the results of the 2 nebulizers are observed. With no impactor, Misty-Ox produces large droplets with a distribution function similar to that of the 6-jet Collison nebulizer. Consequently, when an impactor is used to remove the large droplets, similar distribution functions are observed. Furthermore, it is interesting to note that all 3 nebulizers show similar multimodal peaks under the conditions of these tests.

The use of a fine mass size distribution of PDA for the ESC input is crucial for the production of truly monodisperse particles by the ESC. As shown in Figs 8-10, a Mercer impactor used after the nebulizer to collect the larger particles will eliminate the large droplets generated by the nebulizers and will reduce the multiply charged aerosol in the MDA stream classified by the ESC. Moreover, the portion to the right of the modal peak in the PDA mass size distribution (Figs 8-10) must be used for particle generation and the initial particle number concentration must be high enough so that a significant number of monodisperse particles are generated.

Table III lists the measured CMD, volume median diameter (VMD), and the number concentration of the PDA particles produced by each nebulizer-impactor combination. As can be seen, the total particle concentration generated by the 3-jet Collison nebulizer is smaller than that by 6-jet Collison and Misty-Ox nebulizer by almost one order of magnitude. When an impactor is used after the nebulizer, the CMD for the 3 nebulizers does not change appreciably, but, a significant reduction in the VMD is observed as the number of stages of the impactor is increased. Since the total particle

number concentration of the 3 nebulizers are not noticeably affected by the presence of the impactor, it is concluded that only large particles are collected from the flow and they are removed effectively.

When the impactor is placed after the nebulizer, the doublets are significantly reduced in the ESC output stream. Figure 11 shows the mass distribution when a 2-stage Mercer impactor is used after the 6-jet Collison Nebulizer to produce singly charged $0.15\text{ }\mu\text{m}$ (6.41 kV) and 1-stage impactor for $0.18\text{ }\mu\text{m}$ (8.57 kV) particles. The results show only small doublet fractions for each case. For $0.15\text{ }\mu\text{m}$ particles, there are more singly charged and fewer doubly charged particles since more stages of impactor are employed. Comparing the results for the ESC voltage of 8.57 kV in Fig. 11 with that in Fig. 6, demonstrates the importance of using the impactor to reduce the doublets.

When a 2-stage impactor is used immediately after the Misty-Ox nebulizer, the doublets are only reduced slightly for $0.05\text{ }\mu\text{m}$ particles (Fig. 12). This is because the portion to the left of the modal peak in the PDA size distribution (Figure 10) is utilized for the size classification by the ESC. However, for $0.05\text{ }\mu\text{m}$ particles, the doublets are only slightly larger than the singly charged particles, so the error introduced by the presence of the doublets is small. On the other hand, for $0.1\text{ }\mu\text{m}$ and $0.18\text{ }\mu\text{m}$ particles, since the portion to the right of the modal peak in the PDA size distribution is used, a fine MDA is generated. It can also be seen by comparing Figs. 7 and 12 that the fraction of singly charged particles is not appreciably changed for $0.05\text{ }\mu\text{m}$ particles while that for $0.1\text{ }\mu\text{m}$ is significantly increased. The fraction increased is smaller for $0.18\text{ }\mu\text{m}$.

The measured CMD, VMD, and number concentration of the MDA particles classified by the ESC with and without an impactor in the system are reported in Table IV. The results are given for different nebulizers at various ESC voltages. Because of the doublets in the MDA stream, the particle size distribution is not log-normal but bimodal. The distribution approaches log-normal when the doublets are removed. To investigate the size dispersion of the MDA particles, the geometric standard deviation (σ_g) based

on a log-normal size distribution can be calculated; these are given in Table IV. According to Table IV, with no impactor, the particle number size distribution is narrow ($\sigma_g < 1.3$) for all ESC voltages. However, for mass size distribution, due to the doublets, the distribution is wider and $\sigma_g > 1.3$. Furthermore, the VMD calculated from the Hatch and Choate equation (1929) for a log-normal distribution of the particle number ,

$$\text{VMD} = \text{CMD} \exp(-3 \ln^2 \sigma_g), \quad (3)$$

is smaller than the measurements. This is an error that results from assuming a log-normal distribution when it is in fact, bimodal. When the Mercer impactor of different stages is used to collect the large droplets, the particle mass size distribution will also become more monodisperse and $\sigma_g < 1.3$. The VMD calculated from equation (3) will then be in close agreement to the measured values.

The measured number concentration of the MDA particles reported in Table IV is in good agreement with the number concentration obtained by adding the number concentrations of the calculated singly and doubly charged particles in Table II. Due to deposition losses in the generation and measurement lines, the measured concentrations are expected to be smaller than the calculated ones. While for 3-jet Collison and Misty-Ox nebulizer this is true, for the 6-jet Collison nebulizer, the measurements are slightly higher.

SUMMARY

Although, different nebulizers produce different droplet concentrations and size dispersions, the findings reported here provide general guidance. Monodisperse singly charged particles separated from a polydisperse stream by means of their electrical mobility, will contain a fraction of multiply charged particles with equal mobility but

different size. The degree of inhomogeneity is more severe for mass size distribution than for count size distribution. The mass fraction of doublets reported here reached up to 48% for 0.18 μm particles. As the size of the singly charged particles increases, the fraction of doublets will also increase. The multiply charged particles may be effectively removed by means of an impactor placed after the nebulizer to remove the larger droplets in the polydisperse stream. The effectiveness of the impactor depends on the flow rate of the polydisperse particle stream.

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Table I- Performance characteristic of a Mercer impactor at a flow rate of 5 lpm.

<u>stage number</u>	<u>number of jets</u>	<u>jet diameter (cm)</u>	<u>cut-size[*] (μm)</u>
0	1	0.5613	8.1
1	1	0.3454	4.0
2	2	0.1778	2.1

* : Aerodynamic diameter of particles which are collected with 50% efficiency.

Table II- Measured number and mass concentrations of singly and doubly charged dry salt aerosol in the MDA stream.

nebulizer type	voltage kv	Number Concentration (#/cc)			Mass Concentration ($\mu\text{m}^3/\text{cc}$)		
		singly charged	doubly charged	ratio	singly charged	doubly charged	ratio
3-jet	0.93	8.91×10^3	1.21×10^3	7.4	0.57	0.22	2.6
	3.26	1.21×10^4	1.03×10^3	9.2	6.29	1.94	3.2
	8.57	2.38×10^3	309	7.7	6.98	6.34	1.1
6-jet	0.93	2.1×10^4	2.1×10^3	10	1.31	0.47	2.8
	3.26	2.02×10^4	3.26×10^3	6.7	10.58	6.24	1.7
	8.57	2.38×10^3	309	7.7	27.4	25.52	1.1
Misty ox	0.93	2.39×10^4	2.41×10^3	10	1.5	0.54	2.8
	3.26	2.19×10^4	3.32×10^3	7	11.11	6.35	1.7
	8.57	7.2×10^3	918	7.8	24.45	18.86	1.3

Table III- Measured median sizes and concentration of PDA particles generated by different types of nebulizers.

Nebulizer <u>type</u>	CMD <u>μm</u>	VMD <u>μm</u>	Concentration <u>(#/cc)</u>
3-jet	0.074	0.332	5.44×10^5
3-jet + 0-stage	0.070	0.320	4.80×10^5
3-jet + 1-stage	0.075	0.313	4.39×10^5
3-jet + 2-stage	0.079	0.249	3.61×10^5
6-jet	0.065	0.351	1.36×10^6
6-jet + 0-stage	0.061	0.238	1.39×10^6
6-jet + 1-stage	0.060	0.198	1.39×10^6
6-jet + 2-stage	0.063	0.163	1.29×10^6
Misty ox	0.068	0.348	1.4×10^6
Misty ox + 0-stage	0.069	0.275	1×10^6
Misty ox + 1-stage	0.066	0.189	1×10^6
Misty ox + 2-stage	0.063	0.148	1.07×10^6

Table IV- Median sizes, geometric standard deviation, and number concentration of the MDA particles classified by the ESC.

nebulizer type	voltage k v	CMD μm	σ_g —	VMD μm	σ_g —	* VMD		concentration #/cc
						μm		
3-jet	0.93	0.043	1.25	0.052	1.36	0.055		1.59×10^3
	3.26	0.083	1.27	0.114	1.53	0.099		573
	8.57	0.179	1.27	0.236	1.46	0.212		875
6-jet	0.93	0.055	1.24	0.067	1.37	0.063		4.46×10^4
	3.26	0.107	1.27	0.14	1.44	0.127		2.98×10^4
	8.57	0.194	1.29	0.258	1.42	0.236		1.10×10^4
6-jet+ 2-stage	6.41	0.142	1.15	0.15	1.16	0.151		7.51×10^3
6-jet + 1-stage	8.57	0.174	1.19	0.193	1.23	0.191		5.65×10^3
Misty ox	0.93	0.050	1.20	0.058	1.32	0.055		2.17×10^4
	3.26	0.102	1.30	0.128	1.38	0.125		1.85×10^4
	8.57	0.183	1.37	0.250	1.40	0.246		4.48×10^3
Misty ox +	0.93	0.050	1.19	0.058	1.32	0.054		1.03×10^4
2-stage	3.26	0.095	1.27	0.111	1.25	0.113		5.74×10^3
	6.41	0.121	1.27	0.144	1.29	0.144		2.05×10^3

* : calculated based on log-normal size distribution

Fig 2

Fig. 2

Fig. 6

mop

Fig. 8

Fig 12

VIII. LAMINAR FLOW AEROSOL GENERATOR

A Condensation aerosol generation system was constructed to produce monodisperse radioactively tagged aerosol particles with diameters from 0.2 μm to 10 μm based on the design of (Prodi, 1972). The system consisted of a pump delivering dry filtered air at 25-40 psi to a single-jet Collison nebulizer containing a very dilute solution of 0.2% saline in doubly distilled water (Figure 1). The atomized salt droplets were passed through a heater and dried by directing the stream through a silica gel desiccator. The stream of resulting nuclei was then led to a bubbler which contained a heated solution of Triphenyl Phosphate (TPP). Temperature was controlled by immersing the bubbler in a thermostated silicone fluid bath (Dow Corning 550 Fluid, Dow Chemical, Midland, MI). Particle size is controlled by the temperature of the TPP. The temperature stability of the silicone fluid bath was regulated to $\pm 0.5^\circ\text{C}$ by means of an Omega Series 4000 temperature controller coupled with an Omega TRP-J probe (Omega Engineering, Inc., Stamford, CT). The quantity of the TPP in the bubbler was approximately 150 g.

In the bubbler, the salt nuclei mixed with the TPP vapor. Vapor concentration in the resulting flow was a function of the oil bath temperature. The flow was subsequently directed through a 1-meter vertical cooling column. At the entrance of the column a heating taped was mounted which was controlled by a voltage regulator maintained at 60V. This insured complete vaporization of the TPP. The vapor in the mixture cooled gradually and uniformly as it traveled down the cooling column and condensed on the nuclei, forming the primary aerosol.

In order to minimize the effects of radial gradients in concentration and temperature which are present in the vaporization zone/condensation chimney and effect aerosol monodispersity, the aerosol stream was divided at the end of the cooling column. Only the central 5% of the aerosol flow was extracted with the remainder

discharged through a HEPA filter. Only the more monodisperse central portion of the aerosol stream was used as test aerosol, and its flow was maintained at positive pressure by a peristaltic pump (Model 1215, Harvard Apparatus Co., Cambridge, MA) operating at 0.15 lpm.

To generate a radioactively tagged aerosol, Technetium 99m labeled sodium pertechnetate was added to the salt solution (Ultra Technetow FM, Mallinkrodt Diagnostics, Maryland Height, MD) in the nebulizer.

MEASUREMENT SYSTEM

The output of the generator was evaluated by two different methods. Initial calibration of the generator was conducted using a Climet 208 Particle Analyzer (Climet Instruments Co., Redlands, CA) with a Canberra Series 30 Multichannel Analyzer (Canberra Industries, Inc., Meridan, CT). This system could only measure particle sizes larger than 0.3 μm . The relatively high concentration monodisperse aerosol from the peristaltic pump ($\approx 10^6 \text{ cm}^{-3}$) was diluted with dry filtered air (at least 5 lpm) because the Climet could only detect particle concentrations of up to approximately $100/\text{cm}^3$. The flow system of the Climet 208 is designed so that the particles passing through the viewing volume are surrounded by a sheath of clean air. It uses an elliptical mirror optical system to detect particles, and was equipped with 0.3 μm sensitivity option. Light scattered by particles passing through the viewing volume is detected by a photomultiplier tube and a voltage pulse is produced for each particle passing through this volume. Amplitude of the pulse is a measure of the particle size.

The Canberra pulse height analyzer was used to measure the voltage pulse height distribution. The Climet provided voltage pulses which had a rise time of 2 μsec , which could be fed directly into the pulse height analyzer analog to digital convertor. The calibration of the pulse height analysis system depended to some extent on the

input pulse width, so the system was calibrated for each conversion gain used with pulses produced by a waveform generator (Mini-Pulser NP-1, Eberline Instrument Corporation, Sante Fe, NM). Voltage pulse amplitude was related to particle diameter on the Climet calibration curve provided by the manufacturer.

The second method for particle size specification utilized an Electrostatic Classifier (ESC, model 3701, TSI Inc., St. Paul, MN) and a condensation Nucleus Counter (CNC, model 3020, TSI Inc., St. Paul, MN) as shown in Figure 2. Particle diameters between 0.01 μm and 1 μm could be measured by this system. No dilution air was needed since the system worked for any particle concentration. However, the flow entering the system was adjusted to match the flow rate required by the ESC. This was easily achieved by placing a by-pass before the ESC. Aerosol flowed into one inlet of the by-pass while filtered room air flowed into the other. In this system, particles were separated according to their electrical mobilities. The particles were size classified by the ESC connected in series to the CNC. Particle size analysis was controlled by a personal computer connected to both the ESC and the CNC. An automated system varied the voltage on the ESC electrode and recorded the measurements of the particle concentration from the CNC; the software program then calculated the particle count and mass size fractions.

The results of particle size distribution in terms of count median diameter (CMD) and mass median diameter (MMA) and geometric standard deviation (σ_g) based on a log-normal particle size distribution are listed in Table I and shown in Figures 3 and 4 as a function of the oil bath temperature. These results were obtained with the ESC-CNC measurement system. The results have been confirmed with the Climet-Multichannel Analyzer system for large particles. Esc't (1986) developed a similar system for the production of large monodisperse aerosol particles. The results reported for oil bath temperatures of 175 °C and 190 °C are in good agreement with the present

results.

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FIGURE CAPTIONS

Figure 1. Schematic diagram of the Laminar Flow Aerosol Generation System.

Figure 2. Particle measurement system for particles between 0.01 μm and 10 μm .

Figure 3. Particle CMD as a function of oil bath temperature.

Figure 4. Particle VMD as a function of oil bath temperature.

Table I-Size distribution and geometric standard deviation (σ_g) of particles generated from a LFAG as a function of oil bath temperature.

Temperature	CMD*	σ_g	VMD**	σ_g
(°C)	μm		μm	
121	0.187	1.25	0.221	1.29
132	0.233	1.22	0.266	1.25
141	0.295	1.18	0.319	1.18
158	0.486	1.22	0.532	1.17
158	0.472	1.21	0.534	1.24
160	0.453	1.26	0.508	1.18
160	0.388	1.37	0.487	1.26
165	0.603	1.16	0.638	1.14
168	0.692	1.22	0.739	1.12
168	0.703	1.18	0.740	1.12
170	0.670	1.26	0.731	1.13

* : count mean diameter

** : volume mean diameter

$$CMD = -17.0138 + 0.378587T - 0.002796T^2 + 6.95518 \times 10^{-6}T^3$$

$$\text{VMD} = -5.128273 + 0.124334T - 9.92165 \times 10^{-4}T^2 + 2.726604 \times 10^{-6}T^3$$

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