

ACKNOWLEDGEMENTS

This work is dedicated to the memory of my father, whose life inspired all who knew him. He encouraged my persuit of knowledge and fostered my desire for excellence. His loving presence is sorely missed, but his spirit is with me always.

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THERESA RENÉE ANTHONY. Factors that Affect the Size Distribution of Nebulized Fluid.

ABSTRACT

A test nebulizer was developed to evaluate the effect of altering nebulizer geometry and flow conditions upon the size distribution of the droplets produced. An Andersen impactor was used to collect the dried residual particles of a methylene blue dye and sodium bicarbonate solution. Residual particle mass was determined by colorimetric analysis in a spectrophotometer. Droplet size distributions were calculated from the amount of methylene blue dye collected on each impactor stage, the cut sizes of the Andersen impactor, the nebulized solution's concentration, and the residual particles' density. The effects of air velocity, liquid velocity, air nozzle diameter, and nebulizer baffle position on droplet size distribution were evaluated. Linear regression was used to develop an empirical relationship between key parameters and the mass median droplet diameter, d_{50} , as well as d_{16} and d_{84} . The model indicates that the median diameter increases from 9 to 22 μ m with decreased kinetic energy loss, decreased air nozzle exit area and, surprisingly, decreased distance separating the baffle and air nozzle.

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LIST OF NOMENCLATURE

Α	flow area of upstream atomizing airstream, m ²
c	concentration of the nebulized solution, g/cc
d ₁₆	16% of droplets are less than this diameter, μm
d50	median droplet diameter, as formed, µm
d ₈₄	84% of droplets are less than this diameter, μm
da	aerodynamic diameter of the dried droplet, μ m
dd	droplet diameter, µm
dp	diameter of the dry particle, µm
Da	air nozzle diameter, cm
D	liquid nozzle diameter, cm
D _{mean}	mean particle diameter of aerosol, μm , as used by Nukiyama and Tanasawa and by Wigg,
h	height of air annulus, cm
L	distance between air nozzle face and the liquid nozzle centerline, cm
MMD	mass median diameter of aerosol, μm , as used by Kim and Marshall
Qa	air flow rate, L/sec
QI	liquid flow rate, L/sec
Pa	air density, g/cc
P1	density of nebulized liquid, g/cc
٥p	density of the dry residual particle, g/cc
PI	liquid density (g/cc)

LIST OF NOMENCLATURE, CONTINUED

- σ_g geometric standard deviation
- σ₁ liquid surface tension (dyne/cm)
- μ₁ dynamic liquid viscosity (poise)
- P liquid kinematic viscosity, centistoke
- Va air velocity leaving the air nozzle, m/sec
- V1 liquid velocity leaving the liquid nozzle, m/sec
- Vr relative velocity, m/sec
- Wa air mass flow rate, g/sec
- W1 liquid mass rate, g/sec
- x distance between air nozzle face and the baffle, cm
- y radius of jet at distance x from nozzle, cm

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FACTORS THAT AFFECT THE SIZE DISTRIBUTION OF NEBULIZED FLUID

BACKGROUND

A nebulizer is a droplet generator that draws liquid from a reservoir through an orifice due to a low-pressure region created by high-velocity air. Nebulizers are used in medical and scientific research to generate mists from a bulk solution. The Laskin, Collison, DeVilbiss, Wright, Retec, and Lovelace nebulizers are most widely used. However, little information is available on the size distribution of the droplets nebulizers generate. Liquid parameters, such as viscosity, density, and surface tension, influence the droplet size distribution (Lefebvre, 1980). In many applications, these fluid parameters are fixed because aerosolization of a specific fluid is required. Furthermore, the air flow rate through nebulizers for inhalation therapy must remain within a certain range to provide optimum patient benefit. With the fluid characteristics and the flow fixed, there is no convenient way to adjust nebulizers to alter the size distribution of the aerosol produced.

Nukyiama and Tanasawa (1939), Gretzinger and Marshall (1961), Wigg (1964), Kim and Marshall (1971), Mullinger and Chigier (1974), Yang and Chin (1990), and others have evaluated other types of aerosol generators to determine the relationship between atomizer dimensions and the mean droplet diameter aerosolized. Air-blast atomizers, also called pneumatic nozzles or twin-fluid atomizers, are used extensively in the combustion industry. The most basic aerosolizer of this type is the plain-jet atomizer, which has separately controlled liquid and air flows through concentric nozzles. The liquid stream is shattered by its interaction with a high-speed gas stream. Several different nozzle configurations including prefilmers, internal mixing chambers, and concentric double air nozzles have been developed to achieve optimally sized fuel droplets for the combustion industry. Nukiyama and Tanasawa (1938), Wigg (1964), and Kim and Marshall (1971) have developed models to estimate the average droplet size generated by this nebulizer type. Lefebvre (1980) has summarized these and other models.

Nukiyama and Tanasawa:

$$D_{\text{mean}} = (585/V_r)^* (\sigma_1/\rho_1)^{0.5} + 597[\mu_1/(\sigma_1^*\rho_1)^{0.5}]^{0.45*} (1000Q_1/Q_a)^{1.5}$$
(1)

Wigg:

$$D_{\text{mean}} = 200^{*}(\nu)^{0.5} W_{1}^{0.1}(1 + W_{1}/W_{a})^{0.5} h^{0.1} \sigma_{1}^{0.2} [\rho_{a}^{0.3}V_{f}]$$
(2)

Kim and Marshall:

$$MMD = 5.36*10^{-3} [\sigma_1^{0.41*} \mu_1^{0.32}] / [(\rho_a^* V_r^2)^{0.57} A^{0.36} \rho_1^{0.16}] + 3.44*10^{-3} [\mu_1^2 / (\sigma_1^* \rho_1)]^{0.17*} (W_a / W_1)^{m*} (V_r)^{-0.54}$$
(3)

where m=-1 if Wa/W1<3

$$m=0.5$$
 if $W_a/W_1>3$

The units for the terms in these dimensional equations are given in the nomenclature.

These models indicate that the average droplet diameter decreases by decreasing the liquid surface tension (σ_1), decreasing the liquid viscosity (μ_1), or increasing the liquid density (ρ_1). This diameter also appears to be related to a ratio of liquid to air flows, either volumetric or mass, raised to a power. The physical characteristics of the atomizers do not appear to affect greatly the mean diameters, as only the height of the annulus (h) and the flow area of the upstream atomizing airstream (A) appear in these models.

Although concentric nozzle configurations are used widely in the combustion industry, most nebulizers in medical and aerosol research applications have perpendicular air and liquid streams. This study was undertaken to evaluate the effects of altering the air velocity, liquid velocity, air nozzle diameter, and baffle position of a nebulizer with perpendicular air and liquid streams on the size distribution of droplets the nebulizer produces. Since only one liquid solution was nebulized, the effects of liquid density and surface tension were not evaluated.

APPARATUS

Figure 1 presents a schematic diagram of the nebulizer used here. The nebulizer was comprised of two fluid nozzles set perpendicular to each other. A baffle was placed perpendicular to the axis of the air jet and downstream of the liquid injection port to separate large droplets from the aerosol. This baffle was either the wall of the drying compartment or flat disk placed closer to the atomization point.

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Nozzle diameters were based on the dimensions of a Laskin nebulizer as reported by Drew et al. (1978). Three interchangable air nozzles were evaluated, the smallest diameter being roughly half and the largest being twice the diameter of the Laskin nozzle, 0.089 cm. The liquid nozzle diameter was fixed at 0.178 cm.

The position of the air nozzle relative to the liquid nozzle was determined by jet theory and experimentation. Theory states that the centerline velocity of a jet is 98% of the velocity at the jet face when $L=4.44D_a$ (Baturin, 1974). Here, the L/D_a was held constant so that with changing nozzles similar dynamics were ensured for each nozzle; thus, the L dimension was changed as different nozzle diameters were evaluated. The resulting L/D_a values for the small, base, and large nozzles were 3.47, 3.57, and 3.57, respectively. The centerline velocities at these distances were essentially the same as the jet velocity as it exited the nozzle. Since velocity, and hence kinetic energy, of a jet is maximum at its centerline, the two nozzles were aligned so that the air nozzle centerline would lie flush with the tip of the liquid nozzle.

Flat circular disks were used as baffles and were set at distances from the air nozzle of ten and twenty times the air nozzle diameter, $x=10D_a$ and $x=20D_a$. The value of $x=10D_a$ was taken as a reasonable upper limit for the distance between an impaction jet and target according to Mercer (1973), whereas the value of $x=20D_a$ represents a value beyond this limit, for which the effect of impaction should diminish. The diameters of the baffles were based on jet theory. The radial distance (y) that the jet of

air spreads as it leaves the orifice is given by $y/D_a = 2.2$ for $x = 10D_a$ and $y/D_a = 4.5$ for $x=20D_a$ (Baturin, 1974). Here, baffle radius was set to a minimum of twice the jet radius. Table 1 provides the x/Da and baffle radius values used for this study. In addition, the effect of no baffle was determined. In this case, the distance between the nozzle face and the closest perpendicular surface was 12 cm, the distance to the wall of the drying chamber.

	T	at	le	: 1	L	
2	ff	0	D	-		

D _a (cm)	x/D _a	radius (cm)
0.046	10 20	0.244 0.476
0.089	10 20	0.476 1.746
0.178	10 20	1.746 1.746

The chamber volume was selected based on estimated drying time of the aerosolized droplets. According to Green and Lane (1964), a 30 μ m water droplet will evaporate in 19.2 seconds at 20^oC and 80% relative humidity, and according to Lefebvre (1980), a 10 μ m particle will evaporate in 1 second in 90% relative humidity. Since the experiments in this study were conducted at temperatures between 20 and 22^oC and at relative humidities between 12 and 46%, drying times less than 19.2 seconds were expected for the maximum droplet size expected. For an air flow of 28.3 Lpm and a chamber volume of 13.8 L, average residence time was 29 seconds and should have been sufficient to dry the droplets to particles.

EXPERIMENTS

Table 2 summarizes the conditions under which tests were run. Seven conditions were tested for each of the three baffle positions, with at least one duplicate run for each condition. Only one of the three variables (D_a, V_a, V_l) was varied in each run, while the other two were held at the base values. The values for the velocities were established by determining the operating ranges of the system. Maximum air velocity was set to the highest value possible through the smallest diameter nozzle, limited by a maximum pressure of 15 psig through the system. Minimum air velocity was determined by the lowest value through the largest nozzle that would nebulize the liquid stream.

Figure 2 shows the setup of the nebulizer and chamber. The nebulizer was connected to compressed air and the liquid fed to the apparatus using a syringe pump. The nebulizer was contained entirely by the drying chamber with Tygon connections plumbed through the chamber walls.

The base liquid velocity was determined by the amount of liquid that was naturally aspirated by the base nozzle at the base air flow. This natural liquid aspiration rate was 0.18 cc/sec; the corresponding liquid velocity was 0.052 m/sec. Velocity was then decreased and increased by a factor of 2.7 to 2.8. Slower liquid flows could have been achieved since the liquid was metered into the apparatus via a syringe pump.



Figure 2: Experimental Set-Up

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Run	D _a (cm)	L (cm)	V _a (m/sec)	V ₁ (m/sec)
1	0.089 (Base)	0.318 (Base)	125 (Base)	0.052 (Base)
2	0.089 (Base)	0.318 (Base)	75 (Low)	0.052 (Base)
3	0.089 (Base)	0.318 (Base)	200 (High)	0.052 (Base)
4	0.089 (Base)	0.318 (Base)	125 (Base)	0.019 (Low)
5	0.089 (Base)	0.318 (Base)	125 (Base)	0.149 (High)
6	0.046 (Low)	0.159 (Low)	125 (Base)	0.052 (Base)
7	0.178 (High)	0.635 (High)	125 (Base)	0.052 (Base)

Table 2 Experimental Conditions for Each Baffle Position

However, the range of liquid flows used better represents the flow that would occur if the nebulizer siphoned liquid from a reservoir. The reservoir method of liquid delivery was not attempted because a known liquid flow was desired; further, the effect of concentrating solution in the reservoir was avoided. Significant amounts of the liquid were not nebulized but rather ran down the side of the nozzle, primarily at the high metering rates. The nebulized liquid was an aqueous solution of 14 g of sodium bicarbonate and 1 g of methylene blue dye in 1000 mL of deionized water.

The nebulizer was usually run for six minutes; however, longer aerosolization times were required with small liquid flows, and shorter times were necessary with large liquid flows. After the aerosolization period, the liquid and air flows to the nebulizer were stopped, but the flow through the impactor continued for another six minutes to purge the chamber of all residual particles. This allowed more than 12 air changes in the chamber and was sufficient to collect all residual particles.

A seven stage Andersen impactor was inverted and attached at the top of the chamber; the impactor fitted tightly and was sealed to the chamber with clay to ensure that all air entering the impactor would come from the drying chamber. The plates used in the impactor were coated with approximately 0.8 mL of a 1:100 silicone grease in hexane solution to reduce particle bounce. Used plates were removed from the impactor, placed in petri dishes, and washed with a known volume of deionized water to dissolve the methylene blue particles. The solution was analyzed with a Gilford/Beckman spectrophotometer at 660 nm. From a calibration curve, the concentration was related to the absorbance (concentration=0.079*absorbance, $R^2=0.998$) and used to determine the mass of particles on each plate.

To determine the droplet size distribution, an analysis of dried particles was performed similar to the method of Gretzinger and Marshall (1961). The diameters of residual particles collected on the impactor were defined by the cut diameters of each stage. Then, with knowledge of the concentration of the nebulized solution and the density of the residual particles, the size of the generated droplets was determined using (Mercer et al., 1968):

$$d_d = d_a [c]^{-1/3} [\rho_p]^{-1/6}$$
(4)

These calculations assume the collected particles were solid spheres. The flow of 28.3 Lpm was drawn through the impactor by vacuum. Since the air flows through the nebulizer were between 1.2 and 18.6 Lpm, two 1 cm holes were drilled in the chamber to provide make-up air to the system.

Cumulative size distribution curves for the generated droplets were used to determine the mass median diameter (d_{50}); d_{16} and d_{84} were used to analyze the spread of the distributions.

RESULTS

Table 3 summarizes the d_{16} , d_{50} , and d_{84} values for each run. Figure 3 provides the cumulative size distribution for the Run 1 condition (Base D_a , Base V_a , Base V_l) at a baffle position of $10D_a$, representative of the tests performed.

DISCUSSION

Terms of interest in evaluating the size distributions included those from Eqs. (1-3). The loss of kinetic energy, a function of $V_a^2/(1+W_l/W_a)$, appears in several equations for the mean and median droplet diameter (Wigg, 1964). As kinetic energy increases, more energy is available to break the liquid into droplets; hence, one would predict smaller droplets with more kinetic energy. The Weber number, a dimensionless parameter (We= $\rho_a V_r^2 d_d / \sigma_1$, with V_r = relative velocity and d_d = dropet diameter)

Baffle	Run	d ₁₆ (μm)	d ₅₀ (μm)	d ₈₄ (μm)
None:	4			
	1	7, 5.5, 6.2, 6.2	12.5, 10.5, 12,12	20.5, 16.5, 19, 19
	2	28.5, 9.4, 9	17, 16, 17.5	23, 23, 18
	3	4.7, 3.7, 4.6	10, 7.5, 9	18.5, 15.5, 1
	4	5.9, 5.6, 8	12, 11.5, 14	20, 16.5, 25
	5	5.8, 6, 6.2	10.5, 12, 12	14.5, 16.5, 1
	6	9.5, 10	15.5, 16.5	23, 24
	7	5.4, 5.4	9.4, 9.4	14.5, 14.5
at 20D _a :	1	7.9*	15.5*	24.5*
	2	7, 7.5	15, 17	24, 25
	3	5.8, 5.8	14.5, 14.5	22, 22
	4	5.3, 5.2	16, 13.5	36, 26
	5	7, 5.9	17.5, 16	29, 31
	6	11, 11.2	17.5, 18	24.5, 25.5
	7	4.6, 4.4	9,9	19, 18
at 10D _a :		10.05	00.17	05.07
	1	12, 9.5	20, 17	35, 27
	2	12, 13.5	19, 19	28, 27
	3	8, 6.2, 6.2	19, 12, 14	40, 18, 24
	4	11, 12, 12	22, 22.5, 20.5	46, 42, 36
	5	9.8, 9, 7.5	16, 14.5, 14.5	25, 21.5, 22
	6	11.5, 11.5, 11.5	18, 17, 18.5	28, 25, 25
	7	6.5, 7	14, 14	20, 21.5

Table 3 Experimental Results

The duplicate run for this condition was an outlier and was not used.





relates the droplet diameter to velocity squared. With the nebulizer, the relevant velocity is that of the air, so d_{50} was evaluated as a function of V_a^{-2} . Another parameter evaluated was obtained from impaction theory, which suggests that $d_{50}=f(D_a^3/Q_a)^{0.5}$. Additional parameters evaluated include the ratio of nozzle diameters (D_a/D_l) , the ratio of baffle distance to air nozzle diameter (x/D_a) , and the mass flow ratio (W_l/W_a) .

Thus, the following values were hypothesized to affect d₅₀ and were investigated:

$$d_{50} = f[(V_a^2/(1+W_l/W_a), V_a^{-2}, (D_a^3/Q_a)^{0.5}, D_a/D_l, x/D_a, W_l/W_a, Q_a, Q_l]$$

Median Diameter

Since the same fluid was used throughout this study, the effects of fluid properties such as viscosity, density, and surface tension were not considered. Furthermore, the liquid nozzle diameter was fixed at 0.178 cm throughout the study, so that the D_a/D_l term depended only on D_a .

Linear regression was used to develop a relationship between the parameters studied and the median droplet diameter. Both linear and exponential models were investigated. The terms that had the best agreement between measured and estimated d_{50} included the kinetic energy loss term ($V_a^2/[1+W_l/W_a]$), the air nozzle diameter, and the baffle position term (x/D_a) . Optimization of this model was performed to determine the exponents for these terms that maximized R². The best equation was:

$$d_{50} = (17.3 \pm 0.7) - (47.8 \pm 7.6) [V_a^2/(1 + W_1/W_a)]^{0.5} - (132 \pm 33) D_a^2 + (54 \pm 6.4) (D_a/x)$$

$$R^2_{adi} = 0.742$$
(5)

with d50 in µm, Va in m/sec, Da and x in cm.

Figure 4 is a plot of expected versus measured d_{50} . The ranges of values for terms in this model are in Table 4. The equation states that the mass median diameter of generated droplets is a function of the loss of kinetic energy to the 0.5 power, the area of the air nozzle, and the inverse of the distance between the baffle and the nozzle.

Term:	47.	${V_a^2/(1+W_l/W_a)}^{0.5}$	132D _a ²	54(D _a /x)
Range inv	vestigat	ed	5. A. A.	
in this stu	dy:	0.33 - 1.16	0.28 - 4.17	0.2 - 5.4

Table 4 Range of Values in d₅₀ Model

The first term states that as more energy is available the liquid is broken up into smaller droplets. This term has a minimal affect on the d_{50} in that its magnitude and spread of its contribution is relatively small, as seen in Table 4. Wood reported that the mean aerosol diameter was related to the square root of $(1+W_l/W_a)/V_a^2$ (unpublished research, quoted by Wigg, 1964), as was found here.



Figure 4: Estimated versus Measured d50

Gretzinger and Marshall (1961), Wigg (1964), and Claire (1954) have found that the nozzle diameter does not greatly affect the size of the droplets generated by air-blast atomizers. In contrast, Mullinger and Chigier (1974) stated that increasing D_a results in an increase in spray size. However, this study found that the particle diameter decreased with increasing D_a . The variability in the contribution by this term is greater than that of the kinetic energy term.

The effect of baffle position is counter-intuitive. One would expect the baffle to remove the larger droplets while the smaller droplets would pass the baffle unaffected; thus, one would expect d_{50} to decrease as x decreased. However, liquid collected by the baffle flowed to its outer edges where the liquid may have been re-entrained or re-atomized. The closer the baffle, the more pronounced this effect. Thus, the smallest d_{50} values were achieved when re-atomization and re-entrainment were least, namely when the baffle was the farthest away. Results might have been different if the baffle had a different shape.

Spread of the Distribution

The cumulative size distributions were not log-normally distributed. When plotted on log-probability paper, the cumulative distributions appeared to have two linear portions (Figure 3). A greater slope was found for the smaller particles indicating a smaller geometric standard deviation, and a smaller slope was found for larger particles indicating a larger geometric standard deviation.

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To address the spread of the distribution, d_{16} and d_{84} were evaluated as a function of d_{50} . First-order power functions were fit to the data to develop equations for d_{16} and d_{84} :

$$d_{16}=0.534*d_{50}$$
 $\sigma_{g)Lower}=1.88$ (6)
 $d_{84}=1.62*d_{50}$ $\sigma_{g)Upper}=1.62$ (7)

Figures 5 and 6 are plots of expected versus measured d₁₆ and d₈₄.

Mercer et al. (1969) state that the geometric standard deviation of a nebulized aerosol has little variation. The best correlation between the experimental conditions and d_{16} or d_{84} was between these diameters and the median diameter, d_{50} . Like Mercer et al., this study found little variability in the dispersion of droplets produced by atomization.

CONCLUSIONS

The study revealed that decreasing the diameter of a droplet produced by a nebulizer can be accomplished by increasing the kinetic energy of the system, increasing the air nozzle diameter, or moving a flat baffle further away from the point of aerosol generation. The nebulizer was capable of producing droplets with mass median diameters ranging from 9 to 22 μ m without altering the liquid nebulized.

The work reported here is relevant to nozzle sizes and fluid flows appropriate for small nebulizers and can be useful for medical work, aerosol research, and other



Figure 5: Estimated versus Measured d16



Figure 6: Estimated versus Measured d16

applications where small droplets must be produced. The flow dynamics of an enclosed nebulizer would be different than the open system evaluated in this study, and the effect of the parameters evaluated here might be different.

Additional work should be undertaken to evaluate the effect of baffles with different shapes, for example spheres, cones, or tear-drop shapes. The flat circular baffle tested in this study created a source for generation of larger droplets at the edges that might be eliminated by using a baffle of a different shape.

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APPENDIX

CUMULATIVE SIZE DISTRIBUTION

This appendix contains the summary size distribution curves for all of this study's experiments. The conditions of the experiment are provided in the title by the code which identifies the nozzle diameter, air velocity, liquid velocity, and baffle position, respectively. For example, B,H,B-10 refers to the test condition with base nozzle diameter (0.089 cm), high air velocity (200 m/sec), and base liquid velocity (0.052 m/sec) with the baffle position at $10D_a$. Replicates of each run condition are included on the same graph, with the legend indicating the sample identification. The d₅₀, d₁₆, and d₈₄ droplet sizes were taken from these curves.





1-3



Cumulative Size Distribution for B,B,H-10d



Cumulative Size Distribution for B,B,H-20d















Cumulative Size Distribution for B,H,B-10d





















