Particle Nucleation in Turbulent Gas Jets

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Experiments were conducted to test the theory of nucleation-controlled growth of spherical particles in a turbulent jet. Dibutyl phthalate (DBP) particles were formed in a bench-scale jet apparatus with nozzle diameters 0.235 cm and 0.375 cm. Size distributions and number concentrations of the particles were measured at different DBP vapor concentrations, jet velocities, and positions. There is evidence that the DBP particle nucleation was confined to the shear layer of the jet and that the particles grew by condensation as they moved away from the nozzle. Trends in the data suggest that for low rates of particle formation in jets, the final aerosol concentration can be predicted from simple scaling laws.

Introduction

Homogeneous nucleation is a temperature- and concentration-dependent process that frequently occurs in turbulent flow. Examples of particle formation by nucleation are emissions from industrial, vehicular and aircraft sources, and process gas mixing. The complexity of turbulent flow fields has made it necessary to make simplifying assumptions, often oversimplifications, in predicting nucleation rates. Turbulent fluctuations have frequently been neglected and calculations based on mean velocity, temperature, and concentration profiles. As methods have been developed to incorporate turbulent fluctuations into models, the effects of these fluctuations have been found to be significant.

A condensable vapor at high temperature may become supersaturated as it undergoes turbulent mixing with a gas at lower temperature. When the vapor attains only moderate saturation ratios such that rates of nucleation are in the range 10^4–10^8 particles/cm^3 s, the resulting particle number concentrations are low. Particle–particle collisions are infrequent; growth is by heterogeneous condensation and the particle concentration changes only by dilution. In such cases, the growth is said to be nucleation-controlled because the final particle size and concentration are determined by the rates and duration of homogeneous nucleation in the initial mixing stage.

This study focuses on a specific flow system of great practical interest, the turbulent jet. As shown in Figure 1, the flow pattern of a turbulent jet has two distinct regions, the initial and similarity regions. The initial region consists of the potential core, a cone of undisturbed nozzle fluid, surrounded by the shear layer. For aerosol formation studies, the shear layer is very important since it is where incipient turbulent mixing, supersaturation, and homogeneous nucleation occur.

Hidy and Friedlander (1964) showed qualitatively that processes occurring in the shear layer of a jet strongly affect particle formation by homogeneous nucleation. Lesniewski and Friedlander (1995) hypothesized that particle formation occurs in the shear layer of the jet, but is quenched as the particles move down the axis. Quenching may occur by three mechanisms: (1) the condensable vapor is diluted by ambient fluid entrained into the jet; (2) the number concentration of newly formed particles grows large enough that condensation suppresses further nucleation; or (3) most of the condensable vapor is depleted by the particle formation.

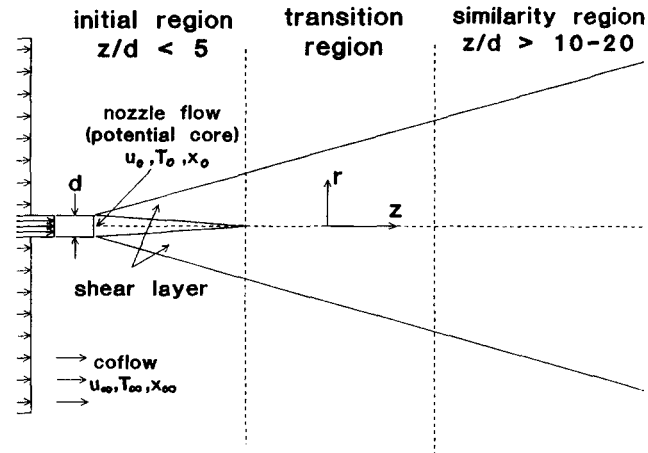

Figure 1. Free turbulent jet (after Abramovich, 1963).
Initial region of the jet extends to $z/d \sim 5$.

Theoretical Background

Lesniewski and Friedlander (1995) derived an expression for the rate of homogeneous nucleation in a fluctuating flow:

$$\bar{I} = \int_0^1 I(\theta, T_0, T_\infty, x_0, x_\infty) \cdot P(\theta)\, d\theta, \qquad (1)$$

where θ is the dimensionless temperature and concentration, I is the instantaneous nucleation rate (taken from classic theory or experimental data), and $P(\theta)$ is the probability density function (PDF) for the turbulent flow. Equation 1 is an advance in nucleation rate calculations, since it accounts for the effects of turbulent fluctuations on the particle formation rate.

Experimental studies (Sreenivasan et al., 1977; Konrad, 1977; Batt, 1977; Broadwell and Mungal, 1991) have shown that mean and rms fluctuation values of temperature and concentration scale with position in turbulent shear layers. Shapes of the measured PDFs were similar across the shear layer in the studies. Konrad (1977) found that the mean and rms fluctuating values were independent of Reynolds number. Lesniewski and Friedlander (1995) used PDF data taken from the literature in their calculation.

The total rate of particle formation in the shear layer is given by the volume integral of the nucleation rate. In the limit of negligible coagulation and vapor depletion, the total number of particles formed per time in an axisymmetric shear layer is (Lesniewski and Friedlander, 1995)

$$\bar{Y} = \frac{2\pi z^3}{3} \int_{\eta_1}^{\eta_2} \bar{I}\eta\, d\eta, \qquad (2)$$

where z is the axial distance, and η is a dimensionless position variable (η_1 and η_2 define the edges of the shear layer). Thus the total number of particles formed per time in the shear layer is predicted to be independent of the jet velocity and proportional to z^3. Since the PDFs are independent of Reynolds number (Re), the particle formation rate is a function only of $T_0, T_\infty, x_0, x_\infty$, and material properties of the condensate.

Lesniewski and Friedlander (1995) calculated the rate of formation of water droplets by homogeneous nucleation in the initial region of a turbulent jet. They compared nucleation rates in the presence of turbulent fluctuations with calculations based on the mean temperature and concentration profiles in the mixing layer. Turbulence significantly affected the number distribution of particles formed by homogeneous nucleation in the shear layer. Across most of the mixing layer, fluctuations led to an increase in the local average nucleation rate relative to calculations based on mean concentration and temperature profiles. But in the region where mean values correspond to the maximum nucleation rate, fluctuations caused the rate to decrease. For the conditions of the calculations, turbulence had a smaller effect on the overall rate of particle formation.

For a turbulent jet, the length of the shear layer is proportional to the nozzle diameter, d. If nucleation is confined to the shear layer, then from Eq. 2 the particle formation rate is proportional to d^3 and independent of u_0. Since $Q \sim u_0 d^2$ and $\bar{Y} = N_s \cdot Q$, we know that $N_s \sim d/u_0$, where Q is the volumetric flow rate of the jet and N_s is the particle concentration in the gas exiting the shear layer. Thus, for a given nozzle vapor concentration and axial position, Nu_0/d should be independent of u_0 and d. Moreover, since the particle concentration downstream of the shear layer changes only by dilution, $N \sim N_s d/z$, the group $Nu_0 z/d^3$ should be constant at any point on the jet axis for a given set of initial gas stream temperatures and vapor concentrations.

Experimental Studies

The goals of this study were to determine the conditions under which nucleation-controlled growth occurs, and to test the predictions outlined in the previous section. The experimental system is shown in Figure 2. A heated, filtered stream of nitrogen (99.9%, <100 ppm H_2O) was passed through a bubbler containing liquid dibutyl phthalate (DBP, 99.9+%, Sigma Chemical Company). The gas stream was again filtered to remove DBP particles formed in the bubbler. The vapor-phase concentration to DBP was continuously monitored by pulling a small side stream of gas through a flame ionization detector (Detector Engineering Technology, Walnut Creek, CA). In previous nucleation studies (Higuchi and O'Konski, 1960; Brock et al., 1986; Okuyama et al., 1987; Strum and Toor, 1992), the vapor concentration was not continously monitored. For this system, on-line measurement of the DBP vapor concentration decreased the uncertainty in the particle concentration by at least a factor of 2. The DBP mole fraction ranged from 1×10^{-4} to 5×10^{-4} in the trials.

The jet entered the nucleation chamber through a small cylindrical nozzle. Two nozzles were used in the study, 0.235 and 0.375 cm in diameter. The nozzle flow temperature was 140°C, and its flow rate ranged from 10 L/min to 20 L/min, corresponding to Reynolds numbers of 4,700–10,000. The chamber was a 6-in. (152-mm) diameter by 18-in. (457-mm) high glass cylinder. Filtered nitrogen entered the chamber concentric to the nozzle at 200 L/min and 26°C. A uniform velocity was achieved by passing the coflow through a packed bed of glass spheres. Background particle concentrations were $<1/\text{cm}^3$, and $x_\infty = 0$.

As the hot nozzle fluid mixed with the cool nitrogen, DBP particles formed by homogeneous nucleation. A sample (1

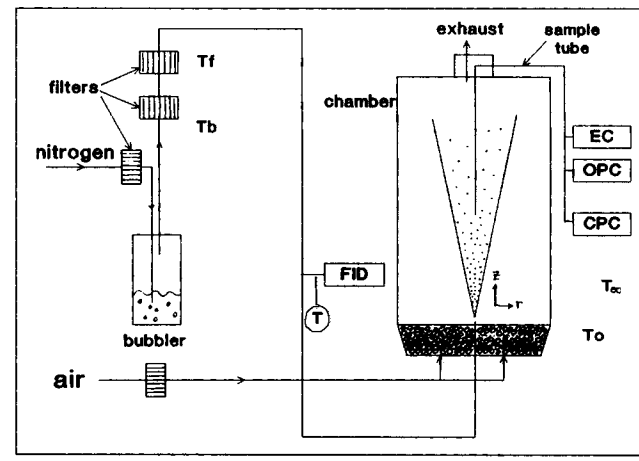

Figure 2. Experimental apparatus.

L/min) was removed from the jet through a probe and its particle concentration measured with a Condensation Particle Counter (TSI model 3010). By moving the probe, the particle concentration in the region downstream of the shear layer could be mapped as a function of position in the jet.

For aerosol-size measurements, a dilution probe was used to prevent particle growth in the sampling tube. Sampling effects were minimized for dilution flow rates of 4–10 L/min and by setting the dilution gas temperature approximately equal to the sample temperature. An Electrostatic Classifier (TSI model 3071) and an Optical Particle Counter (Royco model 226) were used for sizing. For DBP particle number concentrations above 100/cm^3, agreement between data obtained from the two instruments was quite good. The Classifier was less accurate for low particle concentrations. A complete size distribution consisted of Electrostatic Classifier data for $D_p < 0.14$ μm and OPC data for $D_p > 0.14$ μm.

Results and Discussion

Total aerosol number density

DBP particle sizes and concentrations were measured over a range of jet velocities and DBP vapor concentrations. This article gives an overview of the effects of vapor concentration and nozzle diameter on particle concentration. A more complete discussion of the experiments, with additional data on the effect of nozzle velocity and theoretical interpretations, is given by Lesniewski (1997).

As a check of the flow pattern of the turbulent jet, axial and radial velocity and temperature profiles were measured using a hot-wire anemometer and a thermocouple (Lesniewski, 1997). The measurements agreed well with classic jet theory (Abramovich, 1963). Figure 3 shows the measured DBP particle concentration as a function of x_0 on the jet centerline at $z/d = 20$, $T_0 = 140$°C, $T_\infty = 26$°C, and $Re = 4700$. Data observed on four separate days fall on a single curve, demonstrating excellent reproducibility. The abrupt kink in the curve shows that DBP nucleation is activated at $x_0 \approx 1.8 \times 10^{-4}$.

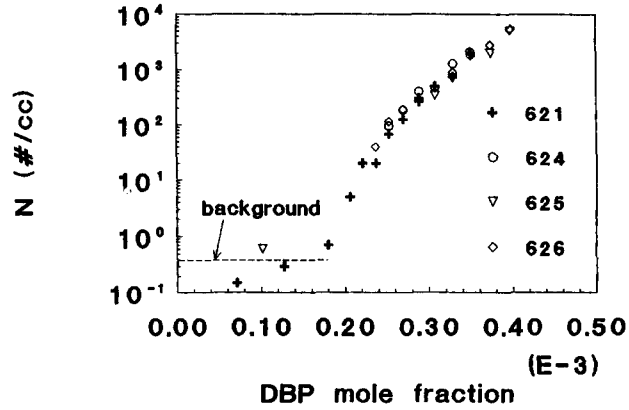

Figure 3. DBP particle concentration as a function of DBP vapor concentration (x_0).

Data from four different trials measured on the jet centerline at $z/d = 20$, $Re = 4{,}700$, $T_0 = 140$°C, and $T_\infty = 26$°C. Data observed on four separate days of experiments demonstrate reproducibility within a factor of 2. Break in the curve shows that DBP nucleation is activated at $x_0 \sim 1.8 \times 10^{-4}$; steep slope of the curve is characteristic of the nonlinear dependence of nucleation rate on the vapor concentration.

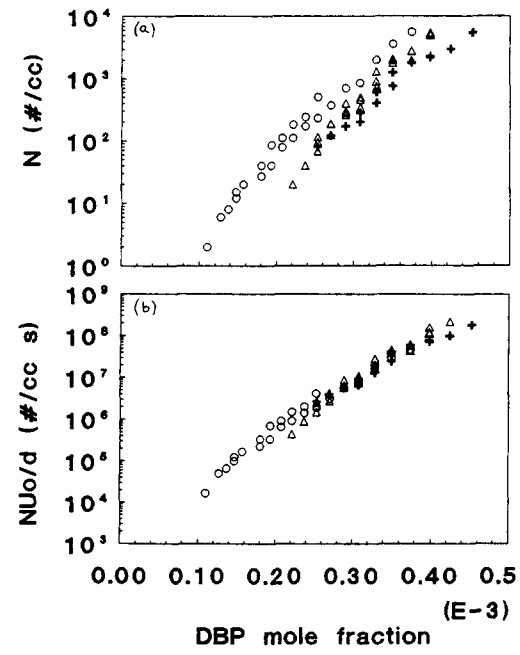

Figure 4. (a) DBP particle concentration vs. DBP vapor concentration for different nozzle diameters and jet velocities.

Measured on jet centerline at $z/d = 20$ with $T_0 = 140$°C and $T_\infty = 26$°C; (b) data from (a) replotted as Nu_0/d vs. x_0. The collapse of data at different values of d and u_0 demonstrates how particle formation scales with volume and residence time in the shear layer. Δ $d = 0.235$ cm, $Re = 4700$; + $d = 0.235$ cm, $Re = 7{,}100$; ○ $d = 0.375$ cm, $Re = 4700$.

The slope of the curve is also characteristic of homogeneous nucleation; small changes in the DBP vapor concentration cause large jumps in the resulting particle concentration.

In Figure 4a, data taken at $z/d = 20$ for different jet velocities and nozzle diameters are plotted as a function of DBP vapor concentration. At a given value of x_0, N decreases as d decreases and u_0 increases, matching the trends outlined in the previous section. For constants T_0, T_∞, and x_0, the group Nu_0/d is constant at any cross section of the jet for nucleation-controlled particle growth. The data of Figure 4a are replotted as Nu_0/d vs. x_0 in Figure 4b. When plotted in this way, the data collapse onto a single curve, suggesting that the DBP particle growth is nucleation controlled.

Another way to test for nucleation-controlled growth is to measure axial number concentration profiles. If nucleation occurs only in the jet shear layer, then for $z/d > 10$, the particle number concentration should change only by dilution. According to jet theory, dilution causes matter to spread like $(z/d)^{-1}$ on the jet centerline. Hence a log-log plot of N vs. z/d shows a slope of -1 if growth is nucleation controlled. A less steep slope means that particles form outside the shear layer or are reentrained in the jet. A slope steeper than -1 signifies that evaporation or coagulation occur in the system.

Axial number density profiles measured at different values of x_0 are shown in Figure 5. For reference, the theoretical profile (a line with slope -1) is drawn. The measurements agree with the dilution profile for $x_0 < 3.5e-4$, providing more evidence that particle growth is nucleation controlled. Deviations in the high number concentration profiles suggest

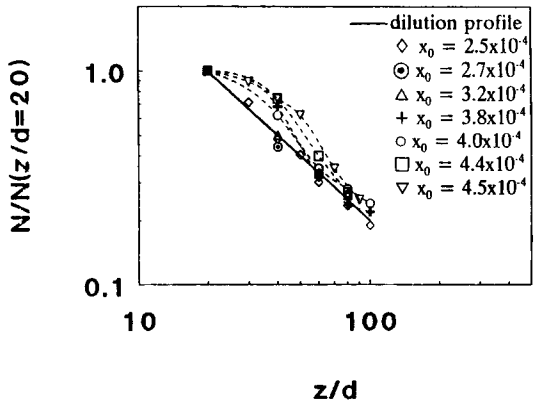

Figure 5. Axial number concentration profiles measured at various x_0 along the jet centerline.

For $x_0 < 3.5 \times 10^{-4}$, measurements agree with pure dilution profile. Observed data fall above the theory for $x_0 > 3.5 \times 10^{-4}$, suggesting that nucleation occurs past the shear layer.

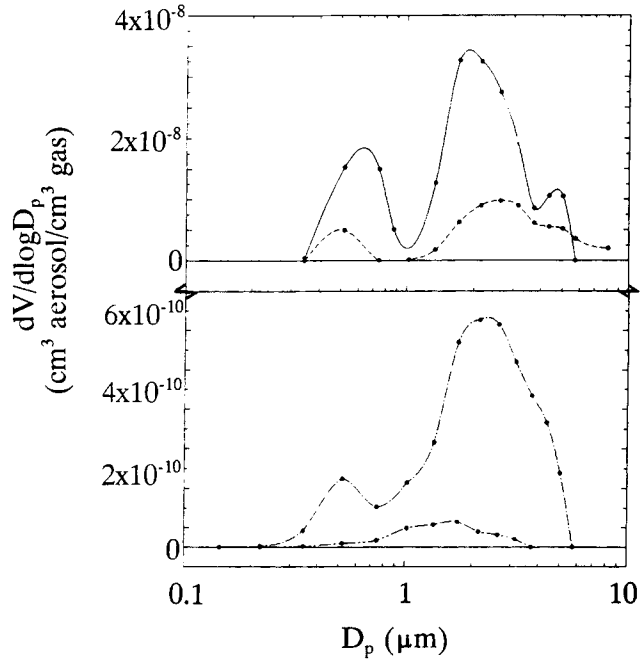

Figure 6. Evolution of the DBP particle-size distribution as x_0 increases.

Lines represent fits to data measured on jet centerline at $z/d = 20$ with $T_0 = 140$°C and $T_\infty = 26$°C. The area under each curve is equal to the total particle volume. At lower vapor concentrations, most of the aerosol mass is found in 1–3-μm particles, which have formed by nucleation in the shear layer and grown by heterogeneous condensation:-· ·-· ·-$x_0 = 2.5 \times 10^{-4}$. At slightly larger vapor concentrations, a submicron mode breaks out, the sign of the onset of particles formation downstream of the shear layer: -·-·-$x_0 = 3.1 \times 10^{-4}$. At intermediate vapor concentrations, the smaller mode grows: ------$x_0 = 4.3 \times 10^{-4}$. The effects of coagulation are seen at large vapor concentrations as the small and large modes grow together and a third mode appears, probably the result of collisions between large particles: — $x_0 = 4.9 \times 10^{-4}$.

that particle formation occurs outside the shear layer for $x_0 > 3.5 \times 10^{-4}$.

An important implication of Figures 4 and 5 is that the quantity $Nu_0 z/d^2$ is constant along the centerline of the jet. Hence in a nucleation-controlled system, the particle number concentration can be predicted at any position in the jet from this simple scaling law. For these experiments, with DBP as condensate, $T_0 = 140$°C and $T_\infty = 26$°C, the scaling law is $Nu_0 z/d^2 = 10^{31} \cdot x_0^{6.45}$, where $Nu_0 z/d^2$ is in #/cm^3 s and x_0 is the mole fraction.

Particle-size distributions

Figure 6 shows the evolution of the DBP particle-size distribution as x_0 increases. The mean diameter data are summarized in Figure 7. At low DBP vapor concentrations, the size distributions are unimodal with count mean diameters 0.4–0.5 μm and mass mean diameters ≈ 3 μm. The distributions are wide, with geometric standard deviations ≈ 1.7. As x_0 increases, the count mean diameter increases. At $x_0 \approx 3.5 \times 10^{-4}$, a second mode develops, suggesting that nucleation occurs outside the shear layer. At $x_0 \approx 4.5 \times 10^{-4}$, the two modes begin to move toward each other. The mass mean diameter decreases and the count mean diameter increases. Eventually, we expect that the distribution would again become unimodal as the asymptotic coagulation limit is reached. Coagulation-controlled growth in jets has been studied by Delattre and Friedlander (1978) and Koch et al. (1993).

Axial size profiles are shown in Figure 8. Steady growth by condensation is evident as the particles travel down the jet. The mass mean diameter and number concentration data can be used to calculate the total mass of particles at any cross section. Figure 9 shows the fraction of total DBP mass in the particulate phase as a function of axial distance from the nozzle. Also shown are the results of Brock et al. (1986) for DBP particle formation and growth in a laminar jet.

For the turbulent jet, less than 10% of the total DBP mass was in the particle phase at 100 nozzle diameters. At the same axial position, the aerosol mass fractions were much higher in the laminar jet; since the nozzle diameters were comparable in size but initial velocity was much lower in the laminar jets of Brock et al. (1986), equivalent axial positions correspond to longer residence times in the laminar jet. The particles have more time to grow by the time they reach a given axial position in a laminar jet than in a turbulent jet.

Effects of stream splitting

An important implication of the theory is that the total particle concentration in the jet is proportional to the nozzle diameter. This property can be exploited to decrease the overall particle formation rate by splitting a large nozzle flow into multiple smaller streams.

Consider a large jet stream ("1") split into n smaller jets with equal diameters. To compare the particle formation rates in the two systems, two additional relations between the streams are needed since their velocities are unknown. The first is that the total mass flow is conserved, $\dot{m}_1 = n \cdot \dot{m}_n$. A second relation, which was implemented experimentally, is that the Reynolds numbers of the large and small jets are the same, $Re_1 = Re_n$ (for the analysis to hold, both streams must be turbulent, $Re > 3{,}000$). Under these conditions, the systems are not dynamically similar; the smaller nozzles have a larger pressure drop.

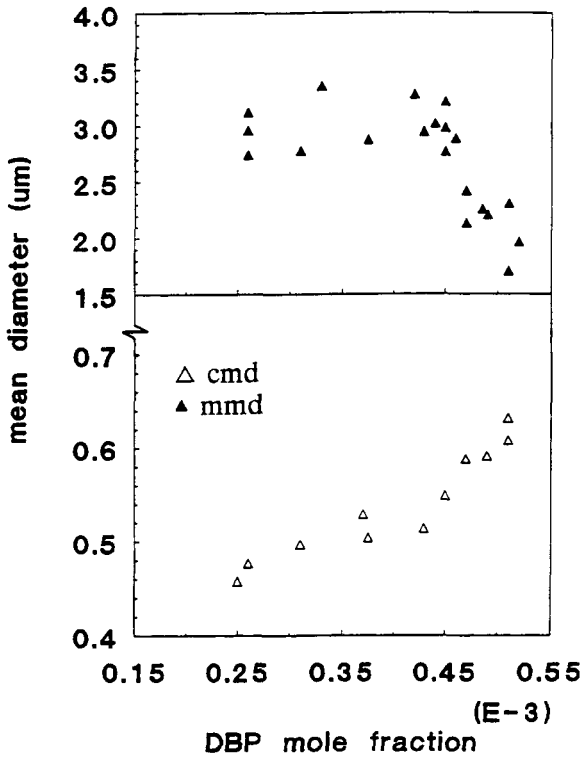

Figure 7. Count mean and mass mean diameters as functions of DBP vapor concentration.

Mean values are calculated from size distributions measured on jet centerline at $z/d = 20$. For $x_0 > 4.5 \times 10^{-4}$, the mass mean diameter decreases and count mean diameter increases as the modes grow together.

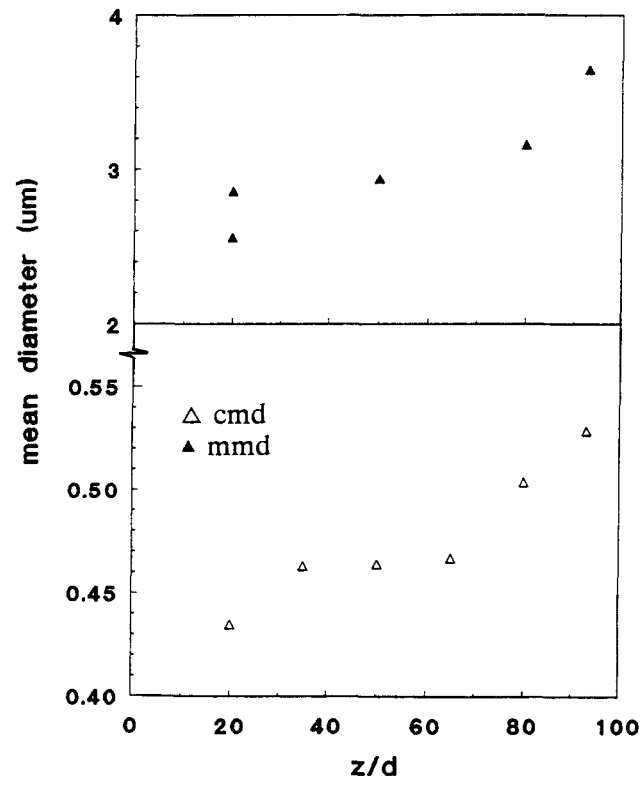

Figure 8. Axial profiles of count mean and mass mean diameters, calculated from size distributions measured on jet centerline for $x_0 = 3.6 \times 10^{-4}$.

Particles grow by heterogeneous condensation for $z/d > 5$.

Using the two relations, we find $d_n/d_1 = u_1/u_n = 1/n$. The total number of particles formed per time in the shear layers of the two systems are Y_1 and $Y_{n\text{tot}} = n \cdot Y_n$, respectively. Since the particles formation rate is proportional to d^3,

$$\frac{\bar{Y}_{n\text{tot}}}{\bar{Y}_1} = n\left(\frac{d_n}{d_1}\right)^3 = \frac{1}{n^2}. \tag{3}$$

The ratio of number concentrations in the two systems is also equal to $1/n^2$. Thus, particle formation in nucleation-controlled jets should be effectively reduced by stream splitting. By splitting a large jet flow into 10 smaller jets, in the case of constant Reynolds number, the overall particle formation rate is decreased by a factor of 100. The price paid for this reduction in particles is the higher pressure drop over the small nozzle array (Lesniewski, 1997).

The stream-splitting principle was tested experimentally by measuring the particle concentration in jets with two different nozzle diameters, keeping the mass flow rate and Reynolds number constant. As shown in Figure 4a, for the same DBP concentration and jet velocity, N decreases as d decreases. For example, at $x_0 = 3.5 \times 10^{-4}$ and $Re = 4{,}700$, the jet with $d = 0.375$ cm has $N = 5{,}400/\text{cm}^3$ and the smaller jet ($d = 0.235$) has $N = 2{,}100/\text{cm}^3$. The ratio of the two concentrations at equivalent throughputs is close to the predicted value $(0.375/0.235)^2 = 2.6$. Figure 10 shows that the stream-splitting correlation holds for $N(d = 0.235 \text{ cm}) > 70/\text{cm}^3$.

Summary

The first measurements are reported of particle nucleation in a turbulent gas jet with continuous vapor-phase monitoring, aerosol-size distribution, and the number concentration measurements at different positions in the jet. For the condi-

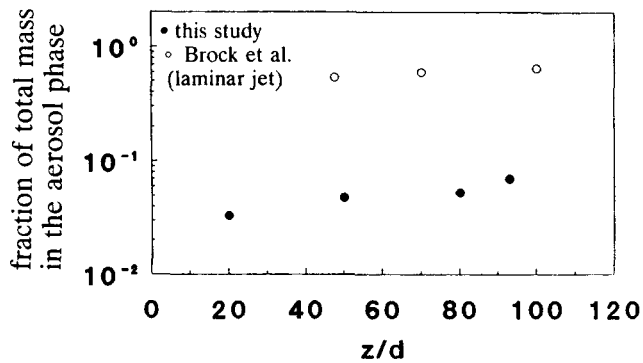

Figure 9. Fraction of total DBP mass in the particle phase vs. axial distance, calculated from the mass mean diameter and number concentration data measured on the jet centerline for $x_0 = 3.6 \times 10^{-4}$.

The data observed in the turbulent jet are about an order of magnitude lower than the data of Brock et al. (1986) for DBP nucleation in a laminar jet for $x_0 = 7.6 \times 10^{-6}$; in the laminar jet, equivalent axial positions correspond to much larger residence times.

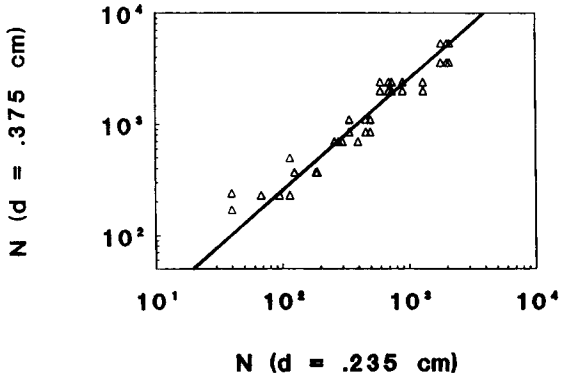

Figure 10. Particle concentration in jet with nozzle 0.375 cm vs. particle concentration in jet with 0.235 cm.

In agreement with Eq. 3 (solid line), $N(0.375)/N(0.235) = (0.375/0.235)^2 = 2.6$. Data shown are measured on jet centerline with $z/d = 20$ and $Re = 4{,}700$. Splitting the large stream into n smaller streams with equivalent total throughput decreases the overall particle formation rate and number concentration by $1/n^2$.

tions of this experiment, dibutyl phthalate (DBP) nucleation is activated at vapor concentrations, $x_0 \approx 1.8 \times 10^{-4}$. Trends in the number density data suggest that particle formation occurs primarily in the shear layer (first five nozzle diameters) of the jet for DBP vapor concentrations less than 3.5×10^{-4}. The group Nu_0/d is constant at any axial position in the jet, and for a given set of temperature and vapor concentration conditions, $Nu_0 z/d^2$ is constant at any position on the centerline.

The count mean and mass mean diameters for the particles varied between 0.4–0.7 μm and 1–4μm, respectively. Distinct changes in the shape of the particle-size distribution were observed as the vapor concentration increased. At low vapor concentrations, the size distribution was unimodal with geometric standard deviation about 1.7. The particles grew by heterogeneous condensation from a few nanometers in diameter in the shear layer to a few hundred nanometers at $z/d = 20$.

At DBP mole fraction of about 3.5×10^{-4}, a second distinct mode forms and the axial number concentration no longer follows a pure dilution profile. Particles in the larger mode form in the shear layer and grow by condensation. The smaller mode is thought to consist of particles formed by nucleation outside the shear layer. Hence the growth is no longer nucleation controlled for mole fraction $\geq 3.5 \times 10^{-4}$. At this vapor concentration, number concentrations at the end of the shear layer reach a level of about 5,000/cm^3.

The count mean and mass mean diameters are nearly constant until the vapor concentration reaches about 4.5×10^{-4}, corresponding to a number concentration of approximately 5×10^4/cm^3 in the gas exiting the shear layer. As x_0 increases further, the two modes grow together; the count mean diameter rises while the mass mean diameter decreases. This effect may be due to scavenging of the fine mode: increased collisions between the large and small particles. It is expected that if the vapor concentration would be increased even further, coagulation would begin to control the particle growth, and the size distribution would again become unimodal.

The results show that the particle concentration is proportional to the nozzle diameter. Hence the ratio of particle concentration in a single jet with large nozzle diameter to the concentration in an array of n smaller nozzles having the same throughput is n. This streamsplitting concept could be an effective strategy for reducing particulate pollution from jet systems by decreasing overall particle formation rates.

Acknowledgments

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Notation

D_p = particle diameter
I = nucleation rate expected in the presence of turbulent fluctuations
n = number of stream splits
T = absolute temperature
u = velocity
V = total aerosol volume per volume of gas
x = mole fraction
ν = kinematic viscosity of gas

Subscripts

0 = jet nozzle (hot) conditions
∞ = ambient (cold) conditions
s = shear layer exit conditions

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