The Morphology of Macroscopic Soot

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ABSTRACT. The morphology of soot collected from a laminar acetylene/air diffusion flame was studied. Collectors included both thermoplastic and impaction sampling from both the laminar and nonlaminar portions of the flow. The soot was viewed with both electron and optical microscopy. Cluster sizes ranged over three orders of magnitude from 50 nm to 400 μm to include some clusters visible to the naked eye. A new method of microscopic analysis, necessary when the clusters were large, was developed to account for the unresolved primary particles. Over the entire size range, the same fractal morphology was found with a fractal dimension of D = 1.8 and within a rather large uncertainty, the same prefactor k = 1.75. Thus, the fractal morphology of soot remains constant from clusters of about 10 primary particles per aggregate to macroscopic clusters of over 50* primary particles.


INTRODUCTION

Considerable effort has been made to study the morphology of soot aggregates that are produced by hydrocarbon flames (Medalia and Heckman, 1967, 1969; Forrest and Wit- ten, 1978; Sannous et al., 1981; Bocrat et al., 1986; Zhang et al., 1988; Magrini and Dobkins, 1990; Chang and Sorensen, 1991; Chatalamopoulos and Chung, 1991; Sorensen et al., 1992a, 1992b; Futri et al., 1993, 1995; Keshi et al., 1992, 1994a, 1994b, 1995;). Consequently, it is now well established that these aggregates are composed of small, or the order of a few tens of nanometers, primary parti- cles which are randomly-interstrated together. These primary particles, or monomers, are roughly spherical and mostly carbon in con- tent. The nondense clusters have a fractal morphology so that the number of primary particles per aggregate N scales with the overall size of the aggregate, which may be quantified by the cluster radius of gyration R, with a power D less than the spatial dimension of three as described by

\[ N = k_s \left( \frac{R}{a} \right)^D \]  

(1)

In Eq. (1), a is the primary particle radius, D is the fractal dimension, and k is the prefactor of the scaling relationship. A summary of past work is given in Table 1 where it is seen that the fractal dimension of the soot has typically been found to be equal to ~1.75. The ability to describe the soot aggregate morphology is important in a number of applications, including under- standing the soot effects, radiation transfer, and kinetics of growth.

Often, soot is emitted from a flame, and under certain circumstances, the individual clusters are large enough to be visible to the naked eye. Examples include soot from

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<table>
<thead>
<tr>
<th>Investigators</th>
<th>Soot Source</th>
<th>Method</th>
<th>( \mu )</th>
<th>( \delta )</th>
<th>Max Size</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mohila &amp; Heckeg (1967)</td>
<td>Carbon Black</td>
<td>TEM</td>
<td>1.5-1.6</td>
<td>13 ( \mu )</td>
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<td>-</td>
<td>-</td>
<td>-</td>
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<td>Marquis &amp; Johnstone (1983)</td>
<td>C(_2)H(_2) Diff.</td>
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<td>1.6-1.8</td>
<td>-</td>
<td>-</td>
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<tr>
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<td>1.4-1.8</td>
<td>-</td>
<td>-</td>
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<td>Zhang et al. (1988)</td>
<td>C(_4)H(_6)/O(_2) premixed</td>
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<td>1.72 \pm 0.16</td>
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<td>-</td>
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<tr>
<td>Ganguly &amp; Mukhopadyay et al. (1991)</td>
<td>C(_2)H(_2)/O(_2) premixed</td>
<td>LS</td>
<td>1.6 \pm 1.8</td>
<td>-</td>
<td>-</td>
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<tr>
<td>Chou &amp; Lee (1991)</td>
<td>C(_2)H(_2)/O(_2) premixed</td>
<td>LS</td>
<td>1.74 \pm 0.08</td>
<td>-</td>
<td>-</td>
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<td>Sisson et al. (1992a,b)</td>
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<td>TEM</td>
<td>1.97-1.97</td>
<td>-</td>
<td>-</td>
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<td>Kopyl &amp; Faeth (1993)</td>
<td>Many fuels</td>
<td>TEM</td>
<td>1.76-1.79</td>
<td>3 ( \mu )</td>
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<tr>
<td>Puri et al. (1983)</td>
<td>C(_4)H(_6) Diff.</td>
<td>TEM</td>
<td>1.74 \pm 1.8</td>
<td>1.5 ( \mu )</td>
<td></td>
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<td>Cal et al. (1993)</td>
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<td>TEM</td>
<td>1.70</td>
<td>-</td>
<td>-</td>
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<tr>
<td>Kopyl &amp; Faeth (1994a,b)</td>
<td>Many fuels</td>
<td>TEM</td>
<td>1.75-1.85</td>
<td>1.5 ( \mu )</td>
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<td>Cal et al. (1995)</td>
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<td>TEM</td>
<td>1.74</td>
<td>2.15 ( \mu )</td>
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<td>Kopyl et al. (1995)</td>
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<td>TEM</td>
<td>1.65</td>
<td>0.5 ( \mu )</td>
<td></td>
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<td>This work</td>
<td>C(_4)H(_6)/O(_2) Diff.</td>
<td>TEM</td>
<td>1.8</td>
<td>0.7</td>
<td>400 ( \mu )</td>
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</table>

acetylene-air diffusion flames, polystyrene flames, and diesel emissions. All of the previous studies summarized in Table 1 studied much smaller soot, typically submicron with the largest on the order of 10 \( \mu \)m. In the context of these studies, it is relevant to ask what the morphology of such large clusters is and how it is the same or different from its smaller precursors. This is of intrinsic interest since morphology is determined by the formation process and is at the foundation of understanding other physical properties, and practical value since this soot is released into the environment.

The purpose of this paper is to present a morphological study of soot obtained from both the luminous and nonluminous portions of a laminar acetylene flame in ambient air. The soot was exsiccated by both electron and optical microscopy, and ranged in size from aggregates of a few primary particles with 23 \( \mu \)m radius up to nearly millimeter-size aggregates, which is a four order of magnitude range. We find that all of this soot can be described as fractal with, remarkably, given the size range, the same fractal dimension of \( D = 1.8 \), and the same prefactor \( k_0 = 1.7 \). In addition, analysis of very large soot clusters for which the primary particles were not resolved required a significant correction which has not been previously described, and hence our analysis will be useful to future studies of very large soot.

EXPERIMENTAL METHODS AND RESULTS

The Flame
A simple diffusion burner was made from a 0.9 cm i.d. brass tube with a screen cap to eliminate flashback. Acetylene passed through this tube at a flow rate of 5.2 cm\(^3\)/s, hence an average flow velocity of 5.9 cm/s at room temperature. The diffusion flame above this tube burned in ambient air. The flame was a bright yellow-white near its base, duller to orange by a height of about 5 cm above the burner lip.
and, thereafter darkened further to a black, laminar flow by 8 cm. This laminar flow continued to approximately 15 cm where it broke into turbulence.

Soot Sampling

Soot was collected by two different methods: thermophoretic and impaction sampling. During the initial period of this work, thermophoretic sampling was used. Average cluster sizes found on these samples were 0.3 μm or less for all heights above burner. Visual observation of the flame at large height showed a graininess which suggested that larger clusters might be present. Thus, impaction was used with the hope that this method's bias for larger sizes would capture any large clusters that might exist; and, in fact, clusters larger by 23 orders of magnitude were found. We believe that these large clusters truly exist in the aerosol and are not artifacts of the impaction collection technique. We base this on the fact that the cluster density on the surface of the impaction sampler was low; thus, the chance for overlap of clusters to make a large cluster is low. In fact, the cluster density for both the impaction and thermophoretic sampling was essentially the same so any overlap problem, or lack of it, would be the same in each method. Furthermore, we expect for both methods that the clusters hit and stick on the probe; they do not move around on the surface, and continue to find each other and aggregate. Finally, we remark that since it is our purpose to study large soot and not to characterize the flame, a sampling bias is of no consequence.

Thermophoretic Sampling on TEM Grids.

Copper electron microscope grids with Formvar coating were placed on a "frog-tongue" probe device (Cai et al., 1993) designed after Debbas and Megaridis (1987). This device injects the grids into the flame for a residence time of 15 ms; the time required for the grid to traverse the flame on entrance and exit was ~3 ms. Grids were held with their face in the vertical plane (parallel to the flame gas flow) and injected at eight different heights above the burner from 1.7 to 25.4 cm. TEM micrographs of the grids were photogaphically enlarged to a net magnification of 20000 x. Figure 1a shows an example. The low surface density of clusters indicates that cluster overlap is not a problem. These photogaphs were scanned, and the digital representation of individual clusters was stored in a personal computer in a 16-level gray scale. The pictures were edited on the computer by visual comparison of the computer image to the photogaph in order to remove all unwanted darkness in the picture background. This image was then converted to a binary format, with the ultimate result that the background was represented as white and the clusters represented as black. Calibration of the pixel elements of the digitized pictures to real sizes was performed to find the conversion factor p = 12.1 nm/pixel side. Programs were written to calculate projected area and radius of gyration (see below).

The average monomer radius was determined by visual measurement of individual monomers and monomers in clusters with a 10 x magnifier equipped with a reticle. Only the thermophoretic flame gas flow, TEM viewed clusters were used in this analysis because the impaction sampled, optical microscope viewed clusters did not have resolved monomers. The monomer radius was measured at all heights above the burner studied. The average monomer radius at a given height ranged from a = 21-28 nm with a very slight height dependency which indicated a maximum monomer radius near a height of 6 cm. However, the combined uncertainty and statistical fluctuation in a was ±3 nm. Thus, this dependency is nearly within the uncertainty. This little or no height dependency is not surprising given the short extent of our luminous flame (~7 cm) and the large extent of our sampling (up to 25.4 cm). Most of the sampling was obtained from the post flame regime where the chemistry (surface growth of oxidation) is essentially done and physics (aggrega-
tion) is the cluster growth mechanism. Because of this, we will use a monomer radius average over all heights of $a = 23 \pm 3$ nm.

Impaction Sampling on Optical Microscope Slides. The sampling device was used again, but with standard glass microscope slides ($25 \times 75$ mm) held with their plane perpendicular to the flow of the flame (i.e., horizontally). Thus, impaction was the major collection scheme. Residence times were again 15 ms. Insertion of the slide face perpendicular to the flow no doubt perturbs the flow, but since our goal is to determine the morphology of macroscopic soot clusters, this perturbation is not a deterrance. Samples were taken between 3.8 and 17.8 cm above the burner.
Soot collected in this manner was significantly larger than soot collected via thermoderivatograph. In fact, some of the clusters were visible to the naked eye. Therefore, an optical microscope was used to take photographs of the impaction clusters. Obviously, the ~23-nm primary particles were not resolved. The net magnification to the photographic print was 72 X. Figure 1b shows an example. These pictures were also scanned into a digital format and visually edited for computer analysis. The conversion factor was p = 3.5 μm/pixel.

**Analysis of Projected Images**

A major problem in the analysis of the morphology of soot clusters lies in the fact that the three-dimensional structures are viewed as two-dimensional projections as a consequence of the microphotography. One way to overcome this is to view the clusters in at least two different projections, and with this stereo technique, reconstitute the true three-dimensional structure. This has been done in the past (Quinton et al., 1987; Koylu et al., 1993), but is untenable for our large soot clusters which have, as we shall see, as many as 10^10 primary particles, and for which the primary particles are not resolved. If, then, the analysis is limited to one projection, and if the density of this projection can give accurate information regarding the total mass along a given projection through the cluster, then a viable analysis of the three-dimensional morphology can be obtained. Such a mass-preserving image is difficult to achieve, however, because the attenuation of the electrons or light which record the projected image is not linearly related to the total mass of soot through which it passed. Furthermore, the response of the photographic film that captures the image in either case is linear only over a small range before it saturates and becomes insensitive to the mass of the cluster above it. This leaves projection of the soot cluster onto the two-dimensional plane into a binary format, a shadow, in which any part of the cluster is the same degree of black as any other, and the background is white. The advantage of this method is that it eliminates the response of the detector, and it is straightforward. What is needed is a quantitative method to convert two-dimensional information into three-dimensional information, and we present such a method below.

**Determination of N. Determination of N**

from the projected area of soot clusters has a long and well-established history (Medallia and Heckman, 1967, 1969; Simon et al., 1987; Megaridis and Dobbs, 1990; Koylu and Faeth, 1992; Cai et al., 1993, 1995; Koylu et al., 1992, 1994a, 1994b, 1995). In general it is found that

\[ N = k_c(A_c/A_p)^a \]  

(2)

where \( k_c \) and \( \alpha \) are constants near unity, and \( A_p \) and \( A_c \) are the projected areas of the cluster and primary particle, respectively. Medallia and Heckman (1967, 1969) first used this form, and found empirically \( k_c = 1.9 \) and \( \alpha = 1.1 \). This has subsequently been corroborated by a number of workers with \( \alpha \) varying by a few hundredths. Recently, Kacoji et al. (1995) analyzed both computer-simulated and real soot clusters, and found \( k_c = 1.15 - 1.16 \) and \( \alpha = 1.09 - 1.10 \). A minor problem with these results is that the limit as \( N \to 1 \) is not preserved because \( k_c \) is not unity. In another simulation, Meaklo et al. (1989) created DLCA clusters with \( D = 1.8 \) and \( N \) up to \( N = 10^5 \), larger than any in any other work that has compared \( N \) to the projected area. They fit their data with

\[ A_c/A_p = 0.4784N + 0.5218 \]  

(3)

This result is equivalent to Eq. (2) with \( k_c = 1.00 \) and \( \alpha = 1.10 \) over the range of \( N = 1 - 100 \) and \( k_c = 1.00 \) and \( \alpha = 1.094 \) over the range \( N = 1 - 1000 \). The slope of a log \( N \) versus log \( A_c/A_p \) graph is \( \alpha \), and Eq. (3) yields a slowly decreasing \( \alpha \) with increasing \( N \). This is consistent with the notion that, for clusters with \( D < 2 \), as \( N \to \infty \), \( N \) should be linear with \( A_c \), i.e., a 

. . asymptotically approaches 1.00, because the
cluster dimension is less than the dimension of the plane onto which it is projected.

Because of these complications, and since in our analysis of the TEM clusters, the range of the number of primary particles is 10 < N < 2000, we use \( t = 1.00 \) and \( a = 1.09 \). This is consistent with Eq. (3) and past work. For the optical clusters, the choice of \( t \) is not immediately evident because there is no a priori value for \( t \). There is no precedent for Eq. (2) in this regime. One option is to use the asymptotic expectation \( t = 1.00 \) and \( a = 1.09 \). This has the advantage of being the same as that used for the TEM clusters. Given the trend in Eq. (3), however, at large \( N \), this will be considered as an upper bound estimate on \( N \). We will also use Eq. (3) directly, which represents an extrapolation by a factor of two in \( N \) space, i.e., \( N \geq 18 \), beyond the data used to generate Eq. (3). Extrapolation can be dangerous, but Eq. (3) appears well behaved in that the overall slope it yields between \( N = 0 \) and \( N = 18 \) is \( a = 1.04 \).

\[ R_x = \sqrt{\frac{1}{N} \int r^2 \, dV} \]

Equation (4) is the radius of gyration of a three-dimensional body given by

where \( \rho(r) \) is the assumed spherically symmetric density. Factored clusters are not spherically symmetric, but for our analysis, we shall make the reasonable assumption that an ensemble of clusters on a TEM grid or microscope slide when viewed from one direction will yield an average spherical symmetry. Then, since \( r^2 = x^2 + y^2 + z^2 \), and since a projection onto a plane eliminates one of the dimensions, it follows from Eq. (4) that

\[ R_{x,3} = \sqrt{\frac{2}{3} R_x} \]

In (5), \( R_{x,3} \) is the true, three-dimensional radius of gyration of the cluster and \( R_x \) is that observed for the projected image.

The factor \( 3/2 \) results from the elimination of one of the three dimensions. Furthermore, Eq. (5) applies to a mass preserving projection. Equation (5) is verified by the computer simulations of Koyh et al. (1995), who found the empirical factor relating the two radii to be \( 1.24 \pm 0.01 \), in good agreement with \( (\sqrt{3}/2)^{2} = 1.25 \).

We have already discussed the difficulty in achieving a mass preserving projection, so we will use a two-dimensional binary representation of the true cluster. It is well established that the fractal dimension of true clusters is less than 2, typically, \( D \) is in the range 1.7-1.8. Thus, \( t \) might be expected to be that the projected image of a cluster onto a plane in a binary format would be mass conserving, i.e., no significant screening or modulation between monomers would occur. It should be stressed that this expectation is for asymptotically large clusters.

For finite size clusters, screening occurs as demonstrated, by Eq. (2) and the empirical fact that \( a > 1.0 \). Thus, the effective fractal dimension in the two-dimensional plane of the binary projection should be different from the fractal dimension of the real, three-dimensional cluster. In the immediately following argument, we will call these fractal dimensions \( D_2 \) and \( D_3 \), respectively.

Consider how the three-dimensional cluster is projected onto the two-dimensional plane. With spherical or circular symmetry, we assume that the density profile of either the three-dimensional fractal cluster or its projection is given by

\[ \rho(r) = r^{D_3 - a}, \quad \text{for } r < R \]

where \( R \) is the perimeter radius and \( D = D_2 \) or \( D_3 \), depending on the spatial dimension of \( a = 2 \) or 3 for the projected or real cluster, respectively.

Then, Eq. (4) yields

\[ R_{x,2}^2 = \frac{D_3}{D_3 - 2} R_x^2 \]

and

\[ R_{x,2,\text{true}} = \frac{D_3}{D_3 - 2} R_x \]

Thus, a relation between the true radius of gyration $R_g$ and the projected radius $R_{g,proj}$ can be determined if we have a relation between $D_2$ and $D_3$.

To determine this latter relation, consider the empirical fact of Eq. (2) that $N_i \sim A^2$, where we now label the number of monomers with a subscript of three to designate this the number in three-dimensional space. We also have by Eq. (1), $N_i \sim R_i^2$. The binary projection has analogous relations such that $N_i \sim R_{i,proj}^2$, which defines $D_3$ but, and here is the key, $N_i \sim A$. Furthermore, by Eqs. (7) and (9), $R_{i,proj} \sim R_{i,proj,3}$. All of these proportionality ties together yield

$$D_3 = D_2 / \pi.$$  \hspace{1cm} (9)

This result is consistent with past work that has measured the fractal dimension of clusters both in terms of three-dimensional quantities and projectional quantities to find that the projectional dimension is typically 10% less than that determined with the three-dimensional quantities (Sensen et al., 1987; Zhang et al., 1990; Cai et al., 1993). It is also consistent with recent simulations by Julián et al. (1994), who also found the projectional fractal dimension to be ~10% less than the fractal dimension of the unprojected clusters. Since $a = 1.1$, we believe that Eq. (9) explains these three observations.

Finally, we use Eqs. (7)-(9) to find

$$R_{g,proj} = \frac{D_3 + 2a^{1/2}}{D_3 + 2} R_{g,proj,3}. \hspace{1cm} (10)$$

For typical values of $D_2 = 13$ and $a = 1.09$, this correction factor is 1.032. Thus, as anticipated and qualitatively explored earlier (Cai et al., 1993), the binary projection yields a remarkably accurate measure of the true, three-dimensional radius of gyration.

**Compact Analysis.** To begin our compact analysis of the clusters, we define the total darkness as

$$D_{tot} = \sum_{x,y} D(x,y)$$ \hspace{1cm} (11)

where $D(x,y) = 0$ or 1 is the darkness of the pixel at position $(x,y)$. Since $D(x,y)$ is binary, $D_{tot}$ is the total number of pixels in a cluster. The cluster projectional area is given by $A = \pi D_{tot}^2$.

To determine the radius of gyration $R_g$ of a cluster, we first calculate the cluster center of mass:

$$\langle x, y \rangle = \frac{1}{D_{tot}} \sum_{x,y} x D(x,y)$$ \hspace{1cm} (12)

and then the radius of gyration:

$$R_g = \frac{1}{D_{tot}} \sum_{x,y} \sqrt{(x - \langle x \rangle)^2 + (y - \langle y \rangle)^2}.$$ \hspace{1cm} (13)

Between 30 and 100 clusters were analyzed to obtain $N$ and $R_g$ for each of the eight heights above the barrier where x-ray was collected for both the TEM and optical samples.

These data sets were then analyzed in accordance with Eq. (11). We found that individual height above the barrier yielded consistent values of $D$ and $R_g$, and the $D$ values were the same between the TEM and optical samples, but the $R_g$ values were different. Because of this, we have grouped together all of the data at different heights for the TEM and optical samples individually, and analyzed pieces two ensembles with Eq. (11). A total of 824 clusters were analyzed. The results are given in Table 2. Table 2 shows fractal dimensions of $1.84 \pm$

<table>
<thead>
<tr>
<th>Sample</th>
<th>$D$</th>
<th>$k_g$ (corrected)</th>
<th>$k_g$ (corrected)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TEM</td>
<td>1.84 ± 0.11</td>
<td>1.84 ± 0.11</td>
<td>1.84 ± 0.11</td>
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<tr>
<td>Optical (a = 1.09)</td>
<td>1.78 ± 0.05</td>
<td>1.78 ± 0.05</td>
<td>1.78 ± 0.05</td>
</tr>
<tr>
<td>Optical (a = 1.02)</td>
<td>1.76 ± 0.05</td>
<td>1.76 ± 0.05</td>
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</table>
0.11 for the TEM sample and 1.78 ± 0.05 and 1.70 ± 0.05 for the optical sample analyzed in the two ways. All of these values are both consistent with each other and previous work (Table 1). The $k_p$ values, however, are drastically different. For the TEM sample, $k_p = 1.66 ± 0.35$, which is comparable but larger than previous work in our lab for soot collected from premixed CH$_4$/O$_2$ flames where we found $k_p = 1.25 ± 0.07$ (Cai et al., 1993). It is also similarly larger than values inferred by Wu and Firestone (1993) from a review of computer simulations which imply $k_p = 1.3–1.5$. It is lower than the values found by Koylu et al. (1994a, 1994b, 1995) of ~ 2.4. We do note, at this time, an understanding of the reason for these discrepancies. In contrast are the very large values of $k_p$ for the optical soot. What is the source of this larger discrepancy? Below, we show that this discrepancy is due, in large part, to the combined effect of the inability to resolve the individual monomers in the optical soot and the fractal nature of the clusters.

Resolution Correlation. In our analysis, the number of primary particles present in the cluster is calculated from the cluster projected area using Eq. (3). This formula works well if the optical sample analyzed by the cluster and its monomers are well resolved. Problems arise, however, at low magnification because the individual monomers are not resolved. This problem does not lie in the relation $N \sim A^d$, but in an erroneous determination of $A$ due to the poor resolution. This, in turn, causes an error in $N$ and hence $k_p$.

To understand this problem, consider the digitized image of a computer-generated DLCA cluster with fractal dimension 1.75 at two different magnifications in Fig. 2. The resolution limit is set by the pixel size. For example, the cluster at magnification 1 might yield $p_x = 30$ microns/pixel, whereas magnification 2, twice that of magnification 1, might yield $p_x = 10$ microns/pixel. The area (e.g., in m$^2$) of the cluster is

$$A = np^d$$  \hspace{1cm} (14)

where $n$ is the number of pixels per cluster, $p$ is the scale of a new structure, then $A_1 = A_2$; hence

$$n_2/n_1 = (p_1/p_2)^d$$ \hspace{1cm} (15)

That is, because the scale is larger by a factor of $p_1/p_2$, there are more pixels in the image of the cluster in 2 by a factor of $(p_1/p_2)^d$. The exponent 2 results because the dimension of the plane is two.

Now, consider the case where higher magnification reveals structure ladders at lower magnification as portrayed in Fig. 2. In this case, the fractal nature of the projection of the cluster implies that Eq. (15) should be modified to

$$n_2/n_1 = (p_1/p_2)^D$$ \hspace{1cm} (16)

where, by Eq. (9), $D = D_f/d$ is the fractal dimension of the cluster when projected into the two-dimensional plane. From (14) and (16), we find

$$A_1/A_2 = (p_1/p_2)^{D-d};$$  \hspace{1cm} (17)
then from Eqs. (1), (2), and (17), we find

\[ k_2 = k_{30} \left( \frac{P_1}{P_0} \right)^{2 \alpha - \beta} \quad (18) \]

where \( D \) is the fractal dimension of the cluster in three dimensions. Equation (18) can be used to compare \( k_4 \) values of the TEM and optical soot samples which were viewed at different magnifications. We remark that this correction can only be used between two regimes of unresolved monomers. Obviously, if the monomers are perfectly resolved at two different scales, Eq. (18) is invalid.

One might expect a correction due to resolution for \( R_q \) as well. We have been unable to calculate such a correction analytically. Simulations similar to Fig. 2 imply a random correction of a few percent for \( R_q \). Given this uncertainty and randomness and the large correction necessary for \( N \), we do not correct the \( R_q \) values.

The scales for our optical and TEM clusters are \( p = 3500 \) and 12.1 mm/pixel, respectively. Thus, using \( \alpha = 1.09 \) and \( D = 1.76 \pm 0.05 \), we find \( k_4 \text{(optical)}/k_4 \text{(TEM)} = 9.7 \pm 2.7 \). Or using \( \alpha = 1.042 \) and \( D = 1.70 \pm 0.03 \), we find \( k_4 \text{(optical)}/k_4 \text{(TEM)} = 8.8 \pm 2.4 \). When these corrections are applied to the two different analyses of the optical sample, the same "corrected" values of \( k_{30} \) also given in Table 2, of 2.6 \( \pm \) 0.9 are found. These values are consistent with the \( k_{30} \) value obtained for the TEM sample. All of the data are plotted in Fig. 3. The large, \( \sim 40\% \) error in the \( k_4 \) for the optical soot is due both to the fact that these data are far from the intercept (hence, a small uncertainty in slope acts as a lever arm on the plot, and \( k_{30} \) is the intercept at \( N = 1 \)), and the corrections of Eq. (18) have an exponential dependence on \( D \) so that the uncertainty in \( D \) is exponentially magnified.

Because of these uncertainties, we consider

![Figure 3](image-url)
the kₙ values for the optical sample to be only semi-quantitative.

CONCLUSIONS

Figure 3 demonstrates that the same morphology describes clusters ranging in size (R_c) from 50 nm to 400 µm, or in terms of N_c the size ranges from 10 to 10⁷, a seven order of magnitude range. This large range of constancy implies that the process by which the clusters are formed is the same over this vast size range. Since D = 1.8, it has been concluded that this process is Diffusion Limited Cluster Aggregation. The constancy of the morphology also implies that other physical properties of these clusters will be interrelated, and any variation would be due to comparisons of cluster length scales to lengths inherent in the given physical property.

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References


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