Specification of the Ruggedness and/or Texture of a Fine Particle Profile by its Fractal Dimension

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(Received August 22, 1977)

SUMMARY

The fractal dimension of a space-filling non-differentiable curve has recently been defined by Mandelbrot. It is shown that the fractal dimension of an indented or convoluted fine particle profile could provide an index for quantifying the ruggedness of the shape of the fine particle. An analogy between the work of Richardson on the problems of coastline specification in geography and the problems of the scientists seeking to specify fine particle structure is drawn. Three different methodologies for measuring the fractal dimension of a rugged and/or textured fine particle profile are suggested. These are (a) random walk strategies using coupled light pens, (b) line scan interception logic, and (c) digital set theory for binary image transforms. Preliminary data for the measurement procedures on the rugged profiles of fine particles and agglomerates are presented.

It is shown that the curve specifying the fractal dimension of the rugged profile of an agglomerate made of clearly discernible subunits which have Euclidian profiles changes its nature when the exploratory step size in the random walk around the profile approaches the order of magnitude of the dimensions of the subunit. This could provide a basis of an algorithm for determining the structure of an agglomerate by an automated iconometric procedure. It is suggested that fractal dimensions along with descriptive parameters from the convex hull of a fine particle profile gives a comprehensive description of the profile. Other potential applications of fractals in fine particle science are briefly discussed.

1. INTRODUCTION

The first procedures developed for qualitatively describing the shape of a fine particle made use of simple linear parameters such as the length, breadth and width of the fine particle. Ratios of these dimensions were then used as coefficients to describe such shape-dependent properties as the elongation ratio (defined as the length to breadth of the fine particle). These pioneer attempts to describe the shape of the profile were developed over a period of years by Heywood [1]. The comprehensive description of the three-dimensional structure of a fine particle has proved to be an intractable problem except for simple structures which approximate to classical geometric shapes. Good reviews of the many types of geometric descriptions and ratios which have been used to describe fine particles have been given by Schädel and Rumpf [2] and Beddow *et al.* [3]. The development of automated microscope systems linked to computer data-processing systems has made it possible to consider methods of specifying the shape of a profile by mathematical procedures utilizing such complex mathematical functions as Walsh functions and Fourier transforms [4 - 6]. Thus Schwartz and Shane developed the concepts of the geometric signature waveform of the two-dimensional profile of the fine particle [7]. The physical basis of this type of signature waveform is illustrated in Fig. 1. A reference point in the profile is used as a rotation pivot for a vector magnitude R which touches the profile of the fine particle. To generate a signature waveform, the vector is assumed to move with constant angular velocity about the pivot point, and a graph of R against θ is regarded as defining a unit element of a harmonic wave. Schwartz and Shane used the centre of the smallest circumscribing circle in defining this signature waveform. Beddow and co-workers have prefered to use the centre of gravity which the profile would have if it were to be treated as a thin lamina fine particle [3]. There is no *a priori* merit in

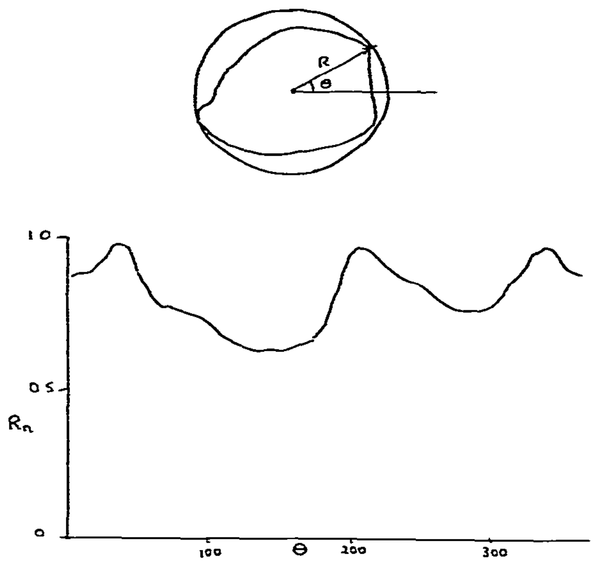

Fig. 1. Geometric signature waveforms such as the Schwartz–Shane geometric waveform of the fine particle shown above are used to describe fine particle profiles. R_n = magnitude of vector R expressed as fraction of the maximum vector.

either of these reference pivot points and many other reference points are possible [8]. The variations in the magnitude of the Feret's diameter of a fine particle profile for a systematic set of varying orientations in space also constitute a useful signature waveform [9]. The Feret's diameter signature waveform, referred to as the FERETS waveform, has the useful property that the waveform generated is independent of any point location within the profile and is related to perimeter of the convex hull. The Schwartz and Shane and Beddow type geometric waveforms are sensitive to error and/or uncertainty in the location of the reference pivot.

The potential uses of geometric signature waveforms for the description of, and the automatic recognition and/or classification of, fine particle shape are undergoing extensive exploration and development [11]. In some techniques for utilizing the signature waveform for a fine particle descriptor, the signature waveform is broken down into its harmonic components using Fourier analysis [5, 11]. For this reason, a list of the constituent waves and their amplitude and phase is referred to as the harmonic shape factor [12].

Although they are proving to be of great utility in some problem areas, the geometric signature waveforms have an inherent limitation in that they are not suitable for the description of the shape of fine particle profiles which contain sharp protuberances or are convoluted. For example, as illustrated in Fig. 2, the presence of a sharp protuberance on a fine particle profile makes it necessary to have high resolution in the examination of the profile by the rotating vector generating the signature waveform. Any subsequent processing of the waveform data by Fourier analysis is complicated by the presence of high frequencies representing the sharp peak superimposed on the signature waveform by the presence of the protuberance. Again, the physical significance of the length of the vector generating the signature waveform as it crosses a re-entrant loop of the fine particle profile is indeterminate. It should be pointed out that one of the advantages of the Feret's waveform is that, since it is related to the con-

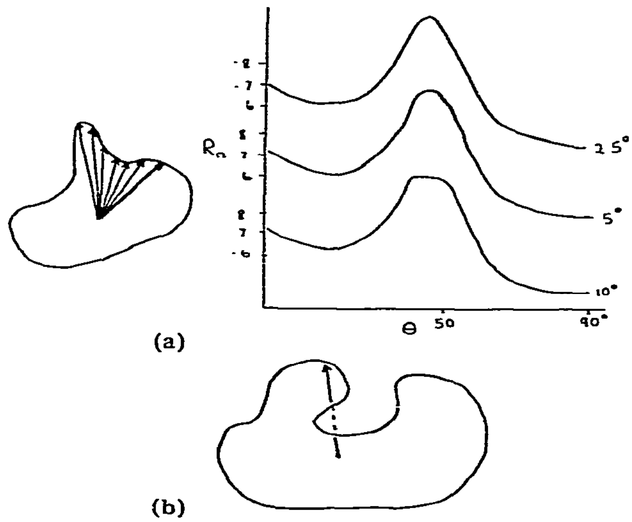

Fig. 2. Fine particle profiles with protuberances and/or re-entrant loops cannot be described in a simple manner by geometric signature waveforms generated by a rotating vector following the profile limits. (a) A protuberance requires high resolution to generate the descriptor waveform and increased mathematical analysis for subsequent data processing in such methodologies as the use of Fourier analysis to describe the waveform. (b) It is not easy to assign physical significance to the magnitude of the generating vector as it crosses a re-entrant loop on the fine particle profile. R_n = amplitude as a fraction of the maximum vector.

vex hull of the profile, all re-entrant loops on the profile are automatically smoothed out to give a description of the gross shape of the fine particle.

The difficulties of describing fine particle profiles with re-entrant loops is a major stumbling block in fine particle science, as pointed out by Orr in his review lecture presented at the Workshop sponsored by the National Science Foundation in 1975 to help assess research funding priorities in fine particle science and technology. In that lecture he states: " Shape is rarely measured, not because of a shortage of so-called shape factor definitions but because no-one has as yet found a completely satisfactory means of measurement especially for particles with re-entrant contours" [13].

The situation has been compounded in recent years for the fine particle scientist by the fact that better methods of imaging fine particles, such as the scanning electron microscope, have revealed that many profiles which appeared to have smooth contours as viewed under the optical microscope were found to have highly textured surfaces when viewed under the scanning electron microscope. (For example, see series of pictures of carbonyl nickel powder reported by Johari and Bhatacharyya [14].)

Hausner was one of the first to attempt to deal with the problems of textured fine particles. He suggested the use of embracing rectangles of minimum area which could be drawn around the profile. In essence, this approach sought to remove the problem of texture and re-entrant loops from the description technology by replacing the profile by equivalent profiles of regular geometric shape [15]. Johnston and Rosen in their recent publication [16] separated the problems of shape and texture description. They measured the area of the fine particle profile using modern automated iconometrics, and calculated the perimeter of the particle and compared this perimeter with that of a circle of equal area. They also evolved a measure they called the edge texture. However, they do not give details in their publications of their quantitative technique for calculating the edge texture [16]. Perhaps the first successful attempt to deal with the structure of convoluted fine particles is that due to Medalia [17], who utilized the concepts of mechanics to describe the distribution of mass in a fine particle profile. He treated a fine particle profile as if it were a thin lamina, and calculated the dimensions of an ellipse having the same radii of gyration about the central principal axes as those which the fine particle profile would have if it were a thin lamina fine particle. He then defined two shape factors for the fine particle profile using dimensions of the ellipse having equivalent mechanical properties. Thus the anisometry is defined as the ratio of the major to minor axis of the ellipse, and the bulkiness is the ratio of the area of the ellipse to that of the profile. Medalia has demonstrated that these shape factors are useful in the description of the behaviour of carbon black flocs. Probably one of the reasons that they have not been more widely adopted in fine particle science is because of lack of information as to the physical significance of these factors for general fine particle systems. Because of the fact that Melalia's shape factors are based upon the mechanical properties of the fine particle, they are often referred to as dynamic shape factors.

Kaye, Naylor and co-workers have attempted to tackle the problem of shape characterization of fine particles using optical computing technology. The basic system that they have described is known as the SHADOW system for shape characterization [18 - 20]. In this system, a silhouette of the fine particle is used to generate a Fraunhofer diffraction pattern using laser light. This diffraction pattern is interrogated using a disc, from which a V-shaped sector has been cut, rotating in front of a photocell. The resultant wave generated in one revolution of the interrogation disc is treated as a signature waveform for the description of the fine particle profile. The name of the system comes from the acronym Shape Analysis by Diffraction Originated Waveform. One of the problems encountered by Kaye *et al.* in the development of the SHADOW system was that again textured profiles generate a complex diffraction pattern in which some of the scattered light comes from the overall shape of the fine particle profile and other light comes from the textured edges of the profile. In some situations, the fact that some of the optical information is coming from the edges of the profile can be exploited to give a measure of the presence of sharp edges in a powder which is to be sintered.

Another procedure suggested by Kaye and Naylor for dealing with complex fine particles such as the carbon black agglomerates is to use the fact that the interaction of the texture and/or shape information is likely to be unique for a complex profile such as a simulated carbon black floc shown in Fig. 3, and therefore one can do direct pattern matching of the diffraction patterns to teach a cybernetic version of the SHADOW system to recognize fine particle shape from diffraction patterns of a comparative series of agglomerates, the diffraction patterns of which have been stored in the memory of the system. For a comprehensive discussion of this approach to the characterization of agglomerates, see [10]. In recent work, Naylor and co-workers are adopting the approach that one can characterize fine particle profiles by using the Fourier transform of the profile of the fine particle, this transform being generated either optically or mathematically [4]. There is a real possibility that, by adopting a strategy in which a smoothed-out profile is used as an object in a SHADOW type system for studying diffraction patterns, rapid efficient shape characterizations of fine particle profiles can be achieved [21].

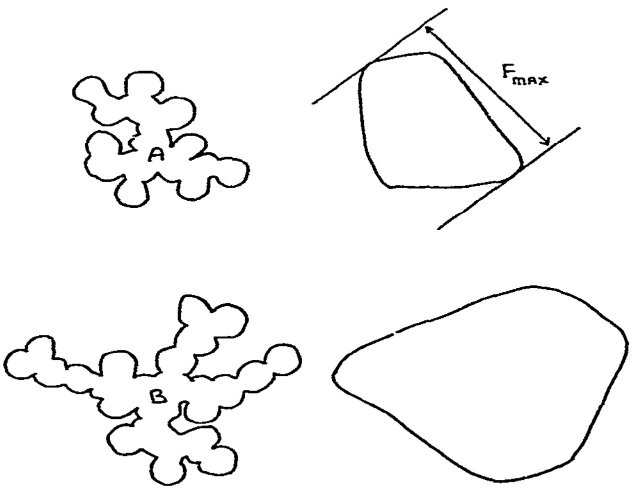

Fig. 3. Simulated carbon flocs reported by Murphy [10]. The convex hull of each profile is shown beside each floc. The gross convoluted real profile is an approximate fractal, but the profiles of the subunits are Euclidean (see Figs. 12, 13). The convex hull is a Euclidean curve which envelopes the profile. F_{max} = maximum Feret's diameter.

From this overview of the various strategies which have been evolved in attempts to deal with the shape of fine particles, it seems that, for other than relatively smooth fine particle profiles which approximate to classical geometric curves, there is no generally applicable adequate technology for characterizing fine particle profiles. Recently, Mandelbrot has published a book in which he discusses the general mathematical problem of a measurement of boundaries in any dimensional system. Of particular interest to the fine particle scientist is the fact that he suggests a mathematical technique for characterizing rugged boundaries. Mandelbrot discusses the work of Lewis Fry Richardson, who attempted to tackle the problem of the precise specification of a length of coastline. The comparison of aerial photographs of an island with those of a fine particle system immediately suggests the analogy that technologies useful in specifying the ruggedness of a coastline should be of use to the fine particle scientist in the characterization of the convolutions of a fine particle boundary. Indeed, it is the analogy between the two problems which suggested to the writer the use of the term "ruggedness index" for the specification of a fine particle convoluted boundary. A reading of the relevant sections of the Mandelbrot book suggests that the crisis in fine particle characterization posed by rugged boundaries arises partly from the fact that the mathematical training of many western scientists is dominated by Euclidean geometry, which is unable to cope with rugged boundaries. Rugged boundaries belong to a class of curves known as "space-filling curves", which in many cases are characterized by the fact that they have no tangents and often no differential function.

In his book Mandelbrot deals with the mathematical properties of non-Euclidean lines. In particular, he suggests the use of a quantity that he defines as the fractal dimension for use in the description of the convolution density, that is, the ruggedness of a non-Euclidean line. It appears that this fractal dimension may be directly usable as a ruggedness index for the description of the convolutions and/or texture of a fine particle profile. From this perspective, the attempts of various workers to replace the physical reality of the fine particle profile by a smooth classical shape can be seen as an attempt to reduce intuitively the dimensional problem of profile specification by replacing the fractal curve of the convoluted profile by its nearest Euclidean equivalent curve.

In Section 2, the basic concepts of fractal dimensions as developed by Mandelbrot are reviewed. A graphical method for evaluating the fractal dimension is described. The normal attribution of a dimensional code (*i.e.* 1, 2 or 3) to a real object is always subjective from the perspective of an observer, and Mandelbrot's comments on this are reviewed. In the same way, the fractal dimension description of a real curve is dependent on the scale of scrutiny used in exploring the boundary. Our quantitative impression of the structure of a curve will depend on the measuring rule used in exploring the structure of a curve. It is quite possible that a real curve, *i.e.* one that exists in nature, may exhibit fractal structure at coarse resolution and Euclidean at very high resolution. Thus, for a real boundary, regions of fractal structure for a range of scrutiny may exist, and both the range and the fractal dimension will be typical of and descriptive of the boundary. Thus one can conceive of a semifractal or multifractal description of a real boundary being defined by linear regions on a log–log plot of perimeter estimates against stride magnitude. In Section 3, preliminary measurements on real and simulated fine particle profiles which indicate the possible existence of aspects of structure describable by fractal dimension are presented. Various strategies for evaluating the ruggedness index of a fine particle profile are briefly explored in Section 4. Section 5 explores briefly further possibilities for the use of fractal dimensions in fine particle science.

2. DEFINITION OF THE FRACTAL DIMENSION OF A NON-EUCLIDEAN LINE

Mandelbrot, in his book Fractals: Form, Chance and Dimension, points out that the classical training in mathematics and physics received by many western scientists glosses over the problems that students have in accepting the dimensionalities of various objects. Students are taught that a line has one dimension, a point has none, a sheet of paper has two, and a solid cube of wood has three. Anyone who has attempted to teach a freshman class of physics students knows that some students immediately have difficulty with the concept that a line has one dimension since they argue that a real line must always have thickness and is two-dimensional. In the same way, they will not accept that a sheet of paper has two dimensions since they claim that the thickness of the paper is obviously a third dimension. Particularly obstinate students insist that points, lines, sheets of paper and cubes all have three dimensions. These students sooner or later conform to the accepted textbook definitions, since this is the way to pass examinations and also because traditional statements have the authoritative backing of those who have already passed their degree. Mandelbrot in his book appears to have some sympathy with student challenges of traditional dimensional descriptions. He points out that the dimensions that one allocates to a spatial object depend to a considerable degree on the perspective from which one is describing an object. Thus, in his book he states that, "Strictly speaking, objects such as a small ball, a veil or a thread, thin though they may be, should all have to be represented by three-dimensional sets" [22] *. Mandelbrot goes on to state that, "However, every physicist knows ...that it is much more useful to think of a veil, a thread or a ball, if they are fine enough, as closer in dimension to two, one and zero respectively." He then states, "In other words, physical dimension inevitably has a subjective basis. It is a matter of approximation and therefore of degree of resolution."

He illustrates this point by considering the various dimensionalities attributed to a ball of wool from outside the ball of wool and then at several scales of scrutiny within the ball of wool. It appears that the major reason we accept traditional dimensional descriptions is pragmatic — such descriptions are useful. Having established that there is subjectivity inherent in our normal treatment of dimensionality of objects, Mandelbrot proceeds to differentiate between the topological dimension of an object and the Euclidean dimen-

*Modern general theories of geometry used to advanced concepts of set theory, and planes, lines and volumes are described in terms of sets of points. For those unfamiliar with this type of geometry, a useful introduction to the relevant concepts can be found in the book Concepts of Modern Mathematics by Steward [23].

sionality. In a presentation such as this, one cannot hope to stick to mathematical rigour in the presentation of abstract ideas. Since the aim of this discussion is only to introduce the reasonableness of a new concept, reasonable analogies will be used in discussing complex ideas. The reader is directed to Mandelbrot's original discussion for the exactitude required in a mathematical presentation. From a reasonable analogy point of view, the Euclidean dimensions of an object correspond to those which we intuitively recognize as one, two and three-dimensional objects. Thus the Euclidean dimension of a polished dense sphere is three. The Euclidean dimension of a sea-coast distributed over a map requires a two-dimensional description. Topology for the layman can be defined as that branch of mathematics which studies relationships between sets which remain true no matter how the space in which those sets are organized is distorted. For example, the two-dimensional profiles of Fig. 4 can be divided into three sets which are topologically equivalent. They are described in mathematical terminology as belonging to genus 0, genus 1, and genus 2, as indicated. (Note: this topological equivalence of various types of fine particle profiles is already in use

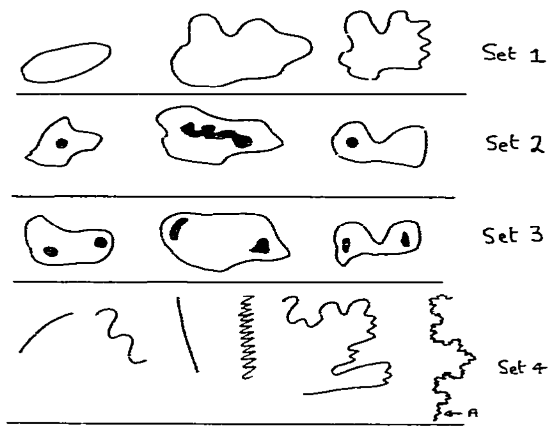

Fig. 4. Topologically equivalent systems if drawn upon rubber membrances can be stretched and distorted so that they can be exactly superimposed on each other. Set 1: Genus 0. Topologically equivalent fine particle profiles. Topological dimensions of profiles = 2. Set 2: Genus 1. Topologically equivalent fine particle profiles. Topological dimension of profiles = 2. Set 3: Genus 2. Topologically equivalent fine particle profiles. Topological dimension of profiles = 2. Set 4: Topological equivalent lines of topological dimension 1. A = triadic Koch curve.

in pattern recognition technology of automated iconometrics [24].)

In Fig. 4 are shown several lines which are topologically equivalent. Theoretically, each one of them can be stretched out and superimposed upon each other. The three lines on the left-hand side represent curves from Euclidean geometry. The next two lines represent arbitrarily twisted lines which have different abilities to cover a given area, and the third wiggly curve is a mathematical curve known as a triadic Koch curve. The Koch curve is an example of a curve which does not have tangents. (Because of the need to use a line of finite width to represent the curve in Fig. 4, one could argue that the Koch curve shown in the diagram can have tangents drawn to it. The reader has to imagine that if one had an infinitely thin pen, the line is drawn in such a way that a sequence of magnifications of the line would reveal that each portion of the line was similar in construction to the loop which is visible in the sketch of Fig. 4.) This Koch curve is also an example of what Mandelbrot defines as a natural fractal. The origin of this word is given by Mandelbrot as coming from the Latin adjective "fractus", meaning "irregular" or "fragmented". A mathematical definition of a natural fractal is any natural pattern representable by a fractal set. This, however, is not particularly meaningful to the engineering scientist, and a useful mental image for helping one to recognize a fractal line is that it is a wiggly line such that scrutiny of the structure of a curve at any scale of magnification would reveal that any one element of the curve is self-similar to any other element of the curve. Perhaps the most familiar fractal to the physicist and engineering student is the Brownian walk depicted in many textbooks and reproduced in Fig. 5. Although such traditional representation of Brownian motion gives a good idea of the random staggering of a colloidal fine particle, the fact that the time-separated positions are linked by finite straight lines gives a false impression of the actual structure of the Brownian motion curve. It is interesting to note that Perrin, in his original discussion of Brownian motion in 1910, warned that detailed scrutiny of the actual Brownian motion revealed the impossibility of drawing tangents to the curve. Thus, in Mandelbrot's translation of Perrin's original paper in French [21, p. 10], Perrin points out,

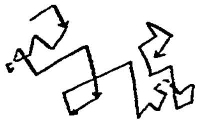

Fig. 5. In the traditional representation of Brownian motion, the successive displacements of a colloidal line particle in sequential time intervals is indicated by a linked string of vectors. Perrin has pointed out that increased levels of scrutiny of Brownian motion reveal that there is a highly randomized track linking any two positions linked in the lower level of scrutiny by a simple linear displacement vector. Perrin also pointed out that no matter how one increased the scale of scrutiny, one would always be looking at a randomized zigzagging trajectory.

"One may be tempted to define an average velocity of agitation (of a colloidal particle) by following a particle as accurately as possible, but such evaluations are grossly wrong. The apparent average velocity varies crazily in magnitude and direction. It gives only a much weakened idea of the prodigious entanglement of the real trajectory. If indeed this particle's positions were marked down a hundred times more frequently, each segment would be replaced by a polygon relatively just as complicated as the whole drawing and so on. It is easy to see that in practice the notion of tangent is meaningless for such curves."

Note that in this quote from Perrin, the essential aspect of a Brownian motion curve is that it is self-similar in that increased scrutiny magnitude reveals self-similar polygons linking time-dependent positions of a colloidal particle.

For curves that meet the requirement of being self-similar mathematically but which have different degrees of structure, Mandelbrot defines the fractal dimension. The fractal dimension of a fractal line is a number between 1 and 2. Intuitively, many people reject the idea of a fractional dimension because they cannot create a mental picture of what an object of 1.24 dimensions looks like. (It is perhaps comparable to the average Canadian family with 2.5 children!) However, as already pointed out, our traditional one-, two-, and three-dimensional descriptions are to some extent subjective artefacts employed because of their utility. The fractal dimension of non-Euclidean objects (although we shall only be concerned in this communication with fractal dimensions of magnitude between 1 and 2, the concept extends to the dimensionalities of lesser and greater magnitudes [see Mandelbrot's book]) describes important spatial behaviour of a geometric system and deserves to be employed in descriptive systems on pragmatic grounds. From an intuitive point of view, the fractal dimension of a self-similar fractal curve actually corresponds to the ability of that curve to cover an area with a dense array of points. Again, in a non-rigorous sense, one can picture a fractal curve of higher fractal dimension covering a plane with points more quickly and/or densely with points. Thus, Brownian motion has a fractal dimension of 2 in that, given long enough, the staggering colloidal particle will entirely cover a Euclidean two-dimensional area. On the other hand, the Koch curve shown in Fig. 4 is not as efficient in covering an area as Brownian motion. It has a fractal dimension of 1.2618. In his book, Mandelbrot gives many examples of different fractal curves with different fractal dimensions [21, 24]. It should be noticed that the topological dimension of the Brownian motion trajectory and the Koch curve is one. From Mandelbrot's discussion it emerges that Euclidean curves are those for which the topological dimension and the fractal dimension are coincident.

The possibility that fractal dimensions may be useful in describing the ruggedness of real curves is demonstrated in Mandelbrot's book by considering the problem of specifying the length of boundaries between countries and the length of coastlines. In his discussion of the ruggedness of coastlines and boundaries Mandelbrot quotes the earlier work of Richardson. The empirical data collected by Richardson for estimates of the length of various boundaries are summarized in Fig. 6. The physical basis of the various estimates over several boundaries discussed in this diagram can be appreciated from the sketches of Fig. 7. One way of estimating the length of a convoluted curve is to step along the curve with a fixed stride, noting how many steps are needed to traverse the curve, as illustrated in the first sketch of Fig. 6. In one sense this constitutes a random walk along the curve. The method of constructing the walk is to swing an arc with a pair of compasses from the last point of intersection between the walk and the curve, and then the next point of intersection becomes the direction of the

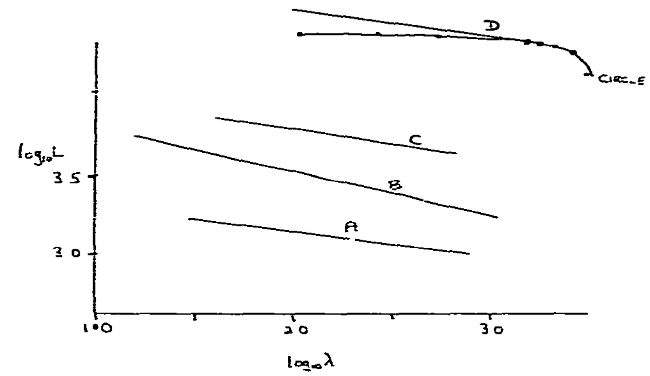

Fig. 6. Richardson's data on the relationship between the estimate of the length of a boundary and the scale of scrutiny used in estimating the length of the boundary. A, land frontier of Portugal; B, west coast of Britain; C, German land frontier (1900); D, Australian coast. λ = stride in kilometers, L = total length in kilometers.

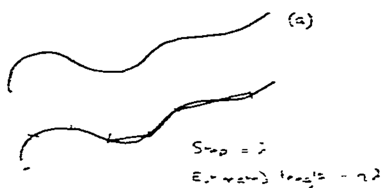

Fig. 7. A reasonable technique for estimating the length of a curve appears to be to make a series of estimates of the length by "stepping" along the curve using a series of decreasing stride or step magnitudes to obtain increasingly better estimates of the curve length. Such a technique yields a finite estimate for a Euclidean curve, but yields an infinitely increasing estimate of length for a fractal curve.

step along the curve. It is a random walk because the direction of sequential steps is unpredictable. By walking along the curve in this way, one in effect replaces the convoluted curve with a zigzagging line which attempts to follow the contours of the curve. We shall refer to the estimate of the length of a curve based on this stepping procedure as a random walk estimate of the length of the curve. It follows then that the smaller the step in the random walk estimate procedure, the more closely the zigzag curve conforms to the structure of the convoluted curve. For a Euclidean curve, such a procedure soon results in the estimated length of the curve reaching a constant value within the limits of experimental error. However, for a fractal curve, such a series of estimates does not reach a limit. In fact, Mandelbrot has shown that if one plots the estimate of the length of the curve against the logarithm of the step size in the random walk estimate, one obtains a linear relationship which theoretically extrapolates to give infinite length for the fractal curve. To illustrate this aspect of the structure of a fractal curve, the experiment summarized in Fig. 8 was carried out. Estimates were made of the length of a triadic Koch curve as reproduced in Mandelbrot's book. In order to normalize the data obtained in this method, the stride magnitudes of the random walk estimate of the length of this curve are expressed as fractions of the straight line shown underneath the length of triadic curve to be evaluated. The actual random walk constructed for a stride length equal to one-tenth of the basic unit is illustrated in Fig. 8. In the lower portion of Fig. 8 the logarithmic plot of the length estimate of a curve against the length of the stride used in the random walk estimate is shown. It can be seen that the data conform to a linear relationship and extrapolate to infinite length. The exact significance of this linear relationship will be discussed later in this communication.

Returning to the data of Fig. 6, we now see that the various boundaries discussed by

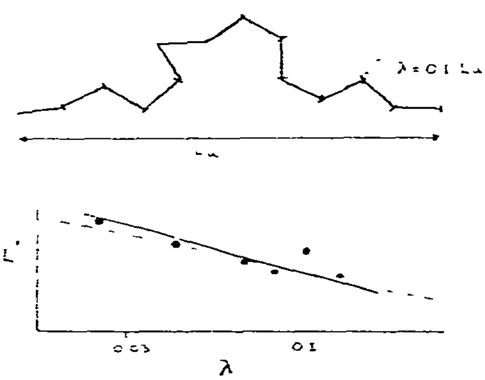

Fig. 8. Mandelbrot has shown that a graph of the log of the length estimate of a fractal curve based on a random walk along the curve against the log of the size of the stride has a slope of $1 - D$, where D is the fractal dimension of the curve. Data in this graph compare the fractal dimensions estimated from a series of random walks of decreasing step and the theoretical value for a true fractal. Curve is "triadic Koch curve" known from theory to have a fractal dimension of 1.2618 [22]. L' = length estimate (log scale arbitrary units), λ = step size (normalized units as defined in the diagram).

Richardson can be estimated using various strides along the boundaries. (In the case of the geographical boundaries, the stride length is in kilometers rather than centimeters and the use of the term stride is intended to be metaphorical.) From Richardson's data, one reaches the seeming paradox that all coastal boundaries and all political frontiers are in fact infinite. As Mandelbrot points out, one finds this a little hard to accept intuitively, but it arises from the fact that if one uses smaller and smaller rulers, eventually one is going around every pebble and then the chips and dents on the pebble until one is forced to weave one's way in and out of the atoms of the pebble. The significance of Richardson's data is that there is no way that one can reach a finite limit for the boundary using physical measuring rules, and one has to state the purpose of the measurement before one can achieve a reasonable estimate for pragmatic purposes. It is interesting to note that Richardson actually discovered this trend in one's estimate of boundary lengths empirically. He did not himself attribute any significance to the slope of the lines linking the data in Fig. 6. It is interesting to note that in Fig. 6 Richardson and Mandelbrot have included data for an ordinary circle, which has a Euclidean boundary. For such a type of boundary, one's estimate of the structure of the circle in terms of random walk theory rapidly approaches a physical limit. Indeed, random walk estimate of the perimeter is actually an extension of Archimedes' original method for finding the value of π by constructing polygons approximating the perimeter of the circle [26]. Mandelbrot has discussed the significance of Richardson's data and pointed out that the slope of the logarithmic plot of perimeter estimate against the logarithm of the stride is actually the quantity $1 - D$, where D is the fractal dimension of the rugged curve making up the boundary. An examination of the data of Fig. 6 shows that the slope of the German land frontier is closer to that of the land frontier of Portugal rather than the other coastlines. Mandelbrot points out that this is because these two boundaries are essentially a series of river segments and that the natural fractal dimension of a river system differs from that of a rugged coastline. Again, the various coastlines differ in their ruggedness because of the various types of erosion and/or rock structure involved in the formation of the coastline. For example, the Atlantic coast of Newfoundland is very much more rugged than that facing the Gulf of St. Lawrence. On the other hand, the ruggedness of Manitoulin Island, which is totally enclosed by Lake Huron, has the same ruggedness on both sides of the island.

The practical significance of Richardson's data is that it shows that naturally occurring boundaries can approximate to mathematical requirement that sub-portions of the curve be self-similar to a sufficient degree to make the concept of fractal dimension useful in describing the structure of that boundary. It therefore appears reasonable to anticipate that the fractal dimension of a rugged fine particle profile may be characteristic of that profile and an adequate description of its convolution and/or texture. What is required is a series of investigations of fine particle boundaries to see if this type of measurement can be abstracted from the boundary and then measurements for various boundaries compared to bring out the physical significance of the fractal dimension measured for such boundaries. Comprehensive studies of this kind are obviously beyond the scope of this initial communication, which is limited to pointing out the basic utility of the concept and the possibility of evolving experimental methodology for making appropriate measurements. In the next section, we explore the results of initial experiments on the measurement of fine particle boundaries using random walk estimates. It is shown that there is every reason to believe that convoluted fine particle boundaries constitute fractal curves to a sufficient degree for the measurement of their fractal dimensions to be useful as a ruggedness index for such boundaries.

3. EXPERIMENTAL MEASUREMENT OF THE BOUNDARY PROPERTIES OF FINE PARTICLE PROFILES

The first experiment which was carried out was intended as a preliminary exploration of the sensitivity of the graphical technique using random walk estimation data for the evaluation of the ruggedness index of a fractal line. For this purpose, estimates were made of the length of a portion of a triadic Koch curve as shown in Fig. 8. To normalize the experimental data, all information is represented as fractions of the straight line drawn underneath the portion of the Koch curve given in Fig. 8. By definition, the triadic Koch curve is a fractal of fractal dimension 1.2618, this value being known from theoretical considerations. In his book, Mandelbrot adopts the convention that fractal dimensions known from theoretical reasons are quoted to four decimal places and empirically determined data to two decimal places. Essentially the same convention is used in this communication, although it should be clearly stated that, because of the simple nature of the experiments reported here, the confidence level to be placed in the second decimal place is not high. Six estimates of the portion of the Koch curve shown in Fig. 8 were made using the random walk estimate technique. The data for these walks are summarized in the graph. It can be seen that a linear relationship can be used to describe the data. It is interesting to point out that the information for the random walk using a stride of 0.1 units appears to be somewhat different from the other points. It should be realized that for such a relatively crude random walk, various estimates of the length of the curve are possible depending upon the starting point along the fractal curve. In other words, the discrepancy for this point is somewhat of an artefact because of the starting point used in the random walk. Generally speaking, when relatively large steps are used to estimate the length of a boundary, considerable variation exists in the estimate, not because of experimental error but because of permitted variation in the magnitude of the estimated perimeter because of the way in which one defines the procedure to be used in making that estimate. The slope of the line linking the various random walk estimates of the Koch curve is 0.32. Therefore the estimate of the fractal dimension is 1.32. Compared to the theoretical value of 1.26 this gives an accuracy of 5% in the estimate of the fractal dimension. Considering the rudimentary nature of the experiments carried out, this is an excellent correspondence of the theoretical and experimental value, indicating that more exhaustive sets of experiments would probably result in an estimate of a fractal dimension sufficiently sensitive to variations in structure to be of use in describing the ruggedness of a curve. For all of the experiments reported in this communication, the error level is approximately 7%. This could be reduced by more exhaustive experiments, but the aim of this communication is to bring a new technique to the attention of scientific workers in the field. More extensive experimental investigations are under way and a complete report of these experiments will be given at a later date.

In Fig. 9, the profile of an actual carbon black fine particle imaged by Medalia is shown. To investigate the possibility of using the fractal dimension to describe such a profile, a series of random walk estimates of the profile were carried out. Obviously, the maximum stride used in such a series of estimates must be smaller than the maximum dimension of the profile for a meaningful estimate to be achieved. The random walk strides used in estimating the perimeter of the profile were normalized with respect to the maximum Feret's diameter of the fine particle profile. The maximum stride used in estimating the perimeter of the profile was just over one-tenth of the maximum Feret's diameter of the profile. It should be stressed that the

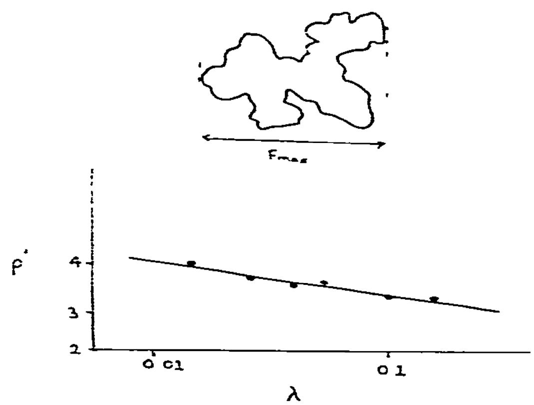

Fig. 9. Data obtained for random walks around a real profile indicate that the fractal dimension of a convoluted profile is probably characteristic of the profile and could be used as a ruggedness index. Profile of a carbon black fine particle imaged using the electron microscope reported by Medalia [17]. F_{max} = maximum Feret's diameter of the fine particle used as a scaling unit in the perimeter–random walk stride graph given above; λ = stride magnitude expressed as a fraction of the maximum Feret's diameter of the profile; p' = estimate of perimeter of profile expressed as multiples of the maximum Feret's diameter of the profile. Slope of data line = 0.18; fractal dimension = ruggedness index = 1.18.

boundary shown in Fig. 9 is a smoothed-out version of the actual profile in which fine detail of the boundary lies within the thickness of the line. However, the thickness of the smoothing line is an order of magnitude less than the smallest stride used in walking around the perimeter, so that this is an acceptable line boundary for the purposes of this experiment. If one wished to increase the precision and/or range of the investigation, one could have worked directly on the electron micrograph and used a wider range of stride magnitudes. The data for a series of random walk estimates of the profile perimeter are summarized in Fig. 9. It can be seen that the data conform to the anticipated linear relationship, indicating that the boundary approximates sufficiently closely to a true fractal for the fractal dimension to be a useful factor in describing its structure. From the graph, the ruggedness index of this particular profile would be 1.18.

It is reasonable to anticipate that carbon black flocs will have a structure close to that of a fractal because of the random way in which they grow in the smoke used in their manufacture. In the same way, fine particles such as agglomerates of crystals will probably be describable by their fractal dimension. Generally, crushed particles will also probably be describable as fractals because of the fact that portions of their surface represent fractures of fractures, etc. The usefulness of the fractal dimension for describing any particular fine particle system would have to be individually explored, but some idea of the generality of the type of curve which can be described by this process can be gained from the data summarized in Fig. 10. In the top portion of this figure is shown a randomly structured profile. (The exact definition of a random line is extremely difficult. All that is intended by the foregoing statement is that the profile was sketched freehand without any preconceived physical constraints, so that a general convoluted shape was achieved.) Random walk estimates of the perimeter of this profile were made using a series of stride magnitudes. The data for this experiment are summarized in the graph of Fig. 10. It can be seen that, even for this first attempt at a random profile, the fractal dimension is a measurable quantity descriptive of the structure of the profile. The slope of the curve yields an estimate of the fractal dimension of 1.16, which is not very

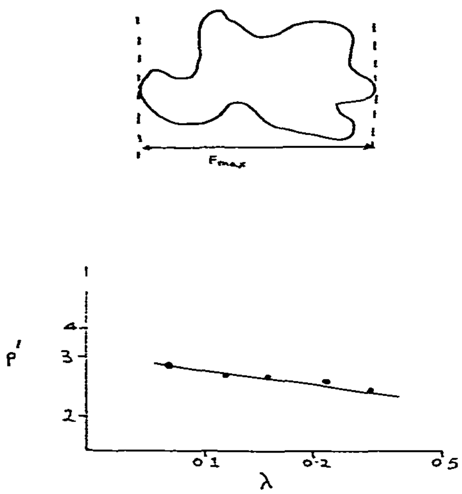

Fig. 10. Even for a randomly sketched profile, the fractal index seems to be a characteristic parameter. λ = stride magnitude expressed as a fraction of the maximum Feret's diameter of the profile; p' = estimate of the perimeter of profile expressed as multiples of the maximum Feret's diameter of the profile; F_{max} = maximum Feret's diameter.

different from that of the carbon black profile of Fig. 9, and again one would not intuitively have described the two as being of grossly different ruggedness, although the carbon black floc appeared to be a little more indented. The numerical difference between the two indices is probably within the range of uncertainty in the experimental data.

An interesting situation arises in fine particle science which has not been considered by Mandelbrot. Often agglomerates of fine particles are constituted of smooth spheres which have been fused together during the formation of larger fine particles. Thus carbon black flocs have this type of structure, and many fly-ash agglomerates are composed of small glass spheres sintered together. This raises the interesting fact that although the overall fine particle profile may be a fractal, the substructure of the obvious units in an agglomerate is Euclidean in its structure. Therefore the plot of the random walk perimeter estimate against the logarithm of the stride should show a discontinuity when the level of scrutiny begins to approach that of the dimensions of the discernible subunit. To test this possibility, measurements were made on the simulated carbon black flocs shown in

Fig. 3. The simulated floc was used in preference to the real one since the diameters of the spheres forming the agglomerates were known exactly. Again, all the measurements are normalized with respect to the maximum Feret's diameter of the agglomerated fine particle system. In Fig. 11 the experimental data for a series of measurements on the two flocs of Fig. 3 are summarized. It can be seen from the data of Fig. 11 that both agglomerates appear to have high fractal dimensions but that the logarithmic nature of the relationship breaks down as the step size approaches the radius of the subunit in the agglomerate. These data would seem to indicate that a plot of the perimeter estimate *versus* random walk step not only gives the fractal dimension for the overall boundary of the profile, but, as smaller and smaller steps are used, any tendency towards Euclidean boundary in the subunits causes the curve to tend to a limit. It also seems reasonable to postulate that any edge texture that differs from the overall agglomerate structure would show up as a line of different slope in the region of very small step perimeter estimations. This possibility is being explored in a more exhaustive study, the results of which will be reported at a later date.

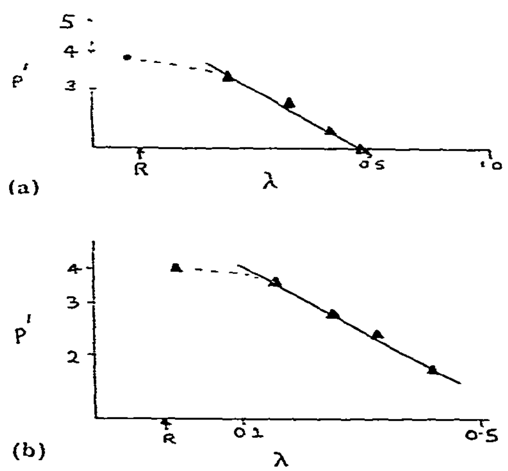

Fig. 11. (a) Agglomerate A, (b) agglomerate B. When the agglomerate with a fractal profile is constituted from subunits having a Euclidean boundary, there is a discontinuity in the fractal dimension curve as the stride of the random walk approaches the magnitude of the subunits. R = radius of sphere from which agglomerate is built; p' = estimated perimeter of profile expressed as multiples of the maximum Feret's diameter of the profile; λ = stride magnitude expressed as a fraction of the maximum Feret's diameter of the profile. Fractal dimension from the slope of the graph: 1.64 for A, 1.60 for B.

From the preliminary data presented in this section, it appears that the fractal dimension could be a useful index of the ruggedness of a profile and that the requirements that the elements of the profile be self-similar could be approximated in many real fine particle situations. In the next section we shall consider several strategies that could be utilized to measure the fractal dimension of a profile.

4. POTENTIAL STRATEGIES FOR THE EVALUATION OF THE FRACTAL DIMENSION OF A FINE PARTICLE PROFILE

In the foregoing sections, the emphasis has been on the estimation of the fractal dimension of a boundary using the random walk strategy based on a series of stride magnitudes. This is a relatively easy technique to implement for graphical evaluation by a human operator. It is not, however, particularly suitable for present automated iconometric systems. One way in which the random walk perimeter estimates could be made is to make use of the light pen feature available in many automated iconometric systems. The light pen enables the operator to participate in the logic procedures for the evaluation of a fine particle. If one were to use two light pens such that the separation of the pens could be altered, one could in fact carry out a random walk around the perimeter as displayed upon a television screen. The algorithm for calculating the number of random steps to complete the perimeter would be relatively easy. The disadvantages of the technique would be basically the slowness of the technique and potential fatigue in the operator. It should be noticed that the actual precision required at any one particular step is not too exacting, since many small statistical errors would average out in the determinations of the slope of the fractal dimension curve. Probably a more realistic alogorithm exploiting the very high speed operations of modern digital computers could in fact track around the profile of the boundary using short search vectors which would constitute the stride of the random walk system.

Another strategy which presents itself as a possibility for measuring the fractal dimension is based upon the intersection frequency of lines placed at random in a given area. Theorems describing the intersection of two lines thrown at random on a field of view have been developed extensively [27]. Practical applications of the intersection frequency of lines placed at random in a field of view have been explored extensively in the subject known as stereology [28]. It now appears that many of the general theorems of this field of mathematics, known as geometrical probability, are actually only true for Euclidean curves, and that if one studies the intersections of Euclidean lines with fractal lines the frequency of intersection will be related to the fractal dimension of the line.

Let us consider a grid of unit length λ. If there are n cells in the grid used to interrogate the profile, the line length of the grid is $2\lambda n$. If L_G is the width of the grid, then

$$n = (L_G/\lambda)^2$$

Therefore the length of the interrogation grid is

$$L_I = 2\lambda(L_G/\lambda)^2 = \frac{2L_G^2}{\lambda} = \frac{2}{\lambda}k$$

where k is a constant for a given search area.

The probability of Intersection P_I is given by

$$P_I = L_I L_C k'$$

where L_C is the length of the curve placed on the grid, and k' is a geometric constant characterization of a square lattice interrogation grid.

It follows that

$$P_I = L_C \frac{2}{\lambda} kk'$$

Let X_λ be the number of intersections of the curve with a grid of space λ. Then X_λ is an estimate of P_I, and

$$X_\lambda = L'_C \frac{2}{\lambda}\alpha$$

where α is a consolidate constant and L'_C is our estimate of curve length. It follows that

$$L'_C = X_\lambda \frac{\lambda}{2\alpha} \qquad (1)$$

Therefore, for a Euclidean curve, a plot of $\log X_\lambda\lambda$ against the $\log_{10}$ of λ should reach a limit for small $\lambda \to 0$.

Since the area-filling capacity of a fractal curve is related to its fractal dimension, it seems reasonable to anticipate that the intersection frequencies for a linear grid superimposed on a fractal curve will be higher than those predicted by considering encounters between Euclidean curves and the grid, by a factor directly proportional to the fractal dimension of the fractal. In a non-rigorous sense, the fractal is a system which spreads out in space by an amount governed by its fractal dimension, so that an ordinary Euclidean line traversing the space occupied by a fractal would be expected to have a higher frequency of intersection with the fractal as compared to those with a Euclidean curve because of the spatial convolutions of the fractal. If this is a reasonable postulate, then eqn. (1) will be modified by a factor proportional to the first power of the fractal dimension. If then one takes logarithms of both sides of the equation, one reaches the conclusion that a plot of the logarithm of the observed intersection frequency times the magnitude of the slide of the interrogation cell against the logarithm of the cell should have a slope equivalent to the fractal dimension. (A rigorous proof of this statement is very difficult. At this time, this proposition is put forward only as a reasonable anticipation, and is not a proven truth. Because of the difficulty of proving general theorems for the meanderings of real fine particle profiles, it is unlikely that a general proof will be established, and one has to accept the possibility of the truth of the proposition from empirical data.)

To explore the possibilities of using grid intersections to evaluate the fractal dimension of an agglomerate, the experiment in Fig. 13 was carried out. A series of grids of various cell size were superimposed on the outline of the agglomerate and its convex hull. The largest grid used had a magnitude of three units of the size illustrated in Fig. 13. Data for the experiment were as follows.

Grid size	Intersection convex hull	Frequency convoluted profile
3 units	39	17
2	42	30
1.5	42	53
1.0	39	86

These data are presented graphically in Fig. 12, and it can be seen that the estimates of

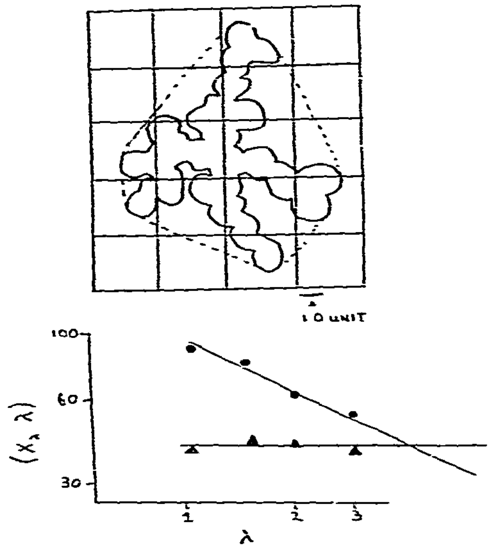

Fig. 12. Estimates of the length of a perimeter by counting intersection frequencies for a series of grids of decreasing cell size demonstrates the fractal nature of the boundary of an agglomerate. --- = convex hull; λ = size of cell used in interrogation grid; $X_\lambda \lambda$ = intersection count × size of cell. From graph, fractal dimension = 1.56.

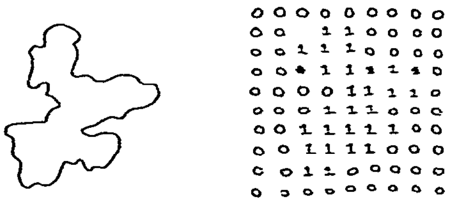

Fig. 13. As explained in the text, a simple three-level mosaic transform of the fine particle at a series of resolutions can be used to deduce the fractal dimension of a rugged fine particle profile. Three-level mosaic transform of the profile: 1, point within the profile; 0, point outside the profile; *, point on the boundary of the profile. Area of profile (estimate) = $\Sigma 1 + \frac{1}{2}\Sigma *$. Intersection frequency of grid with boundary = $\Sigma(1,0) + \frac{1}{2}\Sigma(*1).\ (*0)$.

the perimeter of the convex hull are virtually the same for all grids used in the experiment, whereas the data for the agglomerate profile yield the anticipated linear relationship. The slope of line linking the agglomerate estimate yields an estimate of the fractal dimension of 1.56. This compares with the value of 1.60 determined for the same agglomerate using random walk estimates of perimeter. The discrepancy between these two values is well within the experimental error of the experiments. It therefore seems reasonable to anticipate that grid intersection frequencies will probably yield an estimate of the fractal dimension of a convoluted fine particle profile. This could be a very important factor in the practical implementation of techniques for measuring the fractal dimension, since essentially the grid can be regarded as interrogation of the profile using television scan lines in two directions at right-angles. Therefore present automated iconometric procedures can be directly modified to evaluate the fractal dimension. A series of experiments using different optical magnifications and the same scan line logic should yield the fractal dimension. It can be shown that the square grid is a more efficient interrogation system for a given line length than sets of parallel lines [29]. Sets of measurements using parallel lines should yield an estimate of the fractal dimension, but with less efficiency than a procedure based on grid interrogation.

The possibility that the fractal dimension can be evaluated using line scan logic is very attractive; however, line scan logic still has the problem of thresholding the electrical signal generated as the search beam passes the boundary of the profile. The location of the profile has to be known to within a fair degree of accuracy before intersection frequencies can be evaluated. There is a possibility that an even simpler logic system using a point transformation of the image to be evaluated could be used to estimate the fractal dimension of a rugged fine particle boundary. The physical basis of this logic system is illustrated in Fig. 13. As a first step, the image to be evaluated is transformed into a three-level mosaic in which points lying definitely within the boundary are designated by the symbol 0, and those which probably lie on the boundary by an *. If now we look at this tri-level mosaic, we can deduce that pairs of points in the mosaic which are of the type 0,1; or 1,0 traverse the boundary of the profile grid. All pairs which fall into the category 0,*; *,0; *,1; 1,*; represent potential intersection of the boundary with the grid underlying the mosaic. Statistical reasoning indicates that the sum of half of the pairs in which one point is an * is an efficient estimator of the boundary crossings which are uncertain. Therefore, an unbiased efficient estimator of the number of boundary intersections between the profile and the grid supporting the mosaic used in transforming the profile image is given by the sum of all of the binary pairs which are dissimilar for the symbols 1 and 0, and half of the binary symbol set in which one member of the set is an *. Note that if one were to implement this logic using electronics or electro-optic systems, there is no longer any need to know the precise location of the boundary to be characterized, and only three levels of digitization of any electrical signal are required, with a robust decision being implemented with regard to those points that lie half-way between the on-off signals represented by signals coming from within and without the boundary profile. To evaluate the fractal dimension of the profile, one would basically implement the logic of the grid intersection method using the series of tri-level transformations of different resolution to estimate the perimeter of the convoluted fine particle profile. The possibilities of implementing this logic transformation directly are under investigation, and a full report of the data will be made available at a later date.

5. *DISCUSSION*

In the foregoing sections it has been suggested that part of the difficulty in characterizing convoluted fine particle boundaries arises from their non-Euclidean structure. From the work of Mandelbrot it appears that this type of boundary, and therefore the shape of a fine particle, can be characterized by considering a quantity known as the fractal dimension, which for the fine particle scientist is perhaps better termed a ruggedness index. Preliminary experiments reported in this communication seem to indicate that this is a fruitful area of research for the fine particle scientist. The preliminary experiments reported in this communication have been restricted to the case where the boundaries of the profiles do not contain inclusions, that is, to topological systems of genus 0. However, the mathematics set out in Mandelbrot's book are extended to the case of systems which he describes as having lakes and islands. He also considers problems such as clustering in two-dimensional space of profiles and, therefore, of fine particle systems. Thus his general approach to this subject appears to offer many fruitful avenues to further characterization of rugged fine particle systems. Mandelbrot also discusses the structure of river systems and considers the fractal dimensions of such networks of lines. His mathematics would seem to be applicable to the problem of characterizing the crack structure in unit objects used to explore problems of crushing and grinding theory. It would seem reasonable to suggest that the fractal dimension of a crack structure appearing in a crushed object will be related to the size distribution function arising from the crushing experiment. This possibility is also being explored in current work proceeding at Laurentian University.

ACKNOWLEDGEMENTS

The writer wishes to thank A. G. Naylor, I. Robb and D. T. Wasan for helpful discussions, and Andrew Kaye and Judith Mittermayer for carrying out some of the experimental work reported in this communication. The study was supported by research funds from the National Research Council of Canada.

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