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Project director(s): GOKHALE A M

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Title: EVOLUTION OF MICROSTRUCTURAL DISTANCE DISTRIBUTIONS IN NORMAL & MICROGRAVITY

PROJECT ADMINISTRATION DATA

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Security class (U,C,S,TS) : U
Defense priority rating :
Equipment title vests with: Sponsor

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supplemental sheet

Administrative comments - PROCESSED 3.13 REVISION, ATTACHED

GIT X
GEORGIA INSTITUTE OF TECHNOLOGY
OFFICE OF CONTRACT ADMINISTRATION

NOTICE OF PROJECT CLOSEOUT

Closeout Notice Date 10/31/96

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Project Director GOKHALE A M School/Lab MSE

Sponsor NASA/LEWIS RESEARCH CTR, OH

Contract/Grant No. NAG3-1651 Contract Entity GTRC

Title EVOLUTION OF MICROSTRUCTURAL DISTANCE DISTRIBUTIONS IN NORMAL & MICROGRAVITY

Effective Completion Date 960712 (Performance) 961012 (Reports)

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Comments

USE NASA FORM FOR PATENT.

Subproject Under Main Project No.

Continues Project No.

Distribution Required:

- Project Director Y
- Administrative Network Representative Y
- GTRI Accounting/Grants and Contracts Y
- Procurement/Supply Services Y
- Research Property Management Y
- Research Security Services N
- Reports Coordinator (ODC) N
- GTRC N
- Project File Y
- Other N

NOTE: Final Patent Questionnaire sent to PDPI.
MSAD Program Tasks — Ground-based Discipline: Materials Science

Evolution of Microstructural Distance Distributions in Normal Gravity and Microgravity

Principal Investigator: Prof. Arun M. Gokhale Georgia Institute of Technology

Co-Investigators:

No Co-I's Assigned to this Task

Task Objective:
1. To develop the methodology for estimation of distribution of distances between microstructural features by using digital image analysis and stereological techniques.
2. To apply the methodology to quantify the evolution of microstructural distance distributions during the materials processes such as liquid phase sintering.
3. To quantify the role of gravity in the evolution of microstructural spatial distance distributions during materials processes.

Task Description:
Digital image analysis procedures will be developed and applied to obtain the relative locations of microstructural features observed in metallagraphic planes. Computer codes will be developed and utilized to obtain the two-dimensional distance distributions from the image analysis data. Computer simulations of three-dimensional microstructures and stereology will be utilized to obtain quantitative information concerning the microstructural distance distributions in three-dimensional microstructure. The methodology will be applied to quantify the evolution of microstructural distance distributions of tungsten grains during liquid phase sintering of tungsten heavy alloys in normal gravity and subsequently microgravity.

Task Significance:
An important effect of gravity on microstructural kinetics is through its effect on the spatial arrangements of features in microstructure. This research program will develop the techniques for quantitative characterization of the evolution of microstructural distance distributions during materials processes. The results should be useful to gauge the relative contributions of the intrinsic materials processes and gravity on the evolution of the spatial arrangement of features in microstructures during materials processes.

Progress During FY 1995:
Interactive software has been developed to create a "montage" of a large number of microstructural fields in the memory of the image analyzing computer. The software removes the "edge effect" problems associated with individual microstructural fields, and permits measurements of centroid coordinates and sizes of a larger number of microstructural features in a series of microstructural fields. Software codes have been developed to extract the statistical descriptors of the spatial arrangement of the microstructural descriptors of the spatial arrangement of the microstructural features, such as radial distribution function, pair correlation function, and nearest neighbor distribution function. These procedures have been applied to quantify the evolution of the spatial distribution of tungsten grains in the liquid phase sintered specimens of tungsten heavy alloys supplied by Professor German of Pennsylvania State University. Computer codes are developed to simulate the three-dimensional microstructures to study the spatial characteristics like connectivity of the microstructural features.

STUDENTS FUNDED UNDER RESEARCH:

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Task Initiation: 7/94 Expiration: 7/96
Project Identification: 962-25-05-29
NASA Contract No.: NAG3-1651
Responsible Center: LeRC

Printed: 28 FEB 1996

II-643
BIBLIOGRAPHIC CITATIONS FOR FY 1995:

Journals


Presentations


Dr. Mark Bathea,
National Aeronautics and Space Administration,
Lewis Research Center,
21000 Brookpark Road,
Mail Stop 105 - 1,
Cleveland, Ohio - 44135

Re.: Final report for NASA project NAG3-1651

Dear Dr. Bathea,

Thank you for your recent e-mail. I am happy to know that the copies of the publications from the research done on the above project will serve as the final project report. The research work on this project has been extremely useful for establishing quantitative microscopic and image analysis techniques for characterization of spatial arrangement of microstructural features observed metallographically. This progress has been reported in a number of publications. I enclose the copies of these papers. I am in the process of writing two more papers that deal with applications of these techniques to liquid phase sintering, I will send you the copies as soon as I complete them.

I thank you for all the help and cooperation given to me during the course of this research project. I enjoyed our technical discussions during my visits to the NASA Lewis Center; your input was extremely useful. Thank you for arranging for my visits and my seminar at NASA Lewis Center.

I thank Dr. Tom Glasgow for all the help, cooperation, encouragement, and guidance during this project. It was indeed a pleasure to interact with Dr. Glasgow and you during last two years. Please convey my regards to Dr. Glasgow. Please feel free to give me a call if you need any more information on the research work on this project.

With best regards,

Sincerely,

Arun M. Gokhale
Professor of Materials Science and Engineering

School of Materials Science and Engineering, Atlanta, Georgia 30332-0245 U.S.A., FAX 404-894-9140

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Application of Image Analysis for Characterization of Spatial Arrangements of Features in Microstructure

PASCAL LOUIS and ARUN M. GOKHALE

A number of microstructural processes are sensitive to the spatial arrangements of features in microstructure. However, very little attention has been given in the past to the experimental measurements of the descriptors of microstructural distance distributions due to the lack of practically feasible methods. We present a digital image analysis procedure to estimate the microstructural distance distributions. The application of the technique is demonstrated via estimation of $K$ function, radial distribution function, and nearest-neighbor distribution function of hollow spherical carbon particulates in a polymer matrix composite, observed in a metallographic section.

1. INTRODUCTION

Spatial arrangements of features in a microstructure can be quantified in terms of the distances between the corresponding features. Distribution of distances between features in a microstructure is an important microstructural attribute. The microstructural distance distributions affect the microstructural processes that involve short range, and/or long-range particle-particle interactions (or feature to feature interactions, in general). For example, diverse phenomena, such as formation and growth of microcracks during thermal cycling of metal matrix composites, damage accumulation and fracture behavior of composites, particle coarsening, liquid-phase sintering, microstructural evolution during solid-state transformations where nucleation occurs on the second-phase particles, microvoid induced ductile fracture processes in metals and alloys, creep cavitation, and creep crack growth, crucially depend on the distances between the relevant microstructural features.

The microstructural distance distributions may be significantly altered when a material is processed in reduced gravity or microgravity, particularly if a liquid phase is involved. In such a case, it is desirable to quantitatively analyze the changes in the microstructural distance distributions due to reduced gravity and to quantify and model the role of gravity in the microstructural evolution processes.

Very few investigations deal with the quantitative effect of the spatial distribution of microstructural features on the physical/mechanical properties of materials or on the microstructural evolution during materials processing. An important reason for this is the lack of well-developed practical techniques to quantify the spatial distribution of microstructural features and, as a result, the lack of realistic and flexible quantitative models to represent the spatial distribution in the computer simulation or analytical theoretical studies.

It is the purpose of this article to present an image analysis procedure to obtain the basic experimental data necessary to quantify the spatial arrangement of microstructural features in metallographic sections. To characterize spatial distributions, the necessary basic data consist of centroid coordinates and sizes of the microstructural features of interest. These raw data can be processed to compute the descriptors of spatial order, such as nearest-neighbor distribution, radial distribution, $K$ function, pair-correlation function, or parameters such as short range and long range microstructural gradients. An image analysis procedure was developed to automatically extract centroid coordinates (and feature size, etc.) of the microstructural features observed on different fields of view and referred to the same origin: this is essential to compute the distances between the particles that may not be in the same field of view. The image analysis algorithm provides a practical procedure to acquire the centroid coordinates data of thousands of particles to quantify the short-range as well as long-range affinity (or lack of it) among the features. A simple computer program utilizes these raw data to compute the descriptors of spatial distribution, such as nearest-neighbor distribution and radial distribution.

For population of spherical particles, stereological relationships are available to estimate the three-dimensional (3-D) radial distribution from the corresponding experimentally measured two-dimensional (2-D) radial distribution of the particle sections observed in a metallographic plane. In the case of composites having continuous aligned fibers, the spatial distance distributions of fiber centers observed in a metallographic plane perpendicular to the fibers are sufficient for the characterization of the spatial distribution of the fibers and hence no stereology is necessary in such cases.

In the present study, the image analysis procedure is applied to quantify the spatial distribution of hollow spherical carbon particulates in a polymer matrix composite. The measurements are performed on two specimens having different volume fractions of particulates but the same size frequency function. A brief background on microstructural distance distributions is presented in...
II. DESCRIPTORS OF MICROSTRUCTURAL DISTANCE DISTRIBUTIONS

Let \( N \) be the average number of particle or feature centroids per unit volume of microstructure. Focus on one particle whose centroid is at location \((X, Y, Z)\) in the microstructural space. Draw a test sphere of radius \( R \) around this particle (i.e., having the center at \((X, Y, Z)\)). Let \( K(R) \) be the total number of other particle centroids contained in the test sphere. Obviously, the quantity \( K(R) \) depends on the location of the test sphere (i.e., \((X, Y, Z)\)), the distance \( R \), and the spatial arrangement of the particle centroids in the microstructure. An average value, \( K(R) \), can be obtained by averaging \( K(R) \) over all the particle centroid locations \((X, Y, Z)\) in the microstructural space. The function \( K(R) \) is called the \( K \) function\(^{[19-23]} \) and it gives the average number of particles (strictly speaking, particle centroids) in a sphere of radius \( R \) drawn around the centroid of an arbitrary particle or feature of interest in the microstructure. For a collection of randomly distributed particles, \( K(R) \) is equal to \( (4 \pi R^2/3) \cdot N \); a value higher than this signifies clustering, and a lower one indicates repulsion.

The radial distribution function \( G(R) \) is an alternate descriptor of the spatial arrangement of particle (or feature) centroids. It is defined as follows:\(^{[19,20,21]} \)

\[
G(R) = \frac{1}{(4 \pi R^2 \cdot N)} \left\{ \frac{dK(R)}{dR} \right\} 
\]

The quantity \((4 \pi R^2 \cdot N \cdot dR) \cdot G(R)\) is equal to the average number of particle centroids in a spherical shell of radii \( R \) and \( R + dR \) around an arbitrary particle. For a random spatial distribution of the particles, \( G(R) \) does not depend on \( R \) and it is equal to 1. For a clustered distribution of particle centroids, \( G(R) \) is greater than 1 for small values of \( R \). It is composed of a combination of delta functions for an ordered array of particles.

Note that the radial distribution function is related to the derivative of the \( K \) function (Eq. [1]), and hence it is more sensitive to changes in the spatial order than is the \( K \) function. The radial distribution function is particularly useful to quantify short-range particle-particle interactions. For most of the spatial arrangements of particles, \( G(R) \) approaches 1 for a sufficiently large value of \( R \), and it approaches zero as \( R \) tends to 0.

The radial distribution function contains information concerning the spatial arrangement of particle centers, but it gives no information concerning correlations between sizes and relative locations of the particles. For example, it does not tell us if the small particles are preferentially located near the larger particles, and so on. The pair correlation function contains such information. It is defined as follows:\(^{[21,22,23]} \)

\[
C(\Gamma_1, \Gamma_2, \Gamma) = \left\{ \frac{1}{4 \pi R^2 \cdot n_r(\Gamma)} \right\} \left\{ \frac{d^2Q(\Gamma_1, \Gamma_2, \Gamma)}{d\Gamma_2 \cdot d\Gamma} \right\} 
\]

where \( n_r(\Gamma) \) is the particle size distribution function, such that \( n_r(\Gamma) \cdot d\Gamma \) is the number of particles in the size range \( \Gamma \) to \( \Gamma + d\Gamma \) per unit volume of microstructure. The value of \( Q(\Gamma_1, \Gamma_2, \Gamma) \) is the average number of particle centroids belonging only to the particles of size \( \Gamma \) in a sphere of radius \( R \) centered on a particle of size \( \Gamma \). The value of \( C(\Gamma_1, \Gamma_2, \Gamma) \) is the pair correlation function for the particles of the sizes \( \Gamma_1 \) and \( \Gamma_2 \) separated by distance \( R \). The pair correlation function is equal to 1 for all the values of \( R, \Gamma_1 \), and \( \Gamma_2 \). The pair correlation function is particularly useful in the study of processes, such as particle coarsening, where small particles may be preferentially located near the larger ones.\(^{[5,6]} \)

In the deformation and fracture processes, such as ductile fracture and creep crack growth and fracture,\(^{[12,13]} \) the nearest-neighbor distribution function and average nearest-neighbor distance are the relevant microstructural parameters. The nearest-neighbor distribution function\(^{[16,17,18]} \) is described by the probability density function \( P(R) \), such that \( P(R) \cdot dR \) is equal to the probability that there is no other particle centroid in a sphere of radius \( R \) around a given particle and there is at least one particle centroid in the spherical shell of radii \( R \) and \( R + dR \). The average nearest-neighbor distance \( \bar{R} \) is given by the following equation:

\[
\bar{R} = \int_0^\infty R \cdot P(R) \cdot dR 
\]

For randomly distributed point particles, \( \bar{R} \) is equal to \( 0.554 \cdot \langle N \rangle^{2/3} \). In a manner similar to the nearest-neighbor distribution function, one can define 2nd, 3rd, . . . nth nearest-neighbor distributions.\(^{[22]} \)

The materials microstructures are usually characterized from observations on representative 2-D metallographic sections through the 3-D microstructural space of interest. Thus, the basic microstructural data often pertain to the 2-D metallographic sections. All the preceding descriptors of distance distributions can be analogously defined for the 2-D microstructural sections. The \( K \) function pertaining to 2-D microstructural section, \( k(r) \) is defined as the average number of particle section centroids in a circle of radius \( r \), centered on an arbitrary particle section. The radial distribution function \( g(r) \) pertaining to a 2-D microstructural section is defined as follows:

\[
g(r) = \left\{ \frac{1}{2 \cdot \pi \cdot r \cdot N_a} \right\} \left\{ \frac{dK(r)}{dr} \right\} 
\]

where \( N_a \) is the average number of particle section centroids per unit area of the metallographic section. Note that the lower-case symbols denote the distance distributions in the 2-D sections. The nearest-neighbor distribution function in a 2-D section is described by the probability density function \( p(r) \), such that \( p(r) \cdot dr \) is the probability that there is no other particle section centroid in a circle of radius \( r \) centered on an arbitrary particle section of interest and there is at least one particle section centroid in the circular ring of radii \( r \) and \( r + dr \).
The pair correlation function for 2-D microstructural section \( c(\tau_1, \tau_2, r) \) can be defined as follows:

\[
c(\tau_1, \tau_2, r) = \left\{ \frac{1}{2 \cdot \pi \cdot n_0(\tau_2)} \right\} \cdot \left[ \frac{d^2 q(\tau_1, \tau_2, r)}{dr \tau_2} \right] \quad [5]
\]

\( n_0(\tau_2) \) is the particle-section size distribution observed in the metallographic section. The value of \( q(\tau_1, \tau_2, r) \) is the average number of particle-section centers belonging only to the particle sections smaller than \( \tau_2 \) in a circle of radius \( r \) centered on an arbitrary particle section of size \( \tau_1 \).

If microstructural distance distributions are experimentally measured on representative 2-D metallographic sections, then under certain conditions, the corresponding 3-D distance distributions can be estimated by using stereological relationships. For example, if the particle shape can be assumed to be spherical, then the 3-D radial distribution function \( G(R) \) and the corresponding 2-D function \( g(r) \) measured on a metallographic section are related through the following stereological equation:

\[
g(r) = \left\{ \frac{1}{4 \cdot (\bar{r})^2} \right\} \cdot \int_{0}^{\infty} F(u) \cdot G(2 \cdot u^2) \cdot du \quad [6]
\]

where,

\[
F(u) = 2 \cdot \int_{0}^{u} \left[ 1 - f \left( \frac{u - v}{2} \right) \right] \cdot \left[ 1 + f \left( \frac{u + v}{2} \right) \right] \cdot dv \quad [7]
\]

where \( \bar{r} \) is the average particle radius, and \( f \) is the cumulative size frequency function of the spheres. Note that in Eq. [6], the desired 3-D radial distribution function \( G \) is under the integral sign, and hence numerical solution to this integral equation is necessary for the estimation of \( G \).

### III. IMAGE ANALYSIS TECHNIQUE

To estimate any of the distance distributions in a 2-D metallographic section, it is necessary to measure the \( (X, Y) \) coordinates of the observed particle-section centers and the corresponding section sizes. To obtain statistically meaningful measures, it is essential to perform these measurements on a large number of particle sections distributed over many fields of view (or frames), and hence automatic digital image analysis is essential: for all practical purposes, these measurements cannot be performed manually. In the digital image analysis, the microstructural distances are measured in units of pixels; the absolute sizes are calculated from this information by using the calibration factor, which depends on the microscope magnification. The minimum distance that can be measured is 1 pixel. For estimation of the particle section sizes with a measurement error of less than 10 pct, measurements have to be carried out at a microscope magnification where the particle image sizes are of the order of 10 to 20 or more pixels, so that the measured sizes have an error of less than 1 pixel (i.e., less than 10 pct). At such magnifications, for a number of important classes of microstructures, on the average about 10 to 50 particles are present in an image frame (field of view). Further, in most of the cases, microstructural distances up to about 5 to 10 times the mean free path (average uninterrupted surface to surface distance between the particles through the matrix) have to be measured to obtain meaningful distance-distribution data. Thus, it is necessary to measure the distances between the particles that may not be in the same image frame or field of view (Figure 1). To measure such distances, it is obviously necessary to know the distances between the different image frames on the metallographic plane with an error of less than few pixels. Alternately, a procedure must be developed to obtain the \( (X, Y) \) particle center coordinates of the particles in all the different image frames referred to the same \((0, 0)\) origin; the distance between the centers of the particles situated in different image frames can be then computed by using simple distance formula of coordinate geometry. This approach is adopted in the present work. The limitation on the size of the image frame (typically 512 \( \times \) 512 pixels) also creates another problem due to the associated "edge effect." It is necessary to properly account for the particles that are only partly in the image frame, and their centers may be outside the image frame. Ignoring the particles on the edges of the image frame may introduce a bias of unknown extent in the measured distance distribution functions. This bias due to edge effect can be eliminated for all the practical purposes if a large number of contiguous fields of view (say about 35 to 50) can be precisely "pasted" together by developing an appropriate image analysis software. In the present work, such software is developed for the VIDAS image analysis system supplied by Carl Zeiss, Inc. (Germany). However, the basic algorithm is applicable to any modern image analysis system. Once different contiguous image frames are pasted together, the distance between the consecutive frames is simply equal to length of the image frame (i.e., 512 pixels in the present case), and hence the \( (X, Y) \) coordinates of the particle centers in the different image frames can be referred to the same...
(0, 0) origin by simple translation of the coordinates. The resulting coordinates and the respective particle sizes then form the basic raw data necessary to compute any distance distribution. The algorithm for the image analysis software to paste the image frames and to extract the \((X, Y)\) center coordinates and the particle image size data is as follows.

The first image frame (field of view) is selected arbitrarily and stored in the memory of the image analyzing computer. The right border (having about a 50-pixel width) of this image is pasted on the left edge of the next approximately contiguous live-image frame (field of view), and the microscope stage is moved and adjusted so as to fit the right border of the previous frame to the left edge of the live image within about 10 to 20 pixels; this rough matching is done manually by the operator. At this point, the image of the live frame is grabbed and translated pixel by pixel until it perfectly matches with the right border of the previous field displayed on the screen.* Once a satisfactory match (within one pixel)

*Note that the image can be translated by a preset number of pixels via a combination of commands available in the image analyzer.

is achieved, the second image frame is stored. The same procedure is continued with successive image frames. In this manner, a composite global frame of precisely contiguous 35 image fields (5 x 7 field of view) is stored in the computer; the upper limit on the number of image fields is set by the hard disk size and not by the image-analysis procedure. The successive image fields in the composite global frame are numbered in a serial order. The top left-hand corner of the first frame is designated as \((0, 0)\) and is origin of the coordinate system (Figure 2).

Although the composite global image consisting of 35 fields of view is stored in the computer memory, the image analyzer can characterize only one image frame of the size 512 x 512 pixels at a time, and hence the creation of a composite global image by itself does not eliminate the edge effect. To solve this problem, a measurement frame of one-fourth of the size of 512 x 512 pixel frame is defined. The composite global image is divided into a number of such frames. For example, in Figure 3, the composite image of 16 fields of view is divided into 49 contiguous measurement frames. One-fourth of each of these stored image frames, containing the corresponding part of the measurement frame, is recalled on the analysis screen of the image analyzer; the measurement frame is at the center of the screen. Figure 4 shows this arrangement. In Figure 4, the dashed lines are the boundaries of the measurement frames; the measurements are performed on the measurement frame located in the center and completely inside the analysis screen. The measurements are performed only on those particles whose centroids are inside the measurement frame (for example, dark particles in Figure 4). The image analyzer gives \((X', Y')\) centroid coordinates and particle sizes of these selected particles. At these stage, the centroid coordinates \((X', Y')\) are with respect to the \((0', 0')\) origin at the bottom left-hand corner of the measurement frame, whose location with respect to the fixed origin \((0, 0)\) of the composite global frame (Figure 2) is precisely known. Thus, \((X, Y)\) centroid coordinates with respect to the fixed \((0, 0)\) origin of the composite global frame can be calculated from local \((X', Y')\) centroid coordinates by simple coordinate translation. The \((X, Y)\) centroid coordinates and corresponding particle sizes are stored in a file as a series of triplets.

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*Fig. 2—Coordinate reference for a global image made of 16 contiguous fields of view.

*Fig. 3—Division of composite global image frame made up of 16 contiguous fields of view (solid lines) into a set of measurement frames (dashed lines).

*Fig. 4—The measurement frame for image analysis.
of numbers. The particles, which are partly in the measurement frame and whose centroids are outside the measurement frame under consideration (white particles in Figure 4), are accounted for and measured when the corresponding measurement frames containing their respective centroids are analyzed.* In this manner, centroid coordinates and particle size of different particles are measured, and they are reported only once. There is a band of exclusion of about 100-pixel width all around the composite global image (consisting of 35 fields of view), which does not belong to any measurement frame (region bounded by broken lines and global frame boundaries in Figure 3), and hence no measurements are performed on the particles whose centroids are in this band. The error due to this “global” edge effect is expected to be very small, and it decreases with the increase in the number of fields of view in the composite global image.

The data on the \((X, Y)\) centroid coordinates and the particle section sizes form the input for the computer program used to calculate the \(K\) function, radial distribution function, and the nearest-neighbor distribution function pertaining to the 2-D metallographic section on which the measurements have been performed. To calculate the \(K\) function, the program proceeds as follows. Let \(m\) be the total number of particles on which the measurements are performed, and let \((X_1, Y_1), (X_2, Y_2), \ldots (X_m, Y_m)\) be the centroid coordinates of the particles having the sizes \(r_1, r_2, \ldots r_i, \ldots r_m\). The program calculates the distances between every particle \((X_i, Y_i)\) and all other \((m-1)\) particles. It then computes the number of particle centroids at a distance less than \(r\) from the particle at \((X_i, Y_i)\), and repeats this calculation for all values of the index \(i\), from 1 to \(m\); the average value of this quantity (properly normalized by magnification) is the estimate of the \(K\) function for that value of \(r\). The procedure is repeated for all the desired values of \(r\) to obtain the estimate of the function \(k(r)\) for the 2-D metallographic section. The program also computes the total number of particle sections per unit area of metallographic plane, \(N_s\). These estimates of \(k(r)\) and \(N_s\) together with Eq. [4] are then utilized to compute the radial distribution function \(g(r)\). The pair correlation function can be also estimated by using a similar procedure. The nearest-neighbor distribution can be estimated simply by recording the distances between the particle centroids and respective nearest particles.

### IV. A PRACTICAL EXAMPLE

The electrical conductivity and other physical properties of polymer matrix composites containing hollow spherical carbon particles in a polymeric resin matrix are of interest for certain electrical shielding applications. In such a material, the conductivity of the polymeric matrix is significantly lower than that of the carbon particulate. Thus, the conductivity and other electrical properties of the composite are very sensitive to the spatial arrangement of the carbon particles in the microstructure and the interparticle contacts among these particles. The experimental measurements of the microstructural distance distributions of the hollow carbon particles in the polymer resin matrix have been performed to demonstrate the image analysis procedure developed in Section III. The measurements are performed on this composite because it is easy to fabricate in the laboratory, its electrical properties are sensitive to the microstructural spatial arrangement, and the material is of practical interest for certain aerospace applications.

#### A. Material

The hollow spherical carbon particles were supplied by Carbosphere Inc. (United States). Figure 5 shows the size distribution of these particles. The polymeric resin (araldite) and the hardener were supplied by CIBA GEYG (France). The resin, the hardener, and the particulate powder were mixed in the appropriate proportions, and the mixture was heated in vacuum to degas the material and to decrease the viscosity of the polymer. It takes about 10 hours for the polymer to harden completely, and hence to minimize the particle segregation due to gravitational effect (the specific gravity of particles is significantly lower than the liquid polymer), the molds were slowly rotated during the hardening of the polymer. The specimens having particulate volume fraction of 0.29 and 0.47 were prepared in this manner.

#### B. Materialogy and Image Analysis

The specimens were cut, mounted, and polished on a series of SiC polishing papers, followed by polishing on diamond cloths using automatic polishing wheels. Figure 6 shows a typical microstructure of the composite observed on a random metallographic plane. The carbon particles are round in shape, and there is a range of particle sizes. In Figure 6, the dark regions are the “hollows” created by the intersections of hollow spherical particles with the sectioning plane. The bright regions

![Image Analysis vs. Laser Granulometry](https://example.com/fig5.jpg)

**Fig. 5.—Frequency distribution of the particles.** The volume frequency is the fraction of total particle volume in a given size range. The curves given by image analysis and granulometry are in reasonable agreement with each other.
are due to near-tangential intersections between the carbon particle shells and the metallographic plane. The contrast between the particles and the matrix is not good, and hence a number of image transformations are necessary to develop the appropriate contrast via image processing.125,204

The image analysis and measurements were performed at a microscope magnification of 500 times, yielding a pixel size of 0.325 \( \mu \text{m} \). At this magnification, on the image analyzer screen, the average particle size is about 50 pixels, permitting estimation of particle sizes and interparticle distances with an error of less than 5 pct. In the low volume fraction specimen (29 pct particulate), the measurements were performed on 105 fields of view (at magnification 500 times) spread over three independent composite global image frames (35 fields of view each). In the specimen containing 47 pct carbon particulate, the measurements were performed on 70 fields of view at magnification 500 times spread over two independent composite global image frames, each composed of 35 fields of view. The image analysis procedure was used to obtain the \((X, Y)\) centroid coordinates and the particle section sizes, as described in the last section. The \(k(r)\) function, the radial distribution function \(g(r)\), and the nearest-neighbor distribution function \(p(r)\) were estimated from these data by using a computer program written in the C language. These results are discussed subsequently.

C. Results and Discussion

Figure 7 shows the variation of the function \(k(r)\) with the microstructural distance \(r\) between the particle section centroids observed in a metallographic plane. Recall that \(k(r)\) represents an average number of particle section centroids at a distance less than \(r\) from an arbitrary particle centroid. The value of \(k(r)\) increases monotonically with \(r\), it approaches 0 as \(r\) approaches 0, and it tends to infinity as \(r\) goes to infinity, as expected. At any given value of \(r\), \(k(r)\) is higher for the high volume fraction specimen than for the lower volume fraction specimen. Figure 8 shows the radial distribution function \(g(r)\) for the two specimens plotted against the normalized microstructural distance \((r/\bar{r})\), where \(\bar{r}\) is the average particle

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Fig. 6—Microstructures observed in a metallographic section: (a) 29 pct particulate volume fraction specimen at magnification 200 times; (b) 29 pct particulate volume fraction specimen at magnification 500 times; (c) 47 pct particulate volume fraction specimen at magnification 200 times, and (d) 47 pct particulate volume fraction specimen at magnification 500 times.
Fig. 7—Estimated K-function of the carbon particle sections observed in a metallographic plane.

Fig. 8—Estimated radial distribution function of the carbon particle sections observed in a metallographic plane.

Fig. 9—Estimated nearest-neighbor probability distribution of the distances between carbon particles in the polymer matrix composite.

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REFERENCES

COMPUTER SIMULATION OF SPATIAL ARRANGEMENT AND CONNECTIVITY OF PARTICLES IN THREE-DIMENSIONAL MICROSTRUCTURE: APPLICATION TO MODEL ELECTRICAL CONDUCTIVITY OF POLYMER MATRIX COMPOSITE

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Abstract—Computer simulation is a powerful tool for analyzing the geometry of three-dimensional microstructure. A computer simulation model is developed to represent the three-dimensional microstructure of a two-phase particulate composite where particles may be in contact with one another but do not overlap significantly. The model is used to quantify the "connectedness" of the particulate phase of a polymer matrix composite containing hollow carbon particles in a dielectric polymer resin matrix. The simulations are utilized to estimate the morphological percolation volume fraction for electrical conduction, and the effective volume fraction of the particles that actually take part in the electrical conduction. The calculated values of the effective volume fraction are used as an input for a self-consistent physical model for electrical conductivity. The predicted values of electrical conductivity are in very good agreement with the corresponding experimental data on a series of specimens having different particulate volume fraction.

INTRODUCTION

Physical properties of multi-phase solids often depend on the arrangement of features in three-dimensional microstructural space. For example, fluid flow through a porous solid depends on the connectivity of the pores [1]. Similarly, electrical conductivity of a two-phase composite containing particles of a conductive material in a dielectric matrix critically depends on "connectedness" of the conducting particulate phase.

Two important aspects of the spatial arrangement of the microstructural features are: (i) distribution of distances between particles (or features); and (ii) "connectedness" of the microstructural features in three-dimensional microstructural space. Several rigorously defined and physically meaningful descriptors of inter-particle distance distributions are available (for example, nearest neighbor distribution function, radial distribution function, etc. [2-4]). The choice of a specific descriptor obviously depends on the physical phenomena or property under investigation. Recently, an efficient and practical digital image analysis technique has been developed for statistically reliable estimation of inter-particle distance distributions observed in a metallographic plane [5]. The three-dimensional inter-particle distance distributions can be estimated from the corresponding planar data under certain conditions [6, 7]. On the other hand, practical experimental techniques are not available for estimation of the attributes related to the "connectedness" of microstructural features in 3D space, from the observations on the metallographic sections. Further, the attributes of the "connectedness" that are of relevance to the phenomena, such as electrical conduction, are not well understood. In this context, the topological connectivity of the particle surfaces (i.e. genus or first Betti number) may not represent the relevant aspect of the particle connectivity. This leads to difficulties in establishing meaningful quantitative correlations between the microstructure and electrical conductivity of two-phase (or multi-phase) solids containing particulate of a conducting phase in a dielectric matrix of another phase.

Computer simulation of three-dimensional microstructures is an attractive alternative for studying the spatial arrangement of particles (features), and for developing the microstructural parameter(s) that represents the relevant aspects of the "connectedness". A microstructure based physical model for electrical conductivity of two-phase solids can be developed from the resulting information. This paper reports a new computer simulated model of three-dimensional microstructure containing polydispersed spherical particles that may be in mutual
A brief background on the characterization of spatial arrangements of particles and computer simulation of microstructures is given in the next section. The new computer simulated microstructure model and experimental work are presented in the subsequent sections. Finally, the self-consistent microstructure based physical model for electrical conductivity of a two-phase solid is presented and the predictions of this model are compared with the experimental data on electrical conductivity.

**BACKGROUND**

*Computer simulation of microstructures*

For computer simulation of any modeled microstructure, the first step is the representation of the microstructural space by a set of regularly and closely spaced discrete coordinate points \((X, Y, Z)\). This is equivalent to the generation of a three-dimensional "graph paper". The total number of coordinate points depends on the volume of the space to be simulated, and the distance between the consecutive points. Once the microstructural space is simulated, the particles (or features) are placed in this "box" one by one: the size, shape and location of the particles are chosen according to a set of rules that make up a microstructure model. The most simple scheme is the so called "random hard sphere" model, where particle overlap is not allowed [8]. From three sets of numbers (say, \(1-n\), \(1-m\) and \(1-p\), three numbers (one from each set) are chosen at random. These three numbers (say, \(X_0, Y_0, Z_0\)) constitute the coordinates of the center of the first sphere. Once the radius of this sphere (say, \(R_0\)) is specified (according to some fixed scheme), the equation of this sphere is known, and the first particle is essentially placed in the box. One now chooses the center and the radius of the second "probable" particle by using the same scheme. From the known equations of the surfaces of the first and the second sphere, it can be detected if these two spheres intersect. If they intersect, then the second particle is rejected, new center coordinates and radius are chosen using the same algorithm, and the process is repeated till the second particle is located such that it does not intersect the first one. Using the same logic, the third particle is chosen so that it does not intersect the first two particles. The process is repeated till the space is filled with the required volume fraction of the spheres having preset size distribution. The microstructure is thus represented by a set of equations of the surfaces of non-overlapping spheres. Figure 1 shows a schematic two-dimensional microstructure generated by the "hard sphere" (i.e. hard "circles" in 2D) model. The "hard sphere" model is useful for simulating a microstructure consisting of uniformly distributed spherical particles that do not touch one another. Another popular microstructure model is the Boolean model, where randomly distributed convex particles (for example, spheres) are allowed to freely overlap to any extent [9]. Figure 2 shows a schematic representation of a two-dimensional microstructure generated by the Boolean model. Several more sophisticated models are available in the literature [10, 11]. However, such models are designed for specific applications, and they require powerful mainframe computers.

The particulate volume fraction and size distribution are input parameters for all the simulated microstructures. It follows that the microstructures simulated from different models differ mainly in the
second order attributes such as spatial arrangement of particles, rather than the first order parameters such as volume fraction. Thus, a quantitative comparison of the relevant aspects of spatial arrangement of particles in the true and simulated microstructures is essential for choosing a specific model to represent a given microstructure. The spatial arrangement of particle section centers in a random plane through three-dimensional simulated microstructure can be quantified in a straightforward manner. A comparison with the corresponding attributes measured on a random metallographic plane through true microstructure provides an objective criterion to verify whether a simulation model closely represents the true microstructure. The three-dimensional spatial arrangement of particles in true microstructure can then be approximated by the corresponding arrangement in the simulated three-dimensional microstructure: this is the main objective of the computer simulations in the present study.

Descriptors of spatial arrangement of particle centroids

An important descriptor of the spatial distribution of particle centroids is the nearest neighbor distribution function [2]. It is described by the probability density function $P(R)$ such that $P(R) \cdot dR$ is the probability that there is no other particle centroid in a sphere of radius $R$ around an arbitrary particle, and that there is at least one particle centroid in the spherical shell of radii $R$ and $(R + dR)$. In a metallographic plane (two-dimensional section) through three-dimensional microstructure, the nearest neighbor distribution function of particle section centers is defined by the probability density function $\delta(r)$ such that $\delta(r) \cdot dr$ is the probability that there is no other particle section center in a circle of radius $r$ around an arbitrary particle section, and that there is at least one particle section center in a circular shell of radii $r$ and $(r + dr)$.

Another important descriptor of the spatial distribution of particle centroids is the radial distribution function [2-4]. It is described by the function $G(R)$ such that $\pi R^2 \cdot N_v \cdot G(R) \cdot dR$ is equal to the average number of particle centroids in a spherical shell of radii $R$ and $(R + dR)$ around an arbitrary particle centroid. $N_v$ is the average number of particle centroids per unit volume of three-dimensional microstructure. Therefore, for a completely random distribution of point particles, $G(R)$ is equal to one. Analogously, the radial distribution function in a metallographic plane, $g(r)$, is defined such that $2\pi r \cdot N_v \cdot g(r) \cdot dr$ is equal to the average number of particle section centers in a circular shell of radii $r$ and $(r + dr)$ around an arbitrary particle section. $N_v$ is the average number of particle section centers per unit area of metallographic plane.

A practical and efficient digital image analysis technique is available for experimental estimation of the radial distribution function $g(r)$ and the nearest neighbor distribution $\delta(r)$ in a metallographic plane [5]. These functions can also be calculated for a random plane through simulated three-dimensional microstructure. If the simulated microstructure is a good approximation of the true microstructure, then experimental measurements of $g(r)$ and $\delta(r)$ functions must be in good agreement with those calculated for the simulated modeled microstructure. In such a case, the connectivity and other attributes of connectedness of the particles in true three-dimensional microstructure (which cannot be measured) can be approximated by the corresponding values for the simulated modeled three-dimensional microstructure (which can be calculated). This is the approach adopted in the present investigation.

EXPERIMENTAL WORK

Material

The experiments were performed on a series of specimens of polymer matrix composite consisting of hollow spherical carbon particles dispersed in a hardened polymer resin matrix of Araldite. The particulate volume fraction was varied from 0.16 to 0.47, but the particle size frequency function was the same for all the specimens. The hollow spherical carbon particulate powder was supplied by Carbosphere Inc.; the average particle diameter was 10.6 µm. The liquid polymeric resin (Araldite) and the hardener were supplied by Ciba Geigy. The appropriate proportions of the particulate powder, the liquid Araldite resin and the hardener were thoroughly mixed to ensure uniform distribution of the carbon particles, and the mixture was heated in vacuum to degas and to decrease the viscosity of the polymer. The molds were slowly rotated during the hardening period of the polymer (which is approximately 10 h) to avoid any segregation of the particles due to gravitational effects.

Electrical conductivity measurements

The resistivity (and therefore, the conductivity) was measured by using the well-known "three point"
Fig. 3. Microstructure of a typical composite specimen observed on a representative metallographic plane (particulate volume fraction = 0.29); magnification, 500 x.

resistivity measurement technique. Cylindrical specimens of 14.25 mm diameter and 20 mm length were used for the conductivity measurements. Silver based conductive paint was coated on both the flat ends of the specimens, and a constant electrical current was applied across these two ends. A 1 mm wide circular ring was painted (with conductive paint) at about one fourth of the specimen length from each flat end, and the potential drop was measured across these two segments. The conductivity was calculated from the measured potential drop and the known constant applied current.

Metallography

The specimens were cut, mounted and polished on a series of SiC polishing papers, and then followed by polishing on diamond cloths on automatic polishing wheels. Figure 3 shows a typical microstructure revealed in this manner. Note that the carbon particles are round, and there is a range of particle sizes. In Fig. 3, the dark regions represent the "hollows" created by intersections of polishing plane with the hollow carbon particles; the bright regions are due to the near tangential intersections.

Digital image analysis

In this material, the contrast between the particles and the matrix is not very good, and therefore several image editing steps are necessary to develop the appropriate image contrast for subsequent measurements on the digitized images [12]. The image analysis was carried out on the images at the microscope magnification of 500 x. At this magnification, the observed particle sizes are about 50 pixels, which allows the measurements of particle sizes and inter-particle distances with an error of less than ±5%. The measurements were performed on more than 100 fields of view on each specimen to ensure sampling of at least 3000 particles from each specimen. The centroid coordinates (referred to the same origin) and the particle section size of each particle were measured using an image analysis procedure described in detail elsewhere [5]. From these data, the radial distribution function and nearest neighbor distribution function of the particle section centroids in the metallographic plane were estimated (see Ref. [5] for details). These descriptors quantify the spatial arrangement of the particle sections in the metallographic plane. Figures 4 and 5 show the radial
distribution function and the nearest neighbor distribution function obtained in this manner. The particle section sizes in the metallographic plane were cast into the section size distribution, and the true threedimensional particle size distribution was estimated from these data by using stereological techniques [13]. Figure 6 shows the true particle size distribution estimated in this manner together with the size distribution of the carbon particulate powder measured directly by laser granulometry. An excellent agreement between the two distributions shows that the measurements on about 3000 particles yields an excellent representative sampling of the microstructure.

NEW COMPUTER SIMULATED MICROSTRUCTURE MODEL

In the microstructure of the present composite, the carbon particles are uniformly distributed in the matrix. These particles may (and do) touch one another, but they do not overlap significantly, i.e. they remain rigid spheres. A valid microstructure model must allow for these simple geometric aspects of the true microstructure. It follows that the "hard core sphere" model, where the particles are not allowed to touch one another (see Fig. 1) is not applicable to the present material. The Boolean model [9], which permits extensive overlaps among particles (see Fig. 2) is also not applicable. In the new model described below, the particles are first generated according to the Boolean model, which enables uniform distribution of particles. In the subsequent step, the overlapping particles are pulled apart from one another according to a repulsion law that allows maximum overlap up to one tenth of the particle diameter. This leads to a microstructure having uniformly distributed spherical particles that may have inter-particle contacts but no significant particle overlaps. Figure 7 shows a two-dimensional section through a typical three-dimensional microstructure simulated in this manner. It is worthwhile to compare Fig. 7 with the schematic microstructures shown in Figs 1 and 2. The details of the simulation steps of the new model are as follows.

1. The volume of the three-dimensional microstructural space to be generated is specified depending on the average sphere size; typically, a cubic microstructural volume having edge length of approximately 10-30 times average particle diameter is sufficient to obtain meaningful statistical averages. The simulated microstructural volume is periodic, and therefore, there is no edge effect.

2. The size frequency function of the spheres is specified in terms of a histogram representation having 50 regularly spaced size classes. The absolute number of spheres in a given size class is directly proportional to the particle volume fraction to be simulated.

3. The particles are then generated one by one according to the Boolean model [9], where the particle centroids are randomly located in the microstructural space and particles are allowed to freely overlap.

4. The overlap vector of a pair of overlapping ith and jth particles \( V_{ij} \) is defined as the shortest vector displacement of the centroid of the ith particle necessary to completely separate the overlapping ith and jth particles.

5. The particles are analyzed for overlap one by one. Suppose that there are a total of \( N \) particles and the nth particle is being analyzed, then its overlaps with only the next \((N - n)\) particles in the queue are analyzed; the overlaps with the first \((n - 1)\) particles were analyzed when these particles were under attention. This scheme eliminates redundancy and makes the computation more efficient.

6. Suppose the nth particle overlaps with the ith, jth and kth particles (where, i, j and k > n), then the net displacement vector of the nth particle, \( V_n \) is given by the following equation.

\[
V_n = \frac{V_{ni} + V_{nj} + V_{nk}}{M}
\]

The three displacement vectors on the right-hand side of equation (1) pertain to the overlap vectors for the particle pairs \( n \) and \( i \), \( n \) and \( j \), and \( n \) and \( k \), as defined in step (4). Repulsion strength, \( M \) is one of the parameters of the model. In the present study, \( M \) was set to be equal to 2. Figure 8 illustrates the geometry involved in equation (1).

7. The net displacement vector [step (6)] is calculated for all the overlapping particles in the system.
and the centers of these particles are shifted by the corresponding net vector displacements.

(8) After the overlapping particles are displaced by their corresponding net vector displacements, the overlap vectors of all the overlapping particle pairs (in the new spatial arrangement) are recalculated, and the magnitude of maximum particle overlap, \( \tau \), is calculated.

(9) The maximum permissible particle overlap, \( \epsilon \), in the final microstructure is an input parameter of the model whose value has to be specified. In the present case, \( \epsilon \) was set to be equal to one tenth of the average particle diameter. If \( \tau > \epsilon \) then, the steps (5)–(8) are repeated as the next iteration of the simulation. These iterations are continued until the actual maximum overlap \( \tau \) reaches a value lower than the set maximum value \( \epsilon \); at that point the simulation is terminated. Figure 7 shows a two-dimensional section through a typical three-dimensional microstructure simulated in this manner.

\textit{Adjustable model parameters}

The maximum permissible particle overlap, \( \epsilon \), and the repulsion strength, \( M \), are the adjustable parameters of the model. The rate of convergence of the simulation (i.e. number of iterations after which actual maximum overlap, \( \tau \) is less than \( \epsilon \)) depends on these two parameters. As \( \epsilon \) approaches zero, the necessary number of iterations approaches infinity. In such a limiting case, the simulated microstructure reaches a stage where only tangential \textit{point} contacts between the particles are allowed. A very large value of \( \epsilon \) leads to a simulated microstructure having extensive particle overlaps (as in the Boolean model), which is not realistic in the present context. A value of \( \epsilon \) equal to one-tenth of the average particle diameter yields microstructure that is close to the actual microstructure, and permits completion of simulation.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig7.png}
\caption{Two-dimensional section through typical microstructure simulated from new model. Note that the particles touch, but do not overlap significantly.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig8.png}
\caption{Definition of the net displacement vector.}
\end{figure}
within a reasonable time. The resulting quantitative data on spatial distribution of particles are also in good agreement with those experimentally measured by image analysis. It is necessary to point out that just because the maximum permissible overlap is set at one-tenth of the average particle diameter, it does not mean that all the overlapping particle pairs have this extent of overlap; most of the overlapping particle pairs in the final simulated microstructure are expected to have an overlap significantly less than \( \epsilon \).

The repulsion strength \( M \) is the second adjustable parameter of the present model. The higher the value of \( M \), the greater the number of iterations necessary to reach the final simulated microstructure. The value of \( M \) does not affect the final simulated microstructure significantly. In the present study, \( M \) was set to be equal to 2, which is the minimum value that guarantees that the particles that are in contact in the first step of simulation do remain in contact at all stages of the simulation, as well as in the final simulated microstructure.

**Spatial arrangement of particles in a random two-dimensional section through simulated three-dimensional microstructures**

Once the three-dimensional microstructure is simulated, the coordinates of the centers of particle sections (as well as section sizes) present on a two-dimensional section through the simulated three-dimensional microstructure can be calculated. From this information, the radial distribution function and nearest neighbor distribution of particle section centers on the two-dimensional section through a simulated microstructure can be calculated. If the modeled structure is a reasonable representation of the true microstructure, then the corresponding descriptors experimentally measured on a metallographic plane through real microstructure should be in good agreement with the simulated data. This is a crucial step for the validation of the microstructure model. Figure 9 shows the experimental and simulated radial distribution functions. The simulated functions are in good agreement with the experimental data. Figure 10 shows a plot of average nearest neighbor distance observed on a two-dimensional section versus the particulate volume fractions, for the present model together with some experimental data; the experimental data are in reasonable agreement with the simulated curve. Figure 11 compares the nearest neighbor distribution function on a two-dimensional section through the simulated microstructure, and the experimentally measured distribution on a metallographic plane. Again a reasonable agreement is observed between the experimental and the simulated distributions. Thus, it can be concluded that the spatial arrangement of the particles in the simulated microstructures is a good approximation of the spatial arrangement of hollow spherical particles in the polymer matrix composite specimens under present investigation. We can now go on to analyze the spatial connectivity of the carbon particles (which cannot be experimentally measured) by using the simulated microstructures.

**ANALYSIS OF CONNECTIVITY OF THE PARTICLES**

In the present polymer matrix composite, the electrical conductivity of the dielectric resin matrix is \( 10^{-14} \) S/m, whereas the conductivity of the carbon particles is 200 S/m (these conductivity data were given by the manufacturer): the conductivity of the two phases differ by 16 orders of magnitude. Thus, if electrical voltage is applied across two opposite faces of a specimen, for all practical purposes, the electrical conduction will occur only if there are continuous chains of touching carbon particles extending from...
Fig. 11. Experimental and simulated nearest neighbor distribution function on a two-dimensional section.

one face of the specimen to another face. If there is not a single particle chain connecting the two faces of the specimen, then the conductivity is expected to be zero. Thus, there is a microstructural percolation threshold: there exists a critical volume fraction $f^*$ below which the probability of the formation of a continuous particle chain is almost zero, and consequently the electrical conductivity is also zero. This critical volume fraction $f^*$ depends on the size distribution of the particles.

As the particle volume fraction $f$ increases beyond $f^*$, there are more continuous pathways for the electrical conduction, and more particles participate in the conduction process. In this stage, the conductivity is a direct function of the volume fraction of the particles that actually participate in the conduction process. This is illustrated in Fig. 12. In this figure, the bright particles are not connected to any chain, and therefore they do not participate in the conduction process. The black particles that form continuous chains contribute to the electrical conduction. The cross-hatched particles touch the chains formed by black particles, but they do not contribute to the electrical conductivity. A particle actively participates in the electrical conduction process if and only if from that particle one can travel through the particulate phase in at least two distinctly different directions (each path through different inter-particle contact belonging to the same particle) and reach the two external surfaces of the specimen across which an electrical potential is applied, without ever leaving the particulate phase. Therefore, the most simple microstructural parameter that is relevant to the electrical conduction process is the volume fraction $f_c$ of only those particles that actively participate in the conduction process according to this definition. To the best of our knowledge, there is no experimental technique to identify in the material microstructure the particles that are effective in the conduction process, and therefore the effective volume fraction $f_c$ cannot be experimentally measured. However, such particles can be identified in a computer simulated microstructure, and their volume fraction $f_c$ can be calculated.

It has been shown that the overall spatial arrangement of the particles in our simulated microstructures is a reasonable approximation for that in the corresponding actual microstructures (see Figs 9–11). Thus, the effective volume fraction $f_c$ in the real microstructure can be approximated by its value in the simulated microstructure. Figure 13 shows a plot of the effective volume fraction $f_c$ versus the overall particulate volume fraction $f$. The percolation volume fraction $f^*$ is equal to 0.26. Note that for $f < f^*$, $f_c = 0$. There is a discontinuous jump in the effective volume fraction, $f_c$, at $f = f^*$. As the overall particulate volume fraction $f$ increases beyond the percolation volume fraction $f^*$, the effective volume fraction $f_c$ increases.
The following empirical equation represents the computed data in Fig. 13 quite well
\[ f_c = 2.78(f + 0.21)^{0.4} - 2.0 \quad \text{for } f > f^*. \quad (2) \]

The relationship between the effective volume fraction and overall global volume fraction \( f \), is the necessary input for the self-consistent model for electrical conductivity discussed below.

**MODEL FOR ELECTRIC CONDUCTIVITY**

There are a number of theoretical treatments that predict the bounds on the electrical conductivity of two-phase composites. The most simple treatment is the one due to Voigt and Reuss [14]. The composite has maximum possible conductivity when the two phases are stacked in the form of alternate parallel slabs extending from one end of the specimen to another, and the electrical current is applied parallel to the slabs; the conductivity is minimum when the applied current is perpendicular to the slabs (the two arrangements can be visualized as “series” and “parallel” configurations of the resistors). These bounds depend only on the volume fraction of the two phases. Hashin and Strikman [15] have given tighter bounds than those of Voigt and Reuss, however, these bounds are also completely determined by volume fraction of the two phases.

Landauer [16] has given a model for the estimation of electrical conductivity of two-phase composites where the two phases are randomly distributed and isotropic (i.e. no preferred orientation). Although, Landauer’s derivation is based on the assumption that the particulate has a spherical shape, the resulting expression for electrical conductivity is expected to be a good approximation for any two-phase isotropic microstructure having random distribution of the two phases. Landauer [16] has given the following expression for estimation of the electrical conductivity, \( P \), of a two-phase composite material containing particulate

\[ P = \frac{[(3f_1 - 2) - (3f_2 - 2)]P_1 + [(3f_1 - 1) - (3f_2 - 1)]P_2}{(3f_1 - 2) - (3f_2 - 2) - (3f_2 - 2) - (3f_2 - 2)} + 8 \cdot P_1 \cdot P_2^{1/2}/4. \quad (3) \]

In equation (3), \( P_1 \) and \( P_2 \) are the electrical conductivities of the particulate and the matrix, respectively, and \( f_1 \) and \( f_2 \) are their volume fractions. Obviously, \( f_1 + f_2 = 1 \).

In the Landauer’s model it is assumed that all the particles contribute to the electrical conduction, which is reasonable if the conductivities of the particulate and the matrix are not drastically different. However, in the present composite, the conductivities of the carbon particles and the resin matrix differ by sixteen orders of magnitude. Therefore, a particle can contribute to the electrical conduction if and only if there is a continuous path that goes through two different inter-particle contacts of the same particle, and connects opposite faces of the specimen across which an electrical potential is applied (see Fig. 12). The particles that do not satisfy this condition do not contribute to the conduction process, and hence they can be regarded as a part of the matrix, for estimation of the electrical conductivity. Therefore, for the present composite, the volume fractions \( f_1 \) and \( f_2 \) in equation (3) must be appropriately interpreted: \( f_1 \) should be equal to the effective particulate volume fraction \( f_c \), and \( f_2 \) is equal to \((1 - f_c)\). Further, in the present case, the electrical conductivity of the matrix, \( P_c \), can be equated to zero. With these changes, equation (3) takes the following modified form

\[ P = 0 \quad \text{for } 0 \leq f_c \leq 1/3 \]

\[ P = (3f_c - 1) \cdot P_c/2 \quad \text{for } 1/3 \leq f_c \leq 1. \quad (5) \]

Equation (5) predicts that the electrical conductivity should increase linearly with the effective particulate volume fraction, for \( f_c > 1/3 \), and it should be equal to zero for the effective volume fraction less than \( 1/3 \). Substituting the value \( f_c \) equal to \( 1/3 \) in equation (2) yields, \( f = 0.42 \) for the percolation threshold for electrical conduction. Recall that morphological percolation threshold for the initiation of the formation of continuous particle chains (i.e. \( f^* \)) occurs at the particle volume fraction equal to 0.26. Therefore, the morphological percolation threshold and the percolation threshold for electrical conduction predicted by Landauer’s equation are not equal. The morphological percolation threshold gives the particulate volume fraction at which the continuous particle chains begin to form. At this stage, the conductivity is non-zero in one direction (where a continuous chain has formed), and it is zero in all other directions. However, for isotropic electrical conduction, it is essential to have a number of continuous particle chains randomly oriented in three-dimensional space, and this is achieved only at a volume fraction higher than that for morphological percolation threshold. Landauer’s equation assumes isotropy in electrical conduction, and therefore, it is applicable only after a sufficient number of continuous particle chains are formed. Due to this reason, Landauer’s equation predicts a percolation threshold for electrical conduction \((f = 0.42)\) that is higher than the morphological percolation threshold for the formation of continuous particle chains \((f = 0.26)\).

Equation (5) predicts that for \( f_c > 1/3 \), the electrical conductivity varies linearly with the effective particulate volume fraction \( f_c \). Further the slope of the linear portion of the plot should give the effective conductivity of the particulate phase. Figure 14 shows the plot of experimentally measured electrical conductivity vs the effective particulate volume fraction \( f_c \). The values of \( f_c \) were obtained from the global volume fraction \( f \) by using the curve in Fig. 13, obtained from computer simulations. The data is in good agreement with the predictions of equation (5).
Fig. 14. Plot of electrical conductivity versus effective volume fraction $f_c$.

The conductivity is zero for effective particulate volume fraction less than about 0.33, as predicted. However, the slope of the linear region of the plot yields 43 S/m for the effective conductivity of the carbon particulate phase, which is lower than the reported bulk conductivity of carbon (200 S/m) by a factor of five or so. This is not surprising, because the inter-particle contact resistance is expected to be significantly higher than the bulk electrical resistance. Thus, the effective conductivity of the particulate reflects the conductivity of the inter-particle contacts rather than the bulk conductivity of carbon. It follows that the calculated value of 43 S/m essentially represents the conductivity of the inter-particle contacts, and therefore it is reasonable.

CONCLUSIONS

Computer simulation of a three-dimensional microstructure is useful to quantify the microstructural attributes associated with the "connectedness" that are difficult to measure experimentally. In this study, computer simulated three-dimensional microstructures were used to estimate the morphological percolation volume fraction for electrical conduction, and particulate volume fraction that is effective in the electrical conduction in a polymer matrix composite. These calculated values are successfully utilized to predict the conductive particulate volume fraction dependence of the electrical conductivity in a polymer matrix composite where the matrix is dielectric.

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