Determination of Distribution Patterns of Fillers in Composites by Micromorphological Parameters*

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The nonuniform distribution of fillers in a composite is studied both experimentally and analytically. A set of three parameters are proposed to characterize the distribution patterns of fillers in a particulate composite: homogeneity distribution parameter ξ , anisotropy parameter of the first kind η and that of the second kind ϕ . These parameters are used to examine the distribution patterns of 10 % and 20 % SiC particulate/6061 Al (SiCp/Al) composite. This led us to believe application of the three parameters to actual composites provides useful information about the micromorphology of particulates. The distribution pattern of SiCp/Al composite is also examined from the viewpoint of fractal analysis, resulting in some correlation between the fractal analysis and one of the proposed three parameters.

Key Words: Composite Materials, Micromorphology, Filler Distribution, Fractal Analysis

1. A Composite Consists of the Matrix and Fillers

The macroscopic properties of a composite are known to depend upon the micromorphology of the fillers as well as the properties of the constituents. In predicting the macroscopic properties of the composite, it has often been assumed that the distribution of fillers is spatially homogeneous. In reality, the distribution of fillers in composites is not always homogeneous. Thus, strong efforts to effect nearly homogeneous distribution of fillers have been made by composite engineers with the aim of improving the macroscopic properties of the composites. Nevertheless, one cannot avoid inhomogeneous distribution of fillers in composites mainly due to several factors related to processing. For example, particles tend to be clustered along the grain boundaries of the matrix in a particle metal matrix composite⁽¹⁾ due to slow cooling. The degree of inhomogeneous distribution also depends upon the size of the window (or unit cell) through which the analysis is normally made. In this

respect, locally inhomogeneous distribution of fillers in a composite becomes homogeneous as the window size (L) increases so that many more particles are included within the window. Hence, the issue of inhomogeneous distribution of fillers becomes different from that of fractal analysis⁽²⁾ in that the number of fillers within the window N is proportional to L^D where L is the window size, and D is called the fractal dimension and is usually not an integer but a fraction. In the case of extremely inhomogeneous distribution of fillers, the above relation may not be valid, leading to nonconstant D . In such a case, fillers "cluster", and one can observe the number of clusters in a composite. If those clusters are connected to each other such that a continuous path along the connected clusters can be established within the window, then the macroscopic behavior of the composite can be quite different from that of a composite without such continuous clusters. For example, in a composite where conductive fillers are embedded in an insulative matrix, there exists a threshold volume fraction of fillers at and above which the conductive path along the connected clusters is established, exhibiting a switching behavior of the composite from "insulator" to "conductor"⁽³⁾. Thus, one could purposefully utilize such an inhomogeneous distribution to design a composite with unique properties.

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Recently, Taya⁽⁴⁾ made an attempt to quantify degree of inhomogeneous distribution of fillers in a composite by introducing the inhomogeneity parameter (ξ) when the shape of fillers is a sphere. In Taya's model, frequency (N) of interparticle distance (d) is the key input parameter from which the inhomogeneity distribution parameter ξ is defined. The exact micromorphology of a composite with inhomogeneous distribution of fillers, however, cannot be determined only by such a single parameter. In this study, we will propose additional parameters in describing the micromorphology of filler distribution more accurately, and discuss the accuracy of these parameters by using the experimental data. In section 2, the definition of micromorphology parameters including ξ will be given. An experiment conducted on SiC particulate/aluminum alloy is described in section 3, and the experimental results and discussion are presented in section 4. Finally, the conclusion is given in section 5.

2. Definition of Micromorphology Parameters

In this study, we assume that fillers are of particulate shape, which includes spherical particles and nonspherical (rod shaped) particulates. We will introduce the following three micromorphology parameters:

- (1) homogeneity distribution parameter, ξ ;
- (2) anisotropy parameter of the first kind, η ;
- (3) anisotropy parameter of the second kind, ϕ .

The homogeneity distribution parameter ξ was previously called the "inhomogeneity" distribution parameter⁽⁴⁾, but it is more appropriate to name ξ the "homogeneity" distribution parameter, as will become clear later. The anisotropy parameter of the first kind, η , is for spherical particles, whereas that of the second kind, ϕ , is related to the case of nonspherical particulates. We will define these parameters in the following.

2.1 Homogeneity distribution parameter ξ

Consider a particle composite where the shape of fillers is a sphere and the particles are distributed in two-dimensional space. We now look at the composite through a window (unit cell). A typical view through the window is shown in Fig. 1(a). Next, we measure the interparticle distance d between the nearest neighbors. If this measurement is conducted over all particles within the window, then one can obtain the frequency diagram for interparticle distance, such as shown in Fig. 1(b), where d is measured between the centers of gravity and the total number of particles within the window, N , is set equal to 500. The homogeneity distribution parameter ξ is defined as

$$\xi = \frac{d_p}{\sqrt{A/N}}, \quad (1)$$

where d_p is the peak value in the frequency diagram (Fig. 1(b)), and A is the area of the window. The denominator on the right side of Eq. (1) is a sort of "average interparticle distance" if the particles are distributed homogeneously. Thus, the meaning of ξ is the degree of homogeneous distribution of particles; as $\xi \rightarrow 1$, the distribution becomes increasingly homogeneous, (i. e., consider a simple cubic array of particles, then $\xi=1$) while as $\xi \rightarrow 0$, the distribution becomes localized (all particles cluster at the same point corresponding to the frequency diagram where the Dirac delta function has its peak at $d=0$).

2.2 Anisotropy parameter of the first kind η

Considering a window similar to that in Fig. 1(a), we first calculate the center of gravity G of all particles within the window and take G as the origin of the x - y coordinates of the window (Fig. 2). Drawing a line connecting G and the center of the i -th particle with mass m_i , we can define the orientation angle θ_i . Then, the anisotropic parameter of the first kind η is defined as

(a) definition of interparticle distance d

(b) frequency of interparticle distance d of the nearest neighbors, and the peak value of d

Fig. 1 Particles in a window

$$\eta = \frac{\sum_i m_i \cos 2\theta_i}{\sum_i m_i}, \quad (2)$$

where the summation is taken over $i=1$ to N . If all particles have the same mass, then Eq. (2) can be simplified to

$$\eta = \frac{\sum_i \cos 2\theta_i}{N}. \quad (3)$$

When all particles are distributed along the x -axis, or along the y -axis, then the values of η become $+1$ or -1 , respectively. When all particles are distributed isotropically, then $\eta \rightarrow 0$. Thus η indicates the degree of anisotropy of particle distribution.

2.3 Anisotropy parameter of the second kind ϕ

When the filler is nonspherical particulates, the orientation of nonspherical particulates (hereafter, particulates) is equally important in influencing the overall properties of the particulate composite. The most popular shape of particulates is rod or short fiber; hence we will examine here only the case of the rod-shaped filler. Referring to Fig. 3, the orientation of a rod can be described by the angle θ_i between the rod axis (denoted by $D_{\max}$ in Fig. 3) and the reference axis (the x -axis in the figure). Again, the origin of the x - y coordinate system is taken as the center of gravity of all particulates. Then, the anisotropy parameter of the second kind, ϕ , is defined as

$$\phi = \frac{\sum_i m_i \cos 2\theta_i}{\sum_i m_i} \quad (4)$$

For all particles having the same mass, Eq. (4) is reduced to

$$\phi = \frac{\sum_i \cos 2\theta_i}{N}. \quad (5)$$

The right sides of Eqs. (4) and (5) are identical to those of Eqs. (2) and (3), but the definitions of θ_i in η and ϕ are different.

Fig. 2 Definition of the orientation angle θ_i of the i -th particle with respect to the origin of the coordinate, G which is the center of gravity of all particles within the window

3. Experimental Results and Discussion

3.1 Target composite

In order to apply the micromorphology parameters, we have chosen the SiC particulate/A356 aluminum matrix (SiC/Al) composite as a target composite. This composite was fabricated via a melt-casting route⁽¹⁾, followed by extrusion with the ratio 9:1. The as-extruded composite was solution-treated at a temperature of 530°C. We have used two different volume fractions of particulates: $V_f=10$ and 20 %, and also two different particle sizes: 400 grit (coarse) and 600 grit (fine). As-extruded composite specimens were sectioned along the extrusion direction (longitudinal specimen) and perpendicular to it (transverse specimen). The combinations of the composite specimens discussed above are tabulated in Table 1, where as-cast 10 % V_f SiCp composite with 600-grit (fine) particles is also included. Typical micromorphologies of the as-cast, longitudinal and transverse sections of 10 % V_f SiCp/Al composite with 600-grit (fine) particles are shown in Fig. 4 (a), (b) and (c), respectively. In the figure, black islands denote SiC particulates, and the gray background is the aluminum matrix. It is noted in Fig. 4 that the SiC particulates are of angular shape. Micrographs are taken for all of the composite specimens categorized in Table 1.

3.2 Data reduction process

The micrographs are subjected to an image processor (Excel, Avionics, Co. Ltd.) where a CCD camera thresholds each photo to identify SiC particulate islands against the aluminum matrix. The image processing route is schematically shown in Fig. 5. In determining the orientation angle of a particulate θ_i defined in Fig. 3, we used one of the softwares built into the image processor, i.e., first finding $D_{\max}$, and then measuring the angle between the $D_{\max}$ line and the x -axis (Fig. 3). The image processor provides the

Fig. 3 Definition of the orientation angle θ_i of the i -th particulate (of rodshape) with the maximum length $D_{\max}$

basic data necessary to calculate three micromorphology parameters, ξ , η , and ϕ .

3.3 Results of ξ , η , and ϕ for SiCp/Al composite specimens

Before determining the homogeneity distribution parameter ξ of a given micrograph, one must construct a frequency diagram similar to that in Fig. 1(b). For example, the frequency diagrams corresponding to Figs. 4(a) and (b) are shown in Figs. 6(a) and (b), respectively. The homogeneity distribution parameters of all composite specimens are summarized in Fig. 7, where the cases of as-cast, as-extruded specimens with 400-grit (coarse) particles and 600-grit (fine) particles, are shown by different symbols. It follows from Fig. 7 that in the 20 % V_f composite specimens, particles are distributed more homogeneously than in the 10 % V_f composite specimens, while the effect of particle size on ξ is not clear.

Next, the results of the anisotropy parameter of the first kind η for the cases of as-cast, as-extruded 400-grit and 600-grit composite specimens, are shown in Fig. 8. Please note that despite various angular shapes of SiC particulates, their centers of gravity are used to determine the angle θ_i defined in Fig. 2. Figure 8 indicates that the values of η are all close to zero, suggesting that the distribution of the centers of gravity of SiC particulates of the composite gives rise to nearly isotropic morphology. This does not mean

that the macroscopic properties of the composite are isotropic. In fact, the results of the anisotropy parameter of the second kind, ϕ , shown in Fig. 9, yield clear indication that the as-cast and the transverse sections are nearly isotropic, while the longitudinal one exhibits some anisotropy. The fact that the transverse section gives rise to nearly isotropic morphology of SiC particulates while the longitudinal one exhibits anisotropy along the x -axis (i.e., the extrusion direction) is quite reasonable from the viewpoint of the extrusion process used.

3.4 Parametric study on extreme cases of distribution

To understand the role of the three parameters more clearly, we have purposefully generated the following extreme cases of filler distribution:

- (1) six different distributions of 100 particles in a window, shown in Figs. 10 (a) ~ (f);
- (2) two different distributions of rod-shaped particulates shown in Fig. 11, (a) $\phi=1$ and (b) $\phi=1$.

In Fig. 10, the horizontal flow of (a) $\rightarrow$ (b) $\rightarrow$ (c) $\rightarrow$ (d) denotes the change in the particle distribution due to the change in the anisotropy parameter of the first kind η , while the homogeneity distribution parameter ξ is kept constant. On the other hand, the vertical flow of (e) $\rightarrow$ (c) $\rightarrow$ (f) demonstrates the change in the homogeneity distribution parameter ξ as it increases toward (f), while the anisotropy

Table 1

vol%SiC	Grit Size	Processing condition, cross section
10	400 (coarse)	Extruded, Transverse
		Extruded, Longitudinal
	600 (fine)	Extruded, Transverse
		Extruded, Longitudinal
		As-cast
20	400 (coarse)	Extruded, Transverse
		Extruded, Longitudinal
	600 (fine)	Extruded, Transverse
		Extruded, Longitudinal

Fig. 4 Micrographs of 10 % V_f SiCp/6061 Al composite: (a) as-cast specimen, (b) longitudinal section, and (c) transverse section of as-extruded specimens

Fig. 5 Schematic diagram of image processing system

Fig. 6 Examples of frequency diagram of 10 % V_f composite: (a) as-cast specimen, and (b) longitudinal section of as-extruded specimens

parameter of the first kind η is kept close to zero. Most of the SiC particle data analyzed in the preceding subsection appear to fall into the case between (c) and (f). We tend to believe that the best particle distribution in Fig. 10 is the case with smaller $|\eta|$ and higher ξ , i.e., (f). Thus, extreme cases, Eiffel-Tower-shaped distribution of (a) and (d), ring-shaped distribution of (b), and localized distribution of (e), are to be eliminated. However, there may be cases where the composites with such extreme cases of distribution give rise to unique overall behavior or property; thus it is worth tailoring such a micromorphology. This issue will be discussed later in the next subsection.

If fillers are of rod shape, two extreme cases can be realized despite both possessing the same η value, as demonstrated in Fig. 11 where (a) and (b) denote the case of $\phi=1$ and -1 , respectively, and both η are

Fig. 7 Homogeneity distribution parameter ξ as a function of volume fraction of particulates

Fig. 8 Anisotropy parameter of the first kind η as a function of volume fraction of particulates

approximately equal to 0. Figure 11 demonstrates the necessity of using the anisotropy parameter of the second kind ϕ .

3.5 Fractal analysis

It appears from Fig. 4 that the distribution of particulates in as-cast composite specimens is less homogeneous than in as-extruded composite specimens, and the value of the homogeneity distribution parameter ξ of the as-cast composite (Fig. 4(a)) is

as low as 0.3827. The degree of homogeneous distribution of particulates depends on the window size (L), Fig. 12. This size dependence can be examined from

Fig. 9 Anisotropy parameter of the second kind ϕ as a function of volume fraction of particulates

the viewpoint of fractal analysis. For a given window of L by L , the total number of particulates contained in the window N is counted. If the distribution obeys fractal behavior, then N should be related to L by⁽⁵⁾

$$N(L) = aL^D, \quad (6)$$

where D is a fraction and is called the “fractal dimension”, and a is a constant. The results of $\log N$ and $\log L$ for 10 % V_f as-cast and transverse-section as-extruded composites are plotted in Figs. 13(a) and (b), respectively. It is noted in Fig. 13(a) that the results do not converge to a single straight line throughout the entire size domain, and the values of D range from 1.286 to 1.592, while in Fig. 13(b), the data points lie on a single line, the slope of which provides $D=1.668$. These results indicate that the distribution of the as-cast composite exhibits less fractal behavior with a smaller value of D than the as-extruded composite, suggesting that the particulate distribution in the as-cast composite is less homogeneous than that in the as-extruded composite. For particle composites with homogeneous distribution of particles, the fractal behavior described by Eq. (6) has been observed⁽⁶⁾.

Next we examine ξ dependence on size L by

Fig. 10 Extreme cases of distribution of particles where the effects of ξ and η on the micromorphology are demonstrated: horizontal flow, (a) → (b) → (c) → (d) showing the change in η by keeping $\eta=0.31266$, and vertical flow, (e) → (c) → (f) showing the effect of changing ξ by keeping η nearly zero

$$\phi = 1$$

(a)

$$\phi = -1$$

(b)

Fig. 11 Two extreme cases of distribution of rod shaped particulates where $\eta=0$: (a) $\phi=1$, (b) $\phi=-1$

focusing on the cases of the 10 % V_f as-cast composite (corresponding to Fig. 13(a)) and the transverse section of the 10 % V_f as-extruded composite (to Fig. 13(b)), the results of which are shown in Figs. 14(a) and (b), respectively. It is evident from Fig. 14 that the ξ -log L relation in the transverse section of the as-extruded composite exhibits some plateau over the 2-2.7 region of log L , and while for the as-cast composite, the plateau is difficult to observe, and instead, the values of ξ decrease continuously as log L increases. The above results suggest that the as-cast composite exhibits a strong dependence of ξ on size L . In such a case, the fractal analysis gives rise to a nonlinear relation between log N and log L . Let us examine this nonlinear behavior in more detail. To

Fig. 12 The procedure of fractal analysis where the number of particles N contained in the window of size L is counted

(a)

(b)

Fig. 13 log N -log L relationships: (a) as-cast 10 % V_f composite, (b) the transverse section of as-extruded 10 % V_f composite

this end, we will use a flat Fournier universe with $D=1^{(2)}$, shown in Fig. 15(a). For a given window size L , we count the number of particles within it and plot the results of $\log N$ against $\log L$ as in Fig. 15(b). It can be seen from Fig. 15(b) that the relationship of $\log N$ - $\log L$ exhibits two different regions: flat and steep slope regions, showing a stair (or zig-gag)-shaped curve. The dash-dot straight line shown in Fig. 15(b) is the line connecting the top edges of the stairs, resulting in the slope $D=1$. It can be concluded from Fig. 15 that the flat Fournier universe is fractal not only for each narrow range of L , but also for a long range of L if the zigzag behavior is ignored. The dependence of the homogeneity distribution parameter ξ on size L for the flat Fournier universe is also examined. To this end, we have plotted ξ as a function of $\log L$ in Fig. 16(a), showing that ξ decreases

in a zigzag manner as $\log L$ increases. Figure 16(a) indicates the absence of any flat region, unlike the case of the SiCp/Al composite specimen where some flat regions were observed. Next we examine more closely the results of Fig. 16(a) by replotting them as a $\log \xi$ - $\log L$ relationship, as shown in Fig. 16(b), which shows two different regions: long down and short up hills. It should be noted that the long down and short up hills correspond to, respectively, flat and steep up hill regions in Fig. 15(b). The slope of the long down hills in Fig. 16(b) was found to be -1 . This can be explained Eq. (1). Let us focus on a locally fcc (2-dimensionally face center cubic array) packed region. Then we increase the window size L such that the total number of particles N within the window is kept constant. This process corresponds to the flat region in Fig. 15(b). During this process, d_p and N in Eq. (1) remain constant, while the window area increases with L ; i.e., $A=L^2$. Thus, ξ becomes proportional to L^{-1} , which represents the long down hill region in Fig. 16(b).

In summarizing the above results, we can conclude that for a space consisting of a large number of clustered regions (represented by the flat Fournier universe), the homogeneity distribution parameter ξ decreases rapidly (i.e., becomes more localized) as the window size increases. This trend is coincident with the zigzag behavior in the $\log N$ - $\log L$ graph, and is a departure from normal fractal behavior.

4. Conclusions

The spatial distribution of particulates in a composite was studied both experimentally and analytically. A set of three micromorphology parameters was proposed to assess the degree of inhomogeneous (or homogeneous) distribution, and that of anisotropy of particulates. The data of SiC particulate/aluminum composites were analyzed by use of these three parameters. This method explains well the difference of the micromorphological nature of SiC particulates between as-cast and as-extruded composites, and also between the transverse and longitudinal sections of the composite.

Finally, a standard fractal analysis was conducted on SiCp/Al composites to determine the fractal dimension D and the degree of linearization. Based on the results of the micromorphology parameter study and fractal analysis, we found some correlation between the homogeneity distribution parameter and fractal behavior.

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Fig. 14 ξ - $\log L$ relationships: (a) as-cast 10% V_f composite (b) the transverse section of as-extruded 10% V_f composite

Fig. 15 Flat Fournier universe with $D=1^{(2)}$: (a) particles contained in a 2-D fcc packing expand infinitely, (b) fractal analysis, $\log N$ - $\log L$ relationship

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Fig. 16 Dependence of homogeneity distribution parameter ξ on window size L for the flat Fournier universe of Fig. 15(a): (a) ξ - $\log L$ relationship, (b) $\log \xi$ - $\log L$ relationship

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