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Estimation of contiguity for a monodispersed system of spherical particles

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A new stereological method for the estimation of the contiguity (the degree of contact) for the monodispersed system of spherical particles is reported. Since this method is applicable to point-like contacts, the contiguity of ceramic particles can be evaluated in a metal matrix composite in which the contact is assumed to be point. The contiguity is derived from the geometrical calculation of centre-to-centre distance between particles, evaluation of contact, and consideration of the geometrical probability that two particles in contact are cut simultaneously by a test plane. The contiguity can be expressed by either the number of contacts per unit volume or the number of contacts per particle. Applying this method to a model material (Shirasu-balloon/aluminium alloy composite), the interrelations between the change in the contiguity and some physical properties of the material can be accurately explained.

1. Introduction

In particle-dispersed composite materials, the contiguity of dispersed particles can exert significant influence on properties such as tensile strength and electrical conductivity. There are some well-known methods to evaluate the contiguity of grains or particles stereologically [1]. Most of those methods are concerned with the facial contact between the phase(s) of interest, however. Therefore, they cannot be applied to point-like contacts between rigid particles such as ceramic particles in a metal matrix composite, because two dimensional observations on a test plane never catch the three dimensionally dispersed points, each of which has no volume or area.

Robine *et al.* [2] reported a method to evaluate the number of point-like contacts between monosized spherical particles. They introduced a concept of bi-particle, and found the coordination number (the number of contacts per particle) from a consideration of the plane coordinance which is defined as a function of the euclidean distance between particle sections on a test plane.

In this article, an approach to estimate the contiguity for a monodispersed system of spherical particles stereologically, i.e. by observing two dimensional test section, is reported. The proposed method is based on a consideration of the geometrical probability, and has potential to evaluate point-like contacts more easily. The degree of contact is defined as either the number of contacts per unit volume or the number of contacts per particle. With an appropriate modification, we apply this quantity to a Shirasu-balloon/aluminium alloy composite (SBAC) to examine the relationship between some physical properties of SBAC and the degree of contact of Shirasu-balloons.

2. Contiguity for a monodispersed system of spherical particles

2.1. Estimation of contact from an observation of two dimensional section

As mentioned before, it is generally difficult to catch three dimensionally dispersed points of contact by a two dimensional section plane through a material. If some assumptions are made, however, we can estimate such a contiguity from relatively straight-forward geometrical analysis as follows.

First, we assume that the particles are spheres with the same radius, r_0, and are not deformed by contacting. Then we consider a two dimensional test plane through the sample in which the particles assumed above are dispersed. On the test plane, circular features of various sizes are seen as in Fig. 1a. Let the sectional radii and the coordinates of the centres of circle i and j be r_i, r_j and (x_i, y_i), (x_j, y_j), respectively, then the distances (z_i, z_j) between the centres of particles and the test plane are written as

$$\begin{aligned} z_i &= (r_0^2 - r_i^2)^{1/2} \\ z_j &= (r_0^2 - r_j^2)^{1/2} \end{aligned} \quad (1)$$

As the particle centre corresponding to each circular feature possibly exists either above or below the test plane (as C_i or C_i' in Fig. 1b), there are two possible centre-to-centre distances ($C_i - C_j$ or $C_i' - C_j$) between two particles. These two distances are written as

$$d^{\pm} = \{(x_i - x_j)^2 + (y_i - y_j)^2 + (z_i \mp z_j)^2\}^{1/2} \quad (2)$$

where d^+ and d^- indicate the centre-to-centre distance between two particles whose centres are existing

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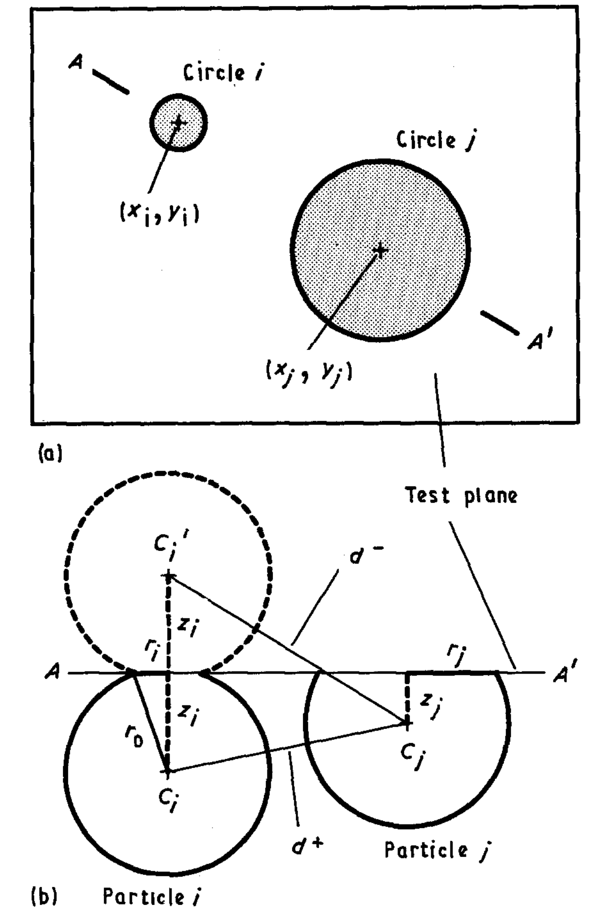

Figure 1 (a) Particles observed on a test plane, and (b) their profile view which is normal to the test plane. There are two cases of centre-to-centre distances (d^+ and d^-) between two particles seen on a test plane (b). Assume that all particles have the same radius, r_0.

in the same side ($C_i - C_j$) and in each side ($C_i' - C_j$) of the test plane, respectively. It is apparent that d^+ is always smaller than d^-.

Now, we can know the probability c_{ij} of contact between the ith and jth particles by comparing $2r_0$ with $d^{\pm}$. The theoretical criteria for determining c_{ij} are summarized in Table I. If two particles contact with each other, either d^+ or d^- equals $2r_0$. Converses seem to be not always true. In the case of $d^- = 2r_0$, the particles necessarily contact with each other, so $c_{ij} = 1$. In the case of $d^+ = 2r_0$, the situation is somewhat complex. If the centres of particles are in the same side to the test plane, they contact with each other. But if the centres of particles are in each side of the test plane, they do not contact. And the probability for the centres of particles to be in the same side to the test plane seems to be 0.5. The probability that two particles are in each side of the test plane and $d^+ = 2r_0$ is negligible, however, comparing with the probability that both particles are in the same side to the test plane and $d^+ = 2r_0$. Therefore, unity is assigned to c_{ij} when $d^+ = 2r_0$.

The theoretical criteria in Table I seems to be scarcely applicable to practical particles, because practical particles are not of the same size and truly spherical in a strict sense. So, it might be reasonable to assume a particle to be interpenetrated by another particle when d^+ or d^- is smaller than $2r_0$. Such an assumption might well simulate practical contacting

TABLE I Theoretical criteria for determining the probability of contact, c_{ij}, between two particles

Condition	c_{ij}
$d^- \geqslant d^+ > 2r_0$	0
$d^- > d^+ = 2r_0$	1
$d^- > 2r_0 > d^+$	0
$2r_0 = d^- \geqslant d^+$	1

TABLE II A practical criteria for determining c_{ij}

Condition	c_{ij}
$d^- \geqslant d^+ > 2r_0$	0
$d^- > 2r_0 \geqslant d^+$	0.5
$2r_0 \geqslant d^- \geqslant d^+$	1

with some degree of local flattening or coalescence, although it means that the volume of the particle decreases by contacting. Thus, we introduce a new assignment for practical c_{ij} as in Table II. In these criteria, 0.5 is assigned to c_{ij} when $2r_0$ is between d^+ and d^-, because in this case, the probability for the centres of both particles to be in the same side to the test plane is considered to be 0.5.

Using the criteria of Tables I or II, we can assign the probability of contact, c_{ij}, for every pair of particles, and thus obtain the apparent total number of contacts C_T by summing up c_{ij} for all possible pairs of particles as

$$C_T = \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} c_{ij} \tag{3}$$

where N is the number of particles observed on the test plane.

Next we examine the spatial range within which the contacts are estimated with the above method. As shown in Fig. 2, the particles which are observed on a

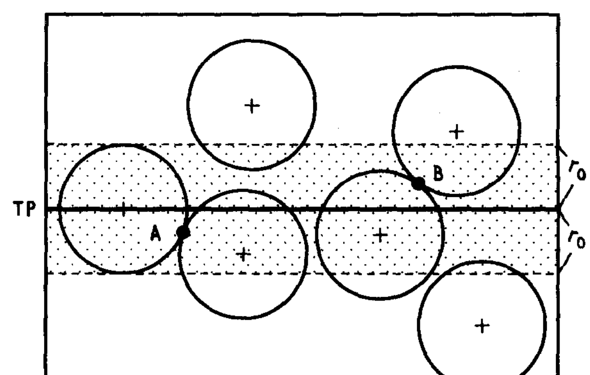

Figure 2 Profile view of particles cut normal to a test plane (TP) and through the centre of each particle (+). The test plane is shown as a bold line. All particles which are cut by the test plane have their centres within the distance r_0 from the test plane. Also, the contact point (as point A) must be within the distance r_0 from the test plane if both of participating particles are cut by the test plane. But not all the contact points within this range can always be estimated (as the case of point B), since both of participating particles are not always cut by a test plane simultaneously.

test plane must have their centres not farther than r_0 from the test plane. Therefore, the contacts estimated with the above method must also occur within the distance r_0 from the test plane. But it is noted that all the contacts within this $2r_0$ range cannot be estimated. Such an example is also shown schematically in Fig. 2. While the contact point A is derived from the two particles observed on the test plane, the point B comes from the contact of one particle observed and another not observed on the test plane.

Then, we have to introduce the probability of the detectable contact occurrence in order to estimate the total number of contacts occurring within the distance r_0 from the test plane.

2.2 Probability of the detectable contact occurrence

In order to obtain the probability, P, of the detectable contact occurrence, we have to consider the following two kinds of probabilities: (1) the probability P_1 for a contact to occur within the distance r_0 from the test plane, and (2) the probability, P_2, for such a contact to be detected from the section of the particles (i.e. the probability for both particles i and j to be cut by the same test plane).

Suppose that particle j is in contact with particle i, the centre of particle j is on the spherical surface radius which is $2r_0$ and the centre coincides with that of particle i. And probabilities P_1 and P_2 vary depending on the distance z_i of the centre of particle i from the test plane (Fig. 3). Because of the symmetry of configuration, it is enough to consider the centre of particle i below the test plane only.

Consulting Fig. 3, the probabilities P_1 and P_2 are seen to be expressed as the ratio of the surface area generated from rotating the arc $j_0 j_3$ (the total of dotted and bold curves) and arc $j_1 j_2$ (bold curve only) around the z-axis, respectively, to the surface area of sphere with radius of $2r_0$. Now let us consider the situation shown in Fig. 3c and define angles ϕ_0, ϕ_1 and ϕ_2 as in the figure, the surface areas S_1, S_2 (corresponding to P_1 and P_2, respectively) and S_T (surface area of the sphere of radius $2r_0$) are

$$S_1 = 8\pi r_0^2 \int_{\phi_0}^{\pi} \sin\theta \, d\theta = 8\pi r_0^2 (1 + \cos\phi_0) \quad (4)$$

$$S_2 = \begin{cases} 8\pi r_0^2 \int_{\phi_1}^{\phi_2} \sin\theta \, d\theta = 8\pi r_0^2 (\cos\phi_1 - \cos\phi_2) & (0 \leqslant z_i < r_0) \\ 0 & (r_0 \leqslant z_i \leqslant 2r_0) \end{cases} \quad (5)$$

$$S_T = 4\pi (2r_0)^2 = 16\pi r_0^2 \quad (6)$$

And from the definitions

$$\cos\phi_0 = \frac{(r_0 - z_i)}{r_0} \quad (7)$$

$$\cos\phi_1 = \frac{(r_0 - z_i)}{2r_0} \quad (8)$$

$$\cos\phi_2 = \frac{-(r_0 + z_i)}{2r_0} \quad (9)$$

Putting Equations 7–9 into Equations 4 and 5 and dividing them by Equation 6, we obtain the probabilities P_1 and P_2 at each z_i as

$$P_1 = \frac{8\pi r_0 (2r_0 - z_i)}{16\pi r_0^2} = 1 - \frac{z_i}{2r_0} \quad (10)$$

$$P_2 = \begin{cases} \dfrac{8\pi r_0^2}{16\pi r_0^2} = \dfrac{1}{2} & 0 \leqslant z_i < r_0 \\ 0 & r_0 \leqslant z_i \leqslant 2r_0 \end{cases} \quad (11)$$

Since z_i is expected to be distributed uniformly from 0 to $2r_0$, the probability, P, of the detectable contact occurrence is derived from P_1 and P_2 as

$$P = \frac{\int_0^{2r_0} P_2 \, dz_i}{\int_0^{2r_0} P_1 \, dz_i} = \frac{r_0/2}{r_0} = \frac{1}{2} \quad (12)$$

According to the consideration above, we can estimate

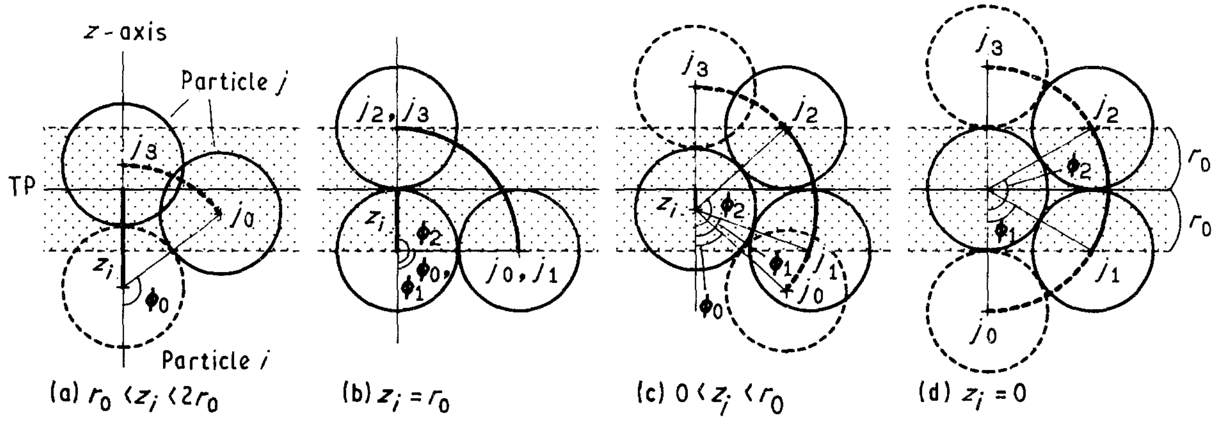

Figure 3 Sections through both centres of contacting particles i and j and normal to the test plane (bold lines). The traces of the centre of particle j make spherical surfaces of radius $2r_0$. The probability of estimating the contact between particles i and j from the observation of the test plane is proportional to the surface area generated by rotating arc $j_1 j_2$ (bold curve) around the z-axis. And the probability for contact to occur within the distance r_0 from the test plane is proportional to the surface area generated by rotating arc $j_0 j_3$ (both bold and broken curves) around the z-axis.

the real number of contacts within the distance r_0 from the test plane by dividing C_T by P (= 0.5).

Now we can express the degree of contact as follows:

1. The number of contacts per unit volume, C_V, is

$$C_V = \frac{C_T}{2r_0AP} \tag{13}$$

where A is the total test area on the section plane.

2. The number of contacts per particle, C_N, is

$$C_N = \frac{2C_V}{N_V} = \frac{4C_Vr_0A}{N} = \frac{2C_T}{NP} \tag{14}$$

where N is the total number of particles observed within the test plane and N_V is the number of particles per unit volume.

3. Applications

3.1. Preliminary considerations prior to application

The reliability of the proposed method was examined by Hirai *et al.* [3]. They applied the method for estimating the contiguity of a computer simulated system of monodispersed spherical particles, and proved that the estimated contiguity, C_N, coincides with the virtual contiguity of the simulated system. They also showed that the estimated contiguity through the practical criteria in Table II agrees well with that obtained through the theoretical criteria in Table I. On the grounds above, we use the criteria in Table II to determine the contiguity of practical particles. There are, however, two problems which might be unfavourable to the direct application of the proposed method to practical materials.

First, the shape of practical particles is not completely spherical while the proposed method for the evaluation of the contiguity assumes particles to be spherical. Therefore, the proposed method may approximately be applied to practical systems of spherical particles. In such a case, we may use the equivalent area diameter ($2(A_i/\pi)^{1/2}$, where A_i is the sectional area of the particle) and gravity centre instead of geometrical diameter and geometrical centre for the particle seen on a two dimensional test plane.

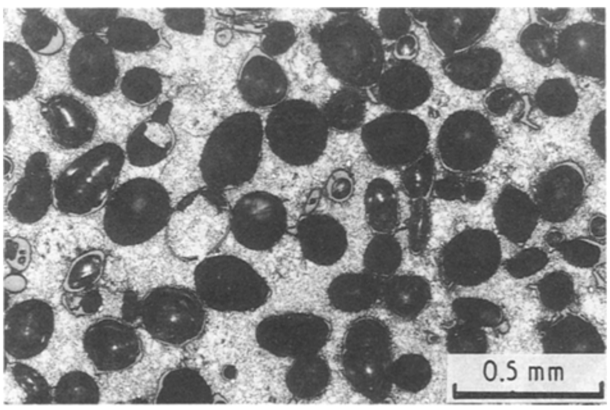

Figure 4 Photomicrograph showing microstructure of Shirasu-balloon/aluminium alloy composite (SBAC).

Second, a system of particles usually has a size distribution. The proposed method deals exclusively with a monodispersed system of particles. Thus, the proposed method may not be applicable to a polydispersed system of particles in a strict sense. According to the principle for the evaluation of the contiguity proposed here, however, the existence of smaller particles tends to increase the calculated contiguity while the existence of larger particles has a tendency to decrease the calculated contiguity. Thus, both effects might work to cancel out each other. Accordingly, when we adopt the mean diameter as a representative diameter of polydispersed particles, the obtained contiguity would not be so different from the real contiguity of the system.

3.2. Results of the application to SBAC

We tried to apply the proposed method to evaluate the contiguity of Shirasu-balloons (SB) in Shirasu-balloon/aluminium alloy composites (SBAC). SB are hollow microballoons made from volcanic ash called Shirasu. SB sieved between 0.149 and 0.210 mm and Al–12%Si alloy were used as dispersed particles and as matrix, respectively. SBAC was prepared as follows: SB was mixed with powdered Al–12%Si alloy, prepacked in a mould, and pre-heated at 723 K. Then molten Al–12%Si alloy at 973 K was poured and squeeze-infiltrated into the mixture of SB and Al–12% Si alloy powders at about 6.5 MPa. The volume fraction, V_V, of SB was controlled from 0 to 60% by the mixing ratio of SB to Al–12%Si powder. Representative microstructure of SBAC is shown in Fig. 4, and representative physical properties are also shown in Fig. 5a and b.

As seen in Fig. 4, SB is almost spherical. The roundness ($4\pi A_i/p_i^2$, where A_i and p_i are the sectional area and the perimeter length of the section of particle, respectively) of SB is measured on the test plane of SBAC with a computer-controlled digital image analyser, the result is shown in Fig. 6. The average of the roundness is about 0.84 and the standard deviation is about 0.11. It is known that the roundness of a circle measures about 0.90 under the same analyser [4], so that SB can well be approximated to spheres. At this point, it may be valid to apply the proposed method to the estimation of the contiguity of SB in SBAC.

Ten test areas (1.20 mm × 1.28 mm) were selected at random from each test plane, and processed by a digital image analyser to measure the equivalent area diameter and the gravity centre of each image of particle. The measured equivalent area diameters were first used to know the size distribution of SB by the Schwartz–Saltykov's method [5]. The obtained size distribution of SB is shown in Fig. 7. It is noticed that the frequency of particles of smaller size includes larger errors due to the nature of this type of analysis. The mean diameter of SB is 0.159 mm and the standard deviation is 0.053 mm.

Based on the mean diameter, the contiguity of SB was first calculated for each test area, and was averaged for each specimen later. The obtained C_V and C_N

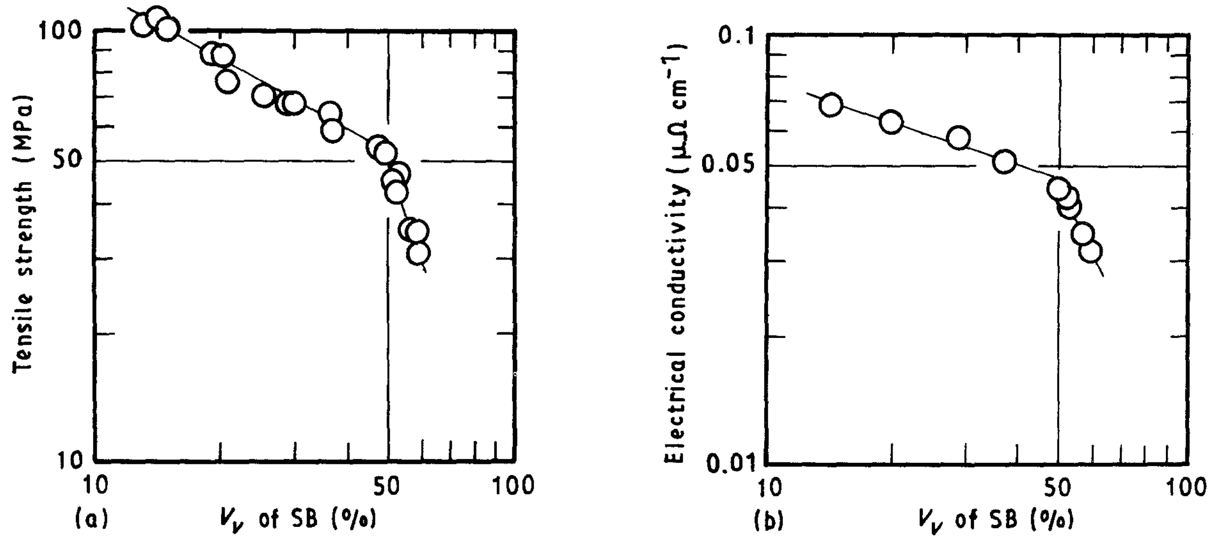

Figure 5 Representative physical properties of SBAC in terms of the volume fraction (V_V) of Shirasu-balloons (SB): (a) tensile strength, and (b) electrical conductivity.

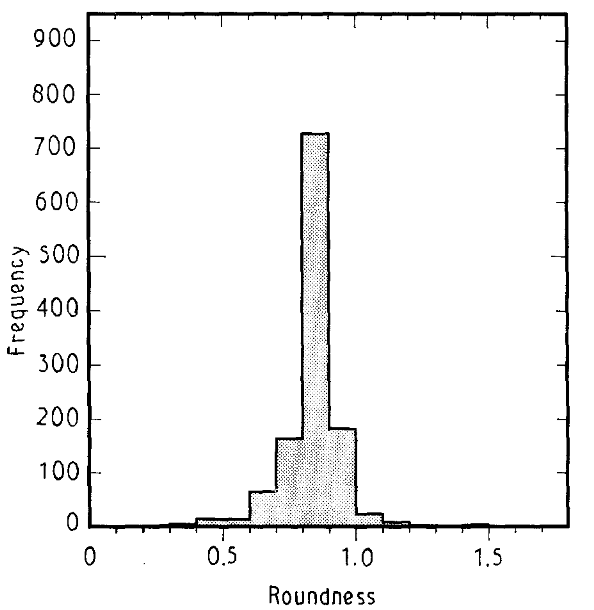

Figure 6 Histogram showing the frequency of the roundness of SB. The roundness of each SB is measured on the test plane of SBAC with computer-controlled digital image analyser. Total number of particles = 1209; mean value of the roundness = 0.838; standard deviation of the roundness = 0.11.

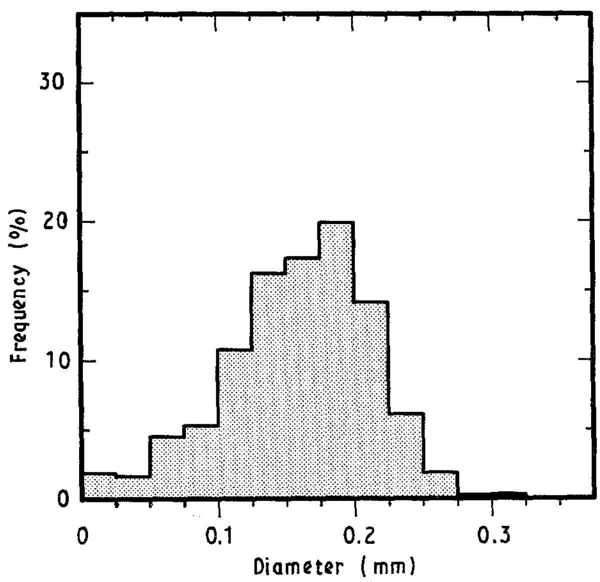

Figure 7 Histogram showing the size distribution of SB assessed by the Schwartz–Saltykov's method [5]. The equivalent area diameter obtained by digital image analyser was used as a sectional diameter of SB. Total number of particles = 2815; mean diameter = 0.159 mm; standard deviation of the diameter = 0.053 mm.

are shown with the volume fraction of SB in SBAC in Fig. 8a and b, respectively. It is noticed that both C_V and C_N increase linearly with the volume fraction of SB in these figures, but beyond 50% of SB the contiguity increases at a higher rate. This behaviour is consistent with the changes in some physical properties of SBAC shown in Fig. 5: tensile strength (Fig. 5a) and electrical conductivity (Fig. 5b). It is obvious that tensile strength and electrical conductivity of SBAC decreases with increasing SB, because SB damages these properties. The volume fraction cannot explain the drastic change in property at around 50% SB, but the contiguity can. Therefore, it can be said that the degree of contact must be one of the parameters which plays an important role in the properties of metal–particle composite.

Gurland [1] reported the relations between the

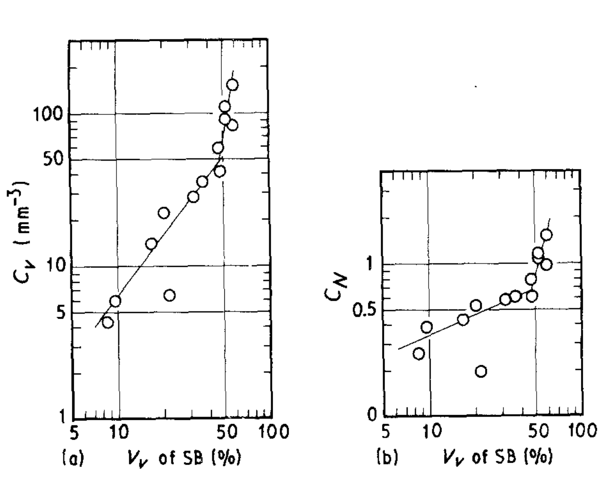

Figure 8 Figures showing the relation between the contiguity (C_V or C_N) and the volume fraction, V_V, of SB in SBAC. (a) C_V versus V_V; (b) C_N versus V_V.

number of contacts per particle, C_N, of silver and the electrical resistivity (reciprocal of electrical conductivity) of a silver–Bakelite composite in terms of the volume fraction of silver particles. He showed a drastic change in the resistivity and C_N at about 40 vol % of silver, which is different from the change in the electrical conductivity of SBAC and C_N at about 50 vol % of SB. The reason for this is as follows: Gurland's conductors are silver dispersoid while ours are aluminium matrices, i.e. 40 vol % means that silver particles begin to contact each other while our 50 vol % means that aluminium matrices begin to become discontinuous.

4. Conclusions

In this article we introduced a new method to estimate the contiguity of a monodispersed system of spherical particles, in order to evaluate point-like contacts. First we calculated the three dimensional centre-to-centre distances of particles, compared them with the diameter of the particle to assign the probability of contact for each pair of particles, and summed them up to obtain the apparent total number of contacts within the distance r_0 (radius of the particle) from the test plane. Next we derived the probability for the contacts to be detected from this method. Dividing the apparent total number of contacts by the probability, to get the real number of contacts within the distance r_0 from the test plane, we could express the contiguity in either: (a) the number of contacts per unit volume, or (b) the number of contacts per particle. With appropriate modifications, we applied this procedure to Shirasu-balloon/aluminium alloy composite and concluded that the contiguity is one of the more useful parameters to describe some properties of metal matrix composites.

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