EVALUATION OF PARTICLE SPATIAL DISTRIBUTION IN PARTICLE DISPERSED COMPOSITES

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1 Introduction

Particle reinforced metal matrix composites (PMMCs) are emerging as an important class of engineering materials due to their superior mechanical properties and amenability to the conventional metal working processes. Over the last decade, experimental and numerical studies [1-4] have been indicated that the spatial distribution of second phase particles played an important role on the mechanical properties. Several kinds of method [5-8] have been suggested to describe the spatial distribution of second phase particles. However, the developments of techniques for characterizing the spatial distribution of second phase particles have not been enough. The descriptors for the spatial distribution of particles have to be sensitive to their arrangements. Simple scalar descriptors such as volume fraction have been successful in understanding and predicting material behaviors of PMMCs using the simple formula, 'rule of mixtures'. Therefore, simple scalar descriptors for the spatial distribution, which links to material behaviors, are desired. In general, the spatial distribution of second phase particles is experimentally evaluated in 2-dimension by analyzing scanning electron microscope (SEM) or optical microscope (OM) images. The spatial distribution in 3-dimension has been also evaluated by serial sectioning method or synchrotron X-ray micro-tomography, but these techniques are time-consuming and expensive. Computer modeling is a powerful tool for analyzing the spatial distribution of second phase particles in 3-dimension, and some researchers [5-9] have investigated the statistical relationship between 3- and 2-dimensional spatial distributions. If the simple statistical relationship between 3- and 2-dimensional spatial distributions is derived, material behaviors of PMMCs may be predicted only by the 2-dimensional spatial distribution, which is evaluated by analyzing SEM or OM images. We have suggested the quantitative method to evaluate the spatial distribution of second phase particles using the relative frequency distributions of 3-dimensional local number, LN3D, and 2-dimensional local number, LN2D. In addition, we demonstrated that randomness of overlap-permissive second phase particles was correctly evaluated in 3- and 2-dimensions by this method [10]. In the present work, we applied this method to overlap-forbidden second phase particles.

2 Definition of LN3D and LN2D

The number of GCs of second phase particles in the measuring sphere, whose center is put at GC of a noticed particle, was defined as LN3D. The radius of the measuring sphere was determined so that the number density of the sphere including GCs of 13 particles corresponds to the whole number density in 3-dimension, where 13 mean the number of a noticed particle and nearest neighbor particles in the measuring sphere when assuming close packed structure. The measuring radius, R_{3D}, is represented by

$$\frac{13}{4\pi R_{3D}{}^3/3} = \lambda_V \quad \Rightarrow \quad R_{3D} = \left(\frac{13}{4\pi\lambda_V/3}\right)^{1/3} = \frac{1.459}{\lambda_V{}^{1/3}}, \tag{1}$$

where λ_V is the whole number density. When GCs of second phase particles have the uniform random arrangement in 3-dimension, we can expected that the relative frequency distribution of LN3D exhibits modified Poisson distribution and the probability of LN3D is represented by

$$P(LN3D = k+1) = \frac{13^k}{k!}\exp(-13) \qquad k = 0, 1, 2 \cdots \tag{2}$$

The average and variance of this distribution are 14 and 13, respectively.

The number of GCs of second phase particles in the measuring circle, whose center is put at GC of a noticed particle, was defined as LN2D. The radius of

the measuring circle was determined so that the number density of the circle including GCs of 7 particles corresponds to the whole number density in 2-dimension, where 7 mean the number of a noticed particle and nearest neighbor particles in the measuring circle when assuming the closest hexagonal structure. The measuring radius, R_{2D}, is represented by

$$\frac{7}{\pi R_{2D}{}^2} = \lambda_A \quad \Rightarrow \quad R_{2D} = \left(\frac{7}{\pi\lambda_A}\right)^{1/2} = \frac{1.493}{\lambda_A{}^{1/2}}, \tag{3}$$

where λ_A is the whole number density. When GCs of second phase particles have the uniform random arrangement in 2-dimension, we can expected that the relative frequency distribution of LN2D exhibits modified Poisson distribution and the probability of LN2D is represented by

$$P(LN2D = k+1) = \frac{7^k}{k!}\exp(-7) \qquad k = 0, 1, 2 \cdots \tag{4}$$

The average and variance of this distribution are 8 and 7, respectively.

3 Computer Experiments

We employ two types of method to arrange spherical particles in computational cell with no overlaps. One is the direct embedding method and another is the particle growth method. When volume fraction was smaller than 0.33, the direct embedding method was employed. In the direct embedding method, a position of a particle was determined with computer-generated uniform random numbers. If overlaps of the particle were detected, the position was discarded and new position was tested. This kind of trials was repeated until number of particles in computational cell amounted to ten thousands. When volume fraction was larger than 0.33, the direct embedding method consumed computational time to search empty space for embedding particles, and therefore the particle growth method become effective. In the particle growth method, particles were uniformly grown with hard sphere molecular dynamics calculation [11]. Perfectly elastic collisions of particles with random velocities were repeated in the cell and empty space was filled with particles which had constant growth rate until the volume fraction of particles amounted to 0.64. Three molecular dynamics calculations were conducted changing initial conditions, velocity distribution and growth rate. In order to eliminate errors close to surfaces of the cell, periodic boundary condition was applied.

LN3D was measured at all particles arranged in the cell. The computational cell was cut by an arbitrary cross-section which was selected by random numbers and LN2D was measured at particles which appeared on the cross-section. This cutting was repeated until number of samples exceeded ten thousands. In the range of volume fraction from 0.0025 to 0.33, one hundred measurements of LN2D and LN3D were conducted at each volume fraction, and relative frequency distribution of LN3D and LN2D were derived from one million samples.

Figure 1(a) and (b) shows relative frequency distribution of LN3D and LN2D, when the volume fraction was from 0.0025 to 0.33. Probability distribution of equation (2) and equation (4) also shown in figures. The distributions become narrower and the peaks shift to the left with increasing volume (area) fraction. This means that the average and variance of the distributions decrease with increasing volume (area) fraction.

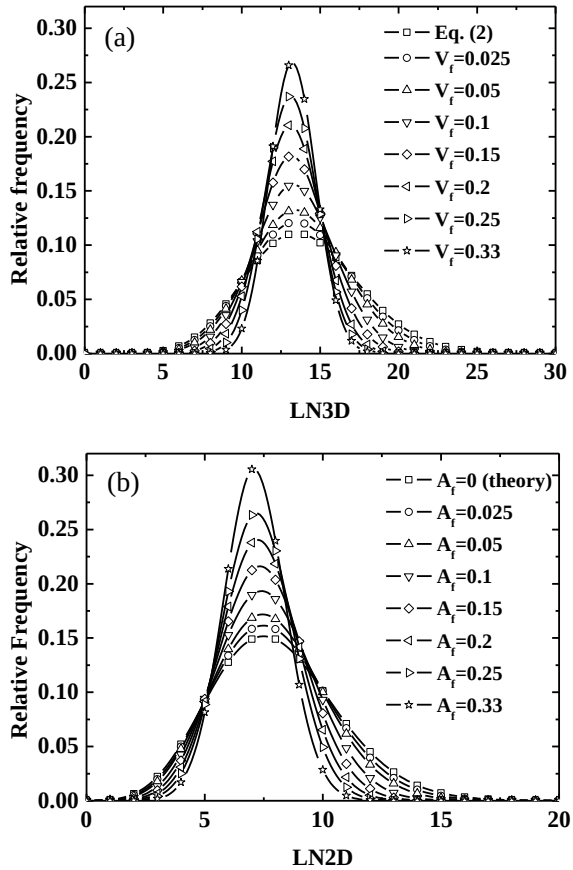

Fig. 1. Relative frequency distribution of (a)LN3D and (b)LN2D from 0.025 to 0.33 in volume fraction.

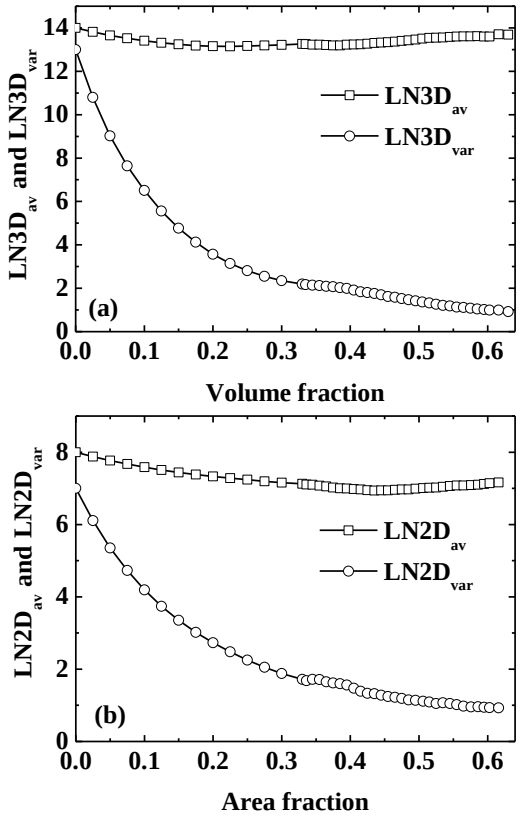

Fig. 2. Average and variance of (a)LN3D and (b)LN2D versus volume(area) fraction.

Figure 2(a) shows plots of average and variance of LN3D, $LN3D_{av}$ and $LN3D_{var}$, versus volume fraction. The theoretical value derived from equation (2) is also plotted at volume fraction of 0.0, where $LN3D_{av}$ and $LN3D_{var}$ were 14 and 13, respectively. With increasing volume fraction, $LN3D_{av}$ vary in the range of 13 to 14 and $LN3D_{var}$ decrease monotonically. These results suggest that there is a uniform spatial distribution which is specific to volume fraction. Figure 2(b) shows plots of average and variance of LN2D, $LN2D_{av}$ and $LN2D_{var}$, versus area fraction. The theoretical value derived from equation (4) is also plotted at area fraction of 0.0, where $LN2D_{av}$ and $LN2D_{var}$ were 8 and 7, respectively. With increasing area fraction, $LN2D_{av}$ vary in the range of 7 to 8 and $LN2D_{var}$ decrease monotonically. Variation of $LN2D_{av}$ and $LN2D_{var}$ is similar to that of $LN3D_{av}$ and $LN3D_{var}$, and it suggests that 2-dimensional spatial distribution of particles which appeared in cross section have a statistical relation with 3-dimensional spatial distribution of particles. Figures 2 also indicate that variance of LN3D and LN2D is sensitive to spatial distribution, and therefore we employ $LN3D_{var}$ and $LN2D_{var}$ as simple scalar descriptors for the spatial distribution in following discussions.

In order to investigate effects of clustered spatial distribution on $LN3D_{var}$ and $LN2D_{var}$, particles were arranged with computer-generated normal random number instead of uniform random number. If standard deviation σ of normal random number decreases, particles tend to be clustered in the center of the computational cell. $LN3D_{var}$ and $LN2D_{var}$ were evaluated at each volume fraction changing σ (0.3, 0.5, 0.8). Figure 3(a) and (b) shows plots of $LN3D_{var}$ and $LN2D_{var}$, respectively, versus volume (area) fraction. The lines of uniform spatial distribution derived from former calculations are also shown in the figures. The deviations of $LN3D_{var}$ and $LN2D_{var}$ from uniform spatial distribution decrease with increasing volume (area) fraction. This means that difference between clustering spatial distribution and uniform spatial distribution becomes small with increasing volume (area) fraction.

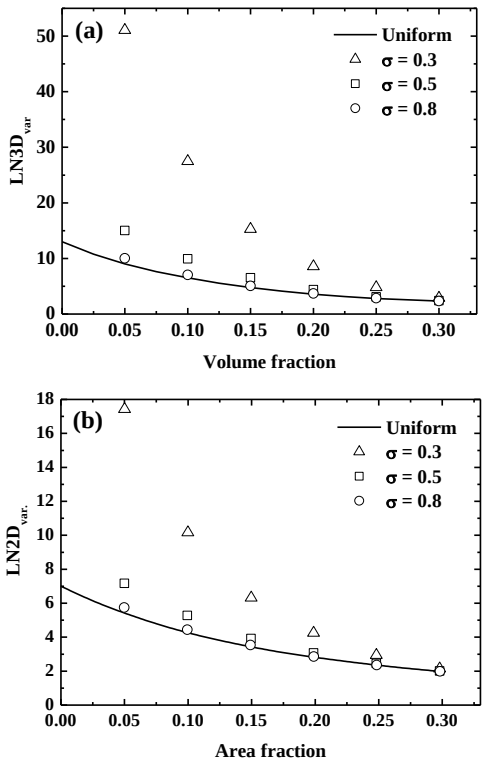

Fig. 3. Variance of (a)LN3D and (b)LN2D versus volume(area) fraction in case of clustered spatial distribution.

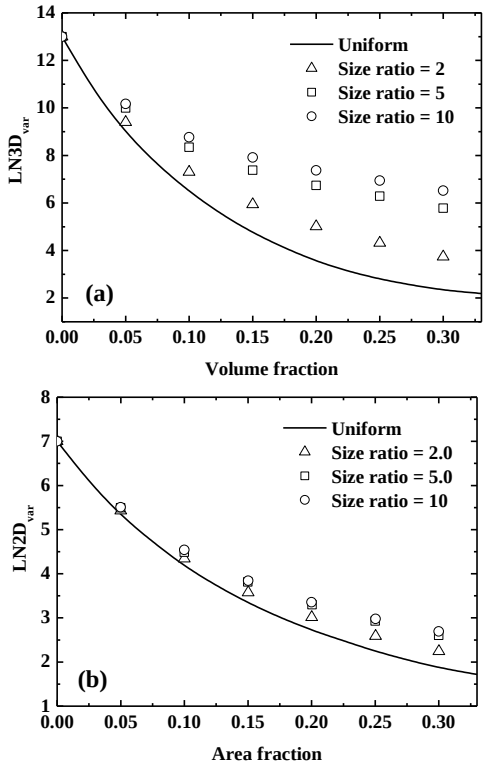

Fig. 4. Variance of (a)LN3D and (b)LN2D versus volume(area) fraction with particle size distribution.

In order to investigate effects of particle size distribution on LN3D$_{var}$ and LN2D$_{var}$, particles with size distribution were arranged in the computational cell. Particle size was divided into ten grades, and numbers of particles in each grade were equal. Ratio of the largest grade to the smallest grade was defined as size ratio. LN3D$_{var}$ and LN2D$_{var}$ were evaluated at each volume fraction changing size ratio (2, 5, 10). Figure 4(a) and (b) shows plots of LN3D$_{var}$ and LN2D$_{var}$, respectively, versus volume (area) fraction. The lines of uniform spatial distribution (mono size) derived from former calculations are also shown in the figures. The deviations of LN3D$_{var}$ and LN2D$_{var}$ from uniform spatial distribution increase with increasing volume (area) fraction and size ratio. Because small particles got into empty space between large particles, clustered arrangement appeared.

4 Evaluation of Particle Spatial Distribution in Al-SiC Composites

The spatial distribution of SiC particles in Al-SiC composites was controlled with changing the relative particles size (RPS) ratio, which is defined as the ratio of the average sizes between Al powders and SiC powders. Details of the material preparation and the SEM image analysis are presented in the reference 12. Figure 5 (a) to (h) show the typical microstructures of the eight samples (Samples 1-8), which were prepared with different RPS ratio and volume fraction. The values of LN2D$_{var}$ which were obtained from the eight samples and the line of uniform spatial distribution which was obtained from computer experiments are plotted in Figure 6. Samples 1-4 were prepared with the same RPS ratio, 1.11, changing volume fraction, 0.05 to 0.2. Samples 2 and 5-8 were prepared with the same volume fraction, 0.1, changing RPS ratio, 1.11 to 10.33. Sample 8 was prepared with the largest RPS ratio, 10.33, and exhibits the largest clustering tendency. Sample 2 was prepared with the smallest RPS ratio, 1.11, and its plot is nearest to the line of uniform spatial distribution.

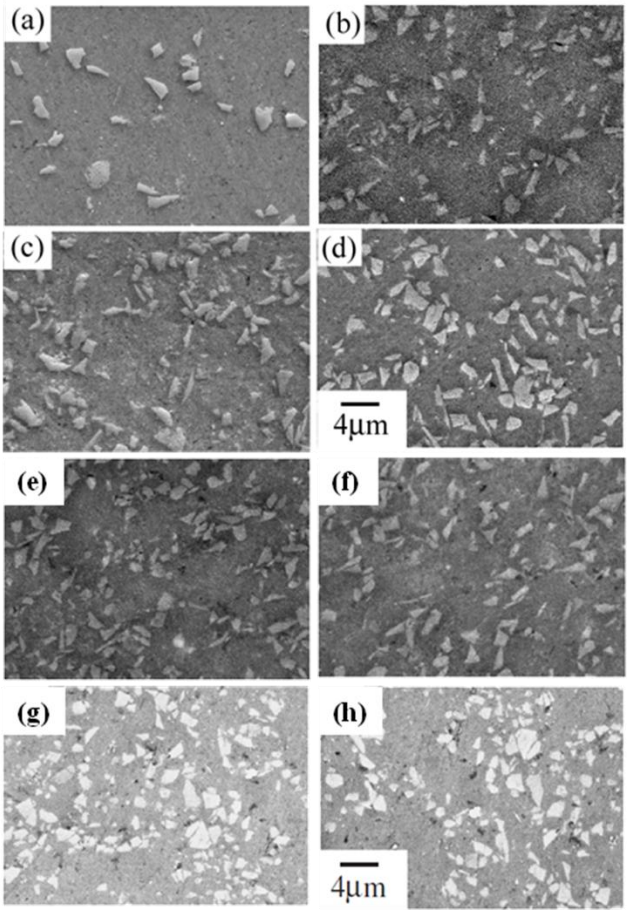

Fig. 5. The typical microstructures of Al-SiC composites. (a) to (h) correspond to Sample 1 to 8.

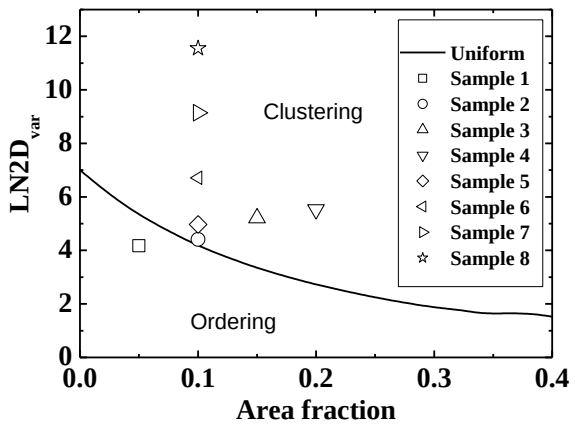

Fig. 6. Variance of LN2D obtained from Al-SiC composites.

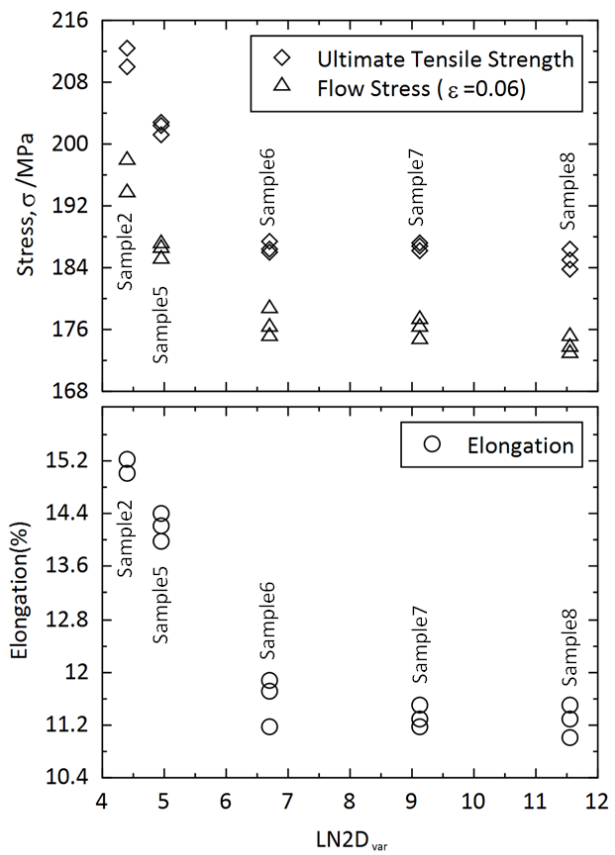

Fig. 7. Mechanical properties of Sample 2, 5, 6 7 and 8.

Figure 7 shows relation between LN2D$_{var}$ and mechanical properties of Sample 2, 5, 6 7 and 8. Ultimate tensile strength, flow stress (ε = 0.06) and elongation of Sample 2 were highest in the samples with the same volume fraction of 0.1. With this kind of a figure, Figure 6, particle spatial distribution in particle dispersed composites can be evaluated quantitatively.

5 Summary

We defined LN3D and LN2D to describe the spatial distribution of second phase particles and applied this definition to overlap-forbidden second phase particles. LN3D$_{var}$ and LN2D$_{var}$ were proper descriptors to evaluate spatial distribution of second phase particles quantitatively. There is a uniform spatial distribution which is specific to volume fraction. Mechanical properties of Al-SiC composites correlate with the spatial distribution of SiC particles. The sample with the uniform spatial distribution of SiC particles had better mechanical properties.

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