

## PARTICLE ARRANGEMENT CHARACTERIZATION BY THE PAIR CORRELATION FUNCTION

Z. Gácsi, J. Kovács, T. Pieczonka

### **Abstract**

*The present paper focuses on the statistical approximation of particle inhomogeneity and on quantifying the degree of particle clustering in the case of aluminum-based SiC ceramic particle-reinforced composites. The composite materials were produced using the powder metallurgy method. SiC powders in different particle sizes (P220, P500 and P800) and quantities (5, 10 and 15 wt.%) were mixed with the aluminum base powder. The powder mixtures were compacted by uniaxial cold pressing and the green samples were isothermally sintered in high purity nitrogen at 610°C for 2 hours. The pair correlation function (PCF) was selected as a mathematical tool to investigate the particle arrangement by using a self-made computer program. The influence of particle size and amount of ceramic reinforcements on the degree of clustering was studied.*

**Keywords:** *particulate composite, powder metallurgy, particle clustering, pair correlation function, image processing*

### INTRODUCTION

Light metal alloys combined with high wear resistant hard ceramic particles are widely applied on several fields of the material science. Embedding these particles in the soft matrix increases the yield strength, the elastic modulus, the thermal resistance and the hardness of the final product but it decreases its thermal expansion. Due to these advantageous attributes, the composites can be properly set up as engine components in the aircraft and automotive industry, or as brake discs for railway brake equipment [1]. Lightweight, high bulk thermal conductivity, low thermal expansion, and excellent thermal resistance make it possible to use them for microprocessor parts in the computer field and for microwave electronic packaging in the microelectronics industry [2].

The mechanical properties of these composites are determined mainly by their microstructure. The main question in particulate composite production is how to achieve a homogenous distribution of the ceramic reinforcements in the matrix. Their volume fraction and mean size mainly influence the arrangement of these particles in the matrix. There is an optimal value of the ratio: mean aluminum powder particle size vs. mean reinforcement SiC particle size, which provides the best properties for specific composition (in terms of constituents concentration given in wt.%) of these composites [3]. On the other hand, the volume fraction of SiC in the composites is also of great importance in cluster formation. The increase of the reinforcement volume fraction can easily lead to the association of its particles in clusters, which in turn can produce more porosity among them. Thus, the higher the degree of clustering the lower mechanical properties of the composite is observed. Therefore, an important problem is to describe the reinforcement distribution in the matrix

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Zoltán Gácsi, University of Miskolc, Physical Metallurgy Department, Miskolc, Hungary

Jenő Kovács, University of Miskolc, Academic Research Group for Materials Science, Miskolc, Hungary

Tadeusz Pieczonka, University of Mining and Metallurgy, Department of Metallurgy and Materials Engineering, Kraków, Poland

by comparable numerical data. "Clustering Probability Maps" were designed to explain the limits for clustering using spherical and mono sized particles for the analysis [4]. These diagrams show the relative particle size ratio (PSR) reinforcement phase to the matrix as a function of reinforcement volume fraction. The different zones indicate the probability for clustering.

## MATERIALS AND METHOD

### Initial powders and mixtures

Atomized, ~99.5% pure aluminum powder produced by FLUKA AG, and different grain-sized ball milled SiC powder prepared by NORTON AS were used as components of the starting materials. The average particle size of aluminum powder was ~20  $\mu\text{m}$ , and the average particle sizes of the SiC powders were ~70  $\mu\text{m}$  (P220), ~14  $\mu\text{m}$  (P500) and ~8  $\mu\text{m}$  (P800). Some more data as to the powders used for composite sample production were published earlier [5]. The powders were blended for 30 minutes in a magnetic laboratory mixer to achieve the proper quality mixtures. Seven mixtures of varying SiC content (5, 10 and 15wt.%) and SiC particle size (P220, P500 and P800) were prepared.

### Pressing and sintering

Rectangular (14x4x4mm<sup>3</sup>) green compacts were fabricated using uniaxial pressing at 400 MPa pressure. No lubricant was used during the compaction. The sintering process was carried out in a push rod NETZSCH 402E dilatometer. High purity (99.999%) nitrogen gas atmosphere was used during the sintering. The technical background of dilatometry investigations, and the dimensional changes occurring during the sintering of analyzed materials have been presented previously [6]. The sample types, the pressing and sintering parameters selected for further examinations are shown in Tab.1.

Tab.1. Sample types, pressing and sintering conditions.

Sample type	SiC grade	Pressing data		Sintering parameters			
		Pressure [Mpa]	Lubricant	Temp. [°C]	Time [min]	Heating rate [°C/min]	Cooling rate [°C/min]
Rectangular samples							
Al-5wt.%SiC Al-10wt.%SiC Al-15wt.% SiC	P220						
Al-15wt.% SiC	P500	400	-	610	120	10	20
Al-5wt.%SiC Al-10wt.%SiC Al-15wt.%SiC	P800						

## CALCULATION OF THE PAIR CORRELATION FUNCTION OF PARTICLES

### Theoretical and methodological backgrounds

Several computer-aided processes were worked out in recent years for numerical description of the particle arrangement and for the degree of particle clustering determination [7, 8]. Because the local inhomogeneity of particles may change within field of view from point to point, the methods have been extended to create the pair correlation function (PCF). Nowadays, this function can be used for quantitative characterization of these microstructures. Different types of arrangement of ceramic particles in the composites have been numerically evaluated using this function. Test circles of radius  $r$  were drawn around the particles. Let  $\lambda$   $K(r)$  be the average number of other particle centroids contained in the test circle, where  $\lambda$  is the intensity of particle centroids in the field of view. The pair correlation function ( $g(r)$ ) was presented by Ohser [9] as follows:

$$g(r) = \frac{1}{2\pi r} \frac{dK(r)}{dr} \quad (1)$$

For estimation of this function the following equation was applied:

$$g^*(r_i) = \frac{\frac{N_i}{A_i}}{\frac{N}{A}} \quad (2)$$

Where  $N_i$  is the number of particles within the  $i^{\text{th}}$  circular ring,  $A_i$  is the area of the  $i^{\text{th}}$  circular ring [ $\text{pixel}^2$ ],  $N$  is the total number of particles in the field of view,  $A$  is the total field of view area [ $\text{pixel}^2$ ].

A simplified method for estimation of the  $g(r)$  function can be seen schematically in Fig.1.

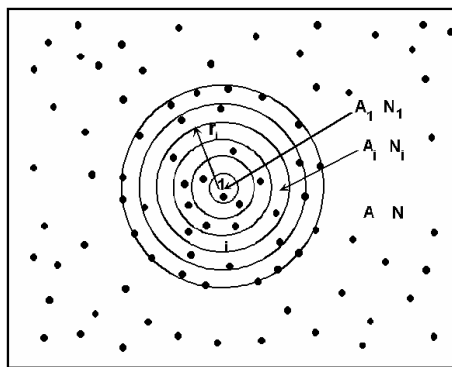


Fig.1. Schematic view showing the principle of the  $g^*(r)$  calculation.

A computerized algorithm for calculation of the pair correlation function has been worked out by the authors. The current investigations were carried out using micrographs of the sintered Al-SiC composites. The microstructures were digitally examined by image analyzer type Quantimet 500 Image Workstation equipped with an optical microscope. Calculation of the pair correlation function of SiC particles observed on the polished transverse sections of the sample was on an algorithm base. The positions of SiC particles in the aluminum matrix were determined by their centroid co-ordinates ( $x$ ,  $y$ ). The resolution of the analyzed images was  $480 \times 480$  pixels and the dimension of the measured frame was  $280 \times 280$  pixels. Concentric circular discs were drawn in the pictures from the

centroid of each particle whose centroid could be found inside the measured frame. The number of reinforcing particles was counted in each circular ring. The computerized method for the PCF calculation is schematically demonstrated in Fig.2. The computations were performed considering the following assumption:

The distance of each particle centroid from the circular disc center should be less than or equal to the circular disc radius.

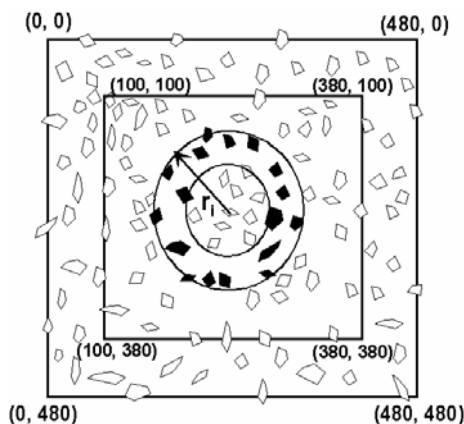


Fig.2. Scheme for computer-aided PCF calculations.

In the case of random particle distribution, the pair correlation function values are nearly 1. If the pattern of the ceramic particles shows a highly clustered array, one significant local maximum appears on the function curve and it is much greater than 1. For areas identified by locally very inhomogeneous particle arrangement, the values of PCF are also above 1. In places where the particles are distributed at a large distance from each other, the function has values below 1. The basic theoretical plots of PCF are presented in Fig.3.

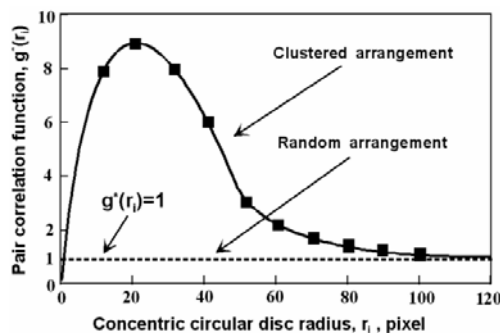


Fig.3. The basic theoretical plots of the pair correlation function [8].

### Calculations on test point patterns

To prove the usage of the algorithm adjusted to determine the reinforcement distribution in composites, the pair correlation function was calculated on test patterns first. Four typical theoretical point arrays (hexagonal, square, random and clustered) were chosen

for the analyses (Fig.4.). The smallest circular disc radius was 10 pixels and this value was increased stepwise by 10 pixels in the case of each concentric disc. The maximum disc radius was 100 pixels and the number of investigated points was 400 in all cases.

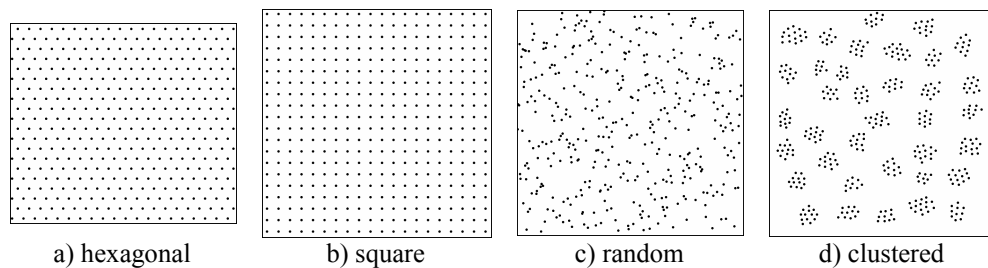


Fig.4. The theoretical test point patterns.

The pair correlation functions calculated on test point patterns as functions of the applied circular disc radii are plotted in Fig.5.

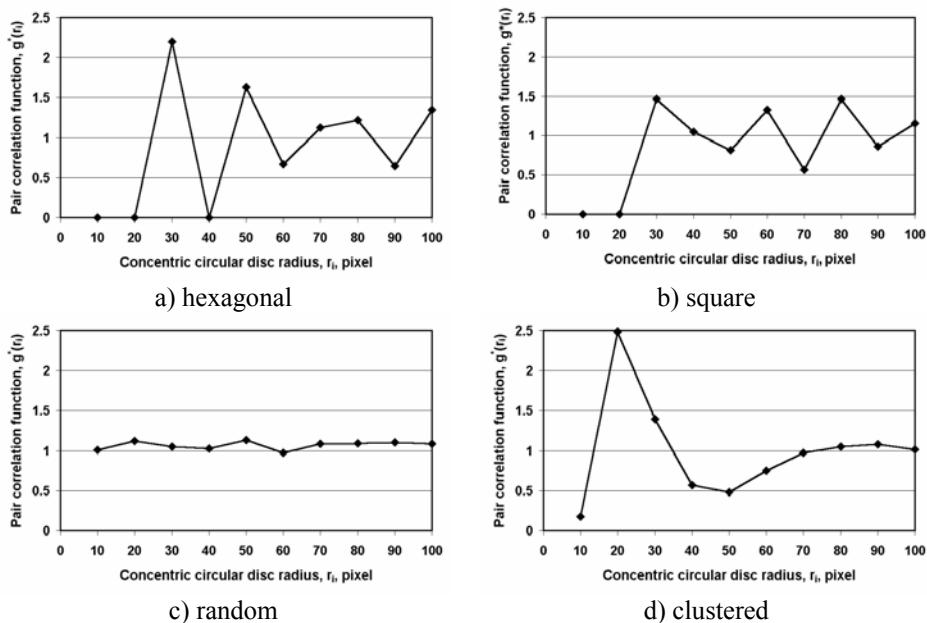


Fig.5. The calculated pair correlation functions.

If many well-determined local maximums can be observed on the pair correlation function curve, the point arrangement corresponds to the hexagonal or square pattern. In the case of hexagonal and square array, the locations of the first, second and third peaks on the curve provide approximately the average distances of the first, second and third neighboring particles. Using this method, the hexagonal and the square arrangement cannot be properly divided. The pair correlation function curve of the random array differs from the others very strongly. In the case of arrangement which accords with the Poisson point process the function curve does not have significant local maximal or minimal values. If the points are settled randomly, the pair correlation function values are almost identical to 1

and its deviations are very small. Regarding the clustered point pattern, there is one significant local maximum on the curve and it indicates the average cluster radius ( $\approx 20$  pixels). The minimal value identifies the mean distance measured from the individual points to the places filled in with few points ( $\approx 50$  pixels) and the second, lower peak shows the mean distance between the individual points and the neighboring cluster centroids ( $\approx 90$  pixels). The spacing between the first maximum and the second lower peak provides the average distance among the neighboring cluster centroids ( $\approx 70$  pixels). The locations of cluster centroids seem to a regular arrangement. The definitions of the characteristic distances determined for the clustered test point pattern are shown in Fig.6.

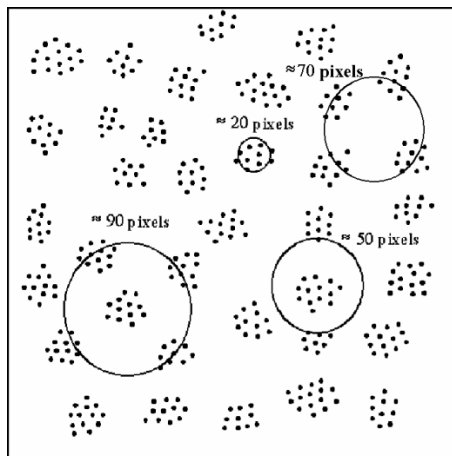


Fig.6. Mean distances in the clustered point arrangement.

## PROGRAM EXECUTION ON THE REAL COMPOSITE MICROSTRUCTURES

### Image processing steps for the determination of centroid co-ordinates of particles

Samples were prepared for quantitative metallographical observation by polishing. The microstructures were analyzed at different magnifications (microscopic and electrical) depending on mean SiC particle size: for P220 at 400x, for P500 at 800x, and for P800 at 2000x. In order to measure properly the SiC particle centroid co-ordinates, an accurate method was developed using a lot of operations (Image Amend-Linear Close, 6 cycles-removes the small black objects and thus eliminates the fine, rounded pores in the matrix from the image Detect-creates the binary image, Binary Segment, 5 steps, 1 filter-separates the touching or overlapping SiC particles from each other, Ultimate Erosion, 5 steps, 1 filter-finds the particle centroids) for image transformations on the gray and binary images (Fig.7).

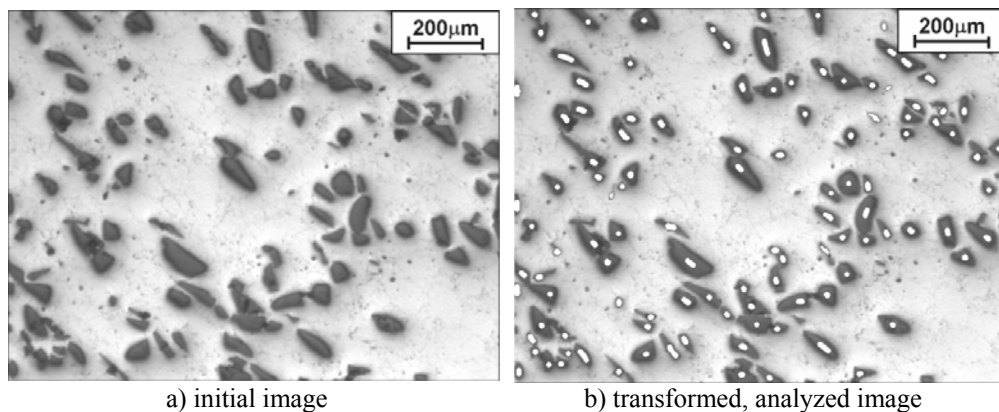


Fig.7. Image preparation for the measurement of centroid co-ordinates of SiC particles.

### The calculated results of the reinforcement distribution

The computer program mentioned above was executed on the investigated composite samples. The smallest circular disc radius was 10 pixels regarding to the finest SiC (P800) sample, 20 pixels in the case of reinforcement grade P500 and 25 pixels concerning the sample containing the largest SiC particles (P220). The maximum disc radius was 100 pixels for each sample. The calculations were performed on 10 fields of view, and around 500 particles were examined for each sample. The pair correlation function curves plotted against the circular disc radii for different SiC particle sizes and for different SiC content in the aluminum matrix are demonstrated in Fig.8.

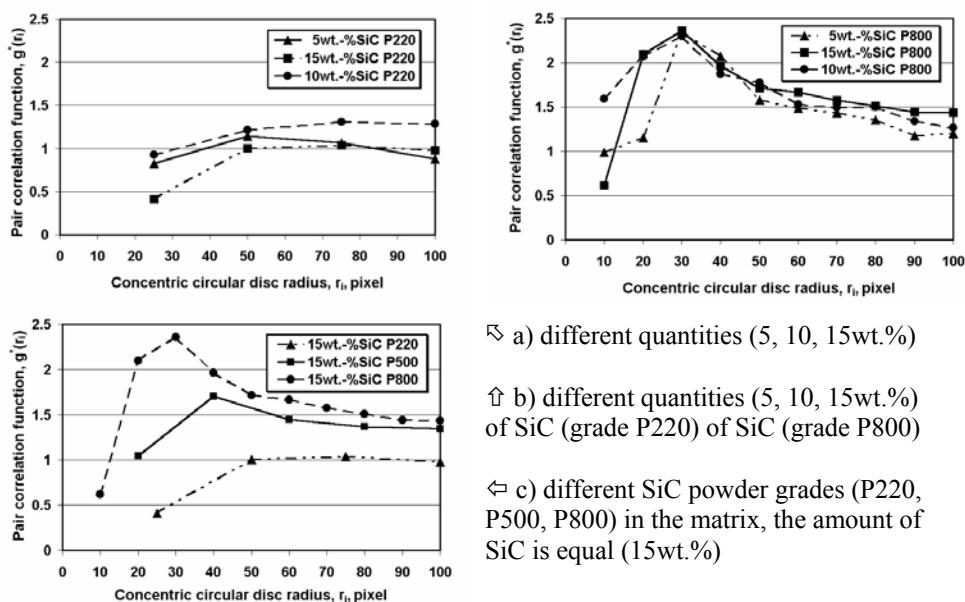


Fig.8. The pair correlation function values calculated on the different composite samples.

The effect of the SiC powder grade (P220 and P800) on the clustering tendency, and thereby on the microstructure evolution, is illustrated in Fig.9.

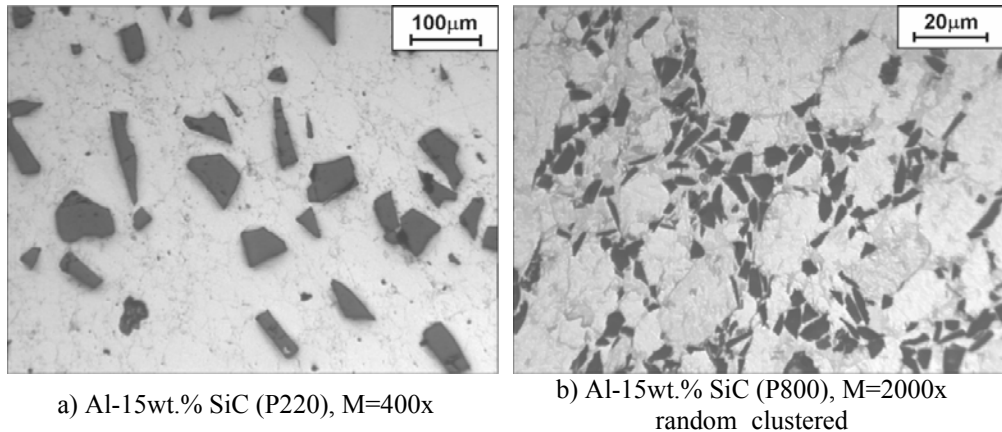


Fig.9. Micrographs of the investigated composite samples.

## CONCLUSIONS

A fully computerized algorithm was developed to characterize the relationship between ceramic reinforcement particle size and its amount, and its inhomogeneous distribution in a metal-matrix composite. It has been proved that for this purpose the determination of the pair correlation function values seems to be useful.

From the results of examinations performed on the test point pattern it could be concluded:

- the hexagonal and the square point array could not be properly separated by using the PCF method, but the mean distances of the first, second and third neighboring points could be calculated,
- the average cluster radius, the mean distance measured from the individual points to the places filled in with few points, the mean distance between the individual points and the neighboring cluster centroids, and the average distance between the neighboring cluster centroids could also be determined,
- in the case of random point array, the pair correlation function values were nearly 1.

Calculations carried out on the real microstructures of the sintered Al-SiC composites allow a conclusion as follows:

- if the ratio of SiC particle mean size to the average aluminum grain size decreases, the degree of clustering increases, when that ratio is much lower than 1, the probability for clustering is higher,
- the use of coarse SiC particles supports the random distribution of reinforcement in the matrix,
- the amount of SiC particles distributed in the aluminum matrix influences the clustering much less than their size,
- the developed method proposed is useful for quality control (in terms of clustering tendency) of metal-matrix composites reinforced by ceramic particles, and thereby allows one to modify the powder metallurgy procedure, if necessary,



- since the finer ceramic particles tend to form a cluster in sintered compacts the use of a polymer binder during powder mixture preparation should be considered.

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