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On the Stereology of the Radial Distribution Function of Hard-sphere Systems

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Most stereological works seem to aim at obtaining informations about the individual particle objects which are distributed in the space of a specimen. This paper, on the other hand, concerns about the radial distribution function of a set of particles. This function characterises the spatial structure of a particle system. Some empirical relationships of radial distribution functions in three-dimensional space to those in two-dimensional section are given for hard-sphere systems. Then, the stereological formulae for radial distribution function given by Hanisch & Stoyan(1981) are examined for hard-sphere systems. The result of comparison indicates some serious discrepancies occur when hard-sphere systems with varying radii are considered.

INTRODUCTION

It seems most works on stereology have been concentrated on the estimation of parameters which characterize the size and shape of individual objects (particles) [for example, see Weibel(1980)]. But it will also be important to develop a scheme which allows us to infer certain parameters of the connectivity between particles. Usually, the connectivity is represented by the term "spatial structure".

Let us consider the case, for example, where two kinds of hard-sphere (non-overlapping sphere) are distributed in a specimen. If we have obtained informations about the individual spheres, such as radii and concentration, could we infer about the inner structure of the system? We have no sufficient information about such a structure yet. In order to obtain such an information, we must measure the interrelationship between particle positions.

For that purpose, there is a quantity called "radial distribution function" (hereafter, we abbreviate it by RDF) which is one of the descriptive measures of spatial structure. This function is usually represented as $\underline{g(r)}$, and is defined in the following manner.

Let ρ_V be the number density (this is usually called numerical density in the standard textbook of stereology) in three-dimensional space. This is equivalent to the mean number of particles per unit volume. Let $\rho_V K_V(r)$ represent the mean number of particles in a sphere of radius r whose center is at an arbitrary particle. Then the radial distribution function in three-dimensional space, $g_V(r)$ is given by the relation:

$$g_V(r) = \frac{1}{4\pi r^2} \frac{dK_V(r)}{dr}. \qquad (1)$$

The two-dimensional radial distribution function, $g_A(r)$, is similarly defined if we properly change the definition of quantities which appeared in 3-D case to fit 2-D case. That is, let ρ_A be the number density in 2-D space, and $\rho_A K_A(r)$ be the mean number of particles in a circle of radius r whose center is at any particle. Then, $g_A(r)$ is given by

$$g_A(r) = \frac{1}{2\pi r} \frac{dK_A(r)}{dr}. \qquad (2)$$

The function $g_V(r)$ is usually used in statistical physics in order to represent the structure of atomic systems such as liquid phase (see, for instance, Rice & Gray:1965). Moreover, the function $K_V(r)$ for a certain system can be experimentally obtained from X-ray diffraction, for example. The functions $g_A(r)$, $K_A(r)$ are used in spatial statistics for the characterization of some spatial data such as distribution pattern of a certain kind of trees (see, for example, Diggle: 1983).

It will be interesting to note that for ideal gases, i.e., for completely random configurations of particles, $g_V(r) = g_A(r) = 1$ holds for any value of r. Deviations from $g_{V[A]}(r) = 1$ represent the existence of correlations between particles at that distance r. For example, if $g(r) > 1$, then the correlation is positive, whereas if $g(r) < 1$, the correlation is negative.

STEREOLOGICAL RELATIONSHIP BETWEEN $g_V(r)$ AND $g_A(r)$

Hanisch & Stoyan (1981) have presented an integral equation which connects $g_V(r)$ and $g_A(r)$ in the case of planar and thin section. Here, $g_A(r)$ is understood to be the RDF for the planar section of a specimen whose RDF is $g_V(r)$. The integral equation is given by

$$g_A(r) = \frac{1}{4[E(\xi)]^2} \int_0^\infty f(u)\, g_V(\sqrt{r^2+u^2})\, du, \qquad (3)$$

where f(u) is given by the following integral

$$f(u) = 2 \int_0^\infty [1 - R_V(\frac{|u-v|}{2})]\,[1 - R_V(\frac{u+v}{2})]\, dv. \qquad (4)$$

Here, $R_V(\xi)$ is the cumulative distribution function of radius ξ of spheres and $E[\xi]$ in eq.(3) is the expected value of ξ.

Note that (3) is the equation for the planar thin section (thickness of section $t = 0$).

RADIAL DISTRIBUTION FUNCTION FOR HARD-SPHERE SYSTEMS

Integral equation (3) has been derived under the following assumption (Hanisch & Stoyan: 1981): the centres x_n of spheres with independently identically distributed radii ξ_n (whose distribution function is $R_V(\xi)$) form a second-order point process of 3-D space strictly stationary under translations and rotations.

Hanisch (1983) used this equation for non-overlapping sphere (i.e. hard-sphere) systems. He applied it to both of fixed radius and varying radii systems.

Let us apply eq.(3) to the following two species hard-sphere system. Let the radii of the sphere of species 1 and species 2 be r_0 and qr_0 $(0 < q < 1)$, respectively, and let the respective corresponding population rates be p and $1 - p$ $(0 < p < 1)$. Figure 1 shows the distribution function $R_V(r)$ of radius for this system. For this system, we can obtain an explicit form of the function $f(u)$. Table 1 shows its result.

Table 1. The explicit form of f(u) for the two species hard-core system whose distribution of radius is given in Figure 1.

(i) $0 \leq q < 1/3$

$0 \leq u < 2qr_0$	$2[2\{q + p^2(1-q)\}r - (2p^2 - 2p + 1)u]$
$2qr_0 \leq u < (1-q)r_0$	$2[2p(1 + q - pq)r_0 - p(2 - p)u]$
$(1-q)r_0 \leq u < 2r_0$	$2p^2(2r_0 - u)$

(ii) $1/3 \leq q < 1$

$0 \leq u < (1-q)r_0$	$2[2\{q + p^2(1-q)\}r - (2p^2 - 2p + 1)u]$
$(1-q)r_0 \leq u < 2qr_0$	$2[2\{q + p(1-q)\}r_0 - u]$
$2qr_0 \leq u < 2r_0$	$2[2\{(1+q) - p^2q\}r_0 - (2 - p^2)u]$

From the equation of Table 1, we obtain

$$f(u) = 2(2r_0 - u)$$

as a special case of single species system by putting q = 1. This derives eq.(25) of Hanisch (1983) as a fixed radius non-overlapping case.

SIMULATION STUDY

We have done a computer simulation for obtaining some samples of hard-sphere systems. The sample was prepared as a random sequential packing of spheres whose $R_V(r)$ is as given in Fig. 1. The explicit procedure for random sequential packing of two species hard-spheres in a finite region is stepwise shown as follows:

(i) The first sphere is put into the region uniformly at random and its radius is chosen to be r_0 or qr_0 with probability p or 1-p, respectively.

(ii) The radius of the n-th ($n \geq 2$) sphere is chosen as similar manner in (i). The centre of the sphere is chosen in the region uniformly at random as a trial position. But if the trial sphere overlaps any of the formerly settled n - 1 spheres, the selection of centre is repeated. Otherwise, the centre is fixed as the position of the n-th sphere.

(iii) The procedure (ii) is repeated until a packing of a specified number of spheres is attained.

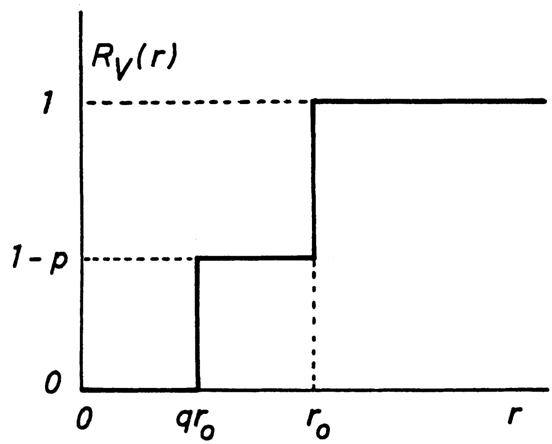

Fig. 1. The distribution function $R_V(r)$ of radii for two-species case.

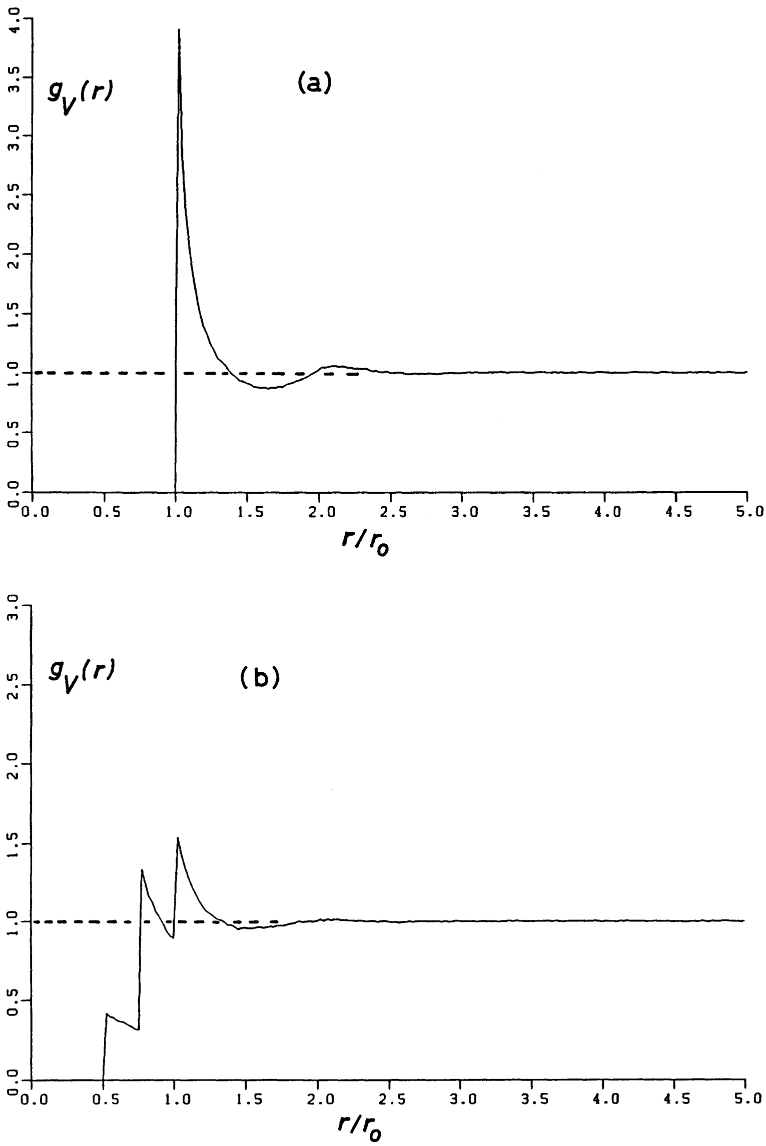

Fig. 2. The RDF $g_V(r)$ for hard-sphere systems : (a) p = q = 1, mean number of spheres = 1000.3, mean volume fraction = 0.362; (b) p = q = 1/2, mean number of spheres = 1501.6, mean volume fraction = 0.339.

The simulations were done for three cases: (a) a single species case (p = q = 1) ;(b) two species case [I] (p = q = 1/2) ; (c) two species case [II] (p = q = 1/4). For each case, samples of 100 packing patterns were generated and $g_V(r)$ was computed from the patterns [see Fig. 2 (a), (b) for the estimated $g_V(r)$]. Then, we evaluated $g_A(r)$ by taking several random sections from each sample pattern. A sample of a random section for the case p=1/4 and q=1/4 is given in Fig. 2.

At the same time, we also evaluated $g_A(r)$ from the eq. (3) where $g_V(r)$ was obtained from the simulation and where f(u) of Table 1 was used. Let us denote $g_A(r)$ evaluated from simulation by $\bar{g}_A(r)$ and the one from eq. (3) by $\hat{g}_A(r)$. The comparison of $\bar{g}_A(r)$ and $\hat{g}_A(r)$ is made in Figs. 4 and 5. Note that for the case (a), $\bar{g}_A(r)$ and $\hat{g}_A(r)$ coincide well for all values of r; whereas for the case (b), they deviate with each other for almost values of r, although the positions of peaks of the graph well coincide. Deviation of $\hat{g}_A(r)$ from $\bar{g}_A(r)$ became much larger for the case (c).

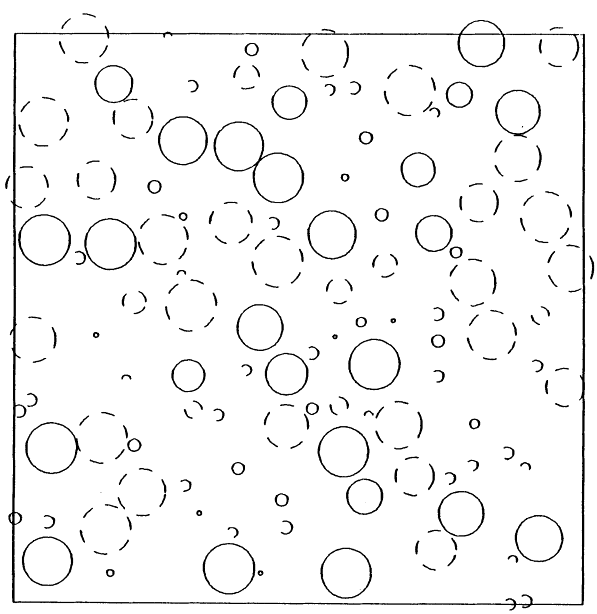

Fig. 3. A sample of random cross section of two-species hard-sphere system (p = q = 1/4). The circles with solid line indicates crossing spheres whose centres are on one side of the section plane and the circles with broken lines represent those ones whose centres are on the other side of the plane.

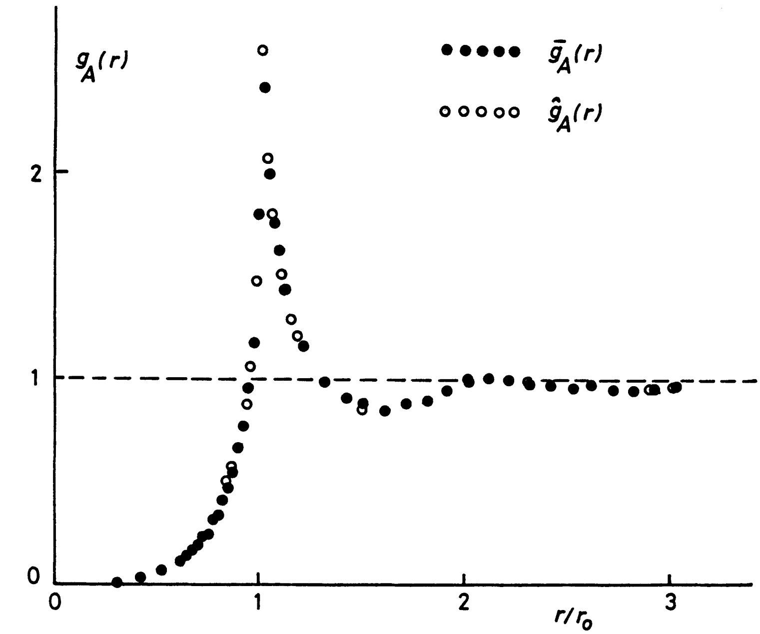

Fig. 4. Comparison between $\bar{g}_A(r)$ and $\hat{g}_A(r)$ for the case (a).

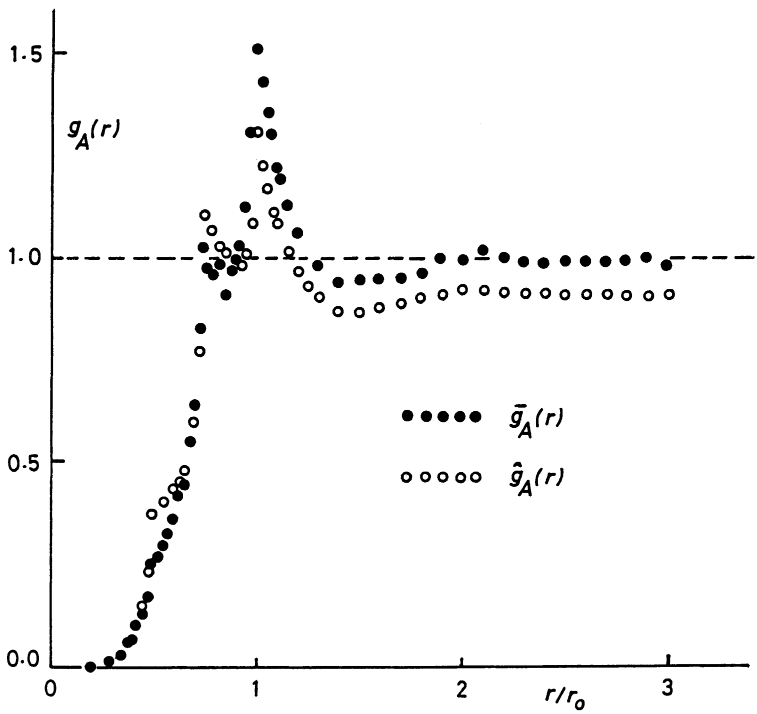

Fig. 5. Comparison between $\bar{g}_A(r)$ and $\hat{g}_A(r)$ for the case (b).

DISCUSSION

The above results indicate that the eq. (3) presented by Hanisch & Stoyan (1981) holds for the hard-sphere system with fixed radius but does not hold for those system with varying radii.

The reason seems to be originated from the assumption which was mentioned above; that is, the assumption of independently identically distributed radii. When the number of radius of spheres is only one, this assumption does not affect the positions of spheres. However, when the number of radius is more than one, the radius of certain sphere at certain position cannot be replaced by another value independently with positions of other spheres according to the non-overlapping property of hard-sphere system. This turns out that in the equation (3), the factor f(u) which contains $R_V(r)$ and those $g_V(r)$ which contains sphere positions cannot generally be separated. It will be easily seen that the same is true also for the case where the radii are continuously distributed.

The derivation of general formula which is useful for the case with varying radii seems very difficult. But some empirical relations derived from simulation studies would help to develop such formula and also to develop stereology of particle correlations.

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3-6

C: I would first like to point out that there is a large 3-D cordinate data set in existence collected on the centroids of osteocyte lacunae collected using Professor Alan Boyde's TSRLM in collaboration with Adrian Baddeley.

On this overhead slide is shown the K(t) function in 3-D. The red line shows the Poisson (i.e. random) distribution. The black lines show the data from several replicates with the 95% confidence intervals in blue. Notice that the data falls below the Poisson at about 25 μm separation (remember in 3-D). This indicates a hand core paching process. This is the only large 3-D cordinate data set in existence, to my knowledge. It should be of some interest to all this morning's speakers. (V. Howard)