Journal of Engineering Physics and Thermophysics, Vol. 68, No. 4, 1995

CONCERNING THE GEOMETRIC LIMIT OF THE DENSITY OF A LOOSE MEDIUM MODELED BY IDENTICAL SPHERICAL PARTICLES

A. B. Shubin

UDC 539.215+532.74

Based on the introduced system of definitions, the statistical-geometric property of a random close packing (RCP) of identical solid spheres (SS) is found that determines the geometric limit of the packing density. The results of calculations using computer models of SS packings show that the magnitude of the geometric limit virtually coincides with a real limiting density of the RCP of SS.

Random packing of identical solid spheres is a convenient idealized model of various disordered systems, in particular simple fluids and granular loose media.

The magnitude of the maximum possible density of a packing of SS (η_{max}) was studied experimentally by many research workers. The values of η_{max} cited in the literature differ only slightly: 0.637 [1]; 0.641 [2]; 0.6366±0.0004 [3]; 0.637±0.003 [4]; 0.646 [5]; 0.639±0.002 [6]. At the present time the most probable experimental value η_{max} = 0.637 is usually used [7]. The theoretical value η_{max} = 0.628 was obtained in [8] employing a number of arbitrary assumptions.

The objective of the present work was to find a purely geometric condition for the existence of a random close packing of SS (within the framework of the system of definitions introduced) and, on the basis of this condition, to estimate the limiting density of this packing.

We consider a uniform packing N of identical SS (N-group), occupying a volume V_0 at time moment t_0, with the value of N chosen as large as desired.

We place randomly in the volume V_0 an additional number k of the same spheres (k-group) at a specified time moment. This arrangement will permit the spheres from the N-group to change their coordinates. Let us affix to the spheres from the k-group a label, which will be their only distinction from the SS of the N-group. The number $k \ll N$ is selected so that at any time moment the probability of contact of one sphere from the k-group with another labeled sphere should be smaller than a certain positive number ε that can be specified to be arbitrarily small. In other words, the number k should be small enough as compared with N to avoid the practical interaction of the labeled spheres with each other.

D e f i n i t i o n 1. Let the k-group of SS be considered to be in equilibrium with a packing of N SS, if on location of this k-group in the volume V_0 the following conditions are satisfied:

a) there are no indications (except for the conventional label) that would make it possible to distinguish between the spheres of the k- and N-groups after the placing of the former;

b) the total density of packing (η) for the system of ($k+N$) spheres remains the same as for the system of N spheres prior to the placing of the k-group.

The requirement b) makes it possible to find the expected value for the change in the total volume of the packing on location of k spheres: $\Delta V = V - V_0 = kv/\eta$, where V is the volume of the packing of ($k + N$) spheres; v is the volume of one sphere.

We now introduce the following additional condition. Let k of SS be located as before randomly in the volume V_0, but with the coordinates of their centers remaining fixed and at any time moment being equal to the coordinates at the moment of placing. This means that these spheres are stationary, and their kinetic energy is equal to zero. By definition, this group of k spheres can be in equilibrium only with such a packing from N of SS,

Ural State Technical University, Ekaterinburg, Russia. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 68, No. 4, pp. 564-568, July-August, 1995. Original article submitted December 10, 1993.

in which the kinetic energy of spheres is also equal to zero. It is obvious that we talk of a close statically stable packing.

It is also obvious that the indicated group of k randomly fixed spheres cannot be in equilibrium with any packing having long-range order (crystalline). Indeed, in this case even the placing of one sphere from the k-group specifies the position of a certain node of the lattice, in which all the other spheres should be located (both labeled and unlabeled). This contradicts the condition according to which the labeled SS are located randomly.

Definition 2. A random close packing of SS is the name given to such a packing of N solid spheres with which a group of k randomly located fixed SS is in equilibrium.

Definition 3. An arbitrarily selected SS from the k-group that is in equilibrium with RCP will be called a trial sphere (TS).

The particular algorithm of the arrangement of the spheres of the k-group is not specified. Therefore, we restrict ourselves to the consideration of only initial (in the volume V_0) and finite (with the volume V) packings that satisfy Definition 2. Suppose the radius of all the spheres is equal to unity ($R = 1$) and, consequently, $v = 4\pi/3$.

Consider an arbitrarily selected SS from the N-group. Let us isolate a spherical layer all the points of which lie at distances in a certain interval $[x; x + dx]$ from the center of this sphere. The volume of this layer is equal to $4\pi x^2 dx$. In this layer we can separate a portion that lies closer to the center of the selected sphere than to the centers of other spheres. In other words, the indicated portion belongs to the Voronoi polyhedron (VP) of the given sphere. Let the mean volume of this portion for all the N spheres at a specified interval $[x; x + dx]$ be equal to $\alpha(x) 4\pi x^2 dx$, where $\alpha(x)$ is a certain function. In this case it is obvious that at a specified density of the packing η

$$4\pi \int_0^{x_{max}} x^2 \alpha(x)\, dx = \bar{u} = 4\pi/3\eta, \tag{1}$$

where x_{max} is the maximum possible distance from an arbitrary point of RCP to the center of the nearest sphere; $\bar{u}$ is the mean volume of VP in an RCP.

The function $\alpha(x)$ has the following properties:

1) when $0 \le x \le 1$, $\alpha(x) = 1$;

2) when $1 < x \le x_{max}$, $\alpha(x)$ is a nonincreasing function;

3) when $x_{max} < x < \infty$, $\alpha(x) = 0$.

Suppose we know that a certain point A belonging to the volume of RCP at a given time moment t lies at a certain distance x from the center of the nearest sphere of the N-group. Further we also know that the RCP volume contains a fixed trial sphere with the center at a certain point O. It is obvious that at the time moment t there exists a spherical volume with center O about which it is known that the given point A is located outside it. Otherwise there would have been the possibility for the overlapping of any sphere from the N-group by the trial sphere. Following [9], we shall call the indicated spherical volume the inaccessible volume (IV) and denote it by $\omega(x)$.

For an arbitrary point A lying at a distance x from the center of the nearest SS of the N-group, the inaccessible volume is defined by the relations:

$$\text{when } x \le 2 \quad \omega(x) = (4\pi/3)(2 - x)^3, \tag{2}$$

$$\text{when } x > 2 \quad \omega(x) = 0. \tag{3}$$

Rather cumbersome considerations, made however without any additional assumptions, permit us to show the following. The expected value for the difference between the finite volume V and initial volume V_0 on placing the k-group cannot be smaller than $k\bar{\omega}$, where $\bar{\omega}$ is the mean value of $\omega(x)$ found for all the points of the volume V_0.

It is obvious that

$$\overline{\omega} = 4\pi \int_0^{x_{max}} x^2 \alpha(x)\,\omega(x)\,dx / 4\pi \int_0^{x_{max}} x^2 \alpha(x)\,dx = 3\eta \int_0^{x_{max}} x^2 \alpha(x)\,\omega(x)\,dx. \quad (4)$$

It is not hard to show that in any statically stable packing the value of x_{max} cannot exceed 2 (at the unit radius of spheres), otherwise the packing would have had cavities exceeding SS in size. Consequently, to specify $\omega(x)$ in an RCP of SS we can use only expression (2), which leads to the relation

$$\overline{\omega} = 4\pi\eta \int_0^{x_{max}} x^2 (2-x)^3 \alpha(x)\,dx. \quad (5)$$

Suppose we randomly select in the RCP volume of an SS I points, each of which at a given time moment is characterized by a certain distance x to the nearest center of the SS of the N-group. The result obtained in the work makes it possible to formulate the following statement: the mean volume of the Voronoi polyhedron in an RCP of SS cannot be smaller than the mean IV for the indicated selection of points (at a large enough I). The mean IV for $I \to \infty$ is defined by expression (5).

Thus, for an RCP of SS (within the scope of Definition 2) the inequality $\overline{\omega} \leq 4\pi/3\eta$ holds. After simple transformations, it is possible to formulate the following property of the DCP. For any density of packing at which the RCP of SS exists, the following relationship is satisfied:

$$\eta \leq \left(3 \int_0^{x_{max}} x^2 (2-x)^3 \alpha(x)\,dx\right)^{-1/2}. \quad (6)$$

Since for $0 \leq x \leq 1$, $\alpha(x) = 1$, it can be transformed to

$$\eta \leq \left(2.1 + 3 \int_1^{x_{max}} x^2 (2-x)^3 \alpha(x)\,dx\right)^{-1/2}. \quad (7)$$

We can easily show that if the function $\alpha(x)$ satisfies the above properties 1)-3), the function $\overline{\omega}(\eta)$ is an increasing function. Two mutually exclusive statements are possible.

1. The real limit for the density of the packing of an RCP of SS is determined by the inaccessible volume factor considered in the work. In this case the following equality holds: $\overline{\omega}(\eta_{max}) = 4\pi/3\eta_{max}$.

2. The real limit for the density of the packing of an RCP of SS is determined by other factors not taken into account in the present work. In this case the following relation holds: $\overline{\omega}(\eta_{max}) < 4\pi/3\eta_{max}$.

We estimate the limiting density of the RCP assuming the validity of statement 1.

If the function $\alpha(x)$ was known at any value of η on the segment $[1; x_{max}]$ in expressions (5)-(7), it would be not difficult to perform a precise calculation of η_{max}. However, a rigorous theoretical determination of this function for an RCP of SS seems impossible. Therefore, we shall avail ourselves of the models that specify a particular form of the functions $\alpha(x)$ and $\omega(\eta)$.

Suppose there exists a model of an RCP of SS (analytical or computerized) that makes it possible to find the value of $\overline{\omega}$ at a given value of η. If there is a value of η at which $\overline{\omega}(\eta) = 4\pi/3\eta$, then this density of packing will be called an estimate for η_{max} within the framework of this model and will be denoted by η_g.

Calculational experience shows that the dependence of the quantity η_g on the form of the model is very weak. This is due to the small sensitivity (at a given η) of the value of a certain integral in expression (5) to a specific form of the function $\alpha(x)$ that satisfies properties 1)-3) for any model. Let us illustrate this by some examples.

1. We considered a specifically rough representation of the function $\alpha(x)$ in the form of $\alpha(x) = 1$ on the entire segment $[1; x_{max}]$. This directly specifies $x_{max} = \eta^{-1/3}$. We can show that the use of such an approximation gives the smallest possible value of η_g at any function $\alpha(x)$ satisfying properties 1)-3). This value can be easily found analytically and is equal to 0.6255...

2. We generated on a computer the "original" crystalline packings of SS with bcc (1729 spheres) and fcc structures (1688 spheres). The radius of all the SS was somewhat decreased, after which the spheres were subjected to multiple random displacements from the nodes of the original lattice. Thereafter, we found the values of $\overline{\omega}$ and η from the data for 50,000 randomly selected points in the volume of packing. We constructed the function $\overline{\omega}(\eta)$ from the results of 25 computational experiments at different values of η. It is the density of packing at the point of intersection of the indicated dependence with the decreasing function $\overline{u}(\eta) = 4\pi/3\eta$ which is the unknown value of η_g. It turns out that the values of η_g are very weakly dependent on the form of the "original" structure, and at the bcc and fcc "original" packings they constitute 0.633±0.002 and 0.635±0.003, respectively.

It can be seen easily that the value of η_g for real models of packings differ from the a fortiori rough approximation (example 1) by only 1.2–1.5%.

On the basis of the calculations made and other experimental data we may state that for any real function $\overline{\omega}(\eta)$) the value of η_g will lie on the interval 0.635±0.005 with probability close to unity. This conclusion relates, of course, also to the RCP of SS satisfying Definition 2.

It is seen that the estimates of η_g obtained within their scatter practically coincide with the value of η_{max} known from experiment for a real DCP of SS. Thus, the available data do not contradict the assumption about the validity of the statement that 1: $\overline{\omega}(\eta_{max}) = 4\pi/3\eta_{max}$. This means that the real limit for the density of packing of an RCP of SS seems to be due predominantly to the geometric factor considered in the work.

Of great interest is the calculation of η_g with the use of various presently available computer models of RCP of SS. Sufficiently adequate models should give coincident values for the limiting density.

NOTATION

η_{max}, limiting density of packing in an RCP of SS; V_0, volume of packing before placing trial spheres; V, volume of packing after placing trial spheres; N, number of spheres in a packing; k, quantity of trial spheres; η, density of packing; x, distance from a given point to the nearest center of SS; ω, inaccessible volume; x_{max}, the largest value of x in a packing; η_g, model estimate of the limiting density of packing.

REFRENCES

1. G. D. Scott, Nature, **188**, No. 4754, 908-909 (1960).
2. R. Rutgers, Nature, **193**, No. 4814, 465-466 (1960).
3. J. L. Finney, Proc. Roy. Soc. London A, **319**, No. 1539, 479-493 (1970).
4. G.A. N. Connell, Solid State Commun., **16**, No. 1, 109-112 (1975).
5. N. I. Topuridze and D. V. Khantadze, Inzh.-Fiz. Zh., **33**, No. 1, 120-125 (1977).
6. Cl. Lemaignan and P. Desre, J. Phys., **41**, No. C8, 305-308 (1980).
7. Yu. A. Buevich, D. E. Dyusembaev, Sh. K. Kapbasov, and E. V. Maksimov, Inzh.-Fiz. Zh., **64**, No. 2, 209-216 (1993).
8. B. S. Radovskii, Izv. Akad. Nauk SSSR, Mekh. Tverd. Tela, No. 4, 195-198 (1972).
9. V. L. Lyuboshits and M. I. Podgoretskii, Usp. Fiz. Nauk, **161**, No. 11, 129-150 (1991).