ACTA STEREOL 1990; 9/2: 219-233
ORIGINAL SCIENTIFIC PAPER

DETAILED CHARACTERIZATION OF A COMPUTER-SIMULATED RANDOM PACKING OF MONOSIZED SPHERES

Bhanu Prasad Pinnamaneni*, Jean-Paul Jernot, Hubert Robine

ISMRa, LERMAT, URA CNRS 1317, 14050 Caen Cedex, France

ABSTRACT

Two computer-simulated random packings of monosized spheres are characterized quantitatively. The wall effect and the anisotropy of the structure are studied. Two mean parameters, the number of particles and the number of contacts per unit volume, are determined as a function of the size of the field of measurement. The pair correlation function is established and the effect of gravity on the stackings is investigated.

Keywords : Simulated packing of spheres, wall effect, anisotropy, pair correlation function.

INTRODUCTION

The context of this work is the three-dimensional quantitative analysis of digitized structures. As a first step, the use of a 3D computer-simulated random packing of spheres is very convenient.

Thus, the aim of this paper is not to perform a general study of 3D packings of spheres, but to characterize quantitatively, in a practical and detailed way, one structure which will be further used for testing 3D transformations.

* Present adress : Confocal Technologies Ltd., Mulberry place, Liverpool L7 7HJ, United Kingdom.

SIMULATIONS OF RANDOM PACKINGS OF MONOSIZED SPHERES

Two random 3D packings of monosized spheres were simulated on a SUN 3 computer using the Fortran program of Visscher and Bolsterli (1972). Each simulation has been performed as follows :

- The simulation is made inside a box ($0 \leqslant x \leqslant 40, 0 \leqslant y \leqslant 40, 0 \leqslant z \leqslant 40$) using monosized spheres of radius r = 1.
- The balls are dropped successively from $z \geqslant 40$ at random x and y coordinates.
- if the ball hits the bottom of the box, it stops.
- if the ball hits one of the previous balls, it rolls downwards to get further contacts up to the stability defined by the "gravitational forces".

It must be noticed that each time a ball is cut by a side of the box in X and Y directions, the outside part of the ball reappears inside the box on the opposite side.

This procedure is used to build two simulations :

a) **Simulation S1** : The bottom of the box is flat and the x and y coordinates of the falling balls are randomly chosen.

b) **Simulation S2** : The simulation S2 is built upon the simulation S1, the top layer of S1 being taken as the bottom of the box. The balls are dropped above the minimum height of the stacking to insure a kind of smooth filling of the box. For that purpose, a square grid (200x200) is placed above the box. At each step the vertical distance h between each point of the grid and the top layer ball is computed and the new ball is dropped from the node for which h is maximum.

In these simulations, the total number of particles is approximately 9000 and the time necessary to perform the simulations is roughly seven hours.

CHARACTERIZATION OF THE SIMULATIONS

Several phenomena will be successively studied on the two structures S1 and S2 :

- the wall effect
- the field size effect
- the anisotropy
- the local fluctuations in the stackings

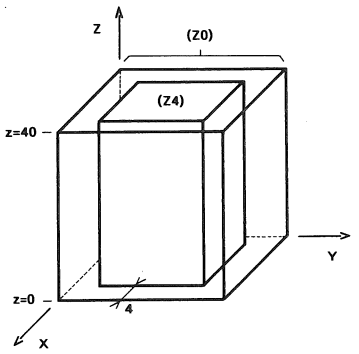

Fig. 1 : Complete, (Z0), and restricted, (Z4), fields sweeping the simulation box along Z direction.

1) **Wall effect**

The plane walls of the simulation box induce some local order on the positions of the balls. This effect is limited to the vicinity of the walls in the three directions X,Y,Z. But, owing to the simulation process, its magnitude should not be the same for these three directions. Therefore, an individual assessment of the wall effect along each axis is required. For instance, the "pure" wall effect along the Z axis will only be obtained after removal of the wall effect along X and Y. This is achieved by the following procedure (Fig.1) :

- The simulation box is intersected by successive parallel planes at increasing altitudes in the Z direction
- Two kinds of fields of measurement are then defined: complete fields (Z0) consisting of the whole intersection with the simulation box and restricted fields (Z4) obtained by removing from (Z0) an outer strip of width 4 units along the directions X and Y. A justification for the value 4 units will be given later on.

The first analysis is the determination of the areal fraction, $A_A(P)$, (or volumic fraction V_V) of particles at increasing values of coordinates. This parameter is calculated from the part of the particle traces which lies inside the field of measurement. The results of analysis in X and Z directions are diplayed on figure 2. The distinction between the complete fields - (X0) and (Z0)- and the restricted ones - (X4) and (Z4)- is not clear-cut but it must be noticed that the mean

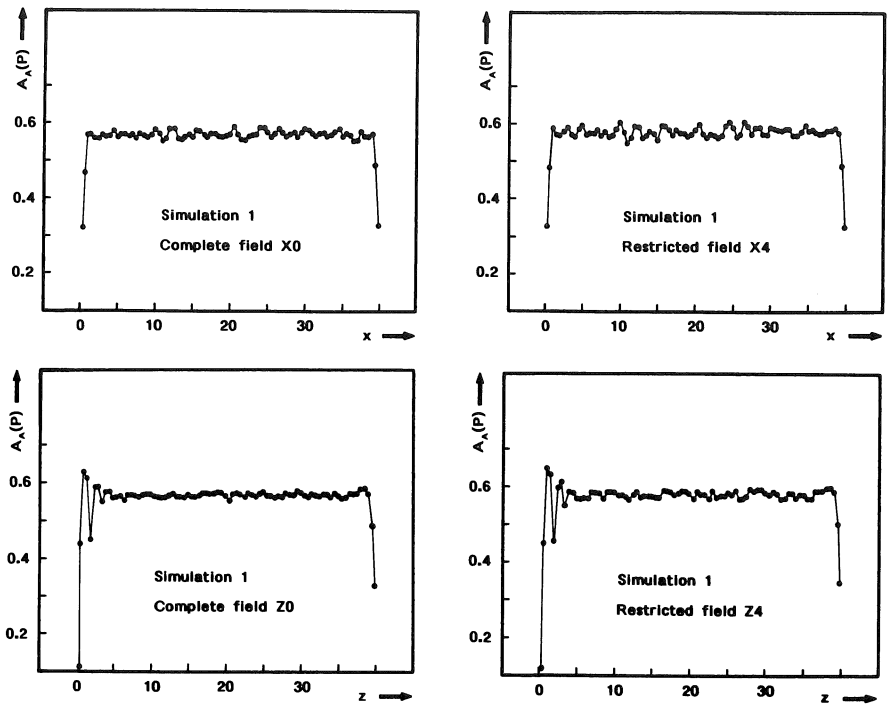

Fig. 2 : Evolution of areal fraction of the particles, $A_A(P)$, in a plane sweeping the simulation box.

value for A_A is significantly lower in the first case (0,569 instead of 0,579). When a comparison is made between the samplings in X and Z directions (e.g.,(X4) and (Z4)) a noticeable difference in the shape of the curves is observed near the origin. The sharp fluctuations of (Z4) must be ascribed to an effect of the plane bottom of the box in simulation S1. The absence of this phenomenon in (X4) is due to the periodic boundaries conditions imposed in the process of simulation for horizontal directions. The fluctuations observed for (Z0) and (Z4) virtually vanish from abscissa 4 onwards. This indicates that the wall effect is limited, in our case, to a shell of thickness four radii, instead of three or four diameters observed in previous works (Benenati and Brosilow, 1962 ; Gotoh et al., 1978a). As expected, the simulation S2, built above the simulation S1, exhibits no wall effect in Z direction.

The second analysis deals with the mean coordination number of particles, N_c. The local value of this parameter is given by twice the ratio between the number of particle con-

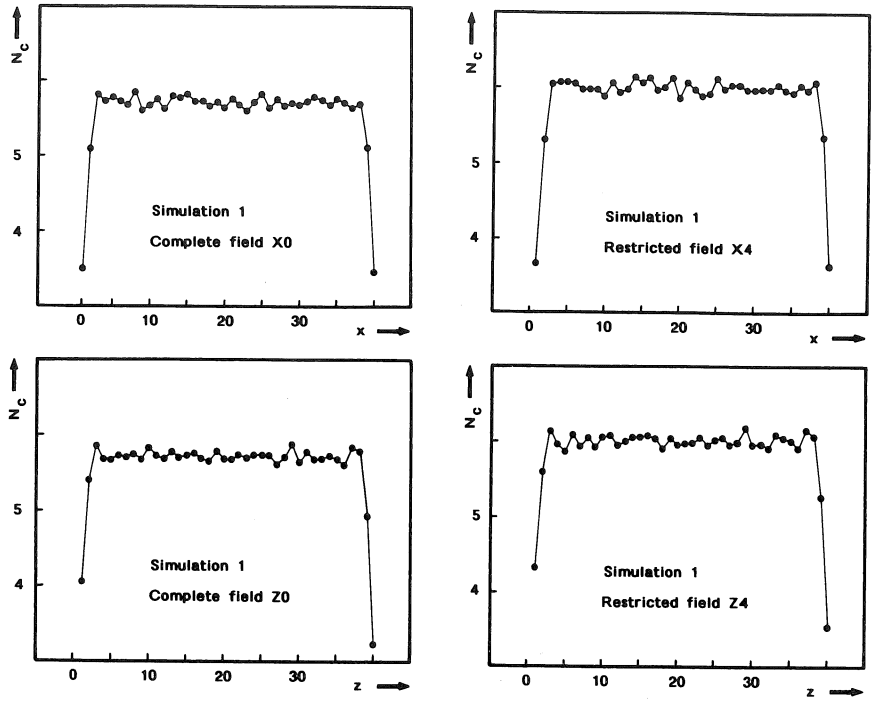

Fig.3 : Evolution of the mean coordination number of particles, N_c, according to their position in the box.

tacts and the number of particle centers in the volume bounded by two successive parallel planes. As can be seen on figure 3, N_c is less sensitive to the wall effect than A_A . Nevertheless, the mean value of N_c increases from (X0) and (Z0) to (X4) and (Z4). Besides, the low value of N_c near the origin is explained by the fact that the particle neighbours are present only in a half-space. The difference between the values of N_c in this domain (~ 3,5 for (XO) and (X4) as opposed to ~ 4,2 for (Z0) and (Z4)), reflects some regularity of the stacking near the bottom plane.

2) **Field size effect**

It is obvious that the measured parameters are meaningful only if the field of measurement is representative of the whole structure. The size of the field must satisfy two opposite requirements : (i) it must be small enough in order to eliminate the wall effects (ii) it must be large enough to be representative of the structure. The simplest way to get an

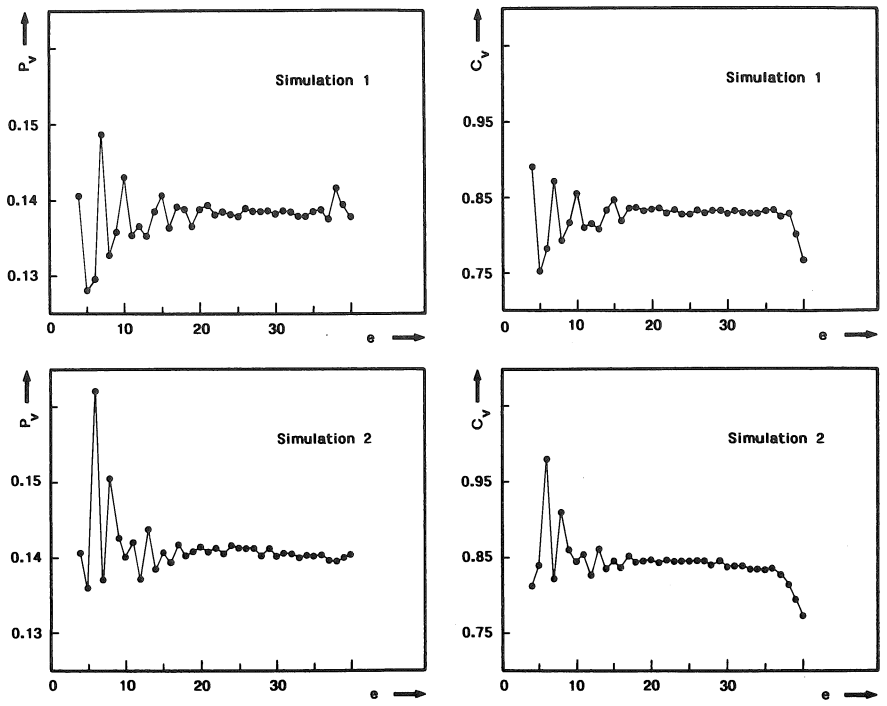

Fig. 4 : Number of particles, P_V, and number of contacts, C_v, per unit volume for increasing sizes of the field (cubic box of edge e)

estimate of the right field size is to measure the parameters of interest in successive boxes of increasing size. The results obtained for P_V and C_v (respectively number of particles and number of contacts per unit volume) are presented in figure 4 for simulation S1 and S2. The decrease in the ordinates of the curves of C_v near their right end is due to the wall effect. Chaotic values are found at the beginning of the curves corresponding to small field sizes.

These results indicate that, for random packings of monosize spheres of radius 1, the minimum field size should be approximately 20. It will then contain about 1000 spheres.

Table 1 summarizes the values found for the main parameters in the largest field without wall effect for simulations S1 and S2.

Table 1 : Global characteristics of the two simulations.

	Number of spheres	V_v	P_v	C_v	N_c
Simulation S1	4537	0.580	0.138	0.415	6.01
Simulation S2	4606	0.589	0.140	0.420	6.00

3) **Anisotropy**

Since the construction of stackings simulate a gravitational field, the resulting structure could possibly present an anisotropy (Tory et al. 1968 ; 1973).

As a simple way to study the spatial orientation of the segments joining two centers of particles in mutual contact, we have measured their projections on each axis (Fig.5). If the structure were isotropic the mean length of the projection, $\bar{h}$, would be equal to 1 (Tory et al. 1973). The mean values of projections in X, Y and Z directions for the two simulations S1 and S2 are gathered in table 2. The expected anisotropy effect is obvious in the Z direction.

Table 2 : Mean lengths of projections of segments joining the centers of touching particles in X, Y and Z directions.

	Simulation S1	Simulation S2
$\bar{h}_X$	0.991	1.001
$\bar{h}_Y$	0.992	1.001
$\bar{h}_Z$	1.092	1.081

The analysis has been taken a step further by establishing the distribution of angles φ_X , φ_Y and φ_Z between the segments and the reference axes X, Y and Z. In the case of an isotropic structure, for an oriented angle $0 \leqslant \varphi \leqslant \pi$, the

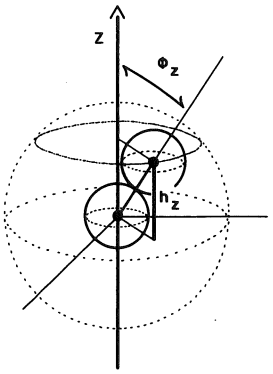

Fig. 5 : Spatial orientation of a biparticle. Projection along Z axis of a segment joining two particle centers.

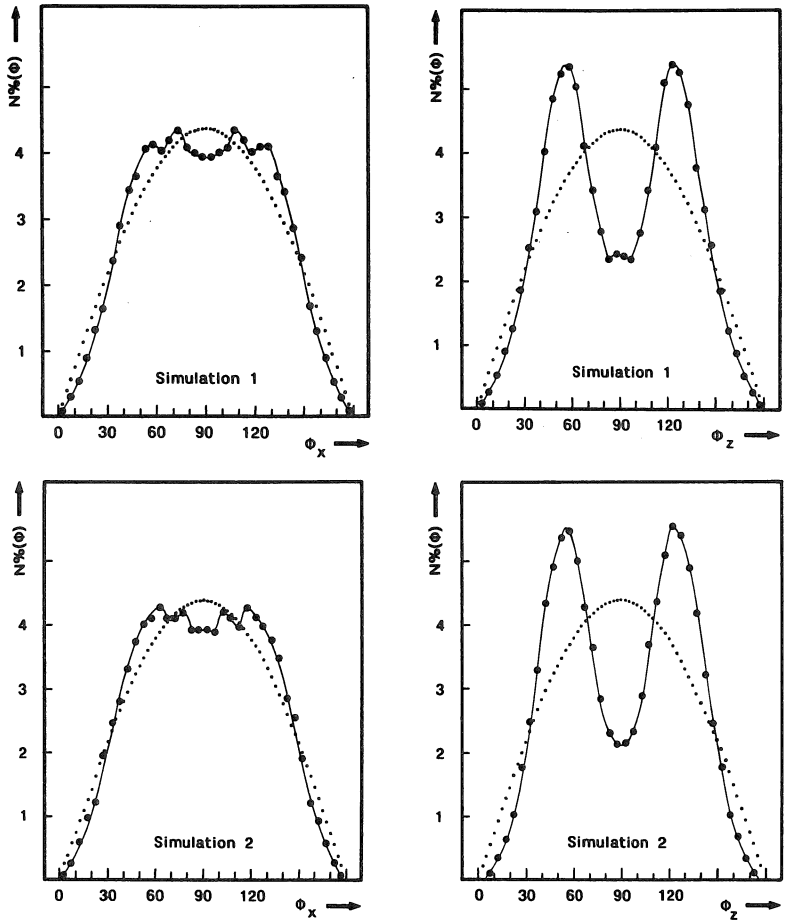

Fig. 6 : Orientation spectra of biparticles along the X and Z directions.

probability for this angle φ to be comprised between φ and $\varphi + d\varphi$ is $1/2 \sin\varphi \, d\varphi$. So, the percentage of orientations within $\pm$ 0.5° of a given value φ should be :

$$N\%(\varphi) \sim 1/2 \sin \varphi \, (\pi/180) \, 100$$

The results are visualized on figure 6 for the two simulations S1 and S2. In each case, the experimental curve deviates from the theoretical one corresponding to the above formula. But the anisotropy is much more striking along the Z direction. In that case, a maximum is found for an angle $\varphi \cong 55°$ (and the symmetrical one $\varphi \cong 125°$). This closely agrees with the previous observations of Tory et al. (1973).

In connection with this result it may be recalled that, according to Bernal (1964), most of the constitutive polyhedra of a random structure are tetrahedra (73%). In the model of flattened tetrahedral units proposed by Gotoh et al. (1978b), the mean height of a tetrahedron equals 1.08 times the particle radius. This is just the value already mentioned as the mean projection length of the segments joining the centers of a biparticle. Moreover, if one assumes that the tetrahedron possesses an equilateral basis and that the most probable orientation of that basis is the horizontal one, one finds 57° as the angle of inclination of the three oblique edges with respect to the vertical axis. The fair coincidence of that value with the abscissae of the peaks on figure 6 provides a strong hint that the most typical geometrical unit in random packings constructed under gravity is a flattened tetrahedron (with no mutual contact between the three particles of the basis). It may be noted however, that the original model of Tory et al., did not imply equal sides for the basis of the tetrahedron, nor horizontality of that basis. This may account for the relatively large width of the peaks in the orientation spectra of figure 6. The aforementioned coincidence between the height of the "typical" tetrahedron and the observed mean length of projections $\bar{h}_z$ may therefore be fortuitous.

4) **Local fluctuations in the stackings**

In order to get more information about the local arrangement of particles, we will first deal with the pair correlation function of particle centers and then with interparticle contacts.

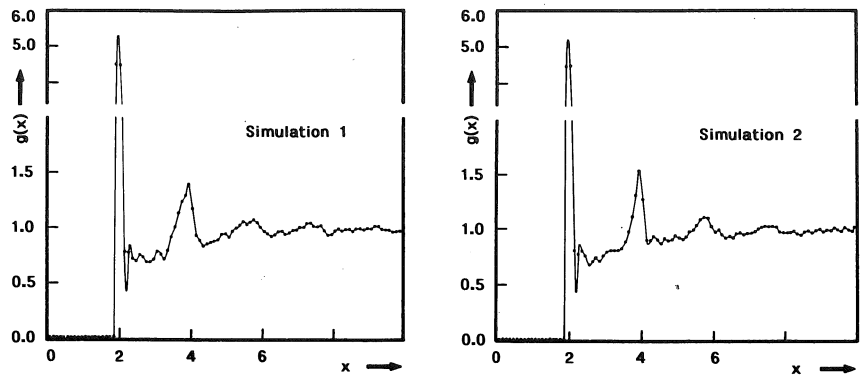

Fig. 7 : Pair correlation function for simulations S1 and S2.

a) **Pair correlation function of particle centers**

The pair correlation function is a basic tool for the study of local fluctuations of density in a random system of points. This second-order property (Diggh 1983; König 1986) may be defined by considering a spherical shell of radius x and thickness dx, centered on one of the points of the system. If the expected number of points whithin that shell is designated as $R_V(x) \cdot dx$, the function $R_V(x)$ is the radial distribution function and the pair correlation function, $g_v(x)$, is given by:

$$g_v(x) = R_v(x) / 4\pi x^2 N_v$$

where N_V is the expected number of points per unit volume.

So, deviations of $g_v(x)$ from unity can be regarded as relative variations of local values of N_V.

Practically, to get $g_v(x)$ from our simulations, three kinds of 3D fields must be defined :

- the complete field which coincides with the whole simulation box,
- the restricted field which excludes all the regions subjected to wall effects,
- the central field which is the domain where any reference particle center must lie in order that its surroundings are accessible to measurement without bias due to boundary effect.

In our case, taking $x \leqslant 10$, the edges of the three cubic fields had respective lengths of 40, 32 and 12 units. The number of

particle centers available for the experimental determination of $g_v(x)$ was roughly 4600 in the restricted field and 240 in the central field.

The results are presented on figure 7. There appears to be no significant differences between the curves obtained for the two simulations. In both cases,clear peaks are observed at abscissae of about 2, 4 and 5.5 units. After the last peak in the region of abscissa 7.5, the curve merges into a continuum indicating the absence of long-range order. These features roughly agree with the data of Matheson (1974) for a simulated packing with $V_v \simeq 0.61$. No bump being discernible at the abscissa $2\sqrt{3}$, the second peak fails to exhibit the splitting classically considered to be a characteristic feature of dense random packings (Finney 1983). This is probably due to the joint effects of the relatively low density of our structure ($V_v \simeq 0.59$ instead of 0.64 for dense random packings) and to its anisotropy. Indeed, it is clear, from the above definitions of R_v and g_v , that these two functions loose some part of their significance when the structure is anisotropic.

b) **Local distribution of contacts**

It is well known that the mean coordination number, N_c , (mean number of contacts around a particle)is usually close to six. This has been established both in experimental (Bernal and Mason 1960, Finney 1970) and computer-simulated (Visscher and Bolsterli 1972, Bennett 1972, Tory et al. 1973, Matheson 1974, Powell 1980, Jodrey and Tory 1985) random packings. A theoretical justification for that result has been given by Bennett (1972).

Table 3 : Distributions of numbers of contacts for the two simulations.

Number of contacts	Simulation S1 Percentage	Simulation S2 Percentage
3	0.11	0.02
4	4.39	3.10
5	24.77	25.58
6	42.76	45.94
7	23.10	22.08
8	4.54	3.19
9	0.33	0.09

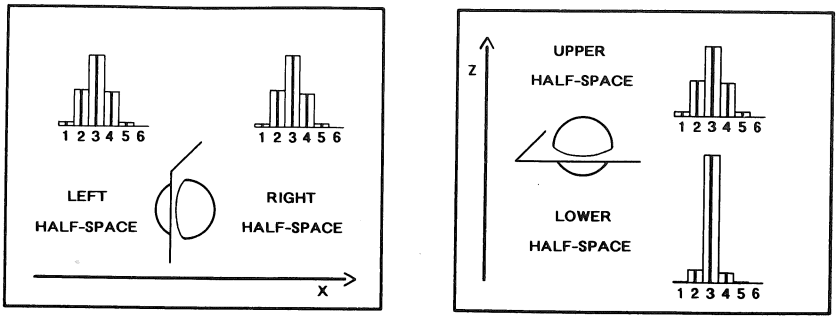

Fig. 8 : Dissymmetry in the particle surroundings with respect to an horizontal plane (simulation S2).

Table 4 : Frequency of contacts around each particle in different half-spaces.

SIMULATION S1

Number of contacts	Percentage left half-space	Percentage right half-space	Percentage upper half-space	Percentage lower half-space
1	3.22	3.26	3.77	0.48
2	24.40	24.32	24.37	10.07
3	45.43	45.46	44.96	79.13
4	23.80	23.29	22.47	9.76
5	2.91	3.42	4.26	0.55
6	0.24	0.15	0.18	0.00

SIMULATION S2

Number of contacts	Percentage left half-space	Percentage right half-space	Percentage upper half-space	Percentage lower half-space
1	2.65	2.35	2.78	0.20
2	24.08	24.71	24.67	8.49
3	47.74	47.74	47.25	83.85
4	23.17	22.84	22.58	7.01
5	2.34	2.21	2.61	0.46
6	0.02	0.15	0.11	0.00

In our case, the mean value obtained for N_c, after elimination of the wall effect, is 6 for both simulations. However,this is only a mean value and the number of contacts around each sphere is not constant, as shown in table 3.

It may be recalled that the simulation process implies that each ball is supported by three neighbours and as a consequence of the stability criterion, these three neighbours will almost always be located below the horizontal plane passing through the ball center. Since the distribution of number of contacts extends over a range from 3 to 9, the dispersion must be due to the balls lying above that horizontal plane (P). Henceforth, the statistical repartition of contacts should not be the same above and below plane (P). On the other hand, there is no reason for the existence of a dissymmetry with respect to a vertical plane. From the data of table 4 represented on figure 8, one can see that the above conjecture is well verified : a very narrow peak is observed for the histogram corresponding to the lower half-space.

This local dissymmetry due to gravity is to be compared to the anisotropic behaviour of electrical conductivity of a real particle stacking of spheres under the influence of pressure (Troadec and Bideau 1981).

CONCLUSION

Although particle stackings of relatively low compacity such as the one studied in this paper probably provide a poor model for the molecular sructure of liquids, they are likely to be useful for understanding the structure and properties of granular materials (Finney 1983).

Stereological methods are presently available in the literature for the determination of both the first-order and the second-order properties of these materials (Hanisch and Stoyan 1981 ; Hanisch et al. 1985 ; Stoyan et al. 1990). A commonplace assumption, in such studies, consists of considering the structure as statistically uniform and isotropic. Though this is not a prerequisite for the determination of properties such as the radial distribution and the pair correlation functions, it greatly simplifies the sampling procedure and the interpretation of the results. From the specific example examined in this paper, however, it appears that even in the simplest structures (monosized spherical particles), it may be quite unsafe to take as granted the fulfilment of the unifor-

mity and isotropy conditions. Both the wall effects and a too small sample size can induce non uniformity. Gravity can result in anisotropy and local dissymmetry.

In most practical situations, departures from uniformity will be easily circumvented, by working on samples of sufficient size and restricting properly the field of analysis. On the other hand, gravity effects, which bear, not on the measurements, but on the structure itself, can obviously not be eliminated.

ACKNOWLEDGMENTS

The authors express their gratitude to William M. Visscher for the original version of the program simulating a random packing of spheres. The support of CNRS and CIES for one of the authors (Bhanu Prasad P.) is gratefully acknowledged.

REFERENCES

Benenati RF, Brosilow CB. Void fraction distribution in beds of spheres. A. I. ch. E. J. 1962 ; 8 : 359-361.

Bennett CH. Serially deposited amorphous aggregates of hard spheres. J. Appl. Phys. 1972 ; 43 : 2727-2734.

Bernal JD, Mason J. Coordination of randomly packed spheres. Nature 1960 ; 188 : 910-911.

Bernal JD. The structure of liquids. Proc. Roy. Soc. London 1964 ; A280 : 200-322.

Diggle PJ. Statistical analysis of spatial point patterns. London : Academic Press ; 1983.

Finney JL. Random packings and the structure of simple liquids. The geometry of random close packing. Proc. Roy. Soc. London 1970 ; 319A : 479-493.

Finney JL. Structure and properties of granular materials : guidelines from modelling studies of liquids and amorphous solids. In: Shahinpoor M, Ed. Advances in the mechanics and the flow of granular materials. Clausthal-Zellerfeld: Trans Tech. Publications, 1983 : 19-39.

Gotoh K, Jodrey WS, Tory EM. Variation in the local packing density near the wall of a randomly packed bed of equal spheres. Powder Technol. 1978 a ; 20 : 257-260.

Gotoh K, Jodrey WS, Tory EM. A random packing structure of equal spheres - Statistical geometrical analysis of tetrahedral configurations. Powder Technol. 1978b ; 20: 233-242.

Hanisch KH, Stoyan D. Stereological estimation of the radial distribution function of centres of spheres. J. Microsc. 1981 ; 122 : 131-141.

Hanisch KH, König D, Stoyan D. The pair correlation function for point and fibre systems and its stereological determination by planar sections. J. Microsc. 1985 ; 140: 361-370.

Jodrey WS, Tory EM. Computer simulation of close random packing of equal spheres. Phys. Rev. A 1985 ; 32 : 2347-2351.

König D. Probabilistic characterization of the inner order of random structures. Proceedings of the first international symposium for science on form. Tokyo : KTK ; 1986.

Matheson AJ. Computation of a random packing of hard spheres. J. Phys. C : Solid St. Phys. 1974 ; 7 : 2569-2576.

Powell MJ. Computer-simulated random packing of spheres. Powder Technol. 1980 ; 25 : 45-52.

Stoyan D, von Wolfersdorf L, Ohser J. Stereological problems for spherical particles - second order theory. Math. Nachr. 1990 ; 146 : 33-46.

Tory EM, Cochrane NA, Waddell SR. Anisotropy in simulated random packing of equal spheres. Nature 1968 ; 220 : 1023-1024.

Tory EM, Church BH, Tam MK, Ratner M. Simulated random packing of equal spheres. Can. J. Chem. Eng. 1973 ; 51 : 484-493.

Troadec JP, Bideau D. Propriétés de transport d'un mélange de sphères conductrices et isolantes : étude des effets de percolation. J. Physique 1981 ; 42 : 113-123.

Visscher WM, Bolsterli M. Random packing of equal and unequal spheres in two and three dimensions. Nature 1972 ; 239 : 504-507.