Computer simulation of random packing of unequal particles

D. He, N. N. Ekere, and L. Cai
Department of Aeronautical, Mechanical, and Manufacturing Engineering, The University of Salford, Salford M5 4WT, United Kingdom
(Received 10 May 1999)

A Monte Carlo simulation model for the random packing of unequal spherical particles is presented in this paper. With this model, the particle radii obeying a given distribution are generated and randomly placed within a cubic packing domain with high packing density and many overlaps. Then a relaxation iteration is applied to reduce or eliminate the overlaps, while the packing space is gradually expanded. The simulation is completed once the mean overlap value falls below a preset value. To simulate the random close packing, a ''vibration'' process is applied after the relaxation iteration. For log-normal distributed particles, the effect of particle size standard deviation, and for bidisperse particles, the effects of particle size ratio and the volume fraction of large particles on packing density and on coordination number are investigated. Simulation results show good agreement with that obtained by experiments and by other simulations. The randomness, homogeneity, and isotropy, which have not been evaluated before for packing of distributed particles, are also examined using statistical measures. [S1063-651X(99)02312-0]

PACS number(s): 81.05.Rm, 05.45.−a, 89.80.+h

I. INTRODUCTION

The random packings of hard spherical particles have long been of interest because they can serve as useful models for many physical and engineering systems, such as the microstructures of simple liquids [1], concentrated suspensions [2], amorphous materials [3], ceramic components prepared by compaction of powders [4], and porous materials. Many experimental [5–9] and analytical [10–14] investigations have been reported. The packing structure, which influences the mechanical, electrical, and thermal properties of such materials, can be characterized by several parameters, such as the packing density, the coordination number, and the radial distribution function. It is an extremely labourious task to obtain these parameters by experiment, while theoretical analysis cannot provide details of the structural data, such as the distribution of coordination numbers and the radial distribution function.

Computer simulation has been proven as a powerful measure in the study of particle random packing and many models have been developed. The simulation models can be classified into two categories, the sequential generation models and the collective rearrangement models. The sequential generation models can be further divided into two types. In the first case, one particle is dropped vertically each time onto the surface of an existing particle cluster which grows upwards [15–18]. In the second case, a particle is centripetally placed on the surface of a center cluster which grows outwards [19–22]. With the collective rearrangement models [23–29], the initial positions of particles are randomly generated within the packing domain with high initial packing density and many overlaps. Then a relaxation process is repeated to separate the overlapping particles. As the overlap approaches a stable value but is still higher than a specified tolerance, the particle sizes are reduced. The relaxation and particle size reduction processes are repeatedly applied and the simulation is completed when the overlap value drops below the given tolerance.

Most of the models developed to date have only been applied to particles of equal size. In practice, however, the particle sizes are most likely to be distributed, such as metal and ceramic powders, paint pigment, and the atoms of a simple homogeneous liquid. For industrial applications, several advantages can be gained by the use of powders with polydisperse particles, such as obtaining higher density components of powders [22] and increasing the fluidity of concentrated suspensions [30–32]. The above facts underline the significance of studying the effect of particle size distribution on the random packing structure. Clarke and Wiley [26] and Jullien and Meakin [18] simulated the random packing of bidisperse systems by collective rearrangement model and by sequential deposition model, respectively. The particle sizes of natural materials are often represented by log-normal distribution [33,34]. Significant contributions to the simulation of random packing of particles obeying log-normal distribution were made by Konakava and Ishizaki [22], Nolan and Kavanagh [28], and by Yang *et al.* [29]. Konakava and Ishizaki moved particles toward the packing center one by one. The packing density obtained by this model decreases with the increase in particle number. The models developed by Nolan and Kavanagh and by Yang *et al.* both belong to the collective rearrangement category. Nolan and Kavanagh presented a repulsive force to relocate the overlapped particles and this repulsive force allows smaller particles to travel longer distance. The sample size of this model was small which might induce significant variation in simulation results. Yang *et al.* only moved the overlapped particles a small distance along the vector sum of overlaps with surrounding particles each time, and did not allow the overlap free particles to move. If a particle was locked in by its surrounding particles, it was allowed to shrink by 10% in diameter. This model was capable of simulating a large number of particles. However, due to the individual shrinkage of particles, the final size distribution was inconsistent with the initial distribution.

In this paper, we present a Monte Carlo simulation model which is applicable to the random packing of spherical particles obeying any specified distribution. This model can be

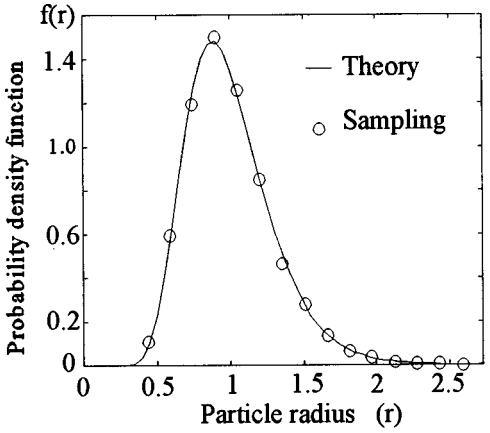

FIG. 1. Comparison of theoretical log-normal distribution with numerical sampling.

classified into the collective rearrangement category. In the next section, we describe the algorithm used for this simulation model. In Sec. III, we present the simulation results of bidisperse particles and log-normal distributed particles, test the randomness, homogeneity, and isotropy of the packing, and discuss the effect of particle size distribution on the packing properties. The final section presents our conclusions.

II. THE ALGORITHM

For random loose packing, the algorithm is composed of three main procedures: particle size and initial position generation, overlap relaxation, and packing space expansion. For random close packing a ''vibration'' process is applied within the relaxation procedure. For convenience, the mean radius of log-normal distributed particles is normalized to be 1.0, and the radii of small particles of bidisperse mixture equal to 1.0.

The particle sizes are generated by the following methods. For bidisperse particles, at a given fraction of large particles P_L, we examine the values of a set of random numbers $(p_1,p_2,\ldots,p_n)$ which obey uniform distribution over (0, 1). If $p_i>P_L$, then $r_i=1.0$, otherwise $r_i=\lambda$. λ is the ratio of radii of large particle to small particle and n is the total number of particles.

For particles obeying log-normal distribution, the probability density function of particle radius r is given by

$$f(r)=\frac{1}{\sqrt{2\pi}\sigma r}e^{-(\ln r-\ln r_0)^2/2\sigma^2}, \tag{1}$$

where $\ln r_0$ and σ are the mean and the standard deviation of $\ln r$. If the mean of particle radii is normalized to be 1.0, then $\ln r_0$ approaches zero. Again, we first generate a set of random numbers $(p_1,p_2,\ldots,p_n)$ obeying uniform distribution over (0, 1). Then we obtain a set of random numbers $(r_1,r_2,\ldots,r_n)$ obeying log-normal distribution by two conversions as $t_i=\sigma(-\ln p_i)^{1/2}\sin(2\pi p_{i+1})$, and $r_i=\exp(t_i)$. Figure 1 is the comparison of theoretical density function with that of ten thousand samples generated by the above method.

The initial position (x_i,y_i,z_i) of a particle is generated within a cubic domain and (x_i,y_i,z_i) obey uniform distribution over $(0+r_i,L_0-r_i)$. L_0 is the initial size of the domain and is decided by the following equation:

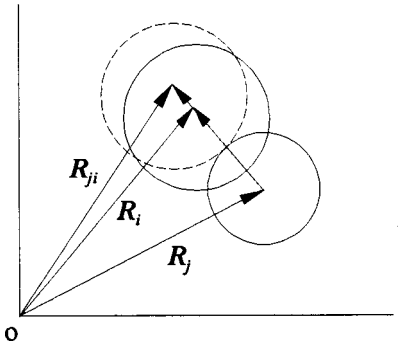

FIG. 2. Separation of particle i from particle j.

$$L_0=\left[\frac{1}{\Phi_0}\sum_{i=1}^{n}\frac{4}{3}\pi r_i^3\right]^{1/3}, \tag{2}$$

where ϕ_0 is initial packing density which we take unphysically higher than the attainable close random packing density thus causing many overlaps. In this study 0.75 is taken as the value of ϕ_0 for random loose packing and 0.86 for random close packing.

The overlap rate between two particle is defined as $(r_i+r_j-d_{ij})/(r_i+r_j)$, where d_{ij} is the center to center distance and $d_{ij}<(r_i+r_j)$. In the rearrangement procedure, the following relaxation technique is applied to relocate particles to reduce or eliminate overlaps. For each particle, particle i, for example, a search of particles which overlap particle i is conducted first. Then from each of the overlapping particles, $j(x_jy_jz_j)$, for example, as shown in Fig. 2, a new position can be calculated by the following equation:

$$\mathbf{R}_{ji}=\mathbf{R}_j+(\mathbf{R}_i-\mathbf{R}_j)\frac{r_i+r_j}{d_{ij}}, \tag{3}$$

where $\mathbf{R}_i$ and $\mathbf{R}_j$ are vectors of the centers of particle i and j, respectively. If particle i is overlapped by n_i particles, then by Eq. (3) n_i positions can be obtained, and the new position of particle i is given by

$$\mathbf{R}_i^l=\frac{1}{n_i}\sum_{j=1}^{n_i}\mathbf{R}_{ij}. \tag{4}$$

Figure 3 shows the two-dimensional case of the relocation of particle i by one step. This relaxation is applied to every overlapped particle. If a particle neither overlaps nor contacts others, it is moved to contact its nearest neighbor. Periodical boundary condition is applied to particles near the boundary. After one iteration each particle has been relocated once. By repeating the iteration the mean of the overlap rates gradually decreases. To avoid any bias the sequence of particle rearrangements is randomized after each iteration.

After a given number of iterations, packing space is expanded to $L=a_lL_0$. a_l is the expanding factor which is dependent on the present mean overlap rate. By repeating the relaxation and expansion procedures, the overlap rate eventually drops below a preset tolerance and the simulation is completed. In this study, 2.0×10^{-4} is accepted as the toler-

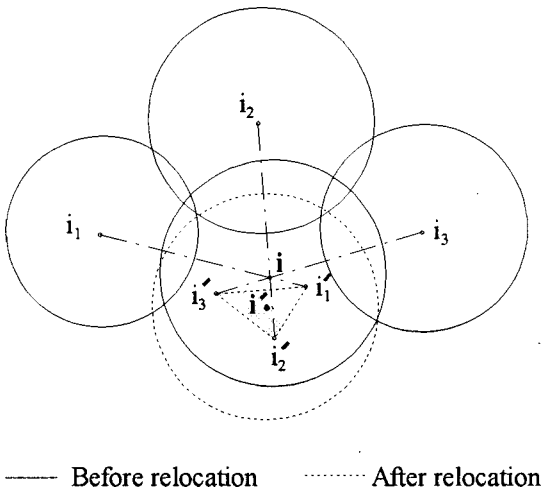

FIG. 3. Relocation of particle *i*.

ance which ensures that the relative error of the final packing density will be less than 10^{-3}.

To simulate the random close packing obtained by vibration, we increase the number of rearrangement iterations. Meanwhile, after each iteration, we apply a ''vibration'' process to randomly disturb the positions of those particles which have a coordination number less than 4 since the particles that form bridges have fewer contacts with others.

III. RESULTS AND DISCUSSION

The above simulation algorithm was applied to simulate the random loose and random close packings of log-normal distributed and bidisperse particles. We observed that the packing density increases with the increase in the number of particles. This agrees with Scott's experimental result [5] but is contrary to the simulation result of Yang *et al.* [29]. When the particle number exceeds 10 000 its influence on the packing density becomes insignificant. In each simulation, therefore, we employed 10 000 particles. With 10 000 particles, it was also shown that the simulation result is very stable. For example, with equal particles, the simulation was run several times and results showed the maximum difference in packing density to be less than 0.001, and the maximum difference in coordination number to be less than 0.05. Figure 4 shows the three-dimensional and two-dimensional cross section views of the simulated packings.

A. Random packing of particles obeying log-normal distribution

With particles obeying log-normal distribution, Figs. 5 and 6 show the effect of the standard deviation of particle radii σ on the random close packing density ϕ_c and random loose packing density ϕ_1. Results of other investigations are included for comparison. With equal particles, a value of 0.6267 to 0.6268 was obtained for ϕ_c which agrees well with the experimental value of 0.625 [7,31] but is about 1–2 % lower than the commonly accepted value of 0.633 to 0.640 [5,23]. However, it should be noted that with 6000 to 20 000 balls, the packing density obtained by Scott [5] was about 0.62 and 0.6334 was the extrapolated value. A value 0.5935 to 0.5938 was obtained for ϕ_1 which lies between the commonly accepted values of 0.59 and 0.60. For random close packing, the trend of the effect of σ on ϕ_c of this study is quite similar to that obtained by Nolan and Kavanagh [28] and by Yang *et al.* [29]. As σ is smaller than 0.15 its influ-

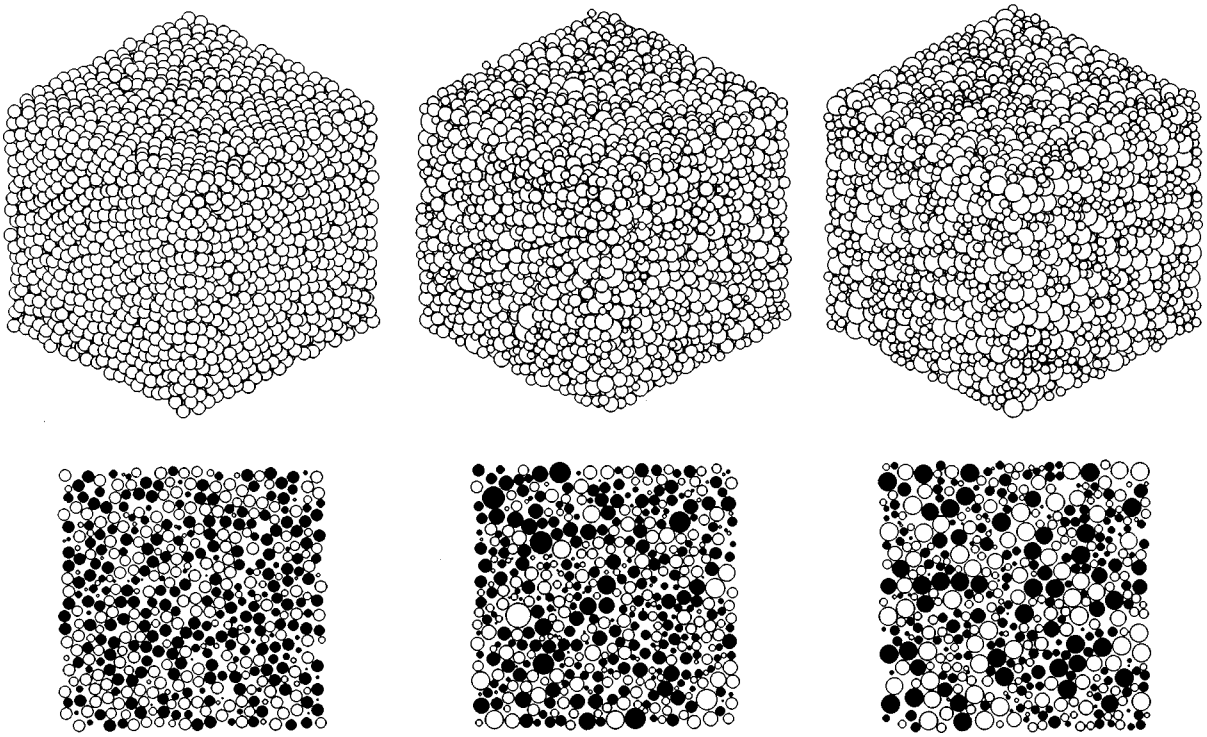

FIG. 4. Three-dimensional and two-dimensional cross section views of particle packings, equal particles (left), log-normal distribution with $\sigma=0.25$ (middle), bidisperse particles with $\lambda=2.0$ and $F_l=0.70$ (right). The centers of particles represented by the dark circles are in front of the cross sections.

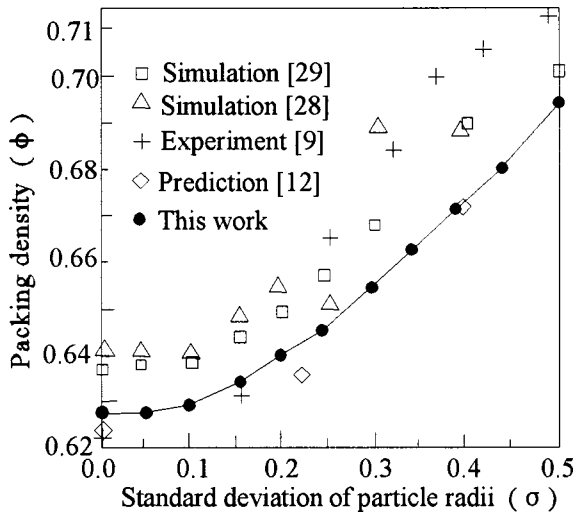

FIG. 5. Random close packing density as function of particle size standard deviation.

ence on ϕ_c is insignificant; when σ is greater than 0.15, ϕ_c gradually increases with σ. It can be estimated that, for $\sigma = 0.1$, the radii of more than 95% particles are expected to fall between (0.8, 1.2). For such a narrow distribution, the ratio of the radii of any two contact particles is close to 1.0. Therefore, a small standard deviation $\sigma < 0.1$ will not cause significant increase in the packing density. For random loose packing, Fig. 6 shows that, for small standard deviation, the packing density of this study is much higher than that obtained by Nolan and Kavanagh [28] and by Yang *et al.* [29]. The trend of the effect of σ on ϕ_1 of this study is similar to that of random close packing. While the results obtained by Yang *et al.* [29] show approximately linear relationship between ϕ_1 and σ.

It is the feature of this simulation algorithm that the exact contacts between particles are very few. Most neighbors are either finely overlapped or separated. If the gap between two particles is smaller than 0.2% of the sum of their radii, we consider that they contact each other. Figure 7 shows the influence of σ on the mean coordination numbers of random close packing (RCP) and random loose packing (RLP). With

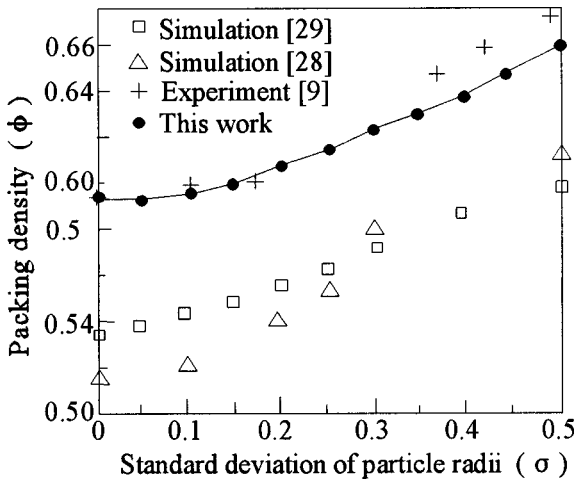

FIG. 6. Random loose packing density as function of particle size standard deviation.

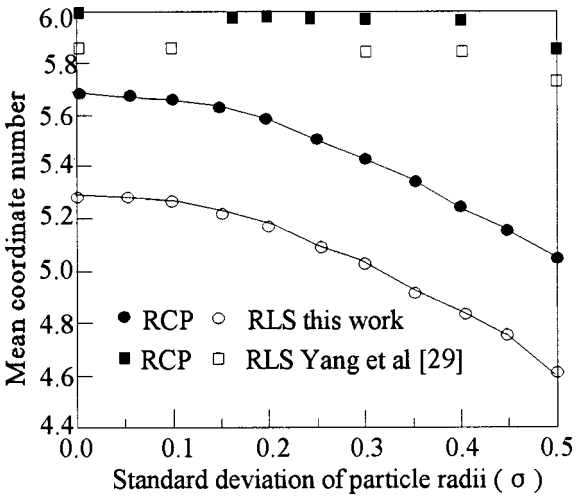

FIG. 7. Mean coordination number as function of particle size standard deviation.

equal particles, the mean coordination number of random close packing of this study is around 5.68 which is about 5% lower than the commonly accepted value of 6.0. The higher value was obtained in the experiments [6,7] and in other simulations [15–18,29], where the packing process was dominated by gravity, i.e., one particle must be supported at least by three others in vertical direction. We did not apply this rule in this study. It was shown that the random packing structure governed by gravity is anisotropic [15,17]. Figure 7 shows that, for random loose packing, the mean coordination number is about 0.4 lower than that of random close packing. As $\sigma > 0.15$, the mean coordination numbers of both close packing and loose packing gradually decrease with the increase in σ. We observed that, at higher value of σ, the coordination number spreads over a wider range. Figure 8 is the comparison of the distributions of coordination number with different values of σ.

B. Random packing of bidisperse particles

With bidisperse particles, Fig. 9 shows the influences of particle size ratio λ and the volume fraction of large particles F_l on the packing density. F_l is defined by $F_l = N_l\lambda^3/(N_s + N_l\lambda^3)$, where N_l is the number of large particles and N_s is the number of small particles. Figure 9 shows that the packing density increases with F_l and approaches a maximum

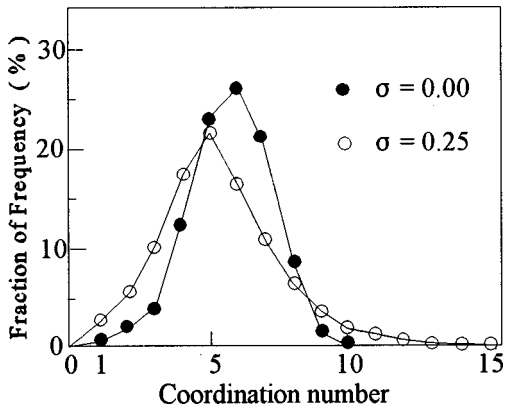

FIG. 8. Comparison of coordination number distributions.

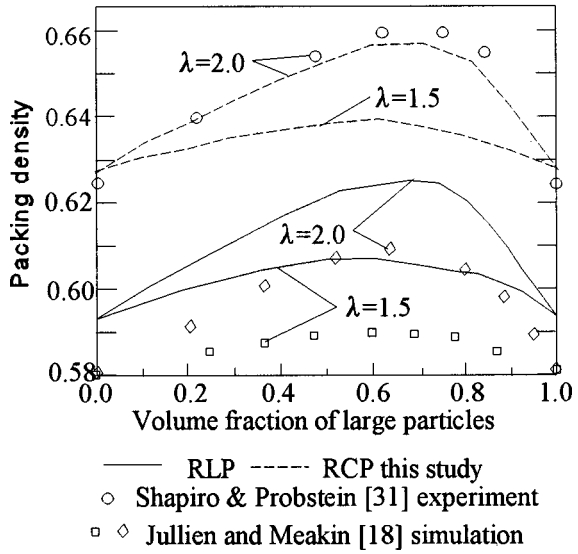

FIG. 9. Random packing density of bidisperse particles.

before starting to decrease with further increases in F_l. The value of F_l which corresponds to the maximum packing density varies with λ. For example, with $\lambda=1.5$ the packing density is maximized at $F_l=0.6$, and with $\lambda=2.0$ the packing density is maximized at $F_l=0.66$. Simulation results also show that, when λ is smaller than 1.5, both F_l and λ have no significant influence on the packing density; where λ is greater than 1.5, the packing density significantly increases with λ. The above observations are in good agreement with that observed by experiment [31] and by other simulation [18].

In a random packing of bidisperse particles, it is expected that the coordination number of a large particle will be higher than that of a small particle. For random close packing with $\lambda=2.0$ and $F_l=0.71$, Fig. 10 shows the distributions of coordination numbers of large particles C_l and small particles C_s, respectively. The range of C_l is from 4 to 15 with a mean of 9.5, while the range of C_s is from 0 to 8 with a mean of 4.1.

C. The randomness, homogeneity and isotropy of the packing

For equal particles, the randomness of the packing can be tested by several methods, such as the radial distribution

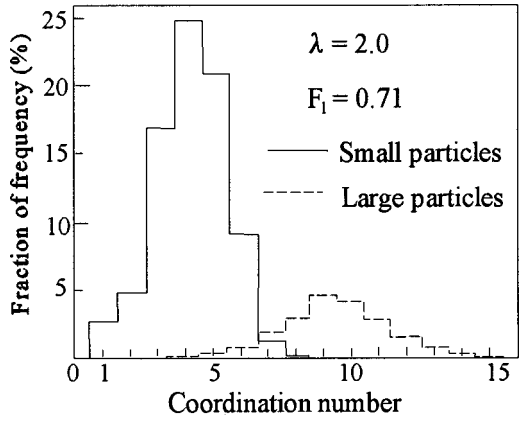

FIG. 10. Coordination number distributions of large and small particles.

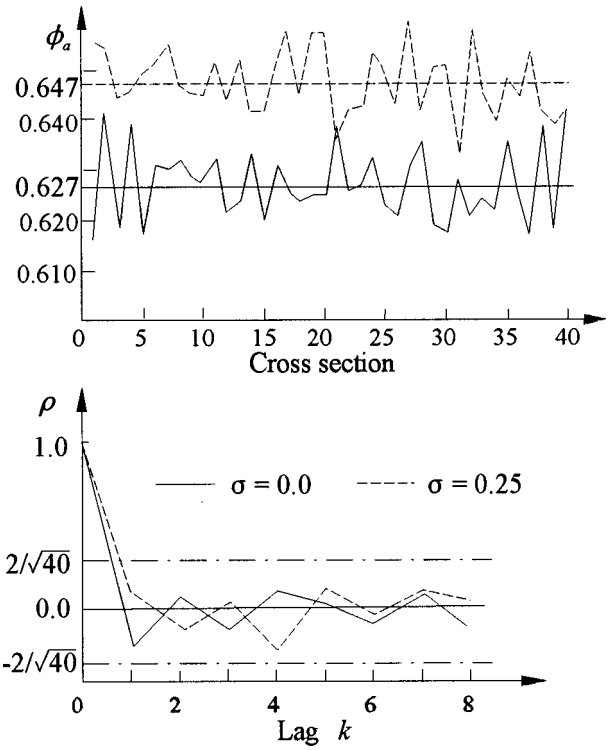

FIG. 11. Area densities on cross sections (top) and autocorrelation coefficients (bottom).

function [20,23] and the Voronoi-cell statistics [26]. However, these methods are not applicable to the packing of polydisperse particles. The time series analysis technique can be used to examine the randomness of the packing. Using this technique, for m observations, $S_1,S_2,\ldots,S_m$, on a discrete time series, the autocorrelation coefficient is defined as

$$P_k=\frac{\sum_{j=1}^{m-k}(s_j-\bar{s})(s_{j+k}-\bar{s})}{\sum_{j=1}^{m}(s_j-\bar{s})^2}, \tag{5}$$

where $\bar{s}$ is the mean of the observation and k is called the lag which should be smaller than $m/4$. If a time series is completely random, then ρ_k obeys $N(0,1/m)$ normal distribution, and over 95% of ρ_k lie between $\pm 2/\sqrt{m}$ [35]. To employ this

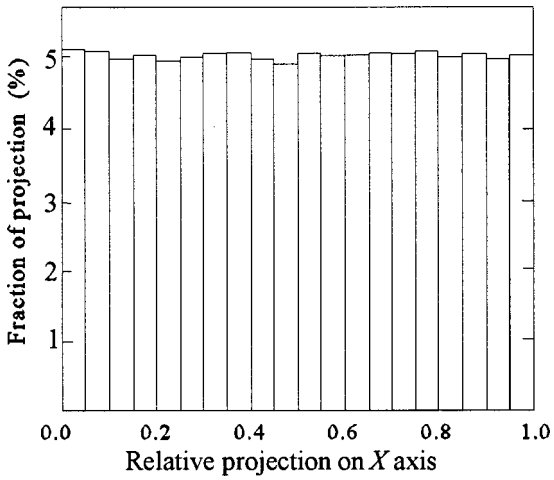

FIG. 12. Distribution of projections on the X axis.

TABLE I. Distribution of particles among subregions.

	Top			Middle			Bottom		
Equal	280	280	290	282	282	285	289	285	272
particles	283	283	284	287	286	287	280	283	294
	283	290	277	280	289	290	290	284	275
Distributed	289	275	267	290	271	279	269	281	281
particles	302	279	268	288	294	269	287	282	282
$\sigma=0.15$	276	276	271	274	290	279	270	279	274

technique, we cut each packing into forty equally separated cross sections and took the area densities on the cross sections as the observations; then calculated the first eight values of ρ_k. Figure 11 is the plot of the area densities on the cross sections together with the plot of ρ_k against k. It can be seen that all the values of ρ_k lie between $\pm 2/\sqrt{m}$ $(m=40)$ and this suggests that the packing is completely random. In contrast, for an ordering packing, such as cubic packing, both the area density on the cross section and the autocorrelation coefficient will be periodical functions of the position of cross section.

To examine the homogeneity, without consideration of the particles at boundaries, we divided each packing into 27 equal cubic subregions and counted the centers of particles in each subregion. The result is listed in Table I. Then the chi-square goodness-of-fit test was applied to examine the hypothesis that the particles are uniformly distributed among the subregions. At 0.05 level the critical value is $\chi^2_{0.05,26}=38.885$. The test statistic for equal particles is $\chi^2=2.611$, and for log-normal distributed particles $\chi^2=7.764$, both being much smaller than the critical value. Therefore, there is no evidence to reject the hypothesis that the particles are uniformly distributed in the packing. This implies that the packing is homogeneous. For particles with high standard deviation, the packing density in each subregion should be used to conduct the hypothesis test. This is necessary since the appearance of a very large particle in a subregion can cause a significant reduction in the number of particles in it. Packing obtained by the sequential central growing models [21] is radially inhomogeneous.

Tory *et al.* [17] proved that, for equal particles, if the projection of two contact particles on any axis obeys uniform distribution over $(-2r,2r)$, then the packing is isotropic. Extending this conclusion to unequal particles, it can be proved that if the relative projections of two contact particles $\Delta x_{ij}=|x_i-x_j|/(r_i+r_j)$, $\Delta y_{ij}=|y_i-y_j|/(r_i+r_j)$ and $\Delta z_{ij}=|z_i-z_j|/(r_i+r_j)$, obey uniform distribution over (0, 1), then the packing is isotropic. The mean and the variance of the projections are 0.5 and 0.0833, respectively. In this study, for equal particles, the mean projections on *X*, *Y*, and *Z* axes were found to be 0.4986, 0.5012, and 0.4999, and the variances 0.0832, 0.0834, and 0.0828, respectively. For log-normal distributed particles with $\sigma=0.20$, the mean projections are 0.4998, 0.5005, and 0.5002, and the variances are 0.0833, 0.0828, and 0.0831, respectively. Figure 12 shows an example of the distribution of the projection on the *X* axis. Hypothesis test suggests that the projections obey uniform distribution. Therefore, the random packing obtained by this algorithm is isotropic. It was shown [16,17] that the random packing obtained by sequential and gravitational addition model is anisotropic.

IV. CONCLUSION

A Monte Carlo simulation model for the random packing of polydisperse spherical particles was developed in this work. The random loose packing and random close packing of log-normal distributed particles and bidisperse particles were studied. The simulation results are in good agreement with both experimental and other simulation results. With particles obeying log-normal distribution, the packing density significantly increases with the particle size standard deviation when it is greater than 0.15, while the coordination number decreases with the increase in the standard deviation. With bidisperse particles, the packing density increases with the particle size ratio. At a given size ratio, the maximum packing density corresponds to a volume fraction of large particles. Examination of the autocorrelation coefficient of the area densities on the cross sections indicated that the packing is completely random. Statistical hypothesis tests also showed that the packing is homogeneous and isotropic. The random packings obtained by this model can represent the structures of amorphous metals, simple liquids, and isostatically compressed components of ceramic and metal powders.

ACKNOWLEDGMENTS

This research was funded by the Engineering and Physical Sciences Research Council of the United Kingdom under Grant No. GR/L20597.

[1] J. D. Bernal, Nature (London) **185**, 68 (1960).
[2] S. Torquato and F. Lado, Phys. Rev. B **33**, 6428 (1986).
[3] A. Angell, J. H. R. Clarke, and L. V. Woodcock, Adv. Chem. Phys. **48**, 397 (1981).
[4] Y. Hamad, K. Ishizaki, and J. Briceno, J. Ceram. Soc. Jpn. **98**, 343 (1990).
[5] G. D. Scott, Nature (London) **188**, 908 (1960).
[6] G. D. Scott and D. M. Kilgour, J. Appl. Phys. **2**, 863 (1969).
[7] J. D. Bernal and J. Mason, Nature (London) **188**, 910 (1960).
[8] G. Y. Onoda and E. G. Liniger, Phys. Rev. Lett. **64**, 2727 (1990).
[9] H. Y. Sohn and C. Moreland, Can. J. Chem. Eng. **46**, 162 (1968).
[10] K. Gotoh and J. L. Finney, Nature (London) **252**, 202 (1974).
[11] J. G. Berryman, Phys. Rev. A **27**, 1053 (1983).
[12] A. B. Yu and N. Standish, Powder Technol. **55**, 171 (1988).
[13] M. Alonso, E. Sainz, F. A. Lopez, and K. Shinohara, Chem. Eng. Sci. **50**, 1983 (1995).
[14] Y. Roualt and S. Assouline, Powder Technol. **96**, 33 (1998).
[15] E. M. Tory, N. A. Cochrane, and S. R. Waddell, Nature (London) **220**, 1023 (1968).
[16] W. M. Visscher and M. Bolsterli, Nature (London) **239**, 504

(1972).
[17] E. M. Tory, B. H. Church, M. K. Tam, and M. Ratner, Can. J. Chem. Eng. **51**, 484 (1973).
[18] R. Jullien and P. Meakin, Europhys. Lett. **6**, 629 (1988).
[19] C. H. Bennett, J. Appl. Phys. **43**, 2727 (1972).
[20] A. J. Matheson, J. Phys. C **7**, 2569 (1974).
[21] G. Q. Lu and X. Shi, J. Mater. Sci. Lett. **13**, 1709 (1994).
[22] Y. Konakawa and K. Ishizaki, Powder Technol. **63**, 241 (1990).
[23] J. L. Finney, Mater. Sci. Eng. **23**, 199 (1976).
[24] W. S. Jodrey and E. M. Tory, Powder Technol. **30**, 111 (1981).
[25] W. S. Jodrey and E. M. Tory, Phys. Rev. A **32**, 2347 (1985).
[26] A. S. Clarke and J. D. Wiley, Phys. Rev. B **35**, 7350 (1987).
[27] G. T. Nolan and P. E. Kavanagh, Powder Technol. **72**, 149 (1992).
[28] G. T. Nolan and P. E. Kavanagh, Powder Technol. **76**, 309 (1993).
[29] A. Yang, G. T. Miller, and L. D. Turcoliver, Phys. Rev. E **53**, 1516 (1996).
[30] M. Z. Sengun and R. F. Probstein, Rheol. Acta **28**, 394 (1989).
[31] A. P. Shapiro and R. F. Probstein, Phys. Rev. Lett. **68**, 1422 (1992).
[32] R. D. Sudduth, J. Appl. Polym. Sci. **48**, 37 (1993).
[33] W. A. Gray, *The Packing of Solid Particles* (Chapman and Hall, London,1968).
[34] H. Rumpf, *Particle Technology* (Chapman and Hall, London,1990).
[35] C. Chatfield, *The Analysis of Time Series* (Chapman and Hall, London, 1989).