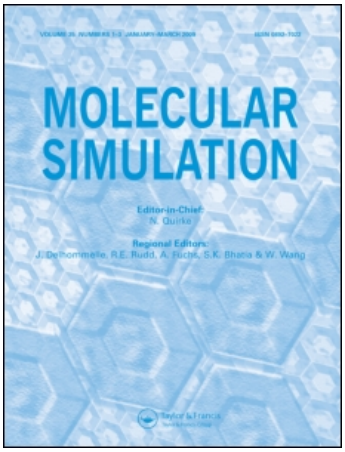

Molecular Simulation
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713644482

The Force-Biased Algorithm for the Irregular Close Packing of Equal Hard Spheres

J. Mościński[ab]; M. Bargieł[a]; Z. A. Rycerz[b]; P. W. M. Jacobs[b]
[a] Institute of Computer Science, Kraków, Poland [b] University of Western Ontario, London, Ontario, Canada

To cite this Article Mościński, J. , Bargieł, M. , Rycerz, Z. A. and Jacobs, P. W. M.(1989) 'The Force-Biased Algorithm for the Irregular Close Packing of Equal Hard Spheres', Molecular Simulation, 3: 4, 201 — 212
To link to this Article: DOI: 10.1080/08927028908031373
URL: http://dx.doi.org/10.1080/08927028908031373

Molecular Simulation, 1989, Vol. 3, pp. 201–212
Reprints available directly from the publisher
Photocopying permitted by license only

THE FORCE-BIASED ALGORITHM FOR THE IRREGULAR CLOSE PACKING OF EQUAL HARD SPHERES

J. MOŚCIŃSKI*†, M. BARGIEŁ*, Z.A. RYCERZ† and P.W.M. JACOBS†

**Institute of Computer Science, AGH, al. Mickiewicza 30, 30–059 Kraków, Poland*
† University of Western Ontario, London, Ontario, N6A 5B7, Canada

(*Received November, 1988*)

We present a "force-biased" algorithm for generating the irregular close packing of hard spheres. The algorithm is partly based on Jodrey and Tory's ideas [9] and incorporates methods from Molecular Dynamics. Packings generated by means of the two algorithms are consistent up to final packing fraction of 0.65, which seems to be the limit density of Jodrey and Tory's method. Significantly higher densities (up to 0.71) can be achieved for small numbers of spheres by the force-biased algorithm. However the shape of the radial and angle distribution functions implies that a partial short-range ordering occurs in packings of those densities.

KEY WORDS: Irregular close packing, computer simulation, molecular dynamics methods

1. INTRODUCTION

Random arrangements of spheres are very useful model systems for a variety of physical and engineering problems. Arrays of closely packed equal spheres are fundamental in the microscopic theory of fluids, glasses and amorphous solids [1–3]. On the other hand sphere packings are crucial in determining the macroscopic granular nature of porous materials and liquid flow characteristics. Random beds of spherical fuel elements are utilized in an experimental pebble bed nuclear reactor [4, 5].

Two different approaches to the irregular close packing problem are generally used:
(a) laboratory-built models in which irregular packings are constructed by shaking together identical steel ball bearings [6–8];
(b) computer-generated sets of spatial coordinates of the spheres obtained by simulating the compression of a hard sphere ensemble [1, 6, 9, 10].
Packing densities obtainable from the laboratory experiments by a careful set of vibrations are about 0.6366 [7]. Studies using computer-based models where both gravitational and frictional effects are absent, have reproduced the value of 0.6366, but have failed to significantly exceed it.

Recently Jodrey and Tory [9, 10] presented an algorithm which generates irregular close packing from a random distribution of points. We named this method the neighbour separation algorithm (NS). The packing fractions they achieved by this method are between 0.642 and 0.649, which were greater than any previous experimental or simulated values. The main feature of this approach is that the diameters of the spheres can vary from one step to another according to the current arrangement of the ensemble. Each sphere has two different diameters, inner and outer. The inner diameter is set after each iteration to the minimum center-to-center distance between

any two spheres and defines the true packing density. The outer diameter has initially a relatively large value giving a packing fraction equal to unity. The outer spheres intersect each other in contrast to the inner ones. Thus in each iteration the worst overlap between the outer spheres is eliminated by separating the spheres until their distance reaches the outer diameter. Simultaneously the outer diameter is slightly reduced using a contraction rate τ :

$$d_{\text{out}}^{(i-1)} = d_{\text{out}}^{(i)} - (\tfrac{1}{2})^{\delta} d_{\text{out}}^{(0)} / (N \cdot \tau), \tag{1}$$

where:

$\delta = [- \log_{10} \Delta\eta]$,
N = number of spheres,
$d_{\text{out}}^{(i)}$ = outer diameter in the i-th iteration,
$\Delta\eta$ = difference between the nominal and actual packing densities,
τ = contraction rate of the outer diameter,
$[\cdot]$ = greatest integer function.

The inner and outer diameters approach each other and their coincidence terminates the procedure.

Using the algorithm description [9] and the source listing of the original Fortran program of Jodrey and Tory we developed and tested a new C-language version of the program for the close packing generation. It was written according to a specified methodology close to the OLYMPUS system standard [11]. The program was also supplied with some new functions as graphical output, input data control, data saving and restart possibility, etc. The innermost part of the program was slightly modified to yield a 30% reduction of the overall computational time compared to the original program.

2. A NEW PACKING ALGORITHM

Using some of the Jordrey and Tory's ideas we have devised and implemented a new algorithm for the sphere packing problem, which subsequently will be referred to as the force-biased algorithm (FB). Following Jodrey and Tory's method, the starting configuration of the algorithm is a set of randomly distributed points with inner and outer diameters. However in our approach in each iteration all of the spheres are moved simultaneously in the direction determined by a "force" acting on each sphere. For every pair of intersecting spheres i and j the individual pair "force" $\vec{F}_{ij}$ is proportional to the overlap area

$$\vec{F}_{ij} = \frac{\varepsilon}{d_{\text{out}}^{(0)}} (d_{\text{out}}^2 - r_{ij}^2) \frac{\vec{r}_{ij}}{r_{ij}}, \tag{2}$$

where:

$\vec{r}_i$ represents the spatial coordinates of the i-th sphere,

$$\vec{r}_{ij} = \vec{r}_i - \vec{r}_j, \; r_{ij} = |\vec{r}_{ij}|$$

and, ε is a scaling factor. Every sphere is moved according to the resultant displacement vector calculated over all overlapping spheres

$$\vec{r}_i = \vec{r}_i + \sum_{\substack{j \\ r_{ij} < d_{out}}} \vec{F}_{ij}. \tag{3}$$

As in Jodrey and Tory's approach the outer diameter is decreased in each iteration using the contraction rate τ

$$d_{out}^{(i+1)} = d_{out}^{(i)} - (\tfrac{1}{2})^{\delta} d_{out}^{(0)} / (2 \cdot \tau). \tag{4}$$

Since one iteration of the force-biased algorithm corresponds to N/2 steps in the Jodrey and Tory's method, Equation (1) differs from (4) in the relaxation constant to preserve the meaning of the parameter τ. The initial value of the outer diameter corresponds to the nominal packing fraction η_0. The convergence criterion of the algorithm was also adopted from Jodrey and Tory. Both algorithms use periodic boundaries.

3. RESULTS OF COMPUTER SIMULATIONS

The packings resulting from both methods were generated by two C-language programs running on the IBM PC/AT turbo supported with the DSI-020 plug-in board. The board consists of Motorola's 68020 CPU, 68881 hardware floating point co-processor, 1MB of RAM and cycle 80 ns. Fortran versions of the programs were used to make part of the calculation on the supercomputer ETA 10-P*108 with cycle 21 ns.

In our computer experiments we tested the relationship between the contraction rate τ and the final packing density. Results of the NS algorithm were compared to the following Jodrey and Tory empirical equation [9]:

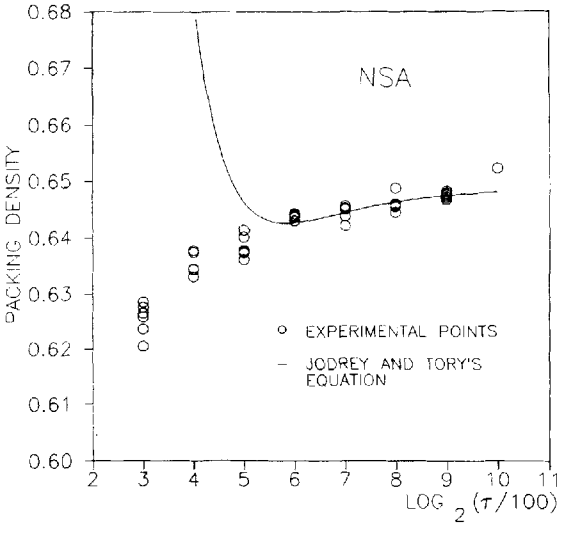

Figure 1 The final packing density vs $\log_2(\tau/100)$, where τ is the outer diameter contraction rate. The open circles correspond to experimental points obtained from the NS algorithm for 256 spheres on the IBM PC/AT with DSI-020. The solid line represents Jodrey and Tory's approximation given by Equation (5).

$$\eta(\tau) = 0.6487 - 69.08/\tau + 196700/\tau^2. \quad (5)$$

Since the above relationship was obtained for τ between 6400 and 51200. it gave proper (i.e. consistent with experiment) values of $\eta(\tau)$ only for τ greater than about 6000 (cf. Figure 1). We also carried out two runs of the NS program with τ equal to

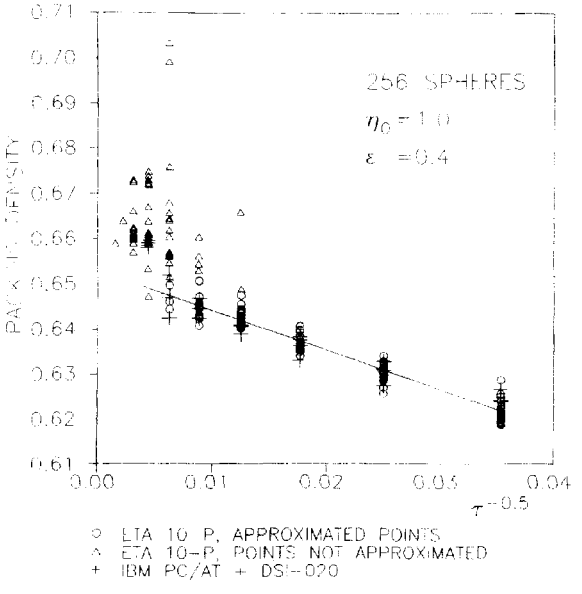

Figure 2 The final packing density vs $\tau^{-0.5}$ for the FB algorithm and 256 spheres. The solid straight line corresponds to the approximation given by Equation (6).

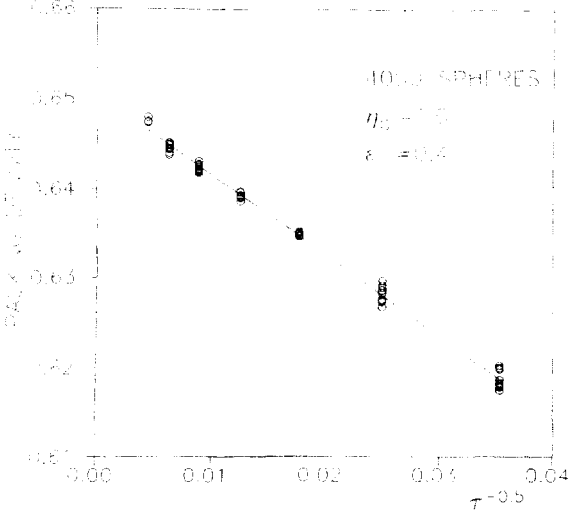

Figure 3 The final packing density vs $\tau^{-0.5}$ for the FB algorithm and 4000 spheres (ETA 10-P). The solid straight line corresponds to the approximation given by Equation (6).

3276800, producing densities of 0.6499 and 0.6490 respectively. This fact suggests that for infinite τ the final packing density tends to a constant limit value about 0.65.

Similar experiments were done for the FB algorithm, partly on the supercomputer ETA 10-P*108 for 256 (Figure 2) and 4000 (Figure 3) spheres. The points for the same tau result from different starting configuration. The main feature of the FB algorithm is that it packs the sphere ensembles to very high densities. The largest packing fraction that we obtained was 0.7034, which to our knowledge had not been achieved so far, especially using periodic boundaries. However such packings are obtained only for small numbers of spheres, thus small and large sphere ensembles should be discussed separately.

For all points presented on Figures 2–3 the radial distribution functions (rdfs) were calculated, plotted and analyzed. It was found that ensembles of 256 spheres fall into two different categories. The first one includes packings with rdfs characteristic of irregular structures and usually with densities up to 0.65. Packings of higher densities are often much more ordered and peaks of their rdfs are in good agreement with those of crystalline structures (fcc or hcp). On the other hand packing densities for large number of spheres are all very close together and their rdfs are characteristic of irregular ensembles.

We calculated a function approximating the final packing density vs. τ, using the MINPACK program (Argonne National Laboratory). While calculating the approximating function parameters for 256 spheres, only those points for which the rdf implied irregularity of its structure were considered. For ensembles of 4000 spheres all points were taken for approximation. As the best fit we take a function of the form

$$\eta(\tau) = A - B / \tau^{1/2}, \tag{6}$$

with the following parameters :

a) 256 spheres

$$A = 0.6529, \quad B = 0.8818$$

b) 4000 spheres

$$A = 0.6506, \quad B = 0.8993$$

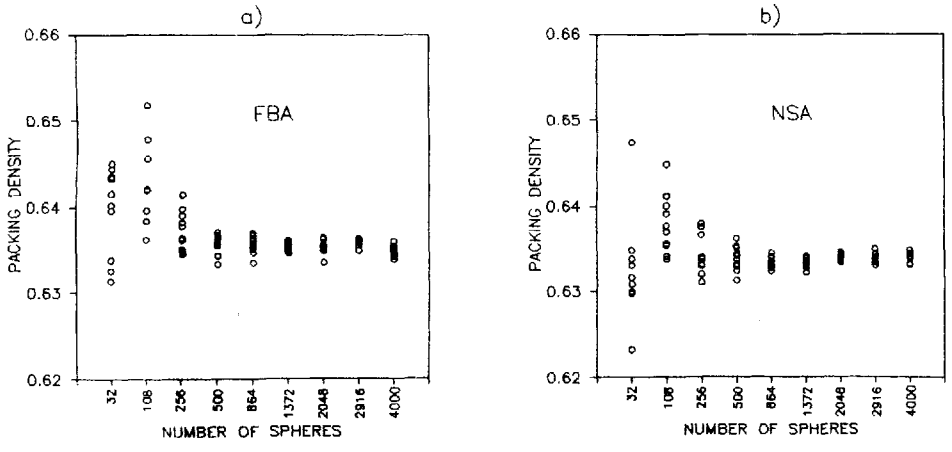

Figure 4 The final packing density vs number of spheres (IBM PC/AT + DSI-020).
a) FB algorithm, $\tau = 3200$, $\eta_0 = 0.8$, $\varepsilon = 0.4$,
b) NS algorithm, $\tau = 1600$.

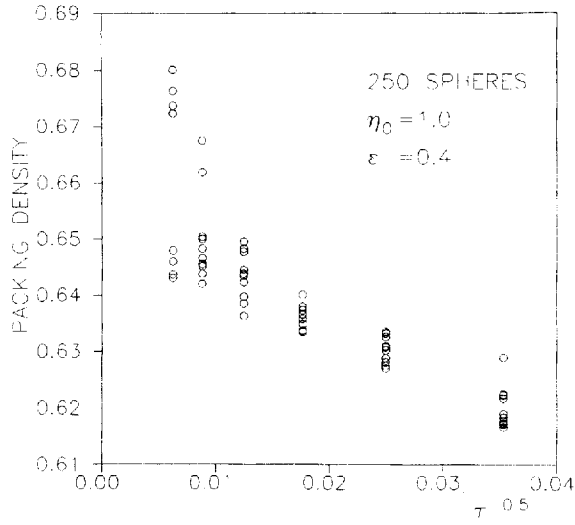

Figure 5 Final packing density vs $\tau^{-0.5}$ for the FB algorithm, 250 spheres and parameters $\varepsilon = 0.4$, $\eta_0 = 1.0$ (IBM PC/AT + DSI-020).

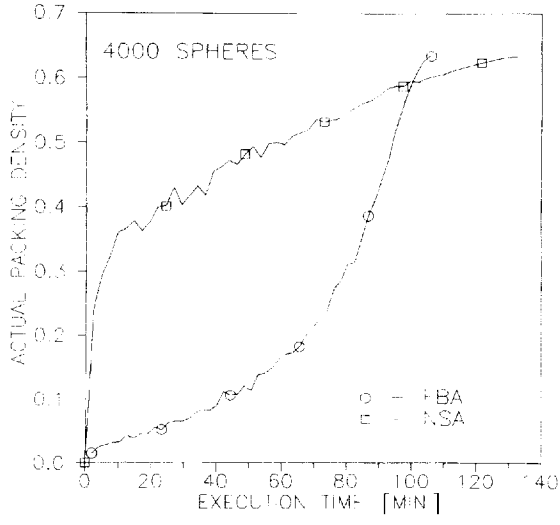

Figure 6 Variations of the actual packing density in computational time averaged over 10 runs for 4000 spheres and parameters : FB algorithm, $\tau = 3200$, $\eta_0 = 0.8$, $\varepsilon = 0.4$; NS algorithm, $\tau = 1600$ (IBM PC/AT + DSI-020).

Due to the approximating function form, the experimental points of Figures 2–3 are plotted against $\tau^{-1/2}$. Densities obtained for 256 spheres (Figure 2) on the ETA-10-P are additionally compared to values calculated on IBM-PC/AT (not approximated).

Some sample runs of the both algorithms with different initial configurations were

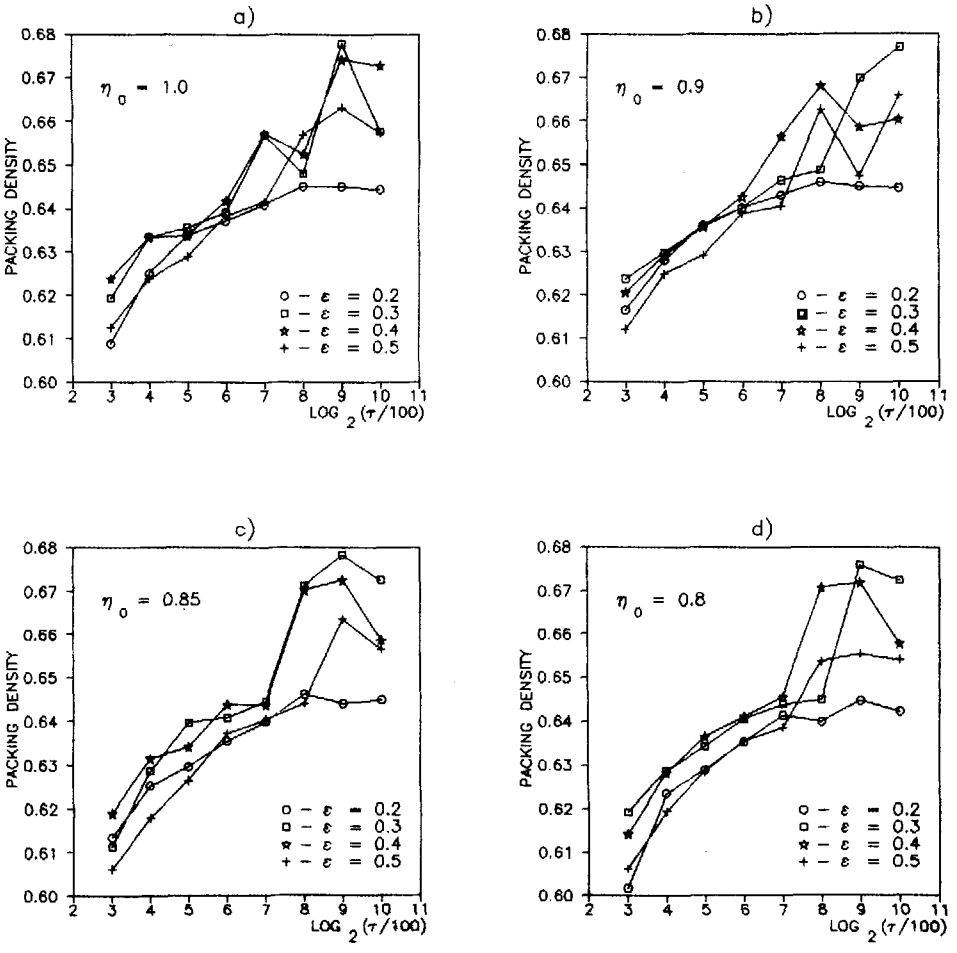

Figure 7 Final packing density vs $\log_2(\tau/100)$ for the FB algorithm, 256 spheres and different values of ε and η_0 (IBM PC/AT + DSI-020).
a) $\eta_0 = 1.0$, $\varepsilon = 0.2, 0.3, 0.4, 0.5$,
b) $\eta_0 = 0.9$, $\varepsilon = 0.2, 0.3, 0.4, 0.5$,
c) $\eta_0 = 0.85$, $\varepsilon = 0.2, 0.3, 0.4, 0.5$,
d) $\eta_0 = 0.8$, $\varepsilon = 0.2, 0.3, 0.4, 0.5$.

carried out for several numbers of spheres (Figure 4). This experiment confirmed a decreasing dispersion of results of larger ensembles. Thus to minimise the influence of the initial configuration on the final results rather large ensembles should be used. Since the number of spheres presented on Figures 2–4 are all numbers which pack, with periodic boundaries, as perfect fcc structures, we also made some tests with 250 spheres (Figure 5), producing again very high packing fractions.

We examined also the variations of the actual packing density during calculation time for the NS and FB algorithms (Figure 6). The curves were obtained for 4000 spheres and averaged over 10 runs. It is easy to see a different behaviour of the both algorithms. In a case of the NS algorithm the actual packing density grows rapidly

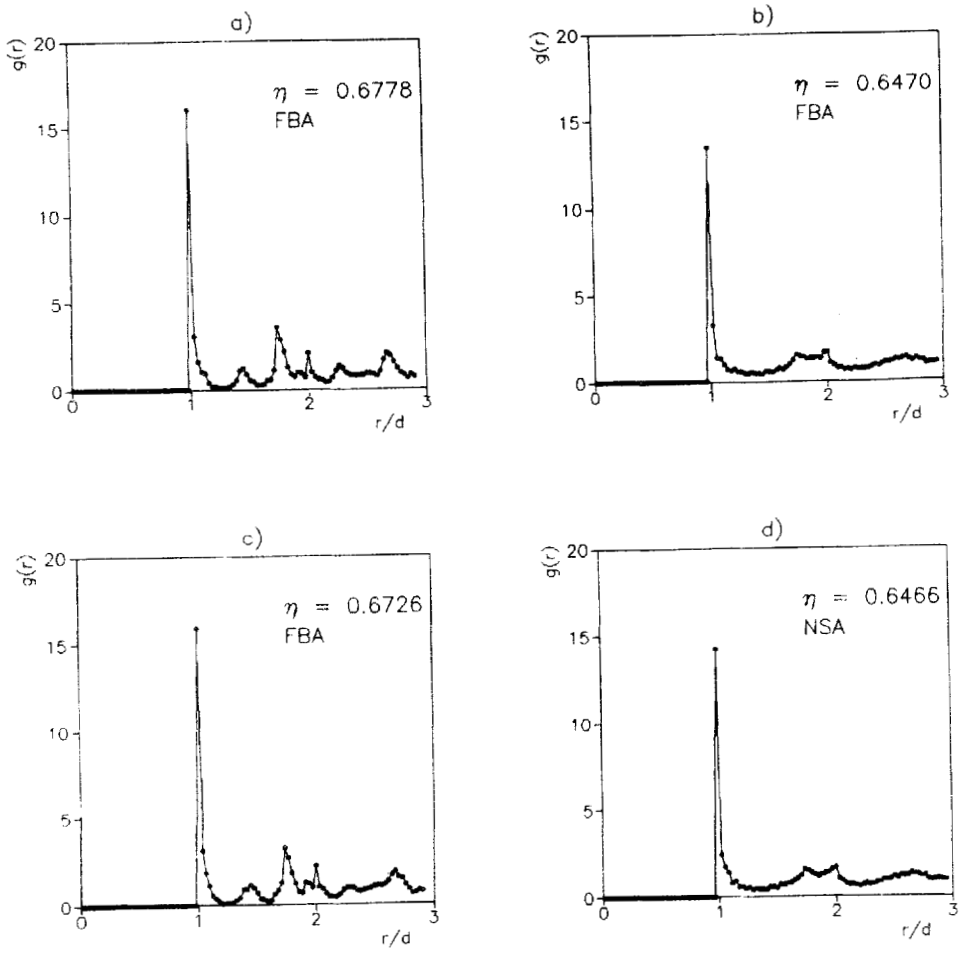

Figure 8 Radial distribution functions for 256 spheres (IBM PC/AT + DSI-020).
a) FB algorithm, $\tau = 51200$, $\eta_0 = 1.0$, $\varepsilon = 0.3$, $\eta = 0.6778$.
b) FB algorithm, $\tau = 25600$, $\eta_0 = 1.0$, $\varepsilon = 0.4$, $\eta = 0.6470$.
c) FB algorithm, $\tau = 25600$, $\eta_0 = 1.0$, $\varepsilon = 0.4$, $\eta = 0.6726$.
d) NS algorithm, $\tau = 51200$, $\eta = 0.6466$.

at the beginning of the calculations after which it changes very slowly, as opposed to the FB algorithm in which the actual packing density grows mainly at the end of the run.

To gain high densities from the FB algorithm it is also important to choose proper values of the scaling factor ε and initial nominal density η_0 (Figure 7). Our tests show, that the best results are obtained for ε between 0.3 and 0.5, and η_0 from 0.8 to 1.0. For τ less than 12800, the final density grows monotonically, but for greater τ the changes are difficult to determine due to the large dispersion of results caused by the small number of spheres used (256). Probably better characteristics would be obtained for

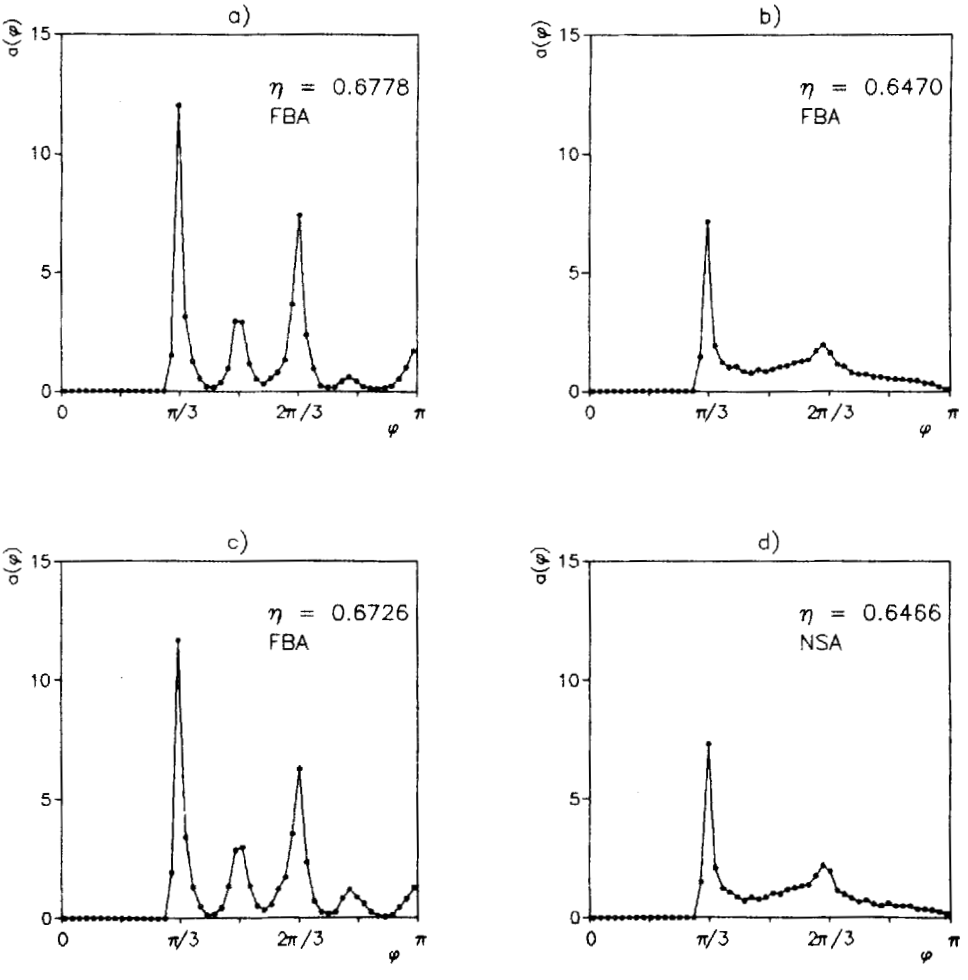

Figure 9 Angle distribution functions for 256 spheres (IBM PC/AT + DSI-020).
a) FB algorithm, $\tau = 51200$, $\eta_0 = 1.0$, $\varepsilon = 0.3$, $\eta = 0.6778$,
b) FB algorithm, $\tau = 25600$, $\eta_0 = 1.0$, $\varepsilon = 0.4$, $\eta = 0.6470$,
c) FB algorithm, $\tau = 25600$, $\eta_0 = 1.0$, $\varepsilon = 0.4$, $\eta = 0.6726$,
d) NS algorithm, $\tau = 51200$, $\eta = 0.6466$.

greater number of spheres, but appropriate calculations require a great deal of computing time, even on a supercomputer.

To compare the geometrical and statistical properties of generated packings, the radial (Figure 8) and angle (Figure 9) distribution functions were calculated and plotted for some final configurations. We defined the angle distribution function $a(\phi)$ as follows

$$a(\phi) = N(\phi, \Delta\phi) / N, \tag{7}$$

where $N(\phi, \Delta\phi)$ is the total number of angles between any two spheres within a distance $d_{out}(1 + \alpha)$ from a common neighbour, included in the interval $(\phi - 0.5\Delta\phi,$

$\phi + 0.5\Delta\phi$). Our results for the NS and FB algorithms are very similar to each other and consistent with rdfs presented in the literature (cf. Figure 8(b) and Figure 5(b) in [12]). However the above is true only for final packing density up to about 0.65. At greater densities the rdfs have considerably different shapes with peaks at values characteristic of regular packings (Figures 8(a) and 8(c)).

The angle distribution functions for four selected runs, shown in Figure 9, were computed for $\alpha = 0.1$. The finite probability that the three spheres will form an equilateral triangle accounts for the sharp maximum at $\phi = \pi/3$. The broad peak at $\phi = 2\pi/3$ probably arises from abutting equilateral triangles. For the high density packing (Figures 9(a) and 9(c)) peaks at $\pi/3$ and $2\pi/3$ are almost two times stronger that those for looser packings (Figures 9(b) and 9(d)). The additional peaks also appear at $\phi = \pi/2$ and $\phi = \pi$.

Both algorithms generate packings in which only two spheres are in real contact, but many spheres are in near contact. Table 1 shows the distribution of near neighbours of the following three runs

a) NS algorithm, $\tau = 51200$, $\eta = 0.6466$,
b) FB algorithm, $\tau = 25600$, $\eta_0 = 1.0$, $\varepsilon = 0.4$, $\eta = 0.6470$ (case 1)
c) FB algorithm, $\tau = 51200$, $\eta_0 = 1.0$, $\varepsilon = 0.3$, $\eta = 0.6778$ (case 2)

Table 1a Dependence of distribution of number of near neighbours on designated maximum spacing between neighbours δ for the NS algorithm

	number of neighbours	*-log δ*							
		2	*3*	*4*	*5*	*6*	*7*	*8*	*9*
number of	0	2	5	9	13	41	169	249	252
spheres with	1	1	1	0	4	36	73	6	4
designated	2	0	0	2	14	37	12	1	0
number of	3	1	1	8	29	67	1	0	0
neighbours	4	14	39	50	62	50	1	0	0
(within δ	5	42	71	86	86	21	0	0	0
of touching)	6	71	61	53	31	4	0	0	0
	7	69	53	37	13	0	0	0	0
	8	44	21	10	4	0	0	0	0
	9	11	4	1	0	0	0	0	0
	10	1	0	0	0	0	0	0	0
	11	0	0	0	0	0	0	0	0
	12	0	0	0	0	0	0	0	0

Near neighbours are defined as those within δ of touching. The distribution of near neighbours of the FB algorithm is shifted in relation to the NS algorithm, what means that for comparable densities (cf. Table 1 (a) and (b)) the FB algorithm produces packings with statistically smaller number of neighbours. A surprising feature of the FB algorithm is that the number of neighbours in a packing decreases with increasing density (cf. Table 1 (b) and (c)). This is probably because a high density packing is much more ordered than a looser ensemble.

While carrying out our calculations on the ETA 10-P we attempted to vectorize the FB algorithm with assistance of the Pacific — Sierra Research Vast-2 v2.23C26 preprocessor. This failed however, due to the purely sequential notation of the algorithm. Consequently the speedup of the program compared to the IBM PC/AT

Table 1b FB algorithm case 1

	number of neighbours	*-log δ*							
		2	*3*	*4*	*5*	*6*	*7*	*8*	*9*
number of	0	1	5	33	187	247	254	254	254
spheres with	1	1	4	30	30	8	2	2	2
designated	2	2	5	22	12	1	0	0	0
number of	3	3	18	29	15	0	0	0	0
neighbours	4	18	46	67	11	0	0	0	0
(within δ	5	65	94	55	1	0	0	0	0
of touching)	6	74	50	18	0	0	0	0	0
	7	48	25	2	0	0	0	0	0
	8	32	7	0	0	0	0	0	0
	9	11	1	0	0	0	0	0	0
	10	1	1	0	0	0	0	0	0
	11	0	0	0	0	0	0	0	0
	12	0	0	0	0	0	0	0	0

Table 1c FB algorithm case 2

	number of neighbours	*-log δ*							
		2	*3*	*4*	*5*	*6*	*7*	*8*	*9*
number of	0	0	5	58	214	248	252	252	254
spheres with	1	2	3	49	17	2	2	4	2
designated	2	4	13	27	11	6	2	0	0
number of	3	5	18	44	8	0	0	0	0
neighbours	4	12	41	37	5	0	0	0	0
(within δ	5	41	72	33	1	0	0	0	0
of touching)	6	78	72	8	0	0	0	0	0
	7	61	26	0	0	0	0	0	0
	8	44	5	0	0	0	0	0	0
	9	9	1	0	0	0	0	0	0
	10	0	0	0	0	0	0	0	0
	11	0	0	0	0	0	0	0	0
	12	0	0	0	0	0	0	0	0

with DSI-020 is only 11. However the FB algorithm seems to be much easier to adapt to vector feature requirements than the NS algorithm of a strictly sequential nature.

4. CONCLUSIONS

We have developed a "force-biased" algorithm for generating the irregular close packing and compared it with other methods used so far, especially with Jodrey and Tory's approach. We concentrated on achieving very dense sphere packings, with special attention given to their phase transition. Packing fractions that we obtained from the FB algorithm (0.7034) are significantly greater than any values achieved so far, especially using periodic boundaries. The execution times of the FB algorithm for smaller densities (up to 0.64) are comparable with Jodrey and Tory's method.

The radial and angle distribution functions of the packings with similar final densities generated by means of the both algorithms are comparable. Approaching

greater densities however (above 0.65) the rdfs as well as angle distribution functions have strong peaks at values characteristic of regular packings as fcc and hcp. A comparison of Table I (a) and (b) shows that the mean distance between spheres packed by means of the NS algorithm is less than when the FB method is applied. Moreover (cf. Table I (b) and (c)) for the FB algorithm the number of neighbouring spheres decreases with increasing density. Thus it seems likely that the high density packing has larger holes between spheres and consequently is much more ordered than the low density ensemble.

Very high densities obtainable using the FB algorithm seem to contradict the Bernal conjecture [13], according to which there is no continuous transition from irregular packings to regular crystalline structures. Taking into account results of Figure 2 which do not show short-range ordering (N = 256) and those in Figure 3 (N = 4000) one can estimate a density limit for irregular (random) packing of 0.651–0.653.

Acknowledgments

J.M. is grateful to the Center of Chemical Physics of the University of Western Ontario for the award of a visiting fellowship. J.M. and M.B. are grateful to Professor Jan Klamut of the Polish Academy of Sciences for arranging partial financial support under project number CPBP 01.12. We would like to thank Professor E.M. Tory of Mount Allison University who kindly supplied us with his program listing. We are also very grateful to Dr. J. Kitowski and Mr. W. Dzwinel of the Institute of Computer Science, AGH, for their co-operation and continuous interest in the problem.

References

[1] J.L. Finney, "Modelling the structures of amorphous metals and alloys", *Nature*, **266**, 309 (1977).
[2] J.D. Bernal, "The structure of liquids". *Proc. Roy. Soc. Lond., A* **280**, 299 (1964).
[3] M. Biasini, "Statistical simulation of the structure of amorphous alloys", *Nuovo Cimento*, **9D**, 353, (1987).
[4] G.A. Tingate, "Parameters influencing the flow of packing of spheres and near spheres in cylindical vessels", *Nucl. Eng. Des.*, **30**, 36 (1974).
[5] L. Bieniasz, J. Mościński, P. Niżegorodcew, and Z. Rycerz, "Monte Carlo simulation of the emergency shutdown system for the high-temperature pebble-bed nuclear reactor", *Ann. Nucl. Energy*, **10**, 299 (1983).
[6] J.D. Bernal, "Geometry of the structure of monoatomic liquids", *Nature*, **188**, 910 (1960).
[7] G.D. Scott, "Radial distribution of the random close packing of equal spheres", *Nature*, **194**, 956 (1962).
[8] G.D. Scott and D.M. Kilgour, "The density of random close packing of spheres", *J. Phys., D* **2**, 863 (1969).
[9] W.S. Jodrey and E.M. Tory, "Computer simulation of close random packing of equal spheres", *Phys. Rev., A* **32**, 2347 (1985).
[10] W.S. Jodrey and E.M. Tory, "Computer simulation of isotropic, homogeneous dense random packing of equal spheres", *Powder Technology*, **30**, 111 (1981).
[11] J.P. Christiansen and K.V. Roberts, "A standard control utility package for initial value FORTRAN programs", *Computer Physics Communications*, **7**, 245 (1974).
[12] J.L. Finney, "Random packing and the structure of simple liquids. I. The geometry of random close packing", *Proc. Roy. Soc. London A*, **319**, 479 (1970).
[13] J.D. Bernal, "A geometrical approach to the structure of liquids", *Nature*, **183**, 141 (1959).