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LETTER TO THE EDITOR

Packing of spheroids in three-dimensional space by random sequential addition

J D Sherwood

Schlumberger Cambridge Research, High Cross, Madingley Road, Cambridge CB3 0EL, UK

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Abstract. Packings of spheroidal particles with semi-axes of length (a, b, b) were generated by random sequential addition (RSA) simulations. As the jamming limit is approached, the volume fraction occupied by particles tends towards an asymptote ϕ_∞, which was determined as a function of aspect ratio $\alpha = a/b$. This asymptote ϕ_∞ has a local minimum at $\alpha = 1$ (spheres), with local maxima at $\alpha \simeq 1.4$ (prolate spheroids) and $\alpha \simeq 0.7$ (oblate). Values of ϕ_∞ agree with results from RSA simulations of sphere packings, but lie below volume fractions obtained in simulations of near-spheres packed under gravity. Volume fractions reported for simulations of spheroids packed under gravity vary widely when the aspect ratio α is very large or small; differences between these results and the predictions of RSA are discussed.

1. Introduction

The macroscopic properties of a granular or fibrous material are strongly influenced by the arrangement of the constituent particles. The arrangement of packed spherical particles has been the subject of many experimental and theoretical investigations [1], but the packing of non-spherical particles in three dimensions has been much less studied. The maximum volume fraction of particles will depend not only on the size distribution and shape of the particles, but also on the method by which the packing is achieved. Experiments to determine the volume fraction of packed rods are reviewed in [2, 3], and Philipse [3] provided an asymptotic analysis for long slender rods. Buchalter and Bradley [4] performed simulations of oblate and prolate spheroids poured into a box under gravity. Coelho *et al* [5] studied the geometrical and transport properties of beds of spheroids, cylinders and cuboids formed by simulations of particles packed one-by-one under gravity.

The simulations described here generated three-dimensional packings of spheroids by random sequential addition (RSA). Two-dimensional RSA, usually described as random sequential adsorption, is reviewed in [6]. Simulations of the adsorption of ellipses and other non-spherical shapes on a plane surface are reported in [7–12], and the adsorption of three-dimensional spheroids on a plane is described in [13]. Three-dimensional RSA studies of sphere packings have been reported [14]. The qualitative features of the packing densities obtained here, as a function of particle aspect ratio, are similar to those observed in [4], but the densities achieved are different. In three dimensions, RSA is not physically realizable, since it requires particles to be placed in positions which are entirely surrounded by particles previously deposited. Nevertheless, it is of interest because of the absence of gravity and of any consequential anisotropy.

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2. Procedure and results

We consider spheroids with axes of length $(2a, 2b, 2b)$ in a cube of side L. The spheroids were placed one at a time in the initially empty cube, which had periodic boundary conditions. The position of the centre of each spheroid was chosen by means of three random numbers uniformly distributed over $(0, L)$. The orientation of the axis of symmetry of the spheroid in polar coordinates (θ, ϕ) was given by two random numbers chosen such that θ fell within $(\theta, \theta + \delta\theta)$ with probability $\sin\theta\,\delta\theta$, and ϕ was uniform in $(0, 2\pi)$. If the newly positioned spheroid overlapped any of those previously placed (or any of their periodic images), all five random numbers were discarded, and the process was started again. If no overlaps were detected [15], the new spheroid was accepted, and fixed in its position. As the packing became denser, the probability that a particle was placed without overlapping became smaller, and the volume fraction ϕ occupied by particles eventually approached an asymptote ϕ_∞. In one dimension (parking on a line) the RSA process is well understood (see [6]). In two dimensions it is known that the area coverage of circular disks dropped on a plane approaches the asymptote with an error $\propto t^{-1/2}$, where t is a time-like variable counting the number of trials [16, 17]. If the particles are not circular, the asymptote should be approached as $t^{-1/3}$, but in practice it can be difficult to observe this approach rate in simulations [7]. Indeed, the large-t behaviour for lines adsorbed on a plane found in [9] subsequently proved to be incorrect when longer simulations were performed [10, 11]. Although the rate of approach to the asymptote has been successfully used to find ϕ_∞ by extrapolation [18], it was here considered more prudent to use simulations with a small box size, and a large number of trials, in order to minimize the extrapolation. For all aspect ratios $\alpha = b/a < 15$, the box size was $L = 15\max(a, b)$, with $t_{\max} = 1614\,431\,772$ trials. For $\alpha = 15$, simulation times were such that it was necessary to reduce the above value of L by a factor of 1.26, and the number of trials was reduced by a factor of 2. A larger simulation size L would reduce finite-size effects, but would require considerable extrapolation, or many more trials, in order to determine ϕ_∞. Buchalter and Bradley [4] found long-range orientational alignment in their three-dimensional packings, but they concluded that translational order was short-ranged, with an average crystallite size less than one semi-major axis length. RSA generates packings with local order determined by the first few particles deposited, but with no long-range orientational ordering, as is easily seen in two-dimensional simulations [9]. Thus, the choice $L = 15\max(a, b)$ should be ample to avoid effects of long-range ordering. Tests to verify that changes in L made little difference to the results which are reported below.

For each value of the aspect ratio, $N = 8$ simulations were performed, and the average volume fraction $\bar{\phi}$ was determined, together with an estimate of the variance $\sigma^2 = \sum_{i=1}^{N}(\phi_i - \bar{\phi})^2/(N-1)$. Results were extrapolated by an amount $\Delta\phi$ by seeking a value of ϕ_∞ such that a plot of $\log(\phi_\infty - \bar{\phi})$ against $\log(t)$ was approximately straight: this assumes that the asymptote ϕ_∞ is approached as some power law t^γ. Results are given in table 1. In general, the amount of extrapolation was smaller than the standard deviation σ at aspect ratios close to 1, and larger at the extreme aspect ratios. The final estimates for ϕ_∞ are shown in figure 1. In the text we quote $\phi_\infty \pm \sigma$, using the standard deviation σ at $t = t_{\max}$.

When the particles are spheres, the value $\phi_\infty = 0.382 \pm 0.003$ obtained here is consistent with the value 0.382 ± 0.0005 found by Talbot *et al* [14]. This is lower than standard results for the volume fraction in loose random packing ($0.609 < \phi < 0.625$) and dense random packing ($0.625 < \phi < 0.641$) quoted in [19], and is also lower than the value $\phi = 0.46$ obtained by Buchalter and Bradley [4] by numerical simulation. If the box size

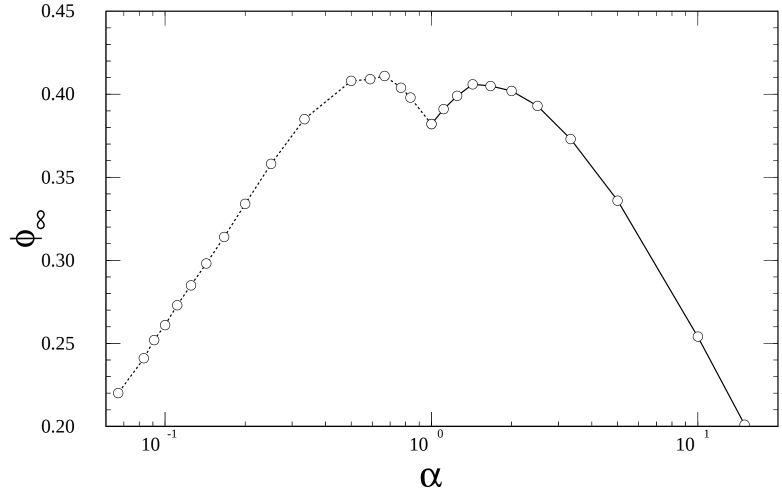

Figure 1. The maximum RSA volume fraction ϕ_∞, as a function of aspect ratio $\alpha = a/b$.

L is reduced by a factor of 1.26, and the number of trials $t_{\max}$ by a factor of 2, we obtain $\phi_\infty = 0.383 \pm 0.004$. If L is reduced by a factor of 2, and $t_{\max}$ by a factor of 8, then $\phi_\infty = 0.387 \pm 0.02$. For oblate spheroids, with $b/a = 10$, simulations in the standard box of side $L = 15b$ lead to $\phi_\infty = 0.261 \pm 0.002$. If L is reduced by a factor of 1.26, and $t_{\max}$ by a factor of 2, we obtain $\phi_\infty = 0.260 \pm 0.002$. If L is reduced by 2, and $t_{\max}$ by 8, then $\phi_\infty = 0.271 \pm 0.003$ after extrapolation by an unacceptably large $\Delta\phi = 0.012$. The major effect of reducing the simulation size is an increase in the uncertainty due either to a higher standard deviation σ or to increased extrapolation $\Delta\phi$. The results given in table 1 suggest that these uncertainties have been adequately controlled by the values of L, $t_{\max}$ used here.

When the particles are slightly non-spherical, ϕ_∞ reaches maxima at aspect ratios $a/b \simeq 1.4$ (prolate spheroids) or $a/b \simeq 0.7$ (oblate). This behaviour has been seen in two-dimensional RSA simulations [8, 9, 12]. It was also observed in the three-dimensional simulations of Buchalter and Bradley [4], although their maxima are much larger ($\phi \simeq 0.49$ oblate, $\simeq 0.48$ prolate). Buchalter and Bradley appeal to arguments based on minimization of gravitational potential energy which cannot be applied here. The corresponding local minimum in the packing fraction when the particles are spherical is not apparent in the results of Coelho *et al* [5]. However, their packing fraction $\phi = 0.598$ for spheres is much closer to accepted values for random packing, and is higher than the values obtained either by RSA, or in [4]. There is therefore less scope for an increase in the packing fraction when their particles become non-spherical.

Buchalter and Bradley [4] found $\phi \simeq 0.29$ for oblate spheroids with $b/a = 8$. This is close to the packing fraction ϕ_∞ obtained by RSA. Coelho *et al* [5] found a much higher packing fraction (approximately 0.6), and explained that this was due to the high degree of orientational ordering of their oblate spheroids. Their volume fractions for beds of disk-like cylinders, which were less ordered than the oblate spheroids, were only slightly higher than those obtained here.

The RSA volume fractions ϕ_∞ for prolate spheroids agree reasonably well with those of spheroids and cylinders reported in [5], and with experimental results from figure 1 of [3]. However, Buchalter and Bradley [4] found $\phi \simeq 0.06$ at an aspect ratio $a/b = 8$, compared with the value 0.28 found by RSA. The origin of their small volume fraction for prolate

Table 1. The estimated maximum packing ϕ_∞ generated by RSA as a function of aspect ratio $\alpha^{-1} = b/a$. The maximum mean volume fraction was extrapolated by an amount $\Delta\phi$ in order to reach the value ϕ_∞. The standard deviation σ is that of the volume fraction at $t = t_{\max}$.

b/a	ϕ_∞	$\Delta\phi$	σ
0.067	0.201	0.004	0.0003
0.1	0.254	0.005	0.0004
0.2	0.336	0.004	0.0006
0.3	0.373	0.003	0.0008
0.4	0.393	0.002	0.001
0.5	0.402	0.001	0.002
0.6	0.405	0.001	0.003
0.7	0.406	0.001	0.003
0.8	0.399	0.001	0.004
0.9	0.391	0.0	0.004
1.0	0.382	0.001	0.003
1.2	0.398	0.001	0.007
1.3	0.404	0.001	0.004
1.5	0.411	0.002	0.005
1.7	0.409	0.001	0.002
2.0	0.408	0.001	0.003
3.0	0.385	0.004	0.002
4.0	0.358	0.002	0.002
5.0	0.334	0.003	0.003
6.0	0.314	0.002	0.001
7.0	0.298	0.002	0.002
8.0	0.285	0.003	0.002
9.0	0.273	0.004	0.002
10.0	0.261	0.003	0.002
11.0	0.252	0.004	0.002
12.0	0.241	0.003	0.001
15.0	0.220	0.003	0.001

spheroids is unknown.

The probability that particles touch in packings generated by RSA is zero, and so there is scope for minor re-arrangement of particles if gravity is switched on. This would tend to suggest that RSA packings of spheres will be less dense than random loose packings created in a gravitational field. However, when the particle aspect ratio is large (or small), the packing volume fraction is small, and only a small proportion of the pore space is contained in narrow gaps between non-touching particles. Minor re-arrangements caused by the application of gravity will lead to only a small change in the packing volume fraction. Differences between RSA and packings made under gravity are more likely to be due to anisotropy caused by the vertical gravitational field.

Philipse [3] observed that the experimental random packing volume fraction of rods with diameter $2b$ and length $2a$ was approximately $\phi \simeq 5.4b/a$ for $a/b > 15$. One of the aims of the current study was to test this asymptote, but computation times became excessive as the particle aspect ratio became either very large or very small. At the maximum aspect ratio $a/b = 15$ studied here, RSA leads to a volume fraction $\phi_\infty = 0.20$, lower than the value 0.36 predicted by the Philipse correlation. If we assume that the effect of changing from Philipse's cylinders to spheroids is merely to reduce the particle volume by a factor 2/3, without changing the particle number density, the amended Philipse correlation predicts $\phi = 0.24$.

In two dimensions the packing of ellipses with aspect ratio $\alpha = a/b$ is identical to that of ellipses with aspect ratio α^{-1}, but this correspondence does not hold between prolate and oblate spheroids in three dimensions. Nevertheless, there is an approximate symmetry between values of ϕ_∞ for spheroids of aspect ratio α and α^{-1}, as can be seen from figure 1. This supports Philipse's suggestion [3] that the curve of volume fraction against aspect ratio should be approximately symmetrical about $\alpha = 1$.

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