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D. J. Daley

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National Institute of Statistical Sciences
19 T. W. Alexander Drive
PO Box 14006
Research Triangle Park, NC 27709-4006
www.niss.org

PACKINGS AND APPROXIMATE PACKINGS OF SPHERES

D. J. DALEY*

School of Mathematical Sciences

Australian National University

Summary

Close-packings of uniformly-sized spheres with centres on various lattices are described, with volume fractions equal or close to the maximum possible $\pi/\sqrt{18}$ (this value has long been 'known' via Kepler's conjecture, and has been proved). Regular packings with two or three sized spheres can push this volume fraction to beyond 80%. The bulk of the paper studies irregular 'packings' of a large sphere by spheres of varying sizes, and attempts to evaluate the influence of factors in the algorithm specifying how the random packing is constructed, in determining the volume fraction of the resultant random set (meaning, the union of all the spheres).

Extrapolation and edge-correction techniques for determining the volume fraction of an infinite array of such balls in an infinitely large sphere are indicated. The paper also investigates questions of inaccessibility of part of the space except to spheres of infinitesimal size. Various questions and problems are recorded also.

The study began from the observation that the volume fraction of aggregate in concrete has a volume fraction in the range 60% to 70%. It is known how to locate spheres on a perturbed lattice and, depending on the perturbation, obtain a volume fraction arbitrarily close to $\pi\sqrt{18}$. If cubes of irregular size but common orientation are used instead of spheres, then the volume fraction can be made arbitrarily close to 1.0 by choosing sufficiently small perturbations (Daley, 2000).

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1. Introduction

This paper investigates the volume fraction of various germ–grain models in which every grain touches at least one other grain. Such touching reflects in part a feature of the particles that constitute 'aggregate' in concrete, namely, that such particles tend to touch each other in such a way that, even without the cured cement that acts as a paste between them, they are largely held in position so as to have virtually no room for movement. We therefore start by discussing the volume fractions of some regular close-packed arrays of spheres (see Section 2), and also 'sparsely packed' arrays (see Section 3), these being indicative of the range for the volume fraction attainable by randomly scattered but packed arrays of similarly sized spheres.

In Section 4 we describe a sequential method of simulating an array of irregularly sized spheres that has the touching property noted above, and in subsequent sections we look at ways of estimating the volume fraction attained by infinitely large such arrays. In a subsequent paper we show how volume fractions arbitrarily close to those attainable by regular arrays of regular spheres or cubes can be attained by a variant of the lilypond growth protocol (see Häggström and Meester (1996); Daley, Stoyan and Stoyan (1999)).

2. Volume fractions of regular packings and related sets

First we describe two close packings of unit spheres in 3-dimensional euclidean space (3-D), where *unit sphere* signifies a sphere of unit diameter, and by 'close packing' we mean that any given sphere touches at least four other spheres that constrain the given sphere from any movement. For both packings the volume fraction V_f, meaning the fraction of space that is covered by spheres, equals $\pi/\sqrt{18} = 0.740480$, which is the maximal volume fraction attainable by arrays of similarly-sized spheres (see e.g. Mackenzie (1998) for discussion of Hales' (1997) proof of this fact, referred to for long as Kepler's conjecture).

Example 1. *Close-packed layers of unit spheres centred on a square lattice.* Locate unit spheres at each of the square lattice points $\{(n,m,0) : n,m = 0,\pm1,\ldots\}$ in the (x,y) plane: call this a 'square-lattice layer' of unit spheres. Place on top of this layer another square-lattice layer, but with its centres at the points $(n,m,0) + (\frac{1}{2},\frac{1}{2},\frac{1}{2}\sqrt{2}\,)$. Observe that each sphere in this added layer 'nestles' on the four spheres with centres at the corners of the square above whose centre the upper-layer sphere is located. Then, when the spheres in the lower layer are fixed in position, so too are the spheres in the upper layer with regard to any rigid motion in the plane of their centres. Now add a third layer on top of the second, with centres at $(n,m,\sqrt{2}\,)$. The spheres in this third layer nestle amongst the spheres in the second layer in much the same way as the second layer spheres nestle on the first layer spheres. Continue the process *ad infinitum*. Then a close packing of spheres ensues, with centres at the points $(n,m,\ell\sqrt{2}\,)$ and $(n+\frac{1}{2},m+\frac{1}{2},\ell\sqrt{2}+\frac{1}{2}\sqrt{2}\,)$ $(n,m,\ell = 0,\pm1,\ldots)$, and since there is a one–one correspondence between the unit spheres and right-parallelipipeds with side lengths 1, 1 and $\frac{1}{2}\sqrt{2}$, it follows that the volume fraction of the spheres in $\mathbb{R}^3$ equals $\frac{4}{3}\pi(\frac{1}{2})^3/\,(\frac{1}{2}\sqrt{2}\,) = \frac{1}{3}\pi/\sqrt{2} = \pi/\sqrt{18}$, as asserted earlier.

In this close-packing it is immediately recognizable that the points $(n, m, \frac{1}{2}\sqrt{2}\,)$ for example are not covered by a sphere, and indeed that we can locate spheres with centres at these points, of common diameter $\sqrt{2}-1$, which touch all six spheres closest to this point (e.g. for the point $(0,0,\frac{1}{2}\sqrt{2}\,)$, these six spheres have their centres at $(0,0,0)$, $(0,0,\sqrt{2}\,)$, and $(\pm\frac{1}{2},\pm\frac{1}{2},\frac{1}{2}\sqrt{2}\,)$). Then, the effect of adding such smaller spheres with centres at these particular interstitial points, which in fact constitute another lattice whose points are in one–one correspondence with those of the original lattice, is to increase V_f by the amount

$$\frac{\frac{4}{3}\pi(\frac{1}{2})^3(\sqrt{2}-1)^3}{\frac{1}{2}\sqrt{2}} = \frac{\pi}{\sqrt{18}}\,(5\sqrt{2}-7) = \frac{\pi}{3(10+7\sqrt{2}\,)}\,, \tag{2.1}$$

hence,

$$V_f = \frac{\pi}{\sqrt{18}}\,(5\sqrt{2}-6) = 0.793104. \tag{2.2}$$

Continuing, we see that the points $(n,m,0)+(\frac{1}{2},0,\frac{1}{4}\sqrt{2}\,)$ and $(n,m,0)+(0,\frac{1}{2},\frac{1}{4}\sqrt{2})$ are not covered by either a unit sphere or a sphere of diameter $\sqrt{2}-1$, being equidistant from four unit spheres, namely a distance $\frac{1}{2}(\sqrt{\frac{3}{2}}-1)$. Inspection shows that to each unit sphere in the close-packing there are two such interstitial points where spheres of diameter $\sqrt{\frac{3}{2}}-1$ can be placed touching but without overlapping any other spheres. Adding them increases V_f by the amount

$$\frac{\pi}{\sqrt{18}}\Big(2\Big[\sqrt{\tfrac{3}{2}}-1\Big]^3\Big) = \frac{\pi}{\sqrt{18}}\Big(9\sqrt{\tfrac{3}{2}}-11\Big) = 0.016812 \qquad \text{to} \qquad 0.809916. \tag{2.3}$$

□

The same volume fractions come from the face-centred cubic lattice, as we now describe.

Example 2. *Close-packed layers of unit spheres centred on a triangular lattice.* Start with a unit sphere with centre at the origin. Then a line of centres at $(n,0,0)$ $(n=0,\pm1,\ldots)$, produces a 'line' of unit spheres, close-packed in 1-D so far as their centres are concerned. By placing similar lines with centres at the points $(n+\frac{1}{2}m,\frac{1}{2}m\sqrt{3},0)$ $(n,m=0,\pm1,\ldots)$, we close-pack unit spheres in 2-D so far as their centres are concerned, thereby forming a close-packed 'triangular-lattice layer' of spheres. Now stack such layers one on top of the other, but with centres offset relative to the layer immediately underneath such that a sphere in the upper layer nestles touching three spheres in the lower layer, such as is achieved by having layers with centres at the points $\big(n+\frac{1}{2}m+\frac{1}{2}\ell,\frac{1}{2}m\sqrt{3}+\frac{1}{2}\ell\sqrt{\frac{1}{3}},\ell\sqrt{\frac{2}{3}}\,\big)$.

It is convenient to describe the centre at

$$\big(n+\tfrac{1}{2}m+\tfrac{1}{2}\ell,\tfrac{1}{2}m\sqrt{3}+\tfrac{1}{2}\ell\sqrt{\tfrac{1}{3}},\ell\sqrt{\tfrac{2}{3}}\,\big) = n\big(1,0,0\big)+m\big(\tfrac{1}{2},\tfrac{1}{2}\sqrt{3},0\big)+\ell\big(\tfrac{1}{2},\tfrac{1}{2}\sqrt{\tfrac{1}{3}},\sqrt{\tfrac{2}{3}}\,\big) \tag{2.4}$$

by its lattice-coordinates or *index* $\langle n,m,\ell\rangle$. In this notation, the centres of the twelve nearest neighbours of the sphere whose centre has the index $\langle n,m,\ell\rangle$, have indexes expressible as $\langle n,m,\ell\rangle+e_j$ $(j=1,\ldots,12)$ where e_j takes one of the twelve values as below, given as pairs for convenience:

$$\langle\pm1,0,0\rangle,\ \langle0,\pm1,0\rangle,\ \langle0,0,\pm1\rangle,\ \langle\mp1,\pm1,0\rangle,\ \langle0,\mp1,\pm1\rangle,\ \langle\pm1,0,\mp1\rangle. \tag{2.5}$$

Note that the distance between the centres at $\langle 0,0,0\rangle = (0,0,0)$ and $\langle n,m,\ell\rangle = \big(n+\frac{1}{2}m+\frac{1}{2}\ell,\ \frac{1}{2}m\sqrt{3}+\frac{1}{2}\ell\sqrt{\frac{1}{3}},\ell\sqrt{\frac{2}{3}}\big)$ equals

$$\sqrt{n^2+m^2+\ell^2+nm+m\ell+\ell n}\ =\ \sqrt{\tfrac{1}{2}[(n+m)^2+(m+\ell)^2+(\ell+n)^2]}\,. \tag{2.6}$$

I assert that to each unit sphere with centre $\langle n,m,\ell\rangle$, there is associated a triplet of interstices into which smaller spheres, described shortly, can be fitted, and that with each such triplet there is associated a well-defined unit sphere, i.e. the association between unit spheres and triplets is one-to-one. The centres for these triplets are located in or closer to the dissecting plane midway between adjacent layers of centres from which the close packing is constructed. Specifically, for the centre at $(0,0,0)$, one element of the triplet is at the point $\big(0,\sqrt{\frac{1}{3}},\sqrt{\frac{1}{6}}\big)$ which is equidistant from the six unit sphere centres denoted

$$\langle 0,0,0\rangle,\ \langle 0,1,0\rangle,\ \langle -1,1,0\rangle,\ \langle 0,0,1\rangle,\ \langle -1,0,1\rangle,\ \langle -1,1,1\rangle, \tag{2.7}$$

i.e. the point with index $\frac{1}{2}\langle -1,1,1\rangle$, this being at a distance $\frac{1}{2}\sqrt{2}$ from the origin and hence distant $\frac{1}{2}(\sqrt{2}-1)$ from the unit sphere with centre at $(0,0,0)$. Each of the other two members of the triplet is the mid-point of a tetrahedron, these two tetrahedra for the unit sphere with centre $(0,0,0)$ being defined by the unit sphere centres with indexes

$$\langle 0,0,0\rangle,\ \langle 1,0,0\rangle,\ \langle 0,1,0\rangle,\ \langle 0,0,1\rangle,\quad\text{and}\quad \langle 0,0,0\rangle,\ \langle 0,0,1\rangle,\ \langle -1,0,1\rangle,\ \langle 0,-1,1\rangle. \tag{2.8}$$

Thus, the centres of the tetrahedra have indexes $\langle\frac{1}{4},\frac{1}{4},\frac{1}{4}\rangle$ and $\langle-\frac{1}{4},-\frac{1}{4},\frac{3}{4}\rangle$ respectively, implying that each of the centres of the tetrahedra is distant $\frac{1}{2}\sqrt{\frac{3}{2}}$ from $(0,0,0)$. At these three points spheres of diameters $\sqrt{2}-1$, $\sqrt{\frac{3}{2}}-1$ and $\sqrt{\frac{3}{2}}-1$ can be 'close-packed' into the interstices we have described. Adding them to the unit spheres increases the volume fraction to

$$\frac{\pi}{\sqrt{18}}\left[1+\big(\sqrt{2}-1\big)^3+2\Big(\sqrt{\tfrac{3}{2}}-1\Big)^3\right]=0.809916;$$

indeed, adding only the largest of these interstitial spheres already increases the volume fraction from $\pi/\sqrt{18}=0.740480$ to 0.793104. Inspection shows we have recovered the volume fractions at (2.2) and (2.3). □

We now describe two more close-packed families of spheres, again with centres at points of a lattice, but they are not the densest such packings, and not all spheres are of the same size.

Example 3. *Unit spheres on the cubic lattice.* Start with unit spheres with centres at the points $\{(n,m,\ell): n,m,\ell=0,\pm1,\ldots\}$ of the cubic lattice. These spheres yield a volume fraction $\frac{4}{3}\pi(\frac{1}{2})^3=\frac{1}{6}\pi=0.523599$. At the centre of each unit cube of the lattice a sphere of diameter $\sqrt{3}-1$ can be located, touching each of the eight unit spheres centred at the closest vertices. Adding such spheres increase the volume fraction by $\frac{4}{3}\pi\big(\frac{1}{2}(\sqrt{3}-1)\big)^3=\frac{1}{6}\pi(6\sqrt{3}-10)\approx 0.205410$, so

the total volume fraction equals $\frac{1}{2}\pi(2\sqrt{3}-3) = 0.729009$, not too much smaller than 0.740480 as in Examples 1 and 2 using close-packed layers of unit spheres. Because these layers of spheres of diameter $\sqrt{3}-1$ are locked in place by the adjacent layers of unit spheres, and because given two adjacent layers of such smaller spheres the enclosed layer of unit spheres would be locked in position, the configuration at this stage is a packing that does not admit any movement of its constituent spheres, in the same way as for the spheres in the close-packings of Examples 1 and 2.

It is tempting to think that we can now place a sphere at the centre of each face of the cube, touching the four spheres centred on the vertices of the face, and therefore with diameter $\sqrt{2}-1$, but because $\sqrt{2}-1+\sqrt{3}-1 > 1$, such a sphere would overlap the sphere of diameter $\sqrt{3}-1$ already located at the centre of the cube. Two possibilities based on accommodating such a sphere ensue. For one, shrink the sphere at the centre of the cube to diameter $2-\sqrt{2}$, so that the total volume fraction is then

$$\tfrac{1}{6}\pi\big(1+(2-\sqrt{2})^3+3(\sqrt{2}-1)^3\big) = \tfrac{1}{6}\pi\big(1+(2\sqrt{2}+3)(\sqrt{2}-1)^3\big) = \tfrac{1}{6}\pi\sqrt{2} = \frac{\pi}{\sqrt{18}}. \tag{2.9}$$

For the other, locate the centre at $(\frac{1}{2}-a, 0, \frac{1}{2})$ say, for some $0 < a < \frac{1}{2}$ so that it is equidistant from the surfaces of the unit spheres with centres at $(0,0,0)$ and $(0,0,1)$ and the spheres of diameter $\sqrt{3}-1$ with centres at $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ and $(\frac{1}{2},-\frac{1}{2},\frac{1}{2})$. The radius r of a sphere centred at such a point is

$$r = \sqrt{(\tfrac{1}{2}-a)^2+(\tfrac{1}{2})^2}-\tfrac{1}{2} = \sqrt{a^2+(\tfrac{1}{2})^2}-\tfrac{1}{2}(\sqrt{3}-1), \tag{2.10}$$

equivalently,

$$a = \tfrac{1}{2}-\sqrt{(r+\tfrac{1}{2})^2-(\tfrac{1}{2})^2} = \sqrt{\big(\tfrac{1}{2}(\sqrt{3}-1)+r\big)^2-(\tfrac{1}{2})^2},$$

hence $(2-\sqrt{3})r+\frac{1}{2}(\sqrt{3}-1) = \sqrt{(r+1)r}$, and thus $4\sqrt{3}\,r^2+6r-1=0$, so, finally,

$$r = \frac{-3+\sqrt{9+4\sqrt{3}}}{4\sqrt{3}} = \frac{1}{3+\sqrt{9+4\sqrt{3}}} = \frac{1}{3+(1+\sqrt{3})\sqrt{3-\frac{1}{2}\sqrt{3}}} = 0.143041.$$

The increment in the volume fraction equals the volume of three such spheres, namely 0.036778, and thus a total volume fraction 0.765787. Note that $a = 0.095647$ so the sphere with radius r does indeed cover the centre $(\frac{1}{2},0,\frac{1}{2})$ of the face. □

Example 4. *Aligned layers of unit spheres on a triangular lattice.* Finally here is an analogue of Example 3 but now via layers of unit spheres whose centres are at the vertices of a triangular lattice as in Example 2. For this example place the layers one above the other, touching in the direction of the z-axis so that the centres are located at $(n+\frac{1}{2}m, \frac{1}{2}m\sqrt{3}, \ell)$ $(n,m,\ell = 0,\pm1,\ldots)$. The points $(n+\frac{1}{2}m+\frac{1}{2}, \frac{1}{2}m\sqrt{3}+\frac{1}{6}\sqrt{3}, \ell+\frac{1}{2})$ are located centrally to six adjacent unit spheres, at the same distance $\frac{1}{2}\sqrt{\frac{7}{3}}$ from each of their centres. Placing spheres of common diameter $\sqrt{\frac{7}{3}}-1$ at these points locks the array of spheres in position, and allows more spheres of the same diameter

to be placed at the other lattice points $(n+\frac{1}{2}m, \frac{1}{2}m\sqrt{3}+\frac{1}{3}\sqrt{3}, \ell+\frac{1}{2})$ because these two sets of spheres do not overlap. These two sets of spheres of diameter $\sqrt{\frac{7}{3}}-1$ increase V_f from

$$\frac{\frac{4}{3}\pi(\frac{1}{2})^3}{\frac{1}{2}\sqrt{3}} = \frac{\pi}{\sqrt{27}} = 0.604600 \quad \text{to} \quad \frac{\pi}{\sqrt{27}}\Big(1+2\big(\sqrt{\tfrac{7}{3}}-1\big)^3\Big) = \frac{\pi}{\sqrt{27}}\Big(\tfrac{32}{3}\sqrt{\tfrac{7}{3}}-15\Big) = 0.782111. \quad (2.11)$$

□

3. Dense sparse-packings

This section, which seemingly digresses from the topic of 'packing', is relevant in giving some perspective on simulations of models that proceed by sequentially placing grains in space. The first example is the antithesis of close-packing, and is best summarized in the following statement.

Conjecture 1. *In $\mathbb{R}^3$, any rigid configuration of unit spheres which precludes the insertion of any more unit spheres without overlapping, has volume fraction $\geq (V_f)_{\min} \equiv \frac{1}{12}\pi = 0.261799$.*

We call such an array a *dense sparse-packing*: we arrived at the conjectured lower bound as follows.

Example 5. Refer back to the close-packed layers of 'square' arrays of unit spheres in Example 1. Recall that the largest spheres that can fit into the close-packing — call them interstitial spheres for convenience — are of radius $\frac{1}{2}\big(\sqrt{2}-1\big)$, and that such largest interstitial spheres are in one–one correspondence with those of the packing. Then if we shrink the radii of the unit spheres of the packing by an amount δ say, the common radius of these largest interstitial spheres can increase to $\frac{1}{2}\big(\sqrt{2}-1\big)+\delta$, and therefore, the infimum of the volume fraction of the shrunken original spheres that precludes the insertion of any spheres of the same radius, is that of an array with radius a_0 say, for which

$$a_0 = \tfrac{1}{2}(\sqrt{2}-1)+\delta = \tfrac{1}{2}-\delta, \qquad \text{hence} \qquad a_0 = \tfrac{1}{2}-\tfrac{1}{4}(2-\sqrt{2}) = \frac{1}{2\sqrt{2}},$$

and therefore

$$V_f \geq \frac{\pi}{\sqrt{18}}\left(\frac{1}{\sqrt{2}}\right)^3 = \frac{\pi}{12}. \quad (3.1)$$

□

Example 6. Applying the same argument to the array of centres in Example 2 also leads to $V_f \geq \frac{1}{12}\pi$. □

The fact that the arrays of centres in both Examples 5 and 6 are 'dense' with respect to close-packing, suggests to us that they are also 'dense' with respect to the limit of 'sparsely arranged' spheres that do not allow any more spheres to be added, as implied in Conjecture 1.

For the purpose of the model described in Section 4 and evaluated by simulation, such dense sparse-packings of spheres are too scattered: we are interested in arrays in which there is at least

one, and maybe infinitely many, 'systems' of spheres each of which touches at least two other spheres. Typically, for our purposes, it is convenient to refer back to Examples 1–4 which are built up from a 'line' of touching spheres, and ask how sparsely these lines may be located so as not to allow any more spheres to be located between them.

Example 7. Taking our cue from the constructions leading to $(V_f)_{\min} = \frac{1}{12}\pi$, we start with the array of Example 1 and rescale the locations of the centres in the y- and z-directions by factors b and c respectively, such that the largest interstitial spheres with centres at $(n, mb, \frac{1}{2}\sqrt{2}\,c)$ for example are of radius $< \frac{1}{2}$. Then the sparsest arrangement of spheres, still with their centres unit distance apart in the x-direction, occurs when for example $(0, 0, \frac{1}{2}\sqrt{2}\,c)$ is distance $\frac{1}{2}$ from $(0, 0, 0)$ and $(\frac{1}{2}, \frac{1}{2}b, \frac{1}{2}\sqrt{2}\,c)$. Then $c = \sqrt{2} = b$, and the volume fraction of the 'lines' of spheres when the largest interstitial spheres are smaller than unit spheres, satisfies

$$\inf(V_f) = \tfrac{1}{2}\big(\pi/\sqrt{18}\big) = 0.370240. \tag{3.2}$$

□

Example 8. To scale the locations of the centres of layered triangularly configured spheres of Example 2, when the line of sphere centres $\{(n + \frac{1}{2}, \frac{1}{2}\sqrt{3}, 0)\}$ is relocated to $\{(n + \frac{1}{2}, \frac{1}{2}\sqrt{3}\,b, 0)\}$ the line of centres $\{(n+\frac{1}{2}, \frac{1}{6}\sqrt{3}\,b, \sqrt{\frac{2}{3}}\,c)\}$ is likewise at a distance $\frac{1}{2}\sqrt{3}\,b$ from the line through $\{(n, 0, 0)\}$ when $\frac{1}{12}b^2 + \frac{2}{3}c^2 = \frac{3}{4}b^2$, i.e. $b = c$. Then the interstitial sphere with centre $\big(0, \sqrt{\frac{1}{3}}\,b, \sqrt{\frac{1}{6}}\,c\big)$ is of radius $< \frac{1}{2}$ when it is distant < 1 from the points $(0, 0, 0)$, i.e. $b = c < \sqrt{2}$. Thus, we obtain the same relation as at (3.2). □

4. Concrete and a hard core model of touching spheres

Concrete is made by hydrating a mixture of aggregate, sand and cement: adding the water brings about chemical change in the cement that 'locks' into position whatever arrangement of the aggregate particles (and sand, and fine cement) is achieved prior to 'setting'. For present purposes we regard the cement as constituting a paste of infinitesimal thickness, and the sand as being indistinguishable from smaller aggregate. Aggregate often consists of pebbles from a river bed and therefore smooth-surfaced and approximately spherical, but may also be crushed rock and more jagged, though this can produce concrete of inferior strength. In practice the aggregate has a volume fraction between 60% and 70%; its complement is largely void. To suggest, even with spherical pebbles, that an approximately regular array is attained, is far from the truth: for one thing a range of particle sizes is used, and for another, their relative arrangement is irregular.

One can regard the effect of mixing dry aggregate (and cement) as arranging the particles as best can be done so as to attain an approximately homogeneous distribution, achieved in part from both the jostling of the mixing process and the help of gravity as the whole is tumbled. Inherent in particle size is an hierarchy: a much larger particle will tend to displace a smaller one a little so as to accommodate itself better — certainly this tends to occur when the mixture is nudged a little.

Therefore, to simulate the arrangement of particles in concrete, it is not unreasonable to allocate 'large' particles according to some partly random homogeneous mechanism, subject to their not overlapping and insisting that every particle touches another; then when no more such particles can be fitted into the space being 'filled', to fit as best one can particles of a slightly smaller size, again until no more will fit; this process is continued as smaller and smaller particles are added to the agglomerate. Throughout, every particle touches at least one other (and likely more, though for simplicity's sake we do not pursue this idea here).

The procedure just sketched simulates a hard-core germ–grain model, i.e. one with non-overlapping spherical grains, in which every grain touches at least one other grain, and there is only one 'cluster' of grains (for our purposes the cluster $\mathcal{C}$ to which a given grain $\mathcal{G}$ belongs, consists of $\mathcal{G}$ and all the grains $\mathcal{G}'$ that $\mathcal{G}$ touches, and all grains $\mathcal{G}''$ that some $\mathcal{G}'$ touches, and so on). In the models that we study we are interested in

(a) the volume fraction, and

(b) the distribution of grain sizes,

in the set Ξ formed by the union of all the grains $\mathcal{G}$, $\mathcal{G}'$, $\mathcal{G}''$, ... In particular, can we discern

(c) what factors affect the volume fraction, and how?

Sequential touching model. Sequentially place in a region, taken here to be a sphere $\mathcal{S}(1)$ of unit radius, spherical grains of somewhat smaller radii, using first spheres of the same radius $R_1 \leq \frac{1}{2}$, then of radii r in the range $R_2 \leq r \leq R_1$, ..., and finally $R_\nu \leq r \leq R_1$, for some decreasing sequence of bounds $R_1 > R_2 > \ldots > R_\nu$, ensuring for all spheres after the first that the sphere being newly located in $\mathcal{S}(1)$ is contiguous with another. As many larger spheres are placed as is reasonably practical to check for possible sites, before considering the placement of any smaller spheres. Let $\mathcal{S}(x, r)$ denote a sphere of radius r with centre at $x \in \mathbb{R}^3$. Since our simulation takes place in a 'large' sphere $\mathcal{S}(1)$, it is convenient to call the smaller spheres of radii $\leq R_1$ *balls*.

Specifically,[1] given a decreasing sequence $\{R_j\}$, the balls have centres x_i and radii r_i that are determined sequentially as follows.

1. Initially place two balls of radius R_1 with their centres at the points $(\pm R_1, 0, 0)$ respectively (assume $R_1 \leq \frac{1}{2}$).

Let n_p denote the number of balls placed at any stage, so after step 1 we have $n_p = 2$. Below, s ranges over $\{1, \ldots, \nu\}$, and $N_{\rm rpt}$ denotes a 'repetition counter'; initially, $s = 1$ and $N_{\rm rpt} = 0$.

2_s. Generate a point at random in $\mathcal{S}(1)$, at $x_{\rm test}$ say. Before locating a ball of radius r with $R_s \leq r \leq R_1$ 'near' $x_{\rm test}$ (see step 3_sa for detail), first check that various conditions are met:

2_sa. First check that $|x_{\rm test}| + R_s \leq 1$, so that a ball $\mathcal{S}(x_{\rm test}, R_s)$ would lie wholly within $\mathcal{S}(1)$. If so, continue at step 2_sb; otherwise, repeat step 2_s.

2_sb. For each ball $\mathcal{S}(x_i, r_i)$ already placed in $\mathcal{S}(1)$, i.e. for $i = 1, \ldots, n_p$, check that $|x_{\rm test} - x_i| \geq r_i + R_s$ (i.e. $\mathcal{S}(x_{\rm test}, R_s)$ would not overlap $\mathcal{S}(x_i, r_i)$); if this fails for any such i, go to step

[1] This model is a variant of one suggested by Alan Karr.

3_sb. If the check passes for all such i, identify the ball $\mathcal{S}(x_{i'}, r_{i'})$ whose surface is closest to $x_{\rm test}$, and go to step 3_sa.

3_sa. Advance n_p to n_p+1 and place a ball of radius $r_{n_p} = \min\left(R_1, |x_{\rm test} - x_{i'}| - r_{i'}\right)$ with its centre at $x_{\rm test}$ if $r_{n_p} < R_1$, or else at

$$x_{n_p} = \alpha x_{\rm test} + (1-\alpha)x_{i'} \qquad \text{where} \qquad \alpha = \frac{R_1 + r_{i'}}{|x_{\rm test} - x_{i'}|}\,. \tag{4.1}$$

For section 5.2, record $t_{n_p} = N_{\rm rpt}$. Then reset $N_{\rm rpt} = 0$ and return to step 2_s.

3_sb. Reject $x_{\rm test}$, advance $N_{\rm rpt}$ by 1, and provided $N_{\rm rpt} < N_s$ for this pre-determined limit N_s on the number of repetitions when seeking to place balls of radius $\geq R_s$, repeat step 2_s. If $N_{\rm rpt} = N_s$, then if $s < \nu$, advance s by 1, reset $N_{\rm rpt} = 0$ and return to step 2_s; else, $s = \nu$, and go to step 4.

4. The volume fraction of the germ–grain realization $\{\mathcal{S}(x_i, r_i) : i = 1, \ldots, n_p\}$ equals $\sum_{i=1}^{n_p} r_i^d$.

Variations of this procedure produce different volume fractions: for example (cf. Table 5 below for 3_sa$'$) we could replace step 3_sa by either of

3_sa$'$. Advance n_p to $n_p + 1$ and place a ball of radius R_s at

$$x'_{n_p} = \alpha x_{\rm test} + (1-\alpha)x_{i'} \qquad \text{where} \qquad \alpha = \frac{R_s + r_{i'}}{|x_{\rm test} - x_{i'}|}\,. \tag{4.1$'$}$$

3_sa$''$. Advance n_p to $n_p + 1$ and place a ball of radius R_s at $x_{\rm test}$.

The volume fraction computed at step 4 is likewise dependent on the stopping rule for the simulation at step 3_sb; in practice we must also limit the number of balls placed (i.e. $n_p \leq n_{\max}$ say), and apply this constraint if it happens before $s = \nu$ and $N_{\rm rpt} = N_\nu$ as in 3_sb.

Table 1 shows some of the results of 20 replicate simulations using $R_{s+1} = 0.95R_s$, $N_s = 10{,}000$ (all s) and a maximum of 10,000 balls placed (in all simulations it was this constraint that was invoked rather than the limit on minimum radius size R_ν). The numbers of balls of maximum radius R_1 and of any radius placed after completion of steps 2_1 and 2_{35} are shown, the volume fraction attained at the end of step 2_{35}, and the volume fraction attained after placing 10,000 balls.

It is evident in Table 1 that the volume fraction attained by placing the largest balls of radius R_1, with mean 0.31545, is far smaller than the value of V_f after smaller balls are allowed (for the record, $R_{35}/R_1 = 0.95^{34} = 0.1840$). It also apparent that the volume fraction seen at the conclusion of the simulation, varies far less than the volume fraction seen after step 2_1, because balls with a range of intermediate sizes fill much of the space, until eventually no voids are larger than the order of the smallest radius R_{35} (= 0.0184 in Table 1) used towards the end of the procedure. The range in the observed $V_f[s \leq 35]$, namely 0.004886, is larger than the range 0.003823 in the observed V_f[10,000 placed]. This is understandable in that the stopping rule for switching minimum sizes from R_s to R_{s+1} induces variability over and above the variation in volume of some 1,000 balls of radius R_{35} for example.

Table 1

Some results from 20 simulations of the sequential touching model
($R_{s+1} = 0.95\,R_s$, $R_1 = 0.1$, $N_s = 10,000$)

Sim'n no.	$\#(R_1)$	$\#(s \le 35)$	$V_f[s \le 35]$	V_f[10,000 placed]
1	320	8297	0.681586	0.690995
2	312	8704	0.682907	0.690081
3	318	9016	0.682689	0.688111
4	318	8610	0.681307	0.688760
5	320	8629	0.681986	0.689602
6	312	8165	0.679729	0.689530
7	314	8429	0.681391	0.689832
8	309	8408	0.679947	0.688858
9	312	8515	0.679977	0.688209
10	308	8015	0.678452	0.689534
11	308	8693	0.681139	0.687988
12	323	8258	0.680448	0.689789
13	315	8689	0.682785	0.689901
14	317	8185	0.681838	0.691811
15	315	8071	0.678021	0.688702
16	313	8527	0.681556	0.689626
17	304	8506	0.680871	0.688952
18	327	8076	0.678782	0.689239
19	314	8402	0.680984	0.689734
20	330	8649	0.682574	0.690056

5. Factors affecting the volume fraction in the sequential touching model

The aim of much of our simulation work has been to attain the largest possible volume fraction by using about 5,000 or 10,000 balls. Doubtless some volume fraction higher than $c.\,70\%$ is attainable by some more complex sequential filling strategy, because close packing yields a higher fraction; we have been guided in part by studying a simple model of the type described. In this model, we can easily identify certain factors that affect the volume fraction achieved in a simulation, and also factors that limit the volume fraction attainable in any model that uses spherical grains (and, in particular, in a model that starts from balls of the same size and requires every ball to touch at least one other ball).

We list here some questions and factors which have arisen in the present context of placing spheres of radii $\le R_1$ in $\mathcal{S}(1)$.

5.1. Edge effects in estimating the volume fraction of homogeneous spheres

It is reasonable to start by asking what influence the largest balls or spheres, of radius R_1, may have on the volume fraction: it is trite to note that it is in the 'channels' around these largest balls that balls of smaller radii are located, thereby raising the volume fraction. To this end we placed as many balls of radius $\frac{1}{30}$ in a sphere $\mathcal{S}(1)$ of unit radius, noting the numbers of such balls that had been placed when for the first time $1000n$ ($n = 1, \ldots, 10$) fruitless attempts at placing another ball occurred as in step 3b. We then repeated the simulation, first with the same $R_1 = \frac{1}{30}$,

and then with increasing sizes of R_1, with the results as shown in Table 2 in which, for example, for $R_1 = 0.05$, $(V_f)_{\rm obs} = 2634 \times (.05)^3 = 2.634/8 = 0.32925$.

Table 2

Numbers of uniform spheres placed at different rejection-number passage times, and V_f attained

Rej.#: R_1	1000	2000	3000	4000	5000	6000	7000	8000	9000	10000	$(V_f)_{\rm obs}$	$(V_f)_{\rm adj}$	$(V_f)_{\rm adj'}$
0.0333	8063	8431	8627	8674	8674	8851	8851	8950	9037	9117	0.33767	0.35184	0.37370
0.0333	7947	8353	8537	8746	8844	8862	8862	8928	8995	9024	0.33422	0.34824	0.36989
0.0350	6847	7113	7364	7457	7627	7627	7697	7697	7815	7815	0.33507	0.34985	0.37286
0.0375	5556	5846	5902	6037	6037	6037	6215	6346	6346	6346	0.33465	0.35050	0.37531
0.0400	4632	4670	4892	4916	4916	5090	5090	5090	5170	5213	0.33363	0.35052	0.37710
0.0425	3803	3984	4099	4099	4188	4188	4310	4319	4319	4319	0.33155	0.34942	0.37769
0.0450	3244	3326	3485	3485	3594	3594	3594	3604	3604	3604	0.32841	0.34720	0.37706
0.0500	2333	2371	2537	2582	2593	2598	2598	2634	2634	2634	0.32925	0.35026	0.38402
0.0550	1764	1847	1847	1892	1892	1915	1977	1977	1981	1984	0.33009	0.35336	0.39114
0.0600	1361	1361	1456	1456	1456	1488	1488	1513	1513	1516	0.32746	0.35275	0.39425

Inspection of the ten columns of data entries in Table 2 together with the volume fraction $(V_f)_{\rm obs}$ attained when simulation stopped, shows two obvious features:

(a) The smaller the radius the larger the observed volume fraction. This increase is due to the reduction in the volume subject to an 'edge effect', and the rest of this subsection discusses this factor.

(b) The larger the rejection number count causing simulation to cease, the larger the observed volume fraction. See also subsection 5.2.

We remark in passing that, from a practical point of view, concrete is always found in a finite space, so the volume fraction observed in concrete will always include an 'edge effect'. However, in order to have any basis for general computation, we should first seek data that relate to no edge effect, because of their generality, and then apply a correction for the edge effect appropriate to the geometry of the particular configuration concerned.

To estimate an edge-correction for data like those giving $(V_f)_{\rm obs}$ as in Table 2, observe that to a first order of approximation, when balls of radius $R_1 \ll 1$ are placed in a sphere $\mathcal{S}(1)$, any ball whose centre is at a distance between R_1 and $3R_1$ of the surface of $\mathcal{S}(1)$, thereby excludes the subsequent location of any ball within a certain larger or smaller portion of $\mathcal{S}(1)$. Consequently, when no more balls can be placed in $\mathcal{S}(1)$, the average distance between the 'surface' of the set Ξ and the surface of $\mathcal{S}(1)$, is about γR_1, say, for some positive constant $\gamma < 1$, implying that the 'effective volume' into which the balls of radius R_1 have been placed is not $\frac{4}{3}\pi$ but about $\frac{4}{3}\pi(1-\gamma R_1)^3 \approx \frac{4}{3}\pi(1-3\gamma R_1)$, and thus

$$V_f \approx \frac{(V_f)_{\rm obs}}{(1-\gamma R_1)^3}. \tag{5.1}$$

For the simulations whose results are as noted in Table 2, the last column, $(V_f)_{\mathrm{adj}'}$, shows the corrected volume fractions using the value $\gamma = 1$ which, both numerically and on further consideration (see next paragraph), is certainly too large: this adjustment of the observed volume fractions produces estimates that increase with increasing R_1; indeed, applying equation (5.1) to the mean observed volume fraction 0.31545 from balls of largest radius $R_1 = 0.1$ in Table 1 gives $(V_f)_{\mathrm{adj}'}$ for homogeneous spheres equal to 0.43272, which lies well outside the range 0.37 to 0.395 of Table 2).

In the limit where the radius R_1 becomes a vanishingly small proportion of the radius 1 of $\mathcal{S}(1)$, we can regard the surface of the latter big sphere as being like a plane relative to balls located close to its surface. In the densest possible configuration of balls near its surface we could have three mutually touching balls with their centres located at a distance $\left(1 + \sqrt{\frac{2}{3}}\right) R_1$ from the surface, and be able to place a further ball inside the sphere so as to touch the surfaces of $\mathcal{S}(1)$ and these three balls (cf. the packing in Example 2). This extreme configuration justifies seeking some constant $\gamma < 1$ in the edge-correction formula at (5.1). Indeed, because any other configuration of balls all at a distance at least $\sqrt{\frac{2}{3}}\, R_1$ from the surface of $\mathcal{S}(1)$ would allow such a further ball to be placed closer to the surface of $\mathcal{S}(1)$, we can argue that we should regard the 'surface' of Ξ as being at an average distance of about $1 - \frac{1}{2}\sqrt{\frac{2}{3}}\, R_1 = 1 - \sqrt{\frac{1}{6}}\, R_1$ from the centre of $\mathcal{S}(1)$. Then $\mathcal{S}\left(1 - \sqrt{\frac{1}{6}}\, R_1\right)$ would have the same volume as the 'space' in which Ξ is located, and (5.1) should hold with $\gamma \approx \sqrt{\frac{1}{6}} = 0.408248$. The edge-corrected estimates $(V_f)_{\mathrm{adj}}$ in Tables 2 and 3 come from equation (5.1) with $\gamma = \sqrt{\frac{1}{6}}$.

Having thus argued that $V_f \approx (V_f)_{\mathrm{obs}} / (1 - \gamma R_1)^3$, it would follow that

$$\sqrt[3]{(V_f)_{\mathrm{obs}}} \approx \sqrt[3]{V_f} - \gamma \sqrt[3]{V_f}\, R_1. \tag{5.2}$$

Then a regression fit of the relation $\sqrt[3]{(V_f)_{\mathrm{obs}}} = a + bR_1$, say, for fitted constants a and b, yields

$$\mathrm{Est}(V_f) = a^3 \qquad \text{and} \qquad \mathrm{Est}(\gamma) = -b/a. \tag{5.3}$$

When applied to the ten points $(R_1, (V_f)_{\mathrm{obs}})$ in Table 2, we obtained $\mathrm{Est}(V_f) = 0.34612$ and $\mathrm{Est}(\gamma) = 0.31502$.

The results in Table 2 suggest that stopping after 10,000 failed attempts to find a point x_{test} at least R_1 from any ball already placed in $\mathcal{S}(1)$, does not necessarily ensure that (almost) none of the interstices then in the agglomerate can accommodate a ball $\mathcal{S}(R_1)$, particularly for smaller values of R_1. Accordingly sets of 25 simulations using a range of the larger sizes R_1 were run, with a more stringent 'rejection number' $N_1 = 50,000$, yielding for each R_1 as in Table 3 the average and standard deviation of the number of balls placed, and the mean observed volume fraction; we also ran replicates of the sizes $R_1 = 0.05\,(0.005)\,0.065$. The larger is R_1, so the more likely we are to fill all interstices that will take a ball $\mathcal{S}(R_1)$; we do not know the size of this error nor how it affects our estimate of $V_f(\cdot)$ (but, see subsection 5.2).

Using regression as around (5.2) now leads to Est$(V_f) = 0.36421$ and Est$(\gamma) = 0.31394$. The larger estimate for V_f arises from the larger value of N_1; the consistency of the estimate for γ is encouraging, but we have not tried to explain why it might be like $\frac{3}{4}\sqrt{\frac{1}{6}}$ or $\frac{1}{10}\pi$.

Comparing these regression results, suggests that we may be able to estimate V_f itself by repeating the replicate studies of Table 3, say, for $N_1 = 12{,}500$ and $25{,}000$, and extrapolating via $1/N_1 \to 0$ (cf. subsection 5.2).

Table 3

Mean numbers of spheres of radius R_1 placed in $\mathcal{S}(1)$ by sequential touching model
($N_1 = 50{,}000$, Mean and s.d. of 25 simulations; edge-correction (5.1) with $\gamma = \sqrt{\frac{1}{6}}$.)

R_1	Mean #	SD #	$(V_f)_{\rm obs}$	SD $(V_f)_{\rm obs}$	$(V_f)_{\rm adj}$
0.100	331.92	4.80	0.33192	0.00096	0.37613
0.095	387.40	5.45	0.33215	0.00089	0.37400
0.090	456.72	6.90	0.33295	0.00101	0.37252
0.085	547.76	7.72	0.33639	0.00095	0.37399
0.080	657.12	9.56	0.33645	0.00098	0.37169
0.075	806.16	9.08	0.34010	0.00076	0.37336
0.070	993.16	7.73	0.34065	0.00053	0.37161
0.065	1247.56	10.76	0.34261	0.00059	
	1247.68	11.71	0.34264	0.00065	0.37142
0.060	1593.92	15.85	0.34429	0.00070	
	1593.36	16.49	0.34417	0.00073	0.37082
0.055	2079.48	16.97	0.34597	0.00056	
	2072.20	16.63	0.34476	0.00055	0.36971
0.050	2781.36	18.82	0.34767	0.00047	
	2775.12	26.69	0.34689	0.00067	0.36945

One possible property to be borne in mind — is it Rankin's problem ? — is work of D. G. Kendall in two papers *c.* 1940s (? *J. London Math. Soc.*) on the 'error' term for the number of points on the square lattice enclosed by a 'large' circle, and the analogous 3-D problem.

5.2. Exploiting the rejection numbers between successfully placing uniform spheres

We turn to problem (b) noted in connection with Table 2, and further exemplified by comparing Tables 2 and 3. It is proper to comment that in Table 3 the observed standard deviation in the number of balls placed in $\mathcal{S}(1)$ reflects two factors:

(a) Genuine variability in the number of grains dispersed according to a specified protocol within a finite region.

(b) The inherent uncertainty associated with randomly searching for possible locations where balls may be placed in a random sequential filling protocol.

Here, factor (a) is irrelevant: it simply emphasizes that the volume fraction V_f of a germ–grain model is indeed a mean: it equals an ergodic limit for suitably defined models. Factor (b) is what we wish to address, noting first that, paradoxically, using smaller values of R_1 should improve $(V_f)_{\rm obs}$ as an estimate because the edge effect noted earlier is reduced, but the number of (tiny) regions

where the centre must be located in order to place the 'last few' remaining balls may increase relative to the number for larger R_1, so on this account any simulation is more likely to make $(V_f)_{\rm obs}$ an under-estimate. Given the nature of extrapolation from regression-based estimators, $\mathrm{Est}(V_f) = a^3$ as at (5.2) is therefore biased downwards from V_f.

In the context of steps 1–4 of the simulation procedure, the study of V_f using only homogeneous balls constitutes the case $s = \nu = 1$, so we can omit the suffix s. Suppose that in between placing the i th and $(i+1)$ th balls there are t_i repetitions of step 2 that result in failure due to a potential overlap with a ball placed previously and hence invoke step 3b. Then, $t_2, t_3, \ldots$ are the consecutive values of $N_{\rm rpt}$ before it is reset to 0 as in step 3a. Assuming (as we must!) that the random number generator is indeed producing points $x_{\rm test}$ that are independently and uniformly distributed in $\mathcal{S}(1)$, each t_i is a geometrically distributed random variable with probability q_i, say of success, i.e. $\Pr\{t_i = n\} = q_i(1-q_i)^n$, $(n = 0, 1, \ldots)$, where the $q_i \equiv q(x_1, \ldots, x_i)$ are themselves random variables that depend on the centres of balls already placed; necessarily, $q_i \downarrow$ for $i \uparrow$, and $N(\omega) \equiv \inf_i\{i{:}\, q_i = 0\}$ determines exactly the observed volume fraction $N(\omega)R_1^3$ of a particular realization. Further, $\mathrm{E}[N(\omega)]$ equals the mean volume fraction for the finite germ–grain sequential touching model, i.e. of whatever corresponds to $(1-\gamma R_1)^3 V_f$ where V_f is the volume fraction in the sequential touching model with $\nu = 1$ in the case of an infinitely large region (equivalently, for arbitrarily small radius R_1 for the balls).

One possible way of estimating $N(\omega)$ is to exploit the random sequence consisting of the conditionally independent geometrically distributed random variables $\{t_i\}$ with (conditional) expectations $(1-q_i)/q_i$. As an approximation, suppose that, conditional on the unknown $N(\omega)$, there is an $\alpha > 0$ such that we can use the relation $q_i \approx [N(\omega) - i]\alpha$ for i sufficiently close to $N(\omega)$. The rationale behind this approximation is that for i close to $N(\omega)$, there are $N(\omega) - i$ isolated regions, with average volume $\alpha \ll \mathrm{vol}\big(\mathcal{S}(R_1)\big)$, in which a centre of a ball $\mathcal{S}(R_1)$ can be placed without the ball overlapping any $\mathcal{S}(x_j, R_1)$ $(j = 1, \ldots, i)$. See also Section 5.6 concerning sets $\mathrm{Sph}(\cdot)$ and $\mathrm{Cvr}(\cdot)$.

¿From these approximations there follows an approximation to the conditional likelihood L_k of the last k observations $t_{n_p - j}$ $(j = 1, \ldots, k)$, namely

$$L_k = \prod_{j=n_p-k}^{n_p-1} q_j(1-q_j)^{t_j} \approx \prod_{j=1}^{k} [N(\omega) - n_p + j]\alpha \Big[1 - [N(\omega) - n_p + j]\alpha\Big]^{t_{n_p-j}}, \qquad (5.4)$$

so that

$$\log L_k = \sum_{j=1}^{k} \log\left([N(\omega) - n_p + j]\alpha\right) + \sum_{j=1}^{k} t_{n_p-j} \log\left(1 - [N(\omega) - n_p + j]\alpha\right). \qquad (5.5)$$

If also the simulation terminates as in step 3b on the N_1 th failure after placing a ball at $\mathcal{S}(x_{n_p}, R_1)$, then in place of (5.4) we should have $\tilde{L}_k = L_k(1-q_{n_p})^{N_1}$, and for $\log \tilde{L}_k$ we should have (5.5) with an extra term $j = 0$ in the second sum and $t_{n_p} = N_1$.

The model here requires $0 \le [N(\omega) - n_p + k]\alpha \le 1$, and the extreme values here cannot be local maxima for the likelihood (or its logarithm), so we can identify the maximum of $\log L_k$ or $\log \tilde{L}_k$ by calculus, regarding $N(\omega)$ as a continuous parameter for convenience. This leads us to seek a solution $(\widehat{N}(\omega), \widehat{\alpha})$ for the pair of equations in $N(\omega)$ and α:

$$\sum_{j=1}^{k} \frac{1}{N(\omega) - n_p + j} = \sum_{j=1}^{k} \frac{\alpha t_{n_p - j}}{1 - [N(\omega) - n_p + j]\alpha} ,$$

$$\frac{k}{\alpha} = \sum_{j=1}^{k} \frac{t_{n_p - j}[N(\omega) - n_p + j]}{1 - [N(\omega) - n_p + j]\alpha} = \sum_{j=1}^{k} \frac{t_{n_p - j}}{\alpha} \left[-1 + \frac{1}{1 - [N(\omega) - n_p + j]\alpha} \right] .$$

Equivalently, with $\mu_j = [N(\omega) - n_p + j]\alpha$,

$$0 = \sum_{j=1}^{k} \frac{1 - (t_{n_p - j} + 1)\mu_j}{\mu_j (1 - \mu_j)} , \tag{5.6a}$$

$$\sum_{j=1}^{k} (1 + t_{n_p - j}) = \sum_{j=1}^{k} \frac{t_{n_p - j}}{1 - \mu_j} > \sum_{j=1}^{k} t_{n_p - j} + \sum_{j=1}^{k} t_{n_p - j} \mu_j . \tag{5.6b}$$

Inequality (5.6b) gives

$$\alpha < \frac{k}{\sum_{j=1}^{k} t_{n_p - j}[N(\omega) - n_p + j]} = \frac{1}{[N(\omega) - n_p]T_0 + T_1} , \tag{5.7}$$

where for $r = 0, 1$, $T_r = k^{-1} \sum_{j=1}^{k} j^r t_{n_p - j}$. Equation (5.6a) implies that

$$\frac{1}{\max_j (t_{n_p - j} + 1)[N(\omega) - n_p + j]} < \alpha < \frac{1}{\min_j (t_{n_p - j} + 1)[N(\omega) - n_p + j]} . \tag{5.8}$$

We have done some preliminary work* on estimating $N(\omega)$ on this basis, with encouraging results.

5.3. The fibre-like structure of the sequential touching model with uniform balls

In contemplating the structure of the tree-like set Ξ that ensues from step 3a of the sequential touching model with $\nu = 1$, it is worth reflecting that the point x_{test}, being uniformly distributed in $\mathcal{S}(1)$, is as likely to be within 0.206 of its surface as within $0.794 = \sqrt[3]{0.5}$ of its centre. Now, in the earlier stages of the growth of Ξ, any random point x_{test} nearer to the surface of $\mathcal{S}(1)$ leads to a ball being placed on the 'outer' side of an existing ball, thereby yielding a 'string' of balls generally heading out towards the surface of $\mathcal{S}(1)$. This means that the tree-like structure

* George Fishman (personal communication, March, 2000) tells me that this is an 'old' problem on which he has notes from about ten years ago, and referred me to both Jodrey and Tory (1980) and Hinde and Miles (1980) *same J.*, **10**, 205–223, though the latter paper is concerned with planar packings and random tesselations. [John Hinde was a chemist, so maybe the 3-D problem is addressed as well ?] Jeff Picka mentioned Jodrey and Tory about two years ago, but a 1985 paper.

of Ξ is initially like individual strands emanating outwards until there is space enough to 'split' and generate more outward-growing strands. If this is indeed the case then, because the fibres are touching in the outward direction but otherwise separated, their volume fraction may well approximate that of sparsely packed lines of balls as in Examples 7 and 8, namely $\frac{1}{2}\pi/\sqrt{18} = 0.370$ (see (3.2)). Whatever else, this is consistent with Est(V_f) from simulations (see especially Table 3).

5.4. Influence on $(V_f)_{\rm obs}$ of the sequences $\{N_s\}$ and $\{R_s\}$

Trivially, by increasing the 'stopping number' N_1 we can only increase $(V_f)_{\rm obs}$. Similarly, as follows from a conservation argument, the more space that is covered by larger balls, the fewer balls in all are needed to attain a particular value for $(V_f)_{\rm obs}$. This is borne out in simulations reported in both Tables 4 and 5 where the same sequence of points $x_{\rm test}$ is used throughout. Comparison of the simulations within either table shows that the use of larger N_s increases the number of balls placed of size at least R_s (in Table 4, with strategy 3_sa), and, less markedly, of size exactly R_s (in Table 5, with strategy 3_sa$'$).

Comparison of the results in Table 4 where $R_{s+1}/R_s = 0.975$, with those in Table 1 where the ratio equals 0.95 and in Table 6 where it equals $0.9025 = (0.95)^2$, shows up a similar result: that the more slowly the minimum radius R_s decreases, so the larger the observed volume fraction attained by the time given smaller-sized balls are placed. Comparison of the simulation with $N_s = 5{,}000$ in Table 4 with the twenty simulations in Table 1 with double the size of N_s and double the difference between 1 and the ratio R_{s+1}/R_s, suggests that the increase in N_s has greater influence in increasing the observed volume fraction. Note however that the strategy 3_sa is in effect, and this tends to minimize the influence of allowing smaller balls to be placed for given $s > 1$.

Table 4

Volume fractions from spheres of radii $R_s \le r \le R_1$ (strategy 3_sa)
($R_{s+1} = 0.975R_s$, $R_1 = 0.1$, N_s as shown)

	min.rad.	(a) $N_s = 5{,}000$		(b) $N_s = 20{,}000$	
s	R_s	$n_p(N_s)$	$V_f[r \ge R_s]$	$n_p(N_s)$	$V_f[r \ge R_s]$
1	0.1000	308	0.30800	320	0.32000
2	0.0975	322	0.32175	345	0.34455
3	0.0951	334	0.33314	354	0.35288
4	0.0927	339	0.33778	354	0.35288
5	0.0904	359	0.35502	375	0.36968
9	0.0817	400	0.38442	422	0.40258
13	0.0738	472	0.42090	491	0.43618
17	0.0667	542	0.44700	543	0.45527
21	0.0603	594	0.46198	655	0.48651
31	0.0468	947	0.52255	993	0.54007
41	0.0363	1494	0.56519	1637	0.58872
51	0.0282	2486	0.60401	2747	0.62799
61	0.0219	4616	0.64323	5077	0.66655
71	0.0170	8727	0.67793	8923	0.69721

When the strategy 3_sa$'$ is used, smaller volume fractions are observed as in Table 5.

Table 5

Volume fractions from spheres of minimum radius (strategy 3_sa$'$)
($R_{s+1} = 0.975R_s$, N_s as shown)

	radius	(a) $N_s = 5{,}000$		(b) $N_s = 10{,}000$		(c) $N_s = 20{,}000$	
s	R_s	$n_p(N_s)$	$V_f[r \le R_s]$	$n_p(N_s)$	$V_f[r \le R_s]$	$n_p(N_s)$	$V_f[r \le R_s]$
1	0.1000	308	0.30800	320	0.32000	320	0.32000
2	0.0975	315	0.31449	322	0.32184	326	0.32556
3	0.0951	325	0.32308	337	0.33474	349	0.34532
4	0.0927	345	0.33900	352	0.34668	361	0.35488
5	0.0904	360	0.35007	369	0.35923	367	0.35930
9	0.0817	412	0.38162	419	0.38912	425	0.39470
13	0.0738	460	0.40260	475	0.41443	489	0.42293
17	0.0667	548	0.43096	547	0.43883	562	0.44570
21	0.0603	638	0.45250	645	0.46318	639	0.46502
31	0.0468	972	0.49704	966	0.50924	1022	0.52015
41	0.0363	1740	0.54549	1664	0.55618	1765	0.56949
51	0.0282	3001	0.58258	3055	0.59864	3038	0.60859
61	0.0219	5226	0.61521	5396	0.63154	5370	0.64293
71	0.0170	9827	0.64784	$> 10^4$	0.66294	$> 10^4$	0.67541

Table 6

Numbers $\Delta_s(n_p)$ of spheres placed between changes in minimum radius
($R_{s+1} = (0.95)^2 R_s$, $R_1 = 0.1$, $N_s = 50{,}000$)

s	$\Delta_s(n_p)$	R_s	$V_f[r \ge R_s]$	av.radius
1	334	0.10000	0.334000	0.100000
2	57	0.09025	0.380886	0.093697
3	43	0.08145	0.407493	0.085213
4	78	0.07351	0.442278	0.076401
5	80	0.06634	0.469027	0.069407
6	118	0.05987	0.497313	0.062120
7	102	0.05404	0.515461	0.056244
8	152	0.04877	0.535499	0.050895
9	232	0.04401	0.558010	0.045952
10	240	0.03972	0.574935	0.041315
11	395	0.03585	0.595750	0.037491
12	397	0.03235	0.611128	0.033835
13	577	0.02920	0.627897	0.030745
14	722	0.02635	0.643157	0.027648
15	1071	0.02378	0.659781	0.024945
16	5000 pts give $(V_f)_{\rm obs} = 0.66447$			

The last column in Table 6 shows the 'average' radius of the $\Delta_s(n_p)$ balls placed at step 3_sa, i.e. while the minimum radius R_s is in force. This average is defined as $\sqrt[3]{\text{av. volume per grain}}$, the average volume being computed over the $\Delta_s(n_p)$ grains concerned. The fact that these 'average'

radii are closer to R_s than R_{s-1} should come as no surprise: because every ball already placed in $\mathcal{S}(1)$ touches another, there are more smaller empty regions (where $x_{\rm test}$ will not be rejected) than there are larger, no matter what the critical cut-off size. Of course, it is also relevant here that $N_s = 50{,}000$: a much smaller size for N_s would give rise to more spheres of radius $> R_{s-1}$ being placed at step 3_sa.

5.5. Variable v. fixed radii: strategies 3_sa and 3_sa′

For the record, we note that following strategy 3_sa and placing at or near $x_{\rm test}$ a ball with the largest radius r possible subject to $R_s \le r \le R_1$, as opposed to using a ball of radius R_s (strategy 3_sa′), leads to higher observed volume fractions (compare Tables 3 and 4).

5.6. 'Exclusion' zone around points of contact of spheres

This subsection is concerned with aspects of the following question.

Question 2. *During steps 2_s of the sequential touching model protocol, what proportion of the volume of $\mathcal{S}(1)$, say, is inaccessible to spheres of radius $\ge \kappa R_s$ for given κ in $(0,1)$?*

Two sets, $\mathrm{Sph}(\cdot)$ and $\mathrm{Cvr}(\cdot)$, are relevant to this question. They are defined in the context of a given compact set $\mathcal{S}$ (e.g. $\mathcal{S}(1)$) that is partly covered by a set X (e.g. $\Xi_i \equiv \bigcup_{j=1}^{i} \mathcal{S}(x_j, r_j)$). Define $Y \equiv X^c \equiv \mathcal{S} \setminus X$, and then

$$\mathrm{Sph}(Y; r) = \{y \in Y : \mathcal{S}(y, r) \subset Y\} \tag{5.9}$$

and

$$\mathrm{Cvr}(Y; r) = \bigcup_{y \in \mathrm{Sph}(Y;r)} \mathcal{S}(y, r). \tag{5.10}$$

In words, $\mathrm{Sph}(Y; r)$ consists of the points y in Y such that a sphere $\mathcal{S}(y, r)$ would lie totally outside X (except maybe for touching the surface of X). The larger set $\mathrm{Cvr}(Y; r)$ consists of all points in any sphere $\mathcal{S}(y, r)$ that has its centre at a point in $\mathrm{Sph}(Y; r)$.

In order to gauge the influence of such sets, we first compute the volume $A(r; R_1, R_2)$ say, of that part of the immediate neighbourhood of the point of contact of two spheres, of radii R_1 and R_2 say, that cannot be covered by spheres of radius larger than r. We evaluate the volume of this 'uncoverable' region, first in the case $R_1 = R_2 = R$ say. Consider the following three elements: the plane tangential to the two spheres, the axis perpendicular to this plane through the centres of the two spheres, and a line through the centre of one of the spheres at an angle θ to the axis directed towards the plane. Let the angled line meet the surface of the sphere at a distance $h(\theta)$ above the plane, at a distance $x(\theta)$ from the axis, so that $h(\theta) = R - R\cos\theta$ and $x(\theta) = R\sin\theta$. Observe that for a sphere of radius r to touch both spheres, a line through the centres of spheres of radii R and r respectively would make an angle θ' with the axis, where $(R + r)\cos\theta' = R$. Then the volume between the two touching spheres of radii R that cannot be covered by a sphere of radius r is

$$A(r; R, R) = \int_{\theta=0}^{\theta'} 2h(\theta)\, 2\pi x(\theta)\, dx - \int_{\psi=0}^{\frac{1}{2}\pi - \theta'} 2r\sin\psi\, 2\pi\left[(R+r)\sin\theta' - r\cos\psi\right] d(-r\cos\psi), \tag{5.11}$$

where the first term corresponds to the region up to a distance $x(\theta')$ from the axis where the centre of a sphere of radius r certainly cannot be placed, and the second term to the region within the same distance $x(\theta')$ of the axis that can be covered by a sphere of radius r placed so as to touch both spheres of radius R. The first term equals

$$\begin{aligned}\int_0^{\theta'} 2R(1-\cos\theta)\,2\pi R\sin\theta\,R\cos\theta\,d\theta &= 4\pi R^3\int_{\theta=0}^{\theta'}(\cos\theta-\cos^2\theta)\,d(-\cos\theta)\\ &= 4\pi R^3\Big[\tfrac{1}{2}(1-\cos^2\theta')-\tfrac{1}{3}(1-\cos^3\theta')\Big]\\ &= \tfrac{2}{3}\pi R^3(1-\cos\theta')^2(1+2\cos\theta')\\ &= \frac{2\pi r^2R^3(r+3R)}{3(r+R)^3} = 2\pi r^2R\big(1+O(r/R)\big).\end{aligned}$$

The second term equals

$$-4\pi r^2\int_0^{\frac{1}{2}\pi-\theta'}\big[(R+r)\sin\theta'\sin^2\psi - r\sin^2\psi\cos\psi\big]\,d\psi,$$

in which the term involving $\cos\psi$ integrates to $\frac{4}{3}\pi r^3\sin^3\left(\frac{1}{2}\pi-\theta'\right) = \frac{4}{3}\pi(r\cos\theta')^3 = \dfrac{4\pi r^3R^3}{3(R+r)^3}$, and the other term equals

$$-2\pi r^2(R+r)\sin\theta'\left(\tfrac{1}{2}\pi-\theta'-\tfrac{1}{2}\sin 2\theta'\right) = -2\pi r^2\sqrt{r(2R+r)}\Big(\sin^{-1}\frac{R}{R+r}-\sin\theta'\cos\theta'\Big).$$

Thus,

$$A(r;R,R) = \frac{2\pi r^2R^3}{(r+R)^2} - \pi r^2(r+R)\sin\theta'\left(\pi-2\theta'-\sin 2\theta'\right), \qquad \text{where} \qquad \cos\theta' = \frac{R}{R+r}\,. \quad (5.12)$$

For small $r \ll R$, the first term is $O(r^2)$ and the rest $O(r^{5/2})$. Table 7 illustrates the growth of $A(r;R,R)$ in r/R.

Table 7

Volume $A(r;R,R)$ of region near spheres of radii R uncoverable by ball of radius $r<R$

r/R	$A(r;R,R)/R^3$	$A(r;R,R)/\frac{4}{3}\pi R^3$
0.1000	0.02683	0.00616
0.2000	0.08710	0.02079
0.3000	0.15324	0.03658
0.4000	0.22164	0.05291
0.5000	0.28925	0.06905
0.6000	0.35458	0.08465
0.7000	0.41699	0.09955
0.8000	0.47619	0.11368
0.9000	0.53216	0.12704
1.0000	0.58497	0.13965

In the close-packings of uniform balls of radius $R = \frac{1}{2}$ in both Examples 1 and 2, each ball touchers 12 others, so the proportion of space that cannot be covered either by the uniform balls of the packing or by much smaller balls of radius $r < \sqrt{\frac{3}{2}} - 1 \approx 0.225$, i.e. for $r/R < 0.45$, equals

$$\frac{6\,A(r;\frac{1}{2},\frac{1}{2})}{\frac{1}{2}\sqrt{2}}\,, \qquad = 0.16 \quad (r/R = 0.3) \qquad \text{or} \quad 0.03 \quad (r/R = 0.1). \tag{5.13}$$

In terms of volume fractions, these examples mean that, given a close-packing like that of Example 1 or 2, in adding balls whose radius is 30% or 10% of the radius of the larger balls, the void cannot be reduced from 26% to less than 16% or 3% of the total space, though these figures are merely bounds that do not reflect the 'inter-stitial voids' that may be created between these smaller balls.

For the record we compute the corresponding functions for $A(r;R,\infty)$, corresponding to a sphere of radius R touching a plane (i.e. approximately, a sphere of radius $\gg R$). With the same functions $h(\cdot)$ and $x(\cdot)$, and much the same argument, augmented now by a 'sliver' of exclusion adjacent to the surface of the plane and a touching sphere of radius r, we have

$$\begin{aligned} A(r;R,\infty) = \int_{\theta=0}^{\theta''} h(\theta)\,2\pi x(\theta)\,dx - \int_{\psi=0}^{\frac{1}{2}\pi-\theta''} 2r\sin\psi\,2\pi\big((R+r)\sin\theta'' - r\cos\psi\big)\,d(-r\cos\psi) \\ + \int_{\psi=\frac{1}{2}\pi-\theta''}^{\frac{1}{2}\pi} r(1-\sin\psi)\,2\pi\big((R+r)\sin\theta'' - r\cos\psi\big)\,d(-r\cos\psi), \end{aligned} \tag{5.14}$$

where $(R+r)\cos\theta'' + r = R$, i.e. $\cos\theta'' = \dfrac{R-r}{R+r}$. Integration much as for (5.12) and collecting terms gives

$$\begin{aligned} A(r;R,\infty) = 4\pi r^2 R\,\frac{R^2(3R-r)}{3(R+r)^3} - \pi r^2\sqrt{rR}\,(6\theta'' - 4\sin\theta'' - \sin 2\theta'') \\ + \tfrac{1}{3}\pi r^3(\sin^2\theta'' + 2(1-\cos^3\theta'') + 4\sin^3\theta''). \end{aligned} \tag{5.15}$$

Inspection shows that in this function the first term is $O(r^2)$ and the others at most $O(r^3)$ for small $r \ll R$.

Details for both the above and the general case $A(r;R_1,R_2)$ with $r \le R_1 \le R_2 \le \infty$ are in the Appendix.

6. A Miscellany

Problem 3. *Suppose that the sequential touching model algorithm is executed with $N_s = \infty$, i.e. at step 2_s, spheres of radii R_s are placed in the region to be covered until no more sites are available, at which point step 2_{s+1} starts, for $s = 1, 2, \ldots$. Is there a sequence of radii $\{R_s\}$ such that the volume fraction $(V_f)_s$ covered by all spheres of radii $\ge R_s$ satisfies $\lim_{s\to\infty}(V_f)_s = 1$, or does the limitation on the number of spheres being countable combined with the curvature of their surfaces force some limit < 1?*

It was partly in response to this question that the quantities $A(r;R,R)$ and $A(r;R,\infty)$ of section 5.6 were computed (cf. also (5.13) as a start).

A common aim of both the simulations reported earlier and of Alan Karr's simulations of a germ–grain model in which successive balls are placed as *remotely* as possible from existing spheres, was to develop some feel for the way in which a volume fraction of between 60% and 70% may be obtainable from spheres. It would appear that the gradation in sphere-size is crucial here — admittedly, these simulations do not prescribe the proportion of volume occupied by various sizes (i.e. the volume fractions for the different sizes). In practice there is a smallest size, so the volume fraction attainable is bounded above by the vacuity fraction of close-packed spheres of the smallest size, even assuming they fill the vacuity fraction of the next size up. For example, referring to Examples 1 and 2, there are fractions there of $1 : \sqrt{2}-1 = 2.414 : 1$ and $\sqrt{2}-1 : \sqrt{1.5}-1 = 1.8099 : 1$ between the 1st and 2nd order, and the 2nd and 3rd order spheres, respectively, that reduce the vacuity fraction by about 20% and 7% respectively, as the 'empty space' changes from being external to spheres all of the one size, to spheres of two sizes (though it is two different types of interstices in the initial spheres that are accommodating the 2nd and 3rd order spheres).

Our algorithm of section 4 can be expected to find 'most' spheres of radius $\geq r'$ say for as long as the r'-vacuity function

$$H(r') \equiv \frac{\ell\big(\mathcal{S}(1) \setminus \Xi; r')\big)}{\ell\big(\mathcal{S}(1)\big)} ,$$

where $\ell(\cdot)$ denotes Lebesgue measure, satisfies

$$(1 - H(r'))^{N_s} \ll 0.0001 \qquad \text{say;}$$

here, N_s denotes the number of failures before the radius is decreased from (say) R_s to R_{s+1}. For the record, Table 8 is a more detailed summary of the simulation reported more briefly in columns (b) of Table 4.

What we want to do is to try and recover the volume fraction of aggregate in concrete, preferably from some 'mix' of aggregate sizes (whether specified by volume or mass), via spherical grains. From this point of view, algorithm with step 3a′ is of lesser interest.

It seems not unreasonable to me that in mixing concrete, that larger items of aggregate should dominate over smaller which, as the mix tumbles, produce a gradation of sizes locally consistent with 'holes' that are large enough to accommodate whatever is local to the region, with preference given to the larger over the smaller, and that every item of aggregate should touch at least one other. Without having the immediate resources to demonstrate that the simulation above achieves just that, it appears reasonable that this situation is approximated by the scheme devised, more particularly by the algorithm with step 3a rather than 3a′ (the interest of 3a′ is more of a mathematical nature: how much larger can the volume fraction be made if we allow ourselves the liberty of choosing the largest possible item, as distinct from the next item to hand; step 3a′ also has the advantage of indicating the extent to which we fail to achieve mixing because it is an indicator of 'larger' holes that are not found by the progression-rule (i.e. increase s)).

Table 8

Evolution of a simulation from the sequential touching model
($R_1 = 0.1,\ R_{s+1} = R_s,\ N_s = 20{,}000$)

s	AllPts	$\Delta_s(n_p)$	$\Delta[(V_f)_{\rm obs}]$	$(V_f)_s$	Radius	s	AllPts	$\Delta_s(n_p)$	$\Delta[(V_f)_{\rm obs}]$	$(V_f)_s$	Radius
1	320	320	0.000000	0.320000	0.10000	37	1317	28	0.002029	0.569110	0.04019
2	345	25	0.024550	0.344550	0.09750	38	1444	127	0.008629	0.577739	0.03919
3	354	9	0.008329	0.352879	0.09506	39	1462	18	0.001123	0.578863	0.03821
4	354	0	0.000000	0.352879	0.09269	40	1535	73	0.004246	0.583109	0.03725
5	375	21	0.016800	0.369679	0.09037	41	1637	102	0.005613	0.588722	0.03632
6	394	19	0.014770	0.384449	0.08811	42	1641	4	0.000213	0.588935	0.03542
7	395	1	0.000692	0.385141	0.08591	43	1809	168	0.007977	0.596912	0.03453
8	414	19	0.012591	0.397732	0.08376	44	1876	67	0.002988	0.599901	0.03367
9	422	8	0.004843	0.402575	0.08167	45	1963	87	0.003533	0.603434	0.03282
10	435	13	0.007367	0.409942	0.07962	46	2089	126	0.004792	0.608226	0.03200
11	451	16	0.008178	0.418119	0.07763	47	2128	39	0.001349	0.609575	0.03120
12	472	21	0.009914	0.428034	0.07569	48	2330	202	0.006584	0.616159	0.03042
13	491	19	0.008151	0.436185	0.07380	49	2510	180	0.005383	0.621542	0.02966
14	498	7	0.002930	0.439115	0.07195	50	2693	183	0.005060	0.626602	0.02892
15	517	19	0.007216	0.446331	0.07016	51	2747	54	0.001387	0.627990	0.02820
16	535	18	0.006263	0.452594	0.06840	52	2891	144	0.003483	0.631472	0.02749
17	543	8	0.002678	0.455272	0.06669	53	3198	307	0.006773	0.638245	0.02681
18	586	43	0.013462	0.468734	0.06502	54	3331	133	0.002765	0.641010	0.02614
19	596	10	0.002816	0.471550	0.06340	55	3541	210	0.004050	0.645060	0.02548
20	637	41	0.010647	0.482198	0.06181	56	3756	215	0.003808	0.648868	0.02485
21	655	18	0.004312	0.486510	0.06027	57	3809	53	0.000856	0.649724	0.02422
22	685	30	0.006836	0.493346	0.05876	58	3973	164	0.002491	0.652215	0.02362
23	716	31	0.006728	0.500074	0.05729	59	4166	193	0.002758	0.654973	0.02303
24	731	15	0.002997	0.503071	0.05586	60	4635	469	0.006210	0.661184	0.02245
25	759	28	0.005354	0.508425	0.05446	61	5077	442	0.005367	0.666550	0.02189
26	784	25	0.004244	0.512669	0.05310	62	5305	228	0.002569	0.669119	0.02134
27	800	16	0.002462	0.515131	0.05178	63	5682	377	0.003914	0.673033	0.02081
28	851	51	0.007326	0.522458	0.05048	64	6052	370	0.003595	0.676628	0.02029
29	884	33	0.004499	0.526957	0.04922	65	6341	289	0.002592	0.679220	0.01978
30	937	53	0.006802	0.533759	0.04799	66	6899	558	0.004654	0.683874	0.01929
31	993	56	0.006307	0.540065	0.04679	67	6983	84	0.000655	0.684529	0.01881
32	1059	66	0.007052	0.547117	0.04562	68	7463	480	0.003485	0.688015	0.01834
33	1076	17	0.001775	0.548893	0.04448	69	8107	644	0.004263	0.692278	0.01788
34	1177	101	0.009307	0.558200	0.04337	70	8614	507	0.003154	0.695432	0.01743
35	1178	1	0.000088	0.558288	0.04228	71	8923	309	0.001777	0.697208	0.01700
36	1289	111	0.008793	0.567081	0.04123	72	9433	510	0.002776	0.699985	0.01657
						73	10000 points give $(V_f)_{\rm obs} = 0.702838$				

Problem 4. *The Surface Skin.* When n spheres of radius R have volume fraction V_f, the amount of space within $R+\Delta$ of the germs is at most $(1+\frac{\Delta}{R})^d v_f$. For example, when $v_f = 0.5$, $(1+\frac{\Delta}{R})^d = 2$ when

$$\frac{\Delta}{R} = 2^{1/d} - 1, \ \approx 0.26 \quad \text{for } d = 3.$$

How can this consideration, not too dissimilar from the motivation behind the function $A(r; R_1, R_2)$ of Section 5.6, be applied to understand observed void fraction?

Problem 5. *For a sequential packing algorithm such as in Section 4, with fixed $R_1, R_2, \ldots,$ is there an optimal ratio R_{s+1}/R_s ? Does the ratio depend on $N_1, N_2, \ldots$?*

For the latter question, compare section 5.NNNNN

It would appear intuitively plausible that sequentially packing spheres into a region such as via the sequential touching algorithm of Section 4, should produce a volume fraction as close as we please to 1. But is this really so? Consider again Examples 1 and 2 where unit spheres are close-packed, occupying 74% of the space. Adding the next largest spheres fills another 5%, or about 20% of the remaining empty space. The third-largest set of spheres that can be put into the empty space that remains fills another 1.6% of the original volume, or about 8% of the then remaining empty space.

Equally, it is salutary to reflect that in a close-packing of space with spheres of diameter 1 cm, the amount of occupied space that is within 0.1 mm of the surface of a sphere is about 6% of the occupied space, equivalent to about 24% of the unoccupied space, suggesting that the amount of *empty* space that is within 0.1 mm of covered space is of the order of 20% of all the empty space. Consequently, it is little wonder that filling as much of the empty space as possible with spheres of about the same size, does nothing like filling up a substantial part of the space that is presently empty. *A considerable part of empty space is close to the surface of at least one grain (and if it is close to two, then it is likely that only tiny grains can be fitted into the void)*; see Table 7.

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Appendix

A. Algebra concerning the excluded volume function $A(r; R_1, R_2)$

In the case of $A(r; R, \infty)$ given at equation (5.14), The first term equals

$$2\pi R^3\big(\tfrac{1}{2}(1-\cos^2\theta'') - \tfrac{1}{3}(1-\cos^3\theta'')\big) = \frac{4\pi r^2 R^3(3R-r)}{3(R+r)^3} = 4\pi r^2 R\big(1+O(r/R)\big).$$

The second term, again from earlier work, equals

$$\begin{aligned}&-2\pi r^2(R+r)\sin\theta''\big(\tfrac{1}{2}\pi - \theta'' - \tfrac{1}{2}\sin 2\theta''\big) + \tfrac{4}{3}\pi r^3\sin^3\big(\tfrac{1}{2}\pi-\theta''\big)\\ &\quad = -4\pi r^2\sqrt{rR}\left(\sin^{-1}\frac{R-r}{R+r} - \frac{2(R-r)\sqrt{rR}}{(R+r)^2}\right) + \frac{4\pi r^3(R-r)^3}{3(R+r)^3}.\end{aligned}$$

The last term equals

$$\begin{aligned}&2\pi r^2\int_{\psi=\frac{1}{2}\pi-\theta''}^{\frac{1}{2}\pi}(\sin\psi-\sin^2\psi)\big(2\sqrt{rR}-r\cos\psi\big)\,d\psi\\ &\quad = 4\pi r^2\sqrt{rR}\,\big(\sin\theta''-\tfrac{1}{2}\theta''-\tfrac{1}{4}\sin 2\theta''\big) - 2\pi r^3\big(\tfrac{1}{2}(1-\cos^2\theta'') - \tfrac{1}{3}(1-\cos^3\theta'')\big).\end{aligned}$$

Thus,

$$\begin{aligned}A(r;R,\infty) &= 2\pi R^3\int_0^{\theta''}(cos\theta-\cos^2\theta)\,d(-\cos\theta)\\ &\quad + \pi r^2\int_0^{\theta''}\big[4\sqrt{rR}\,(\cos\psi-1-\sin^2\psi) - r\sin 2\psi(1-\cos\psi-2\sin\psi)\big]\,d\psi\end{aligned}$$

Now see equation (5.15).

For the general case that $r \le R_1 \le R_2 \le \infty$, the uncoverable volume $A(r; R_1, R_2)$ equals

$$\begin{aligned}&\sum_{k=1}^{2}\int_{\theta_k=0}^{\theta_k'} h_k(\theta)\,2\pi x_k(\theta)\,dx_k - \int_{\psi_1=0}^{\frac{1}{2}\pi-\theta_1'} 2r\sin\psi_1 2\pi\big((R_1+r)\sin\theta_1' - r\cos\psi_1\big)\,d(-r\cos\psi_1)\\ &\quad - \int_{\frac{1}{2}\pi-\theta_1'}^{\frac{1}{2}\pi-\theta_2'}\big(r\sin\psi_2 - [(R_2+r)\cos\theta_2' - R_2]\big)2\pi\big((R_2+r)\sin\theta_2' - r\cos\psi_2\big)\,d(-r\cos\psi_2)\\ &\equiv [1]-[2]-[3] \quad \text{say,}\end{aligned}$$

where for $k = 1, 2$, $h_k(\theta) = R_k(1-\cos\theta)$, $x_k(\theta) = R_k\sin\theta$, and θ_k' are solutions satisfying $0 < \theta_k' < \frac{1}{2}\pi$ of

$$\begin{aligned}(R_1+r)\sin\theta_1' &= (R_2+r)\sin\theta_2',\\ (R_1+r)\cos\theta_1' + (R_2+r)\cos\theta_2' &= R_1+R_2,\end{aligned}$$

$$\begin{aligned}[1] &= 2\pi R_k^3\int_0^{\theta_k'}(1-\cos\theta)\sin\theta\cos\theta\,d\theta = 2\pi R_k^3\big(\tfrac{1}{2}(1-\cos^2\theta_k') - \tfrac{1}{3}(1-\cos^3\theta_k')\big),\\ [2] &= 4\pi r^2\int_{\psi=0}^{\frac{1}{2}\pi-\theta_1'}\sin\psi\big((R_1+r)\sin\theta_1' - r\cos\psi\big)\sin\psi\,d\psi\\ &= 2\pi r^2(R_1+r)\sin\theta_1'\big((at\tfrac{1}{2}\pi-\theta_1') - \tfrac{1}{2}\sin 2\theta_1'\big) - \tfrac{4}{3}\pi r^3(R_1+r)\cos^3\theta_1',\end{aligned}$$

and the integral [3] is evaluated similarly.