Letter

Densest packings from size segregation of particles in geometric confinement

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A correlation between density maximization and size segregation for packings of polydisperse particles in geometric confinement has been discovered, through the derivation of a general solution for the densest-packed zigzag arrangements of polydisperse particles. This solution is a size-graded structure in which the larger a particle the closer it is located to either end of the system, such that the smaller particles in the interior are encapsulated by the larger ones away from it. Any pair of different-sized adjacent particles is a grain-boundary-like configuration that reduces the overall packing efficiency of the system, and this solution corresponds to a minimization of excess-volume contributions from grain-boundary-like configurations of different-sized particles as a result of the clustering of equal- or like-sized particles. Our findings provide new insights into how structural order of polydisperse particles emerges in confined settings.

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Packing problems [1], which concern the optimal arrangements of objects in space, have been studied extensively by mathematicians, physicists, and geologists. From the physicist's point of view, solutions of packing problems can serve as structural models for a variety of condensed matter systems. Examples include the crystalline structures of solids [2], the disordered structures of liquids [3] or glasses [4], and the microstructures of heterogeneous materials [5]. Thanks to the rapid technological development of high-performance computers in the past few decades, computational solutions can now be obtained for a wide range of complex packing problems, of which analytic solutions are elusive. These include a variety of problems concerning the optimal arrangements of particles in geometric confinement [6–19]. In many cases, there exists a complex geometric interplay between the confined particles and the confining boundaries despite the simplicity of the geometries involved. An active area of research in the past two decades concerns the densest-packed structures of equal-sized hard particles in uniform quasi-one-dimensional confinement [8–19]. Among them are a wide spectrum of ordered structures discovered for circles or ellipses within a parallel slit [8–11] or for spheres or ellipsoids within a cylindrical tube [12–18]. Such structures, which are different from their bulk counterparts, are results of a competition between boundary-induced ordering and bulk crystalline ordering [18].

Despite a long tradition of research on bulk packings of polydisperse particles [20–32] and the wide applicability of polydisperse grains in the engineering of high-performance materials [33–36], less attention has been paid to packings of polydisperse particles in geometric confinement. For confined systems, however, packing problems of polydisperse particles could be much more interesting and challenging than those of equal-sized particles, because the coupling of effects of particle dispersity with those of geometric confinement would bring the corresponding packing problems to a new level of complexity. Apart from the additional degrees of freedom that arise from a size variation of particles, the mechanisms of how particles of different sizes interact with the boundaries of a confined system also play a key role in determining the optimal arrangements of particles. In view of the current research interests in confined packings and of the new physics that could potentially arise from an inclusion of particle dispersity, we have carried out an extension of theoretical research on densest packings in quasi-one-dimensional confinement to polydisperse particles, for which we have discovered an unexpected correlation between density maximization and size segregation: For zigzaglike packings of polydisperse circles or spheres within a sufficiently narrow parallel slit or cylindrical tube, there exists a general size-graded solution in which the larger a particle the closer it is located towards either end of the system, such that the smaller particles in the interior are encapsulated by the larger ones away from it. Any pair of different-sized adjacent particles is a grain-boundary-like configuration that reduces the overall packing efficiency of the system, and this general solution corresponds to a minimization of excess-volume contributions from grain-boundary-like configurations of different-sized particles as a result of the clustering of equal- or like-sized particles. Our findings provide new insights into how the structural order of polydisperse particles emerges in confined settings.

Consider a set of N polydisperse disks or spheres packed in a zigzag manner within a parallel slit or cylindrical tube, of slit width or tube diameter D. The integers $i \in [1, N]$ and $j \in [1, N]$ are used as indices to label the particles in ascending order of their diameters d and vertical positions z, respectively:

$$(4 - 2\sqrt{3})D \leqslant d_{i=1} \leqslant d_{i=2} \leqslant \dots \leqslant d_{i=N} \leqslant D \qquad (1)$$

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and

$$z_{j=1} < z_{j=2} < \ldots < z_{j=N}. \quad (2)$$

The smallest possible value of d, only at or above which a planar-zigzag structure can exist [12], can be derived from the cylinder-to-sphere diameter ratio $D/d = 1 + \sqrt{3}/2$ for the zigzag-to-single-helix transition of densest-packed structures of identical spheres in cylindrical confinement [37]. In reality, there exist some very polydisperse granular systems where the interstitial space of the largest particles could be filled up by the smallest ones, but such cases are not considered in this research.

The packing fraction of such a system is defined as the total area or volume occupied by the particles divided by the total area or volume of the confining space. For any given set of particles with a fixed total area or volume, the densest-packed arrangement of particles corresponds to a minimization of the total area or volume of the confining space. As the value of D is fixed, this is equivalent to a minimization of the total length

$$L_{j=1,j=N} \equiv \frac{d_{j=1} + d_{j=N}}{2} + \sum_{j=1}^{N-1} (\Delta z)_{j,j+1} \quad (3)$$

of the system, where [37]

$$(\Delta z)_{j,j+1} \equiv z_{j+1} - z_j = \sqrt{D(d_j + d_{j+1} - D)} \quad (4)$$

is the vertical separation between particle j and particle $(j+1)$. Here we show that, if the particle at each end of the system is one of the two largest particles, a necessary condition for $L_{j=1,j=N}$ to be at its minimum possible value is satisfied:

Take the $j = N$ end of the system as an example, and consider a hypothetical exchange of particle $j = N - 1$ with particle $j = N$. The diameters of particles $j = N - 1$ and $j = N$ before the particle-exchange operation are denoted as

$$d_{j=N-1} = d_{i=a} \text{ and } d_{j=N} = d_{i=b}, \quad (5)$$

respectively. The total length of the system is given by

$$(L_{j=1,j=N})_{\text{bef. exch.}} = \frac{d_{j=1} + d_{i=b}}{2} + \left[\sum_{j=1}^{N-3} (\Delta z)_{j,j+1}\right] + (\Delta z)_{j=N-2,i=a} + (\Delta z)_{i=a,i=b}. \quad (6)$$

After the exchange of particles, the diameters of particles $j = N - 1$ and $j = N$ become

$$d_{j=N-1} = d_{i=b} \text{ and } d_{j=N} = d_{i=a}, \quad (7)$$

respectively, such that we have

$$(L_{j=1,j=N})_{\text{aft. exch.}} = \frac{d_{j=1} + d_{i=a}}{2} + \left[\sum_{j=1}^{N-3} (\Delta z)_{j,j+1}\right] + (\Delta z)_{j=N-2,i=b} + (\Delta z)_{i=b,i=a} \quad (8)$$

for the total length of the system. The corresponding change in $L_{j=1,j=N}$ as a result of this particle-exchange operation is given by [37]

$$(\Delta L_{j=1,j=N})_{\text{exch.}} = F(d_{j=N-2}, d_{i=b}) - F(d_{j=N-2}, d_{i=a}) \quad (9)$$

for

$$F(x, y) \equiv \sqrt{D(x + y - D)} - \frac{y}{2} \text{ and } \frac{\partial F(x, y)}{\partial y} > 0. \quad (10)$$

At any fixed value of x, the function $F(x, y)$ increases monotonously with y, so that the conditions

$$(\Delta L_{j=1,j=N})_{\text{exch.}} < 0 \text{ and } (\Delta L_{j=1,j=N})_{\text{exch.}} > 0 \quad (11)$$

are satisfied for cases of $d_{i=b} < d_{i=a}$ and $d_{i=b} > d_{i=a}$, respectively. The same conclusion can be reached for the other end of the system. By definition, the total length $L_{j=1,j=N}$ of any densest-packed arrangement of particles cannot be further reduced by such particle-exchange operations. This necessary condition of density maximization is satisfied, not exclusively, by any arrangement in which the two largest particles are placed respectively at the two ends, with either (i) $d_{j=1} = d_{i=N-1}$ and $d_{j=N} = d_{i=N}$ or (ii) $d_{j=1} = d_{i=N}$ and $d_{j=N} = d_{i=N-1}$. In the following, we assume that at least one such arrangement of particles corresponds to the minimum possible value of $L_{j=1,j=N}$.

For any such arrangement of particles, if either particle $d_{i=N}$ or particle $d_{i=N-1}$ is brought to the other end such that the two largest particles cluster at either end of the system, the total length $L_{j=1,j=N}$ would increase, resulting in a less dense arrangement of particles. As an example, consider a displacement of particle $i = N$ from the $j = N$ end to the $j = 1$ end. For this particle-displacement operation, the initial and the final conditions are given by

$$d_{j=1}=d_{i=N-1};\, d_{j=N} = d_{i=N};\, \text{“}d_{j=2}\ldots d_{j=N-1}\text{”}=\text{“}d_{i=a}\ldots d_{i=b}\text{”} \quad (12)$$

and

$$d_{j=1}=d_{i=N};\, d_{j=2} = d_{i=N-1};\, \text{“}d_{j=3}\ldots d_{j=N}\text{”}=\text{“}d_{i=a}\ldots d_{i=b}\text{”}, \quad (13)$$

respectively. With the initial total length

$$(L_{j=1,j=N})_{\text{bef. disp.}} = \frac{d_{i=N-1} + d_{i=N}}{2} + \left[\sum_{j=2}^{N-2} (\Delta z)_{j,j+1}\right] + (\Delta z)_{i=N-1,i=a} + (\Delta z)_{i=b,i=N} \quad (14)$$

and the final total length

$$(L_{j=1,j=N})_{\text{aft. disp.}} = \frac{d_{i=N} + d_{i=b}}{2} + \left[\sum_{j=3}^{N-1} (\Delta z)_{j,j+1}\right] + (\Delta z)_{i=N,i=N-1} + (\Delta z)_{i=N-1,i=a}, \quad (15)$$

the corresponding change in $L_{j=1,j=N}$ as a result of this particle-displacement operation is given by

$$(\Delta L_{j=1,j=N})_{\text{disp.}} = F(d_{i=N}, d_{i=N-1}) - F(d_{i=N}, d_{i=b}) > 0. \quad (16)$$

Any arrangement of particles with the two largest particles clustering at either end of the system is thus excluded as a candidate of densest-packed arrangements.

Having the two largest particles placed respectively at the two ends of the system, the total length $L_{j=1,j=N}$

can be expressed as [37]

$$L_{j=1,j=N} = \frac{d_{i=N-1} + d_{i=N}}{2} + \frac{(N-1)\sqrt{D}}{\sqrt{\gamma}} \sqrt{(2\langle d\rangle - D) + \frac{2}{(N-1)}\left[\langle d\rangle - \frac{d_{i=N-1} + d_{i=N}}{2}\right]}, \tag{17}$$

where

$$\gamma \equiv \frac{\langle (\Delta z)^2_{j,j+1}\rangle}{\langle (\Delta z)_{j,j+1}\rangle^2} \tag{18}$$

is the dispersity [38] of $(\Delta z)_{j,j+1}$, and $\langle d\rangle$ is the average diameter of all the particles. A minimization of $L_{j=1,j=N}$ corresponds to a maximization of γ, where the latter requires the spread of values of $(\Delta z)_{j,j+1}$ to be as wide as possible. Here we propose a possible maximization of γ through the clustering of like-sized particles, i.e., demixing, where the upper bound and the lower bound of $(\Delta z)_{j,j+1}$ are maximized and minimized, respectively, through the clustering of the largest particles at the two ends of the system and the clustering of the smallest ones in the middle of the system. Following this approach, we place particles $i = N-2$ and $i = N-3$ at positions next to particle $i = N$ or $i = N-1$. There are two possible ways of doing so: One option is to place particle $i = N-2$ in contact with particle $i = N$ and particle $i = N-3$ in contact with particle $i = N-1$, and the other is to place particle $i = N-2$ in contact with particle $i = N-1$ and particle $i = N-3$ in contact with particle $i = N$. The first option is preferable, not only because it yields the largest possible upper bound of $(\Delta z)_{j,j+1}$, i.e., $\Delta z_{i=N-2,i=N}$, but also because it results in a smaller contribution to the total length $L_{j=1,j=N}$:

$$\Delta z_{i=N-2,i=N} + \Delta z_{i=N-3,i=N-1} \leqslant \Delta z_{i=N-2,i=N-1} + \Delta z_{i=N-3,i=N}, \tag{19}$$

from which the correct inequalities $d_{i=N-3} \leqslant d_{i=N-2}$ and $d_{i=N-1} \leqslant d_{i=N}$ can be derived [37]. An iterative application of the above principle of clustering of like-sized particles to all remaining particles leads us to a theoretical densest-packed arrangement of the form

$$i = \{N-1, N-3, N-5, \ldots, 1, \ldots N-4, N-2, N\}, \tag{20}$$

where the clustering of smallest particles in the middle of the system yields the smallest possible lower bound of $(\Delta z)_{j,j+1}$, i.e., $(\Delta z)_{i=1,i=2}$. Such a general size-graded solution, in which the larger a particle the closer it is located towards either end of the system, can be constructed via a simple method of sequential deposition, as in the case of equal-sized hard spheres in cylindrical confinement [14]: Particles are deposited one by one in ascending order of their sizes. For $i > 1$, they are deposited from alternating sides of particle $i = 1$, with odd-indexed and even-indexed particles deposited respectively from opposite sides. An illustration of this method is presented in Fig. 1(a).

To verify our solution for densest packings of polydisperse particles, we have carried out numerical investigations for a system of $N = 20$ particles with four different diameters: (i) $d_{v=1} = d_{i=1\text{ to }5} = 0.6$, (ii) $d_{v=2} = d_{i=6\text{ to }10} = 0.66$, (iii) $d_{v=3} = d_{i=11\text{ to }15} = 0.84$, and (iv) $d_{v=4} = d_{i=16\text{ to }20} = 0.9$, where each diameter value is shared by five particles. The slit width or tube diameter is set to be equal to $D = 1$. For any pair of adjacent particles and for the sole pair of boundary particles at the ends of the system, there exist 10 possible pairwise combinations of particle diameters. Using the positive integers $N_{\text{end},v,v'}$ and $N_{\Delta z,v,v'}$, the total length of the system is expressed as a dot product that describes the sum of contributions from all possible pairwise combinations of particle diameters:

$$L_{j=1,j=N=20} = \sum_{v,v'}\left[N_{\text{end},v,v'}\left(\frac{d_v + d_{v'}}{2}\right)\right] + \sum_{v,v'}[N_{\Delta z,v,v'}(\Delta z)_{v,v'}], \tag{21}$$

where

$$\sum_{v,v'} N_{\text{end},v,v'} = 1 \text{ and } \sum_{v,v'} N_{\Delta z,v,v'} = N-1. \tag{22}$$

With both $N_{\text{end},v,v'}$ and $N_{\Delta z,v,v'}$ being structure-dependent, the square sum

$$\psi \equiv \sum_{v,v'}(N_{\text{end},v,v'} - N^*_{\text{end},v,v'})^2 + \sum_{v,v'}(N_{\Delta z,v,v'} - N^*_{\Delta z,v,v'})^2 \tag{23}$$

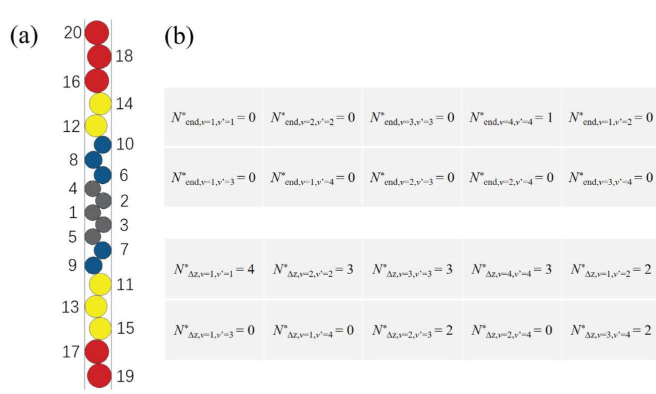

FIG. 1. Theoretical densest-packed arrangement for a system of $N = 20$ particles with $d_{v=1} = d_{i=1\text{ to }5} = 0.6$, $d_{v=2} = d_{i=6\text{ to }10} = 0.66$, $d_{v=3} = d_{i=11\text{ to }15} = 0.84$, $d_{v=4} = d_{i=16\text{ to }20} = 0.9$, and $D = 1$. (a) Schematic illustration of this densest-packed arrangement and the corresponding method of sequential deposition; (b) tabulation of the values of $N^*_{\text{end},v,v'}$ and $N^*_{\Delta z,v,v'}$.

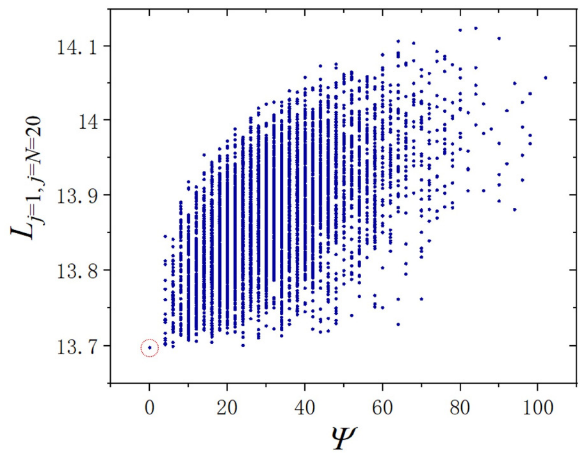

FIG. 2. Plot of the total length $L_{j=1,j=N=20}$ against the order parameter ψ for a system of $N = 20$ particles with $d_{v=1} = d_{i=1\text{ to }5} = 0.6$, $d_{v=2} = d_{i=6\text{ to }10} = 0.66$, $d_{v=3} = d_{i=11\text{ to }15} = 0.84$, $d_{v=4} = d_{i=16\text{ to }20} = 0.9$, and $D = 1$. The theoretical densest-packed arrangement (Fig. 1), which corresponds to the circled datum at $\psi = 0$, has a minimum possible total length of $L_{j=1,j=N=20} = 13.69754$.

is employed as an order parameter to describe the structural deviations of any given permutation of particles from the corresponding theoretical densest-packed arrangement, for which the equalities $N_{\text{end},v,v'} = N^*_{\text{end},v,v'}$ and $N_{\Delta z,v,v'} = N^*_{\Delta z,v,v'}$ hold. This densest-packed arrangement and the corresponding method of sequential deposition are illustrated schematically in Fig. 1(a), and the values of $N^*_{\text{end},v,v'}$ and $N^*_{\Delta z,v,v'}$ are tabulated in Fig. 1(b). Upon a thorough search for all possible combinations of values of $N_{\text{end},v,v'}$ and $N_{\Delta z,v,v'}$, it was found that the theoretical densest-packed arrangement is indeed a structure with the minimum possible value of $L_{j=1,j=N=20}$ (circled datum in Fig. 2). With such an agreement obtained between theoretical predictions and numerical results, we conclude that our solution for densest packings of polydisperse particles has been verified. It should be noted that, while the theoretical densest-packed arrangement is confirmed as a solution to the current problem of interest, the uniqueness of this solution is not guaranteed, because there might exist some other permutations of particles with the same values of $N_{\text{end},v,v'}$ and $N_{\Delta z,v,v'}$.

The tabulated values of $N^*_{\Delta z,v,v'}$ in Fig. 1(b) indicate a preferred clustering of equal- or like-sized particles in the theoretical densest-packed arrangement. To understand the physical mechanism behind, we look into the dense packings of a system of N bidisperse particles, and consider a specific type of particle permutations where each permutation is a repetition of n sets of N/n particles. At a slit width or tube diameter $D = 1$, the particle diameters take on two possible values as follows: (i) $d_{v=1} = d_{i=1\text{ to }(N/3)}$ and (ii) $d_{v=2} = d_{i=[(N/3)+1]\text{ to }N}$. For the s^{th} set of particles ($s \in [1, n]$), we set the particle diameter to be equal to $d_{v=1}$ for particles from $j = [(s-1)(N/n) + (1/3)(N/n) + 1]$ to $j = [(s-1)(N/n) + (2/3)(N/n)]$, and equal to $d_{v=2}$ for particles from $j = [(s-1)(N/n) + 1]$ to $j = [(s-1)(N/n) + (1/3)(N/n)]$ as well as for particles from $j = [(s-1)(N/n) + (2/3)(N/n) + 1]$ to $j = [(s-1)(N/n) + N/n]$. Any considered permutation of particles is thus of the form

$$v = \{[2...2\ 1...1\ 2...2]\ [2...2\ 1...1\ 2...2]...\}, \tag{24}$$

with

$$N_{\text{end},v=1,v'=1} = N_{\text{end},v=1,v'=2} = 0 \quad \text{and } N_{\text{end},v=2,v'=2} = 1 \tag{25}$$

and [37]

$$N_{\Delta z,v=1,v'=1} = \frac{N}{3} - n,\ N_{\Delta z,v=1,v'=2} = 2n$$
$$\text{and } N_{\Delta z,v=2,v'=2} = \frac{2N}{3} - (n+1). \tag{26}$$

The total length of this bidisperse system is given by

$$L_{j=1,j=N} = d_{v=2} + \left(\frac{N}{3} - n\right)(\Delta z)_{v=1,v'=1} + 2n(\Delta z)_{v=1,v'=2} + \left[\frac{2N}{3} - (n+1)\right](\Delta z)_{v=2,v'=2}, \tag{27}$$

which implies [37]

$$\left(\frac{\partial L_{j=1,j=N}}{\partial n}\right)_{N,D,d_{v=1},d_{v=2}} > 0. \tag{28}$$

This inequality holds because any pair of different-sized adjacent particles is a grain-boundary-like configuration that, on average, contributes more to the total length $L_{j=1,j=N}$ than a pair of equal-sized adjacent particles [37] (i.e., it contributes an excess volume [39] to the system):

$$(\Delta z)_{v=1,v'=2} > \frac{(\Delta z)_{v=1,v'=1} + (\Delta z)_{v=2,v'=2}}{2}. \tag{29}$$

A similar effect has been observed for dense packings of ellipses within a parallel slit (or strip) [10], where the packing efficiency of the system is reduced by the presence of grain boundaries between ellipses of mismatched orientations. Since the total length $L_{j=1,j=N}$ increases monotonously with n, the densest-packed arrangement of particles corresponds to the minimum possible value of $n = 1$, at which the value of $N_{\Delta z,v=1,v'=2}$ reaches its minimum and the values of $N_{\Delta z,v=1,v'=1}$ and $N_{\Delta z,v=2,v'=2}$ reach their maxima. The length contributions from the grain-boundary-like pairs of different-sized adjacent particles are minimized and those from the crystallinelike pairs of equal-sized adjacent particles are maximized, resulting in a minimization of excess-volume contributions from grain-boundary-like configurations of different-sized particles. In our size-graded solution for densest packings of polydisperse particles, such excess-volume contributions are minimized because the spatial variation of particle sizes is smoothed out by the clustering of equal- or like-sized particles.

[1] T. Aste and D. Weaire, *The Pursuit of Perfect Packing*, 2nd ed. (CRC Press, Boca Raton, 2008).

[2] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, 2000).

[3] J. L. Finney, Bernal's road to random packing and the structure of liquids, Philos. Mag. **93**, 3940 (2013).

[4] R. Zallen, *The Physics of Amorphous Solids* (Wiley VCH, New York, 2004).

[5] S. Torquato, *Random Heterogeneous Materials: Microstructure and Macroscopic Properties* (Springer, New York, 2002).

[6] B. de Nijs, S. Dussi, F. Smallenburg, J. D. Meeldijk, D. J. Groenendijk, L. Filion, A. Imhof, A. van Blaaderen, and M. Dijkstra, Entropy-driven formation of large icosahedral colloidal clusters by spherical confinement, Nat. Mater. **14**, 56 (2015).

[7] E. G. Teich, G. van Anders, D. Klotsa, J. Dshemuchadse, and S. C. Glotzer, Clusters of polyhedra in spherical confinement, Proc. Natl. Acad. Sci. USA **113**, E669 (2016).

[8] Z. Füredi, The densest packing of equal circles into a parallel strip, Discrete Comput. Geom. **6**, 95 (1991).

[9] G. Bautista-Carbajal, P. Gurin, S. Varga, and G. Odriozola, Phase diagram of hard squares in slit confinement, Sci. Rep. **8**, 8886 (2018).

[10] W. Jin, Y. Wang, H.-K. Chan, and Z. Zhong, Confinement-induced columnar crystals of ellipses, Phys. Rev. Res. **3**, 013053 (2021).

[11] E. Basurto, P. Gurin, S. Varga, and G. Odriozola, Anisotropy-independent packing of confined hard ellipses, J. Mol. Liq. **333**, 115896 (2021).

[12] G. T. Pickett, M. Gross, and H. Okuyama, Spontaneous Chirality in Simple Systems, Phys. Rev. Lett. **85**, 3652 (2000).

[13] A. Mughal, H.-K. Chan, and D. Weaire, Phyllotactic Description of Hard Sphere Packing in Cylindrical Channels, Phys. Rev. Lett. **106**, 115704 (2011).

[14] H.-K. Chan, Densest columnar structures of hard spheres from sequential deposition, Phys. Rev. E **84**, 050302(R) (2011).

[15] A. Mughal, H.-K. Chan, D. Weaire, and S. Hutzler, Dense packings of spheres in cylinders: Simulations, Phys. Rev. E **85**, 051305 (2012).

[16] L. Fu, W. Steinhardt, H. Zhao, J. E. S. Socolarb, and P. Charbonneau, Hard sphere packings within cylinders, Soft Matter **12**, 2505 (2016).

[17] J. Winkelmann and H.-K. Chan, *Columnar Structures of Spheres: Fundamentals and Applications* (Jenny Stanford Publishing, Singapore, 2023).

[18] W. Jin, H.-K. Chan, and Z. Zhong, Shape-Anisotropy-Induced Ordered Packings in Cylindrical Confinement, Phys. Rev. Lett. **124**, 248002 (2020).

[19] H. Liu, Y. Zong, M. Zu, T. G. Masonc, F. Ye, and K. Zhao, Curvature-assisted self-assembly of Brownian squares on cylindrical surfaces, J. Colloid Interface Sci. **605**, 863 (2022).

[20] D. A. Kofke and P. G. Bolhuis, Freezing of polydisperse hard spheres, Phys. Rev. E **59**, 618 (1999).

[21] A. R. Kansal, S. Torquato, and F. H. Stillinger, Computer generation of dense polydisperse sphere packings, J. Chem. Phys. **117**, 8212 (2002).

[22] H. J. Herrmann, R. Mahmoodi Baram, and M. Wackenhut, Searching for the perfect packing, Physica A **330**, 77 (2003).

[23] C. Voivret, F. Radjaï, J.-Y. Delenne, and M. S. El Youssouf, Space-filling properties of polydisperse granular media, Phys. Rev. E **76**, 021301 (2007).

[24] J.-F. Jerier, V. Richefeu, D. Imbault, and F.-V. Donzé, Packing spherical discrete elements for large scale simulations, Comput. Methods Appl. Mech. Eng. **199**, 1668 (2010).

[25] S. D. S. Reis, N. A. M. Araújo, J. S. Andrade, Jr. and H. J. Herrmann, How dense can one pack spheres of arbitrary size distribution?, Europhys. Lett. **97**, 18004 (2012).

[26] M. Maiti, A. Lakshminarayanan, and S. Sastry, Characterization of void space in polydisperse sphere packings: Applications to hard-sphere packings and to protein structure analysis, Eur. Phys. J. E **36**, 5 (2013).

[27] K. V. Tretiakov and K. W. Wojciechowski, Free volume approximation and equation of state for the fcc phase of polydisperse hard spheres, Comput. Methods Sci. Tech. **19**, 59 (2013).

[28] E. Zaccarelli, S. M. Liddle, and W. C. K. Poon, On polydispersity and the hard sphere glass transition, Soft Matter **11**, 324 (2015).

[29] N. Estrada, Effects of grain size distribution on the packing fraction and shear strength of frictionless disk packings, Phys. Rev. E **94**, 062903 (2016).

[30] P. K. Bommineni, N. R. Varela-Rosales, M. Klement, and M. Engel, Complex Crystals from Size-Disperse Spheres, Phys. Rev. Lett. **122**, 128005 (2019).

[31] G. Bareigts, P. C. Kiatkirakajorn, J. Li, R. Botet, M. Sztucki, B. Cabane, L. Goehring, and C. Labbez, Packing Polydisperse Colloids into Crystals: When Charge-Dispersity Matters, Phys. Rev. Lett. **124**, 058003 (2020).

[32] J. Vasseur, F. B. Wadsworth, J. P. Coumans, and D. B. Dingwel, Permeability of packs of polydisperse hard spheres, Phys. Rev. E **103**, 062613 (2021).

[33] F. de Larrard, *Concrete Mixture Proportioning* (Taylor & Francis, Oxfordshire, 1999).

[34] E. V. Shevchenko, D. V. Talapin, N. A. Kotov, S. O'Brien, and C. B. Murray, Structural diversity in binary nanoparticle superlattices, Nature **439**, 55 (2006).

[35] K. Sobolev and A. Amirjanov, Application of genetic algorithm for modeling of dense packing of concrete aggregates, Constr. Build. Mater. **24**, 1449 (2010).

[36] M. Zhang, H. Dong, and Z. Geng, Computational study of particle packing process and fluid flow inside polydisperse cylindrical particles fixed beds, Powder Technol. **354**, 19 (2019).

[37] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevE.106.L042902 for a step-by-step derivation of each equation or inequality.

[38] R. F. T. Stepto, Dispersity in polymer science, Pure Appl. Chem. **81**, 351 (2009).

[39] H. B. Aaron and G. F. Bolling, Free volume as a criterion for grain boundary models, Surf. Sci. **31**, 27 (1972).